

**KFKI-1996-05/C  
REPORT**

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Sz. Bérczi,  
B. Lukács**

**PROCEEDINGS OF THE INTERNATIONAL  
MEETING SPHERULES  
AND GLOBAL EVENTS**

**Hungarian Academy of Sciences  
CENTRAL  
RESEARCH  
INSTITUTE FOR  
PHYSICS  
B U D A P E S T**





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the International Meeting  
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**"SPHERULES AND GLOBAL EVENTS"**

Cs. H. Detre, Sz. Bérczi & B. Lukács (eds.)

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3. CRIP RMKI, H-1525 Budapest, Bp. 114. Pf. 49.

Second International Budapest Meeting  
on Impact and Extraterrestrial Spherules  
Edited by Csaba H. Detre, Szilárd Bérczi, Béla Lukács

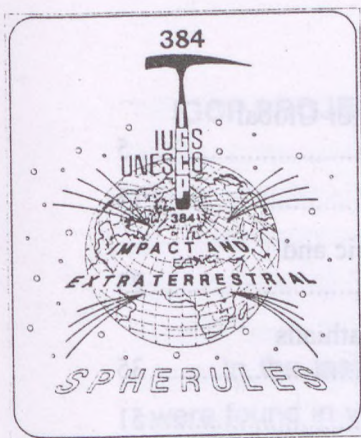
Cs. H. Detre & al. (eds.): Proc. Spherules and Global Events KFKI-1996-05/C

ABSTRACT

This Volume contains the lectures of the 1996 International Meeting "Spherules and Global Events" held on Budapest.

Detre Cs. H. és tsai. (szerk.) Spherulák és Globális Események KFKI-1996-05/C

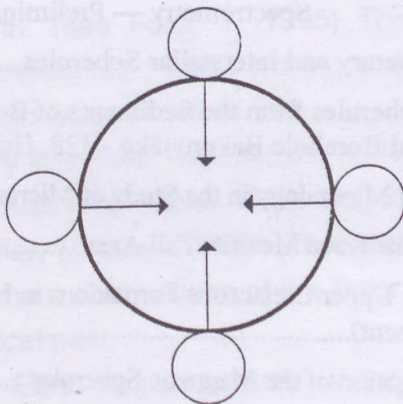
E kötet az 1996-os budapesti "Szférulák és Globális Események" című nemzetközi összejövetel előadásait tartalmazza.



# PROCEEDINGS

of  
the International Meeting

## SPHERULES AND GLOBAL EVENTS



Second International Budapest Meeting  
on Impact and Extraterrestrial Spherules

Edited by CSABA H. DETRE, SZANISZLÓ BÉRCZI, BÉLA LUKÁCS

Organized by  
the Geological Institute of Hungary and the Hungarian Geological Society

1996

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## IGCP PROJECT NO 384 IMPACT AND EXTRATERRESTRIAL SPHERULES: NEW TOOLS FOR GLOBAL CORRELATION

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In the last hundred years microscopic spheroid or deformed spheroid objects were found in various geological formations by micromineralogists and micropaleontologists. The scientists have given them several names, as "spherules", "micro-spherules", "spherulites", "spherolites", "micrometeorites", etc. The investigation of these objects is very difficult, because of their very small size (see later). This may be the reason that they were neglected for a long time. The investigations were quite sporadic and no international organization has been established with the aim of their research.

The Carpathian Basin is a favoured region for spherule studies. Spherules are known from many geological formations there. The Carpathian Basin is one of the deepest sedimentary basins of the world; it is not by chance that it has been nicknamed "Negative Himalaya". The spherules are known to occur in all kinds of sediments, and what is more, probably in all kinds of rocks, even in granite (see *Jakabská, K.; Rozložnik, L. 1989*). The high level of sedimentological research in this region may also play an important role.

**Extraterrestrial spherules** are derived

1. from meteorites that exploded high in the atmosphere : cca. 60—120 km from the surface of Earth. (see *Földi, T. 1995*) There are three main types of spherules derived from meteorites : iron-, stony-, and iron-stony spherule that are identical with the three main types of meteorites.

2. from interplanetary dust (see: *Yamakoshi, K. 1994*)

3. from interstellar dust (see *Miono, S. et al. 1993; Miono, S. 1995; Anders, E.—Zinner, E. 1993; Zinner, E. et al. 1995*). The interstellar spherules can provide data relating to the movement and position of the Solar System within our galaxy, the Milky Way, in the geological past.

The surface of the Earth is permanently under a spherule-drizzle, which has partially been documented in various terrestrial strata.

Spherules also may be formed by **impacts** of crater-producing meteorites, as dissipated melt "glassy spherules", "microtektites" (see *Bouška, V. 1993a; Glass, B.P. 1978; 1990*). A second type of impact generated spherules is those produced by vaporization and condensation of the impacting body (e.g. *Bohor, B.F.—Betterton, W. 1990*). The best documented spherule-producing crater is found in Estonia, the "Kaali" crater (see *Shymankovich, S.—Kolosova, T.—Raukas, A.—Tirmaa, R. 1993*). In some instances terrestrial volcanic (see *Filimonova, L. G. et al. 1981; Filimonova,*

L. G. 1982) diagenetic, biogenic or industrial origin may be supposed as well (see Ausset, P. et al. 1994).

We cannot consider impossible the clouds of spherules to have originated from the active impact activity of the Solar System (see Hoyle, F.—Winchamansinghe, N. C. 1968). These clouds were gathered by the planets or went "down" spirally into the Sun according to the Poynting-Robertson Effect (see Yamakoshi, K. 1994, p. 5., 7., 14.). It is possible that in the early time of the Solar System more spherules landed on the surface of the Earth than in the recent time spitting through the atmosphere (see Crozier, W. D. 1962). In early times the meteorite impact activity on the surface of the Earth was stronger, producing more terrestrial impact-originated spherules.

In spite of their different origins the spherules are identical as to their spherical or deformed spherical form. The diameter of the spherules ranges between 1 $\mu$ m—3000 $\mu$ m, with an average of about 100 $\mu$ m.

**The first step in the world-wide investigations is to find reliable methods to distinguish the various genetic types. It seems that the impact and extraterrestrial types would be the global tools for correlation, because the related events are globally detected. (Great impacts, traversing meteorite and extraterrestrial dust clouds).**

During the history of the Earth the global occurrence of spherules seems to have altogether five abundance peaks (by extrapolation of certain sporadic local result):

**Late Devonian** (see Wang, K. 1992; Claeys, P.—J. G. Casier—S. V. Margolis 1992; Bouška, V. J. 1993a, 1993b; Claeys, P.—J. G. Casier 1994),

**Permian-Triassic boundary** (see Miono et al. 1993; Hadnagy, Á. 1994; Miono, S. 1995),

**K/T boundary global spherule layer** (see Bohor, B. T.—Betterson, W. J. 1991; Smit, J. et al. 1992; Pollastro, R. M.—Bohor, B. F. 1993; Albertao, G. A. et al. 1994; Xiao Zhifeng et al. 1994),

**Upper Eocene** (see Hazel, J. E. 1989; Glass, B. P.—Hill, C. M.—York, D. 1986; Keller, G. 1986; Kennett, J. P. 1977; Molina, E.—Gonzalvo, C.—Keller, G. 1993; Premoli Silva, I.—Cocconi, R.—Montanari, A. [eds.] 1988). A large impact-generated spherule-occurrence in Upper Eocene deep sea sediments from the western North Atlantic, Caribbean, Gulf of Mexico, equatorial Pacific, and eastern equatorial Indian Ocean, correlated using biostratigraphy is known (see Wuchang Wei 1995)

**Pleistocene** (see Glass, B. P.—Kent, D. V.—Schneider, D. A.—Tauxe, L. 1991; Li Chun-Lai—Ouyang Zi-Yuan—Lin Wen-Zhu 1994).



The origin of the impact and extraterrestrial spherules are independent from the terrestrial happenings, therefore are excellent tools for nail through various facies.

The complex comparative studies of the spherules and their host-sediments are very important for a global correlation programme (see *Detre, Cs. H.—Don, Gy. 1995b*).

A very important task of the investigations will be the field re-working of the meteorite fall areas, such as the Kaali crater (Estonia) and the Ries crater area (Germany), accompanied by collection of small fragments of meteorites and spherules. The field re-working of a no crater-producing meteorite fall (happened in 1857) area near to the village Kaba (East Hungary) is connected with detailed geological mapping and produced meteorite debris, micrometeorites and spherules. The stratigraphical evaluation of the spherule-bearing formations requires a "bed by bed" study. Field works of this type were realized in the Apuseni Mts (Romania) (see *Udubaşa, G.—Arsenescu, V. 1987; Hadnagy, Á. 1995*), East Carpathians (Romania) (see *Grigorescu, G.—Baltras, A. 1981*), as well as Gemeric granite region (Slovakia) (see *Jakabská, K.—Rozložník, L. 1989*) and in certain regions of Hungary in the last years.

In Hungary the successive collection of spherules from boreholes was started in early 1994 Permian, Triassic, Jurassic, Upper Cretaceous, Miocene, Pliocene, Pleistocene samples from many boreholes in Transdanubia and in the Great and Little Hungarian Plain (see *Detre, H. Cs. 1994; Kákay-Szabó, O. 1994; Gyuricza, Gy. 1994; Rálich-Felgenhauer, E. 1994; Siegl-Farkas, Á.—Wegreich, M. 1994; Szöör, Gy.—Korpás-Hódi, M.—Don, Gy.—Beszedá, I. 1994*). Spherule-bearing layers available for limited correlation have been found in the Lower Triassic (see *Rálich-Felgenhauer, E.—Rózsa, P.—Braun, M.—Beszedá, I.—Szöör, Gy. 1995*) in Upper Triassic (see *Dosztály, L. 1994*) in Upper Cretaceous (see *Bodrogi, I. 1994; Siegl-Farkas, Á. 1995*) in Middle Miocene, (see *Dávid, Á. 1995*) in Upper Pliocene (see *Rózsa, P.—Braun, M.—Szöör, Gy. 1995*). In the Carpathian Basin which is covered by vast sediment layers a very important branch of the spherule investigation is the research of hidden meteorite craters (see *Detre, Cs. H.—Don, Gy. 1995a*).

The research of the spherules has involved a large interdisciplinary collaboration, including stratigraphy, geochemistry, (micro)mineralogy, paleomagnetic research, planetology, meteoritics, meteorology, glaciology (see *Zbik, M.—Gostin, V. A. 1994*), ocean floor research (see *Glass, B. P.—Zwart, M. J. 1979; Yamakoshi, K. 1994*), and sedimentology in general. Radiometric research will certainly gain in importance in the future investigation of spherules.

From the point of view of extraterrestrial geology micrometeorites and extraterrestrial spherules are at the present time, the only real tools for

interplanetary correlation, since in all likelihood they have to occur on every planet of the Solar System (see *Bérczi, Sz.—Lukács, B. 1994*).

For this purpose a new IGCP project (No.384) is suggested by eight proposers: A. Bevan E., Papp (Australia), Cs. H. Detre (Hungary), B. P. Glass (USA), K. Jakabská (Slovakia), A. Raukas (Estonia), G. Udubaşa (Romania), Ouyang Ziyuan (China): "Impact and Extraterrestrial Spherules: New Tools for Global Correlation". All investigators of spherules are asked to kindly reconsider their contacts all over the world and suggest us names and addresses of possible participants. **It is obvious that the investigation of the spherules has to be global. This project is aimed at developing and organizing these global investigations. By way of this project the first international organization of a new scientific field will be organized.**

Recommended type areas for the world-wide investigations, according to the hitherto informations about the investigations:

1. Baltic and Scandinavian area (l.c.)
2. Central Europe (l.c.)
3. Mediterranean region (see *Funiciello, R.—Fulchignoni, M. 1969*)
4. Central (see *Glass, B. P. et al. 1991*) and Southern Africa (see *Koeberl, Ch. et al. 1993*)
5. Central and Eastern Asia (see *Li Chun-Lai et al. 1993, 1994; Ouyang Ziyuan; Xiao Zhifeng et al. 1994*)
6. North America (see *Glass, B. P.—Zwart, M. J. 1979; Pollastro, R. M.—Bohor, B. F. 1993*)
7. Caribbean region (see *Glass, B. P.—Zwart, M. J. 1979; Bohor, B. F.—Glass, B. P.—Betterton, B. J. 1993*)
8. South America: (see *Albertao, G. A. 1994*)
9. Australia (see *Glass, B. P. 1971; Wasson, J. T. 1990*)
10. Antarctica (see *Zbik, M.—Gostin, V. A. 1994; Kurat, G. et al. 1994; Tazawa, Y.—Fukuoka, T.—Yamanouchi, E.—Miyano, Y.—Endo, K.—Kohno, M.—Fujii, Y. 1995*)

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# RELATIONAL DATA MODEL FOR A GIS DATABASE

## Proposed for the IGCP 384 Project

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### Abstract

The "Impact and Extraterrestrial Spherules: New Tools for Global Correlation" IGCP 384 project requires the collection, storage, organisation and analysis of a large amount of spatially referenced data. Geographical Information Systems are complex computer software packages best suited for this purpose. First this paper gives a short introduction and concise definition of the most important concepts and terms used in relation to spatially referenced relational databases and Geographical Information Systems. In the second part the paper describes the suggested relational model for the spherule database of the IGCP 384 project.

### Introduction

The data model presented in this paper is a preliminary one. A data model, suitable for most researchers in the project to carry out spatial and non spatial analysis, can be developed only with the continuous exchange of information. Researchers working on scientific problems related to the "Impact and extraterrestrial spherules" project are invited to contribute to the further development of this data model by informing the GIS group about their requirements. For the purpose of common understanding concise definition and short explanation is given for the most important terminology used in the GIS literature.

## 1. Definitions

### 1.1. Data

#### 1.1.1. Data in general

Data is information coded in a mathematical, verbal, or any other pre-defined language. The information describing different properties of the same real object will result in different data.

#### 1.1.2. Geo-referenced or geo-coded data

Each data object has a spatial location expressed as a two dimensional (2D) coordinate pair (for example Easting-Northing, strike-dip), or as a three dimensional vector (for example

drillhole collar location as Easting-Northing, with the depth of core sample) in some previously defined geo-referenced coordinate system.

### 1.1.3. Spatially referenced data

Built from geo-coded data points. Describes the location and shape of geo-referenced features, and those features' spatial relationship to each other.

#### 1.1.3.1: Shapes of geo-referenced features

These can be: points, lines or area. Points: for example individual impact crater locations. Lines: geological faults, fluvial systems. Area or closed polygons: lake shoreline, mapped rock outcrops.

#### 1.1.3.2: Spatial relationships

For example distance; elevation difference; "next to" relationship; "identify the nearest feature of a given type" relationship between data objects or features.

#### 1.1.4. Descriptive data or attributes

Any kind of descriptive information – called attributes – about features can be stored, together with the unique ID) of the feature. This unique ID ensures that the required information can be selected from the database and retrieved with the relevant feature. The descriptive information can be any data type the computer system is able to handle. For example: text files, digitalised photos stored as raster files, hand drawings as vector files, sound recordings etc.

#### 1.1.5. Linking spatially referenced data with descriptive data in a GIS

Integrating these two data types opens new ways of analysis. The information in the tabular – mostly, but not necessarily relational – database can be queried in a new, spatial context (for example: what is the closest outcrop to a given location, with more than 50 km<sup>2</sup> area?) through displaying the spatially referenced data on the computer screen, and using its spatial relationships. On the other hand, various up-to-date digital maps can be created according to user needs, by selecting and using the appropriate information from the tabular database. This integrated system is called Geographical Information System, GIS. These systems are developed for the purpose of handling and analysing together spatially referenced data with its nonspatial attributes kept in a database.

## 1.2. Data models

### 1.2.1. Definition of a data model

Data model means storing data with their relationships, connections in a mathematical structure, with well defined mathematical procedures. Most widely used is the relational data model, developed by Codd in 1970. Advantage of a predefined data structure is that many user

can use the same data simultaneously. The data is stored only once, and every authorised user can use the part he needs, knowing the general structure of the database. Subsets can be extracted, reorganised, according to individual needs.

## 1.2.2. Elements of a data model

### 1.2.2.1. Object

The “object” as used in the literature, has two level meaning. It can mean object category, or the individual member of the object category. Object category is an abstract category of real things defined for the purpose of a certain database. For example “meteorite” could be defined as an object category in the Spherule GIS. Mathematically an object is a set or class, and the real objects that make up the set are called its elements or members. For example the “Kaba meteorites” could be defined as members of the “meteorite” object class in Spherule GIS. The database object category can be specified by a condition for membership in it (for example define “meteorite” as an object category which is covered by the Western Australian Meteorite Act), or it can be simply defined by its members without abstract definition (the objects in the “meteorite” object class will be called “meteorites”).

### 1.2.2.2. Attribute

The “attribute” as used in the literature, has two level meaning similarly to the usage of “database object”; attribute can mean the attribute-type as well as the individual occurrence of an attribute. Attribute is an abstract set of features describing some properties of objects (for example mineral hardness on a scale of 6, presence or abundance of a certain element on a binary scale of yes or no, or geographical latitude in the scale of degrees, minutes and seconds). If a certain set of attributes uniquely identifies an individual object, it is called a key. If one or a set of attributes uniquely identifying an object can't be found, they need to be assigned to each of the objects (unique identifier, ID).

### 1.2.2.3. Relation

The word: “relation”, similarly to the words “object” and “attribute” is also used meaning “relation-type” as well as “the individual value of the relation-type”. Relation is an abstract description of a relationship between individual objects (for example a rock sample – an element of the “meteorite” object class – belonging to the “Kaba” group of meteorites). This is a one-to-many relationship, as there are several meteorites in that group, but each sample belongs to one and only group of meteorites.

Possible relationships between objects:

one-to-one (1:1)

one-to-many (1:N)

many-to-many (N:M)

The data model consists of the objects, their attributes and relations in an abstract, formalised way.

### 1.3. Database systems

#### 1.3.1. Definition of a database system

A database system is nothing more than a computerised record-keeping system. A database system is usually a complicated software package for building, maintaining and using the database. The user of a system can perform a variety of operations on electronically stored records. In the process of data definition and maintenance the user can add new, empty data files to the database, insert new data into existing files, retrieve data from existing files, update data, delete data, remove existing files. In the process of using the database a well defined set of queries can be executed on the data files.

#### 1.3.2. Other tasks of a database system

Data security: each user can have a password and a set of permissions to access certain parts of the database. Unauthorized access is a crime, and can be penalised seriously.

Data integrity: data which doesn't satisfy certain requirements either in format or content, can be automatically rejected.

#### 1.3.3. Languages for database systems

According to the two main functions, there are two main languages of a database system: the data definition language and the query language. They either work independently as a programming language (ie. dBase), or need a real programming language in the background to function (ie. SQL = Structured Query Language).

#### 1.3.4. Spreadsheets

For simple tasks spreadsheets can be used instead of large database systems. These tasks can be, for example; entering, organising and retrieving the data.

### 1.4. Geographical Information System

If every data point is spatially referenced in a database system, than with a package of appropriate software, called Geographical Information System (GIS) the data can be displayed on a computer screen in a geographically correct position, in a specified cartographic projection. The data can be transformed and re-displayed in any other projection. With the appropriate selection from the database any other stored auxiliary information (text, images, even sound recordings) can be displayed together. The data can be queried spatially. For example: which are the spherules found within a certain geographic region, on a given rock type, at elevations higher than 500 m, within 50 km of a recent riverbed?

## 2. Spherule GIS data model

The proposed data model evolved during an early stage of data collection. The model needs to be re-assessed and corrected continuously, to incorporate new objects, attributes and relations found by researchers working on the project.

2.1.1. The *objects* of our data model are hard rock pieces from the Cosmos, from the surface or from the depth of the Earth. They could be spherules, cosmic dust, meteorites, rock samples, drill core material, among others. At this early stage we store all of these material in the same database and analyse them the same way.

2.1.2. The *attributes* of our data model are those properties of the objects which we want to record, store and analyse. As there are a large number of these properties, we organise them into tables. Within one table we store attributes of a common theme. Our aim is to organise the attributes in a way that any database query would open the least possible number of tables at once. This has a large effect on the speed and performance of the GIS.

Each table has the numeric identifier (ID) as the *key attribute* included. This numeric ID links tables together, and also provides the link to the spatial co-ordinates, Easting and Northing.

The numeric ID, Easting and Northing together provides the link between the non-spatial database and the spatial GIS software, enabling spatial display and analysis of all non-spatial attributes, linking them to the location of origin of samples.

2.1.3. *Relations* used in defining data dictionaries. For example

Material\_table = ID + Type + International\_type + Size + Mass + [Minerals] + Other  
where

“Type” = {spherule | cosmic\_dust | meteorite | rock\_sample | drill\_core\_material}.

Here

+ signifies a logical AND relationship

{1 | 2 | 3} signifies a logical OR relationship.

[Minerals] signifies a “one-to-many” (1:N) relationship

## 2.2. List and description of tables:

Name of Table	Description
ID	unique identifier, Easting, Northing
Horizontal location	horizontal location of origin of sample
Vertical location	vertical location of origin of sample
Chronology	geological, biostratigraphic and paleomagnetic horizon of inbedding layer at the location of origin of sample
Material	important minerals, rocktype, or meteorite-type of the sample
Probe	laboratory analysis of chemical composition of the sample
Catalogue	international code, catalogue and collection where the sample kept
Literature	important literature describing the sample
Results:	results achieved during research related to IGCP 384 in-determination of age, origin or other important properties of sample

### ID\_table

Name	Description	Data type
ID	Unique identifier for each sample	Text 10 characters
Easting	Location data in a common coordinate system	Double precision floating or real
Northing	Location data in a common coordinate system	Double precision floating or real

### Horizontal\_location\_table

Name	Description	Data type
ID	Unique identifier for each sample	Text 10 characters
Deviation	Horizontal deviation	Double precision floating or real
Name	Location name	Text 20 characters
Description	Description of location	Text 60 characters

### Vertical\_location\_table

Name	Description	Data type
ID	Unique identifier for each sample	Text 10 characters
Deviation	Vertical deviation	Double precision floating or real
Elevation	Elevation of location	Double precision floating or real
Description	Depth of sample below location of sample	Double precision floating or real
Description	Description of location	Text 60 characters

### Chronology\_table

Name	Description	Data type
ID	Unique identifier for each sample	Text 10 characters
International age	International age of inbedding layer	Text 40 characters
Hungarian age	Hungarian age of inbedding layer	Text 40 characters
Biostrata	Bio-stratigraphical horizont	Text 40 characters
Paleomag	Paleomagnetic horizont	Text 40 characters

### Material\_table

Name	Description	Data type
ID	Unique identifier for each sample	Text 10 characters
Type	Type of sample {spherule   cosmic dust   meteorite   rock sample   drill core material}	Text 40 characters
International type	International classification of sample	Text 40 characters
Size	Largest diameter of sample in cm	Double precision floating
Mass	Mass of sample in grams	Double precision floating
Minerals	Important minerals identified in the sample	Text 40 characters
Other	Other important material in the sample, un/identified, problematic etc.	Text 40 characters

### Probe\_table

Name	Description	Data type
ID	Unique identifier for each sample	Text 10 characters
Lab	Laboratory measurement made by	Text 40 characters
Date	Date of analysis	Date
Technique	Experimental technique used	Text 10 characters
Elements	{List of elements analzsed}	Text
Accuracy	Accuracy of analysis for each element	Double precision floating
Detection_limit	Detection limit of the used analytical technique for each element	Text

### Catalog\_table

Name	Description	Data type
ID	Unique identifier for each sample	Text 10 characters
Catalog	Catalog numbers of the sample	Text
International code	International code of sample	Text
Collection	Collection where the sample kept	Text
Photo	Photo documentation available	Text

### Literature\_table

Name	Description	Data type
ID	Unique identifier for each sample	Text 10 characters
Volume	Volume and name of journal	Text
Author	Name of author	Text
Title	Title of paper or book	Text
Abstract	Short abstract of paper	Text
Note	Anz other note	Text

### Results\_table

Name	Description	Data type
ID	Unique identifier for each sample	Text 10 characters
Origin	Suggested origine of spherule or other material	Text
Age	Suggested age of spherule or other material	Text
Other	Anz other result	Text

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# Astronomical significance of microspherule recovered from Paleozoic and Mesozoic radiolarian chert in Japan.

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## abstract

Several authors<sup>1),2),3)</sup> have discussed what would happen if the Solar System were to pass through a dense interstellar matter, however conjectured triggers of the glacial epoch are not confirmed and it seems that their hypothesis are scarcely yet convincing for the lack of direct imprint upon the Earth. A possible evidence of the interstellar grain similar to carbonaceous chondrite has been suggested for the microspherules that have been recovered in Mesozoic-Paleozoic bedded chert at southwest Japan. Can interstellar dust penetrate directly into the Solar System? Are periodic mass extinction caused by interstellar molecular clouds? This article summarizes the several results concerning microspherule in radiolarian bedded chert and an attempt to provide a new scenario to substitute the impact and volcanic eruption hypothesis to explain the mass extinction.

## 1. Introduction.

In Japan the most appropriate geological horizon for the ancient microspherule studies are distributed around the Shimanto belt and Mino-Tamba belt. The bedded chert might have been formed along subduction zone of Pacific Ocean plate. The typical outcrop of cretaceous chert at Yokonami peninsula, Kochi, Jurassic-Triassic chert at Kagamihara along the Kiso River and Permian-Triassic chert at Sasayama section are shown in Fig.1, respectively. Sedimentary rock generally contains volcanic ash, cosmic dust and skeletons of small marine animal etc. Numerous magnetic microspherules were

recovered from these chert as shown in Fig.2.

## 2. PIXE analysis and observation of internal texture of recovered microspherules.

In our previous studies<sup>4),5)</sup>, the ratio Ti/Fe vs. Cr/Fe obtained from PIXE analysis of microspherules provided a useful index for distinguishing whether terrestrial or extraterrestrial in origin. The relative elemental concentrations of Ti/Fe, Cr/Fe, Mn/Fe, Co/Fe and Ni/Fe in the microspherules are closely comparable to those in carbonaceous chondrite.

The author developed a new method to divided the microspherule into two under the stereo microscope as shown in Fig.3. It is clearly shown that the cavities might be unable to take such shapes by corrosion of metallic nucleus, as compared to microspherules recovered in recent Pacific Ocean sediment(see Fig.4). Eventually, so far as we can judge, it is conjectured that such cavities were formed by escape of  $\text{CO}_2$  or vaporizing water.

Even more remarkable, the abundance of microspherules show an abrupt increase to hundred times more in the Triassic chert adjacent to P/Tr boundary as well as in the Tr/J boundary as compared to the ordinary mean value as shown in Fig.5. From these findings, ancient cosmic dusts could account for interstellar grains something like carbonaceous chondrite.

## 3. Origin of microspherule and giant molecular cloud.

The recent millimeter radio waves astronomy from interstellar carbon monoxide has revealed clearly the presence of molecular cloud in the spiral arm. Fig.6 shows the distribution of the clouds in the galactic plane viewed from the perspective of an observer located 2 kpc above the Sun obtained by Dame et al<sup>6)</sup>. Now we can envisage that it is definitely possible for Solar System to pass through the interstellar cloud. Furthermore, recently S. Minami<sup>7)</sup> of Osaka City University reported a surprising simulation. His result showed vanished ionized cavities surrounding the solar system as soon as neutral hydrogen gas was injected. This indicates the possibility that interstellar dust can enter the solar system against Lorentze force.

There now appears to be a close correlation between geological

rhythms and cosmic rhythms. Hence, we postulate an alternate scenario for climatic change on the Earth and mass extinction when the Solar System traverses an interstellar cloud as follows,

- 1) The Sun would accrete enormous amount of hydrogen molecule.
- 2) also hydrogen and ignition of dust grain would remove the oxygen in the Earth's atmosphere.
- 3) the duration of the shielding by cloud is estimated to sustain for a period of approximately  $10^5$  -  $10^6$  years.
- 4) this could trigger a glaciation upon the Earth.

A number of paleontologists have pointed out that the mass extinction over geological time was not an instantaneous event. So far the evidence of findings on microspherule presumed to interstellar origin should be consistent with the hypothesis that the Solar System encountered an interstellar molecular cloud.

#### 4. Solar luminosity evolution and size distribution of microspherule enhancement over geological time.

The theory of stellar evolution predicts that the solar luminosity was originally only about 70% of its current value. Unfortunately, the evidence do not exist upon the Earth. Dust particles in Solar System are stationary due to the balance between radiation pressure and solar gravitational attraction. Then, if solar luminosity gradually increased, mean size distribution of microspherule over geological time should varied depend on luminosity. In fact, preliminary results of size distribution are indicating a gradual increase. Thus if the correlation described above could be rationalized according to the standard solar model, it should contribute to a new understanding of the solar luminosity evolution and its relation to terrestrial climate. This possibility needs further investigation.

#### 5. Summary.

A decade before, most of theorists generally believed that the Solar System started out a clean state and spends most of its life without interaction with interstellar medium. A recent discovery made by E.Anders et al<sup>8)</sup> would ultimately lead to the overthrow of this concept. Also Ulysses<sup>9)</sup> group found the streaming of interstellar dust. By itself, this is not very surprising since the late Prof.

N.Onuma of Tsukuba University of Japan already suggested the possibility of interstellar origin of carbonaceous chondrite and penetration of interstellar dust into Solar System in 1978. Now, it should be better to take into account that the Earth would be collecting the interstellar dust from various region of Galaxy as it encounters with molecular cloud. Further studies of ancient microspherule should be provide a new understanding of the structure of the Galaxy and it seems very worth-while.

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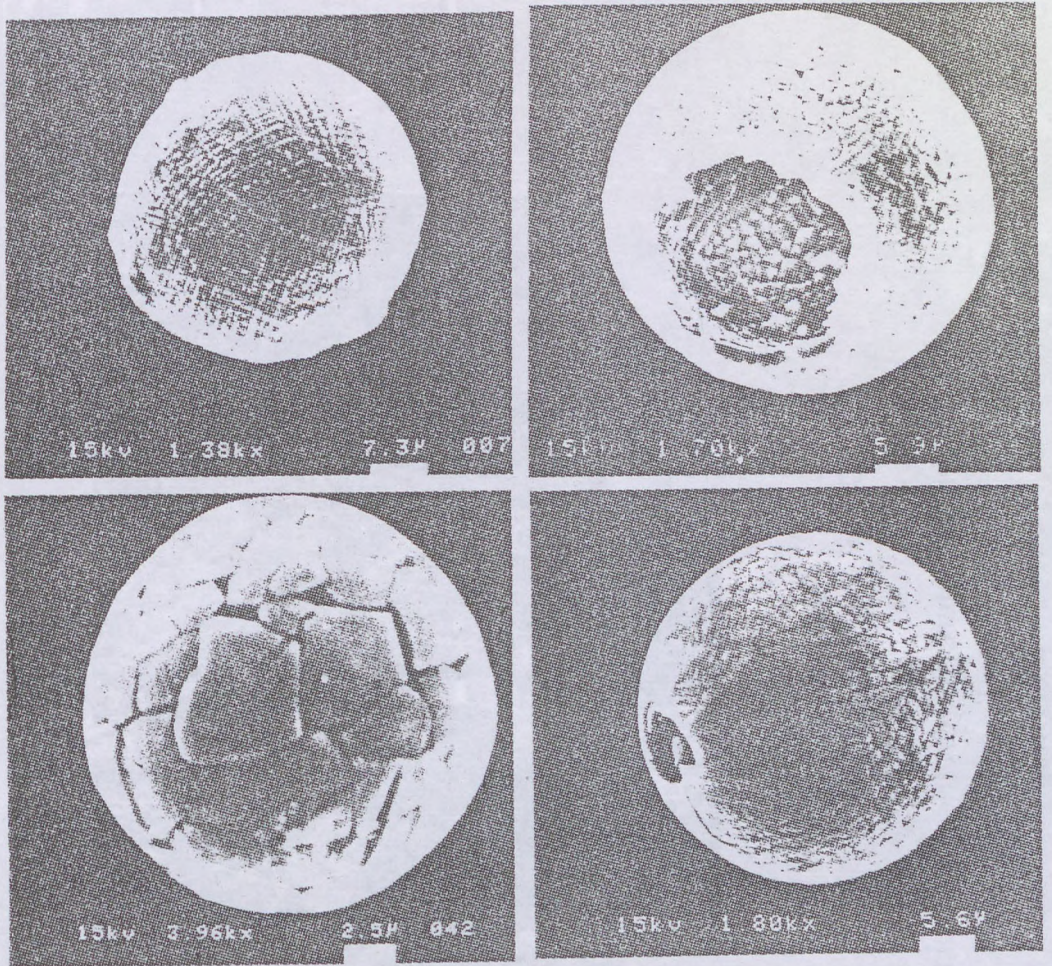


Fig.1. Typical microspherules recovered from Paleozoic and Mesozoic bedded chert.

Fig.2. (continued)

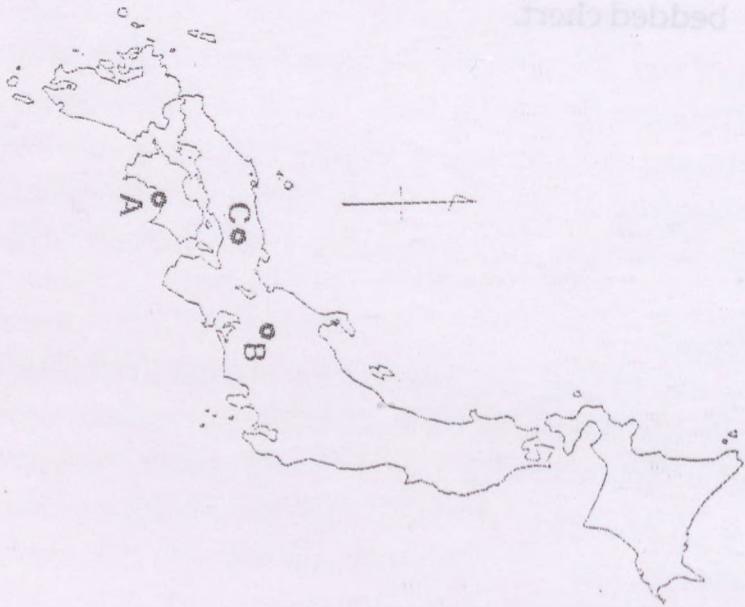


Fig. 2. Outcrop of Cretaceous chert at Yokonami peninsula, Kochi(A), Jurassic-Triassic chert at Kagamihara along Kiso River, Gifu (B) and Permian-Triassic chert at Sasayama, Hyogo(C) respectively.

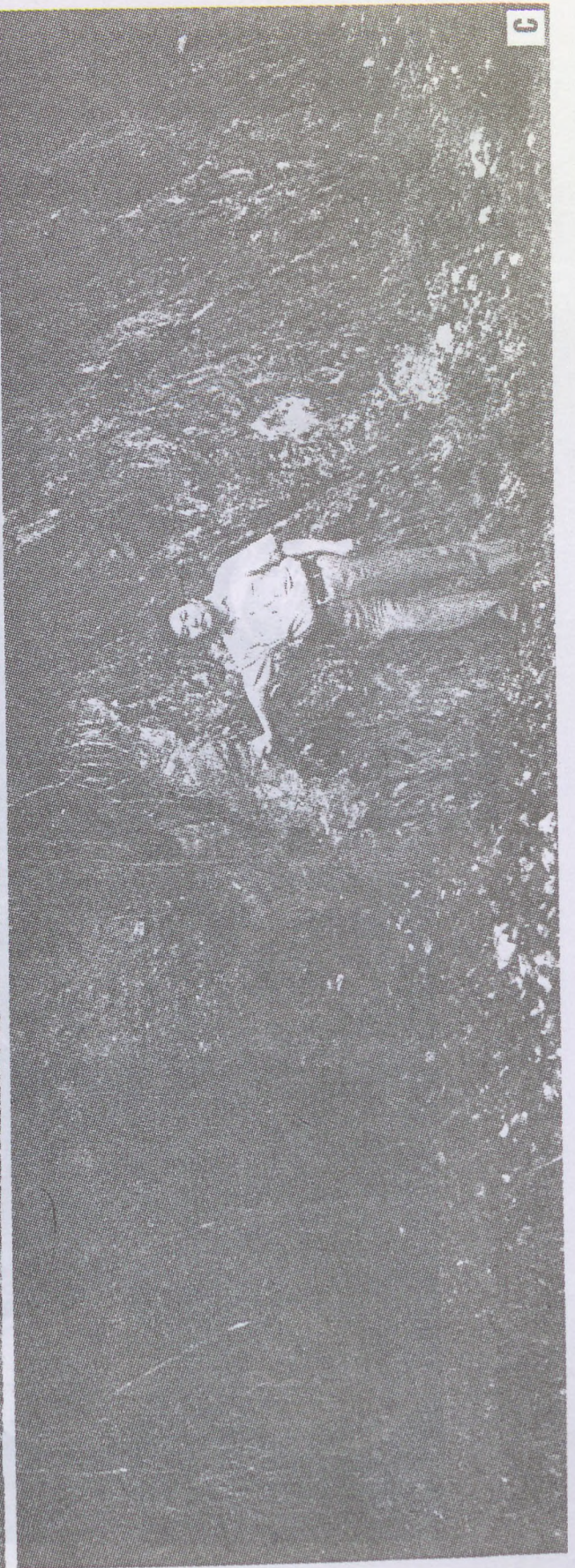


Fig.2. (continued)

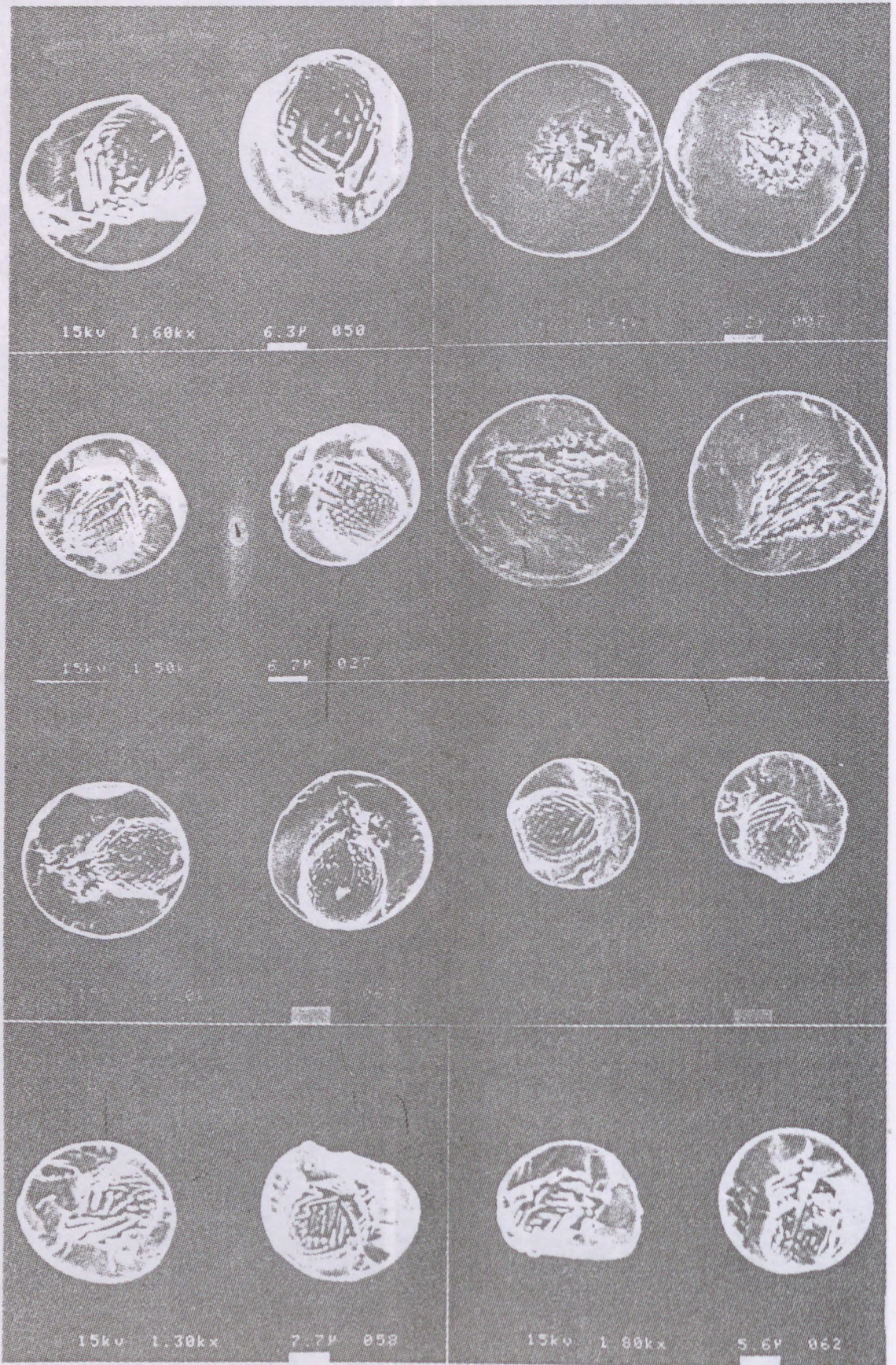


Fig.3. Internal texture of microspherules divided manually under stereo microscope.



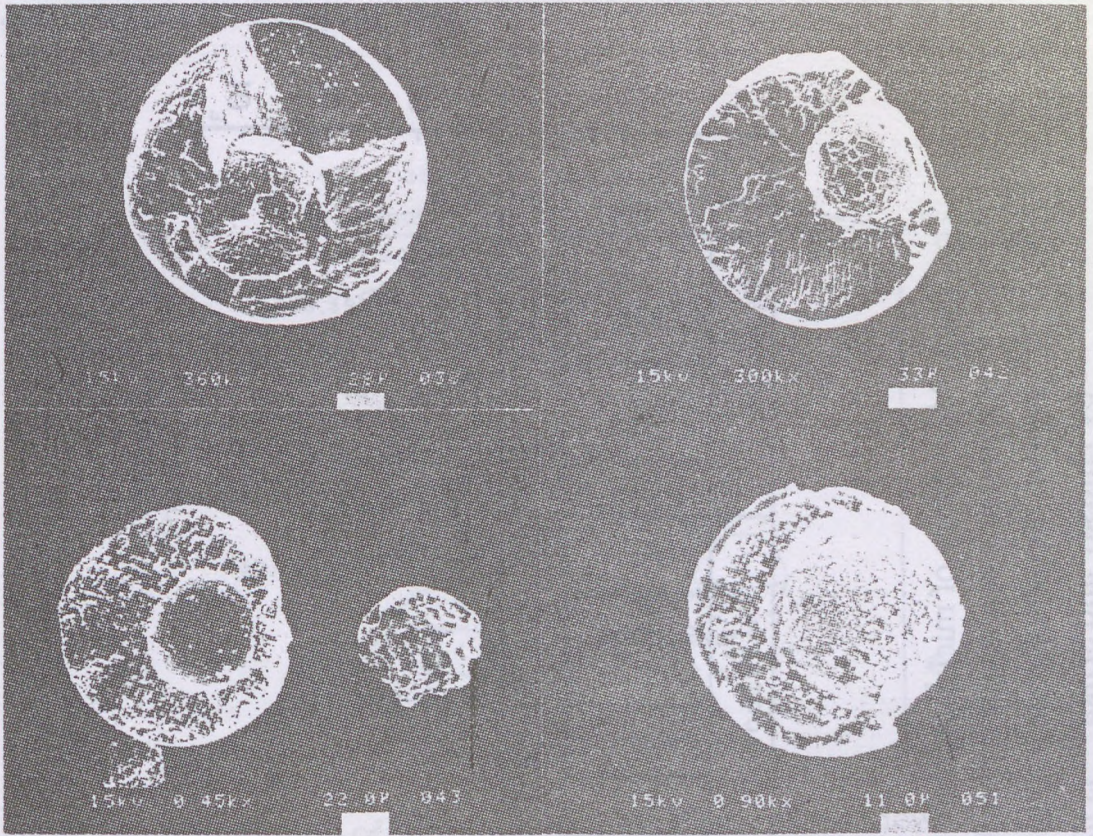


Fig.4. The microspherules including metallic nucleus recovered from recent Pacific Ocean deep sea sediment.

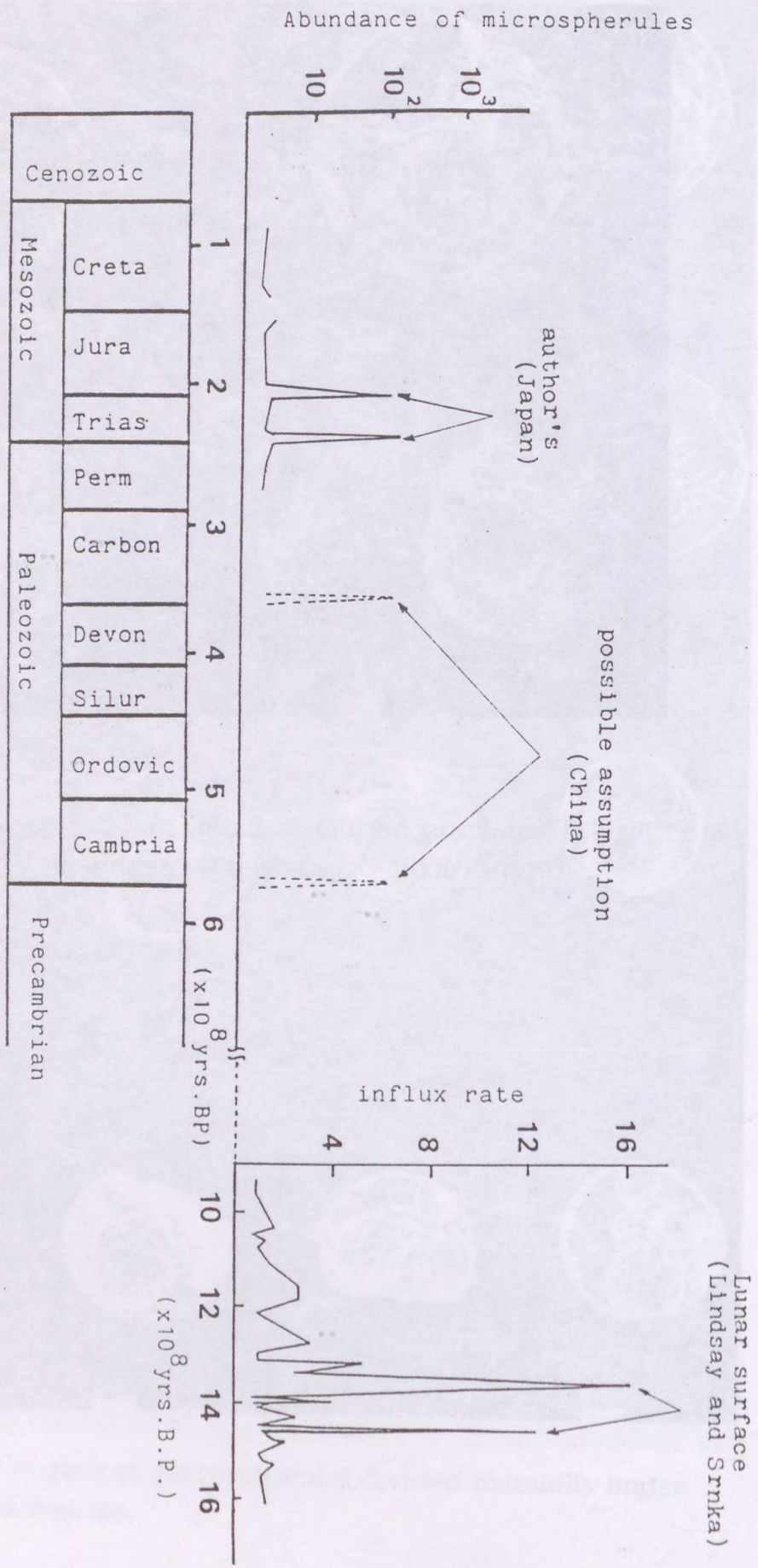


Fig.5. Abundance of microspherule over past geological period. Dotted line represent a possible assumption from IGCP 203 report. Result of past interstellar dust on the Lunar surface by Lindsay and Srnka<sup>(10)</sup> is illustrated in the same figure.

SPHERICAL ACCESSORIES IN GENERIC GRANITES OF THE WEST  
CARPATIANS (SLOVAKIA)

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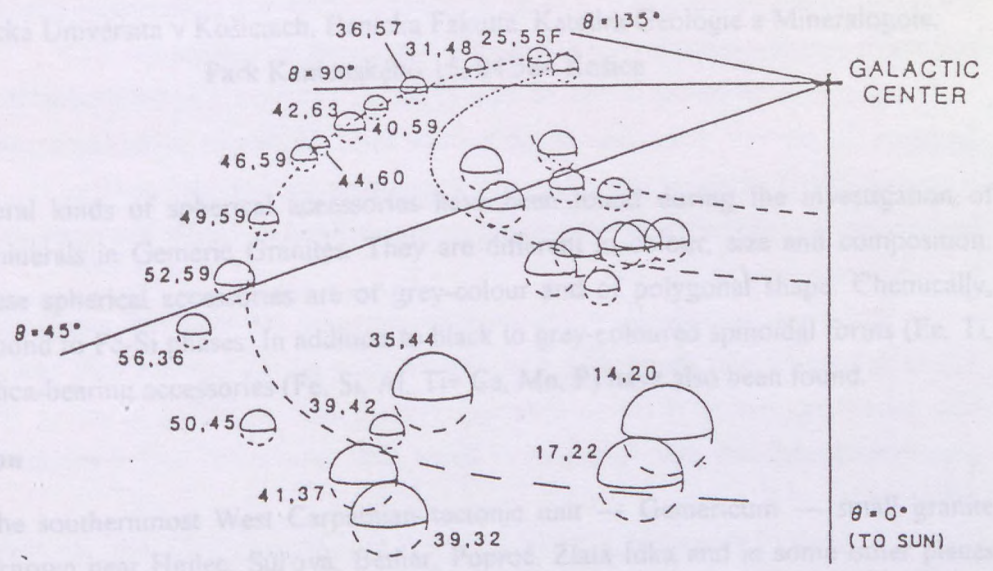


Fig.6. The distribution of the cloud in the Galactic plane viewed from perspective of an observer located 2kpc above the Sun to the direction of Sagittarium and Scutum arm.(after T.M. Dame et al)

These granites have been designated as "Generic Granites" (Báňas et al. 1970; Gubal, 1977; and in-bearing from various localities and by various authors: Dželo et al. (1974); Bagdasaryan et al. (1977); Kováčik et al. (1979); Katar-Rybní (1979); Černý et al. (1980). These indicate Permian, Jurassic and Cretaceous ages. Accordingly the Generic Granites might be polyphase intrusions which were subject also to hydrothermal processes.

Morphological and physical properties of the spherical accessories

The spherical accessories (spherules) have been found in an investigated sample of the Generic Granites from the localities: Beňar, Yafová and Kóber (Fig. 1). The spherules occur in the heavy fraction sized 0.12-0.09 mm. They were locked for also in thin sections where they were eventually discovered (Fig. 2).

Under ordinary binocular microscope, several types of spherules can be recognized. The most abundant are gray spheroidal bodies, followed by black-bare-black ferruginous and "bluish" ones. Brown-black (aluminoferritic) and dark gray (aluminous) spherules form almost perfect spheres, whereas the gray ones are of very different shapes deformed or spheroidal.

The gray bodies — as observed under the scanning electron microscope (SEM) — are sometimes spherical (Fig. 3). On their surface, however, there are often small conical or



# SPHERICAL ACCESSORIES IN GEMERIC GRANITES OF THE WEST CARPATHIANS (SLOVAKIA)

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## Abstract

Several kinds of spherical accessories have been found during the investigation of accessory minerals in Gemic Granites. They are different in colour, size and composition. Most of these spherical accessories are of grey-colour and of polygonal shape. Chemically, they correspond to Fe-Si phases. In addition to black to grey-coloured spinoidal forms (Fe, Ti, Al) some silica-bearing accessories (Fe, Si, Al, Ti+ Ca, Mn, P) have also been found.

## Introduction

In the southernmost West Carpathian tectonic unit — Gemicum — small granite bodies are known near Hnilec, Súľová, Betliar, Poproč, Zlatá Idka and in some other places (Fig. 1). These granites have been designated as "Gemic".

The discovery of the fact that the Gemic Granites are tinbearing (Baran et al., 1970) triggered detailed research of their petrological and geochemical properties (Varga, 1975, Gubač, 1977, Tauson et al., 1977, Rub et al., 1977 and others) concerning their crustal origin and tin-bearing character. Recently, more abundant data were obtained on their isotopic age from various localities and by various methods Bojko et al. (1974), Bagdasaryan et al. (1977), Kovách et al. (1979), Kantor-Rybár (1979), Cambel et al. (1980). These indicate Permian, Jurassic and Cretaceous ages. Accordingly the Gemic Granites might be polyphase intrusions which were subject also to hydrothermal processes-

## Morphological and physical properties of the spherical accessories

The spherical accessories (spherules) have been found in all investigated samples of the Gemic Granites from the localities: Betliar, Súľová and Hnilec (Fig. 1). The spherules occur in the heavy fraction sized 0.12—0.09 mm. They were looked for also in thin sections where they were eventually discovered (Fig. 2).

Under ordinary binocular microscope, several types of spherules can be recognized. The most abundant are gray spheroidal bodies, followed by black-blue-black ferruginous and "titanium" ones. Brown-black (alumoferrosilicium) and dark-gray ("aluminium") spherules form almost perfect spheres, whereas the gray ones are of very different shapes designated as spheroidal.

The gray bodies — as observed under the scanning electron microscope (SEM) — are sometimes spherical (Fig. 3). On their surface, however, there are often small conical or

tumour-shaped protrusions or holes (Fig. 4), which are frequently intergrown (Figs. 5, 6). Potato- (Fig. 4) and pear-shapes (Fig. 7), even with some sort of "branches" (Fig. 8, 9), are common. When magnified several thousand times, the crust of the gray spherules exhibits polygonal disintegration (Fig. 10). Such a type of crust usually covers materials which undergo rapid cooling and shrinking — e.g. meteorites or amorphous alloys.

The gray spherules in thin-section (Figs. 2, 11) were always present as inclusions in feldspars (in perthitic orthoclase or albite-oligoclase). Under the polarization microscope, the cross-sections of spherules show tumours and cavities on their surface (Fig. 11). They are not transparent, in reflected light with bright orange reflection. Under the binocular microscope their surface is lustrous.

The black-blue-black and brown-black spherules are mostly of regular spherical (Fig. 12) or spheroidal oval shape (Fig. 13). Their surface is very lustrous with blue-black shades. The superficial texture of some spherules reveals their crystalline structure (Fig. 14). The brown-black and dark gray spherules of larger size, "aluminium" or "aluminium-ferrosilicate" are sporadic.

Because of their small number and small size, we only 0.02 mm, the chemical analysis of the spherules is difficult. The properties of the gray spherules have been determined more accurately, because they are relatively more abundant and therefore also X-ray diffraction analysis could be carried out.

### Chemical composition and texture of the gray spherules

The chemical composition of the gray spherules electron microprobe analysis, JEOL — Super Probe733 supplemented by a microparticle analyser and the control computer PDP-11/04 (Geological Institute of Dionýz Štúr, Bratislava) is given in Tabs. 1, 2.

Table 1

#### Microprobe analyses of gray spherules in a section from Hnilec Granite

Points Elements%	1	2	3	4	5	6
Fe	82.9586	81.1299	81.2779	82.5038	82.8449	81.5273
Cr	0.2406	0.2130	0.2165	0.1104	0.2086	0.2623
Mn	0.3811	0.4385	0.3896	0.3261	0.3439	0.4029
Al	0.0582	0.077	0.1240	—	—	0.1063
Si	6.8245	6.6625	7.0283	7.3920	7.0569	7.0076
Σ	90.463	88.5216	89.0363	90.4543	90.4543	88.9035

1, 2, 3, 4, 5, 6 — points of records in sections.

Microprobe analyses carried out in the Geological Institute of D. Štúr, Bratislava

In the gray spherules, Fe and Si are by far the most predominant elements. Other elements — (Cr, Mn and Al) are much less abundant. Eventual occurrence of oxygen and carbon is not excluded. (The sum of analyses given in Tab. 1 is only 88—90%.) However, the stoichiometry of ferrosilite or fayalite requires more than 10% oxygen and less Fe. Or is the residual 10—12% represented by carbon and the composition corresponds to a phase between moissanite and cohenite?

Table 2

**Semiquantitative analysis of gray spherules**

-Locality Elements%	Hnilec	Hnilec	Súl'ová
Fe	77.955	75.371	81.50
Cr	0.479	0.483	—
Mn	0.323	0.356	0.34
Si	21.020	23.389	17.55
Cu	0.222	0.401	0.10
Σ	99.999	100.00	99.49

Semiquantitative analyses only an energy dispersion analytic unit attached to REM-JEOL-JSM-35CF (Department of Material Science, Metallurgical Faculty, Institute of Technology, Košice). Copper content may be due to reflection from copper base.

The chemical composition of the gray spheroidal bodies is similar to that of ferrosilicium. Ferrosilicium produced by the firm Knapsack has the following composition: 66.35% Fe and 33.35% Si. If our gray spherules are of Fe—Si character, then their Fe/Si ratio is higher than that corresponding to ferrosilicium. The Fe/Si ratios given mainly in Tab. 2 best correspond to the formula  $Fe_2Si$ . Ferrosilicium with the formula  $Fe_2Si$  contains 79.91% Fe and 20.09% Si. Analyses in Tab. 2 show that our spherules contain 75—81% Fe and 17—23% Si. Nevertheless, Tab. 1 does not exclude also other compounds, mainly with carbon and oxygen. Distribution of Fe, Si, Mn and Cr in the gray spherules observed in the section in Fig. 15a under the scanning electron microscope by means of EDAX — JEOL — JSM — 35 CF (Department of the Science on Metals, Metallurgical Faculty, Institute of Technology, Košice) indicates a homogenous material with no zonation and inclusions (Fig. 15b).

*X-ray diffraction analysis of gray spherules*

The X-ray diffraction analysis has been carried out by Ždimera (Faculty of natural Sciences, Charles university, Prague) by means of the Debye-Scherer method with diffracted radiation registered on a flat film. The analysis was carried out under the following conditions: radiation Co/without filters, exp. 92.30 hrs, chamber Ø 114 — MÜLLER MIKRO. The results can be seen in Tab. 3. The same sample has been investigated by means of X-ray diffraction analysis Derco (ATNS Košice). He used the Bragg-Brentano method on the apparatus

Mikrometa II under these conditions: radiation FeK  $\alpha$ ; filter Mn, voltage 25 kV, current 12 mA, displacement of the arm of the goniometre 2°/min, movement of paper 600 mm/hr, subduing T/10, screen 10 -5, sensitivity  $3 \times 10^2$ .

The X-ray diffraction analysis (Fig. 21) suggests a mineral phase with lower degree of crystallinity. The main plane (100) is developed very well, whereas the other secondary ones are developed only very poor. This fact confirms the presence of a crystalline mineral phase of the following types: FeSi, Fe<sub>2</sub>Si, FeSi<sub>2</sub>, Fe<sub>3</sub>Si, with the best approximation to the phase FeSi.

The assumption that our gray spherules are related to some phase in the Fe — Si system is confirmed also by an X-ray diffraction pattern (Fig. 21) obtained in the same way as the record in Fig. 22. The main planes of both these materials are very close.

### *Characteristics of the other spherules*

The most abundant gray spherules are sporadically accompanied by black, and/or brown-black and dark gray ones. These could not be studied by X-ray techniques because of their insufficient amount. Semi-quantitative analyses have been carried out by means of electron microanalysis by the method of energetic dispersion from the surface of the spherules coming from the Gemic Granites of the localities Hnilec, Súl'ová and Betliar. The following types are present (ranked according to their abundance):

Group of *ferruginous spherules* — on the basis of 11 measurements they average 98—99% Fe, 0,2—0,9% Mn, 0,5% Ti (only traces of other elements).

Group of *titanium spherules* — characterized by very small size and following composition: 55% Ti, 19% Fe, 10% Mn, (1—2% Al, K, Ca).

Group of *aluminium spherules*: 91,98% Al, 5,56% Fe, 0,97% Ti, 0,42% Mn.

Group of Fe-Al-Ca-Si spherules they have a very variable composition.

Table 3

### **X-ray diffraction analyses of spherules from Hnilec Granite and ferrosilicium of the firm Knapsack, % tabular values**

Spherule type	Phase	d (Å)	Intensity
Ferruginous	FeSi	100	Very strong
		110	Weak
Titanium	Ti	100	Very strong
		110	Weak
Aluminium	Al	100	Very strong
		110	Weak
Fe-Al-Ca-Si	FeSi	100	Very strong
		110	Weak



X-ray diffraction analyses of spherules from Imilec Granite and ferrosilicum of the firm Knapsack, % tabular values

Fe <sub>2</sub> Si ASTM 26 1141		Fe <sub>3</sub> Si ASTM 11 616		FeSi (3574)		FeSi 1:1(3572)		FeSi <sub>2</sub> (3715)		Rank		M e a s u r e d		v a l u e s			
d(mm)	I	d(mm)	I	d(mm)	I	d(mm)	I	d(mm)	I	No.	gray spherules	d(mm)	I	gray spherules	d(mm)	I	ferrosilicum by Knapsack
0.2821	40	0.325	40	0.316	15	0.317	60	0.51	30	1	3	0.7560	1	3	0.7560	1	1
0.1988	100	0.283	40	0.259	10	0.258	60	0.237	63	2	2	0.3267	2	2	0.3267	2	2
0.1625	10	0.199	10	0.200	100	0.224	40	0.189	30	3	2	0.2817	2	2	0.2817	2	2
0.1406	60	0.170	40	0.182	40	0.200	100	0.170	8	4	1	0.2194	1	1	0.2194	1	11
0.1256	20	0.162	20	0.141	1	0.183	90	0.184	100	5	10	0.1993	10	10	0.1993	10	100
0.1149	100	0.141	100	0.135	10	0.149	40	0.178	15	6	6	0.1700	1	1	0.1700	1	14
0.09553	100	0.1145	100	0.124	4	0.135	60	0.170	8	7	7	0.1631	1	1	0.1631	1	8
		0.1145	100	0.119	20	0.129	20	0.143	1	8	8	0.1410	5	5	0.1410	5	8
		0.0995	100	0.112	4	0.124	60	0.134	13	9	9	0.1271	1	1	0.1271	1	8
				0.105	1	0.119	90	0.130	1	10	10	0.11535	9	9	0.11535	9	100
				0.102	2	0.112	80	0.127	8	11	11	0.0999	7	7	0.0999	7	100
				0.097	5	0.108	90	0.117	7	12	12	0.0986	1	1	0.0986	1	14
				0.088	1	0.105	80	0.108	20	13	13						8
				0.083	2	0.103	90	0.106	5	14	14						8
								0.095	2	15	15						100
								0.092	2	16	16						100
								0.085	1	17	17						14
								0.078	1	18	18						14
										19	19						14
										20	20						14

Table 4

## Semiquantitative analysis of silicate spherules from Súľ'ová Granite

Elements %	1	2	3
Al	43.52	7.99	1.27
Fe	23.20	34.16	52.36
Si	23.29	34.29	10.95
Ti	8.56	6.72	5.27
Mn	—	0.80	0.29
Ca	0.79	9.70	25.09
K	0.49	0.34	0.19
Mg	—	6.00	0.34
P	—	—	4.24
Σ	99.76	100.00	100.00

Semiquantitative analyses carried out on an energy dispersion analytic unit attached to REM-JEOL-JSM-35 CF (Department of Material Science, Metallurgical Faculty, Institute of Technology, Košice).

as is shown by Tab. 4. The high Ti content is very surprising in this "silicate" composition.

The group of black and brown-black spherules as a whole has an unclear chemical composition because their semiquantitative analysis does not establish the presence of oxygen and carbon. Therefore in the case of the iron spherules we do not know if they consist of magnetite or wüstite or native iron. The lustrous surface of black-blue spherules with some crystalline structure on the surface (Fig. 14) resembles magnetite spherules rounded in placers, alluvium or beach. Some ferruginous spherules, however, have a very regular surface — either without regular texture manifestations (Fig. 16) or represented by polygonal disintegration of the spherule surface similar to the surface of a tortoise shell or football ball (Fig. 17).

Similarly, also in the case of titanium-rich spherules (Fig. 18) we may only suppose that they consist of ilmenorutile (?).

The chemical composition of the spherules rich in Al (Fig. 20) or Al-Fe-Si-Ti and/or Fe-Si-Ca-Al-Ti-Mg or Fe-Ca-Ti P-Al (Fig. 19) can be expressed even less exactly.

The latter might have been contaminated by remnants of other minerals on their surface. It is worth nothing that the Al and Al-Fe-Si-Ti spherules (Figs. 19, 20) are the biggest whereas those rich in Ti and Fe (Figs. 16, 17, 18) are the smallest.

#### *Occurrences of spherical bodies in the world and their genetic interpretation*

Spherules are abundant on the Earth surface as well as on the Moon and are dealt with in very extensive literature. Spherules found on the Earth usually regarded as extraterrestrial, meteoric-intrastellar dust fallen onto the Earth surface. The meteoric origin of these spherules is proved by their relatively abundant occurrence on the surface of polar icesheets,

meteorological balloons and in deep sea clays (Yabuki, 1972; Parkin et al., 1977; Bownlee, 1979; Herr et al., 1980; Lozoyaya, 1981). Spherules formed by melting of meteorites in the course of their fall through the atmosphere or impact spherules formed by the impact of large meteorites can be considered as meteoric, too. The latter might contain also some material of terrestrial origin. The composition of the meteoric spherules is very variable. They are metallic (Fe, Fe-Ni), spinelid (magnetite, chromite, wüstite and others), glassy (Si-Ca-Fe-Mg-Al) and even graphite spherules. Spherules were found also in volcanic rocks (Filimonova et al., 1991; Filimonova, 1982). These authors discovered tiny spherical, egg-shaped and platy bodies having dimensions of up to 0.2 mm amidst shoshonite andesites and potassium rhyolites of the Southern Sikhote-Alin Belt. The authors assume a genetic relationship between the metallic spherules and volcanogene hydrothermal accumulations of oxides and sulphides.

They regard the presence of the spherules in the volcanic rocks as an indication that the magma was ore-bearing — i.e. a prospecting manifestation.

The origin of the terrestrial spherules is derived from the separation of two immiscible melts under subcrustal conditions. This assumption is supported by the results of experiments (Manakov—Sharapov, 1983). According to Ashikhmina et al. (1987) the spherical glassy state of the matter is a common natural state of magma between melt and crystals — between the liquid and the solid states.

Ferrosilicium alloys have been found on the Earth not only in meteorites but also in sedimentary rocks along with missanite (Novosyolova—Sokhor, 1983). Novgorodova et al. (1983) identified  $\text{Fe}_3\text{Si}$  (suessit) that occurred in the cavities of andesite-basalt porphyres. According to the above authors the formation of  $\text{Fe}_3\text{Si}$  was accompanied by the formation of titanium carbide and both were created in a strongly reducing environment.

Artificial balls represent the third group of the spherules. These originate during forest fires and mainly by the combustion of coal. Their composition and overall character are the same as those in the two preceding genetic groups. That is why it is difficult to distinguish extraterrestrial, terrestrial and artificial spherules from each other.

In Czechoslovakia, spherical bodies were studied by Čílek (1985), Čílek et al. (1983) who, among other things, described magnetite and silicite balls from sediments as well as crystalline of the Bohemian Massif. Veselský et al. (1983) described spherical wüstite (?) from West Carpathian granites. Likewise on the world-wide scale, also the genetical interpretation of the spherules from the Gemeric Granite is problematic.

Discussion about the character and origin of spherules in the Gemeric Granites

The most abundant — gray spherules of the Gemeric Granites most probably consist of a ferrosilicium material which is indicated by chemical analyses but mainly by X-ray analysis. The formation of our spherules from the phase  $\text{FeSi}$  and/or  $\text{Fe}_2\text{Si}$  would require heating of the  $\text{FeSi}$  melt to a temperature of at least  $1200^\circ\text{C}$  and its rapid cooling by water. Also an abrupt decrease in pressure is necessary. In nature such PT conditions may best arise by the impact of

extraterrestrial bodies (and/or impacts) onto the Earth surface into water. This is indicated also by frequent occurrence of droplike shapes of our gray spherules (Figs. 7, 8, 9). The other: ferruginous, titanium, aluminium and silicate spherules can hardly be explained in this way. The black spherules rather resemble placer minerals. Nevertheless, they might have also originated from a stellar dust fallen onto the Earth surface and then the spherules might have got, along with the impact (?) ferrosilicium ones, from sediments to the melt of the Gemic Granite where they remained preserved. Meteoric Fe-spherules, however, typically contain Fe-Ni phase, which is absent in ours. On the contrary our spherules contain a distinct Fe-Ti-Mn-Cr association.

Surprisingly, no traces of Sn-W-Mo-Nb-Ta have been found in the spherules, although the Gemic Granites are a potential carrier of these metals. All these facts support the version that the spherules came into the magma of the Gemic Granites from alluvial or beach sediments.

Because of the preliminary character of the results presented and the difficulties in distinguishing the spherules formed in various ways, we cannot exclude also the origin from the initial stages of the formation of the Gemic Granite magma.

Ashikhmina et al. (1987) thoroughly investigated lunar as well as terrestrial glassy balls and came to the conclusion that natural glasses formed when the chemical balance of the magmatic liquid phase is disturbed. It is a protocrystalline state with unfinished level of crystallinity. High-dispersion colloidal aggregation takes place by which multivalent forms of the elements  $Fe^0$ ,  $Ti^0$ ,  $Si^0$  are formed. This state resembles the physico-chemical character of our spherules.

Regardless of their meteoric or terrestrial origin, the spherules found in the Gemic Granites are undoubtedly important from the petrological point of view.

By means of geochronological methods it has been determined that the granite near Betliar is younger than the Hnilec and Súľová Granites. However, the occurrence of spherules found at the localities Betliar, Hnilec and Súľová suggest that the bodies of these three massifs were formed from the same magma, more or less simultaneously.

The goal of this research is to prove the common origin of Gemic Granites resulted also from previous work about accessory minerals (K. Jakabská, 1994).

Acknowledgements: The authors thank Doc. RNDr. V. Bouška, DrSc. and Dr. A. Ždimera of the Faculty of Natural Sciences, Charles University, Prague for aid provided during their research. We are obliged to RNDr. J. Krištin, Csc. of the Laboratory of Electron Microanalysis, Geological Institute of Dionýz Štúr, Bratislava and to employees of the Department of the Material Science, Metallurgical Faculty, Institute of Technology, Košice for analyses. Also thanks Dr. J. Derco, Csc. of ATNS Košice for X-ray analyses.

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Fig. 1. Schematic map of Gemeric Granite occurrences.

Explanations: 1—granite outcrops, the most important localities are marked; 2—geophysically determined isolines of granites extent at depth (Šefara — Plančár et al. 1978 — simplified and supplemented by L. Rozložník, 1987).

- Fig. 2. Section of a gray spherule in Betliar Granite, in the central part nicols II, magn. 66x
- Fig. 3. Spherical shape of a gray spherule, Betliar Granite, magn. 420x
- Fig. 4. Potato-shaped gray spherule with conical protuberances, Hnilec Granite, magn. 360x
- Fig. 5. Gray spherule intergrowth, Hnilec Granite, magn. 360x
- Fig. 6. Gray spherule intergrowth, Betliar Granite, magn. 300x
- Fig. 7. Gray pear-shaped spherule.

Explanations: Depressions probably represent traces after broken off protuberances or traces after contacts with other spherules, Betliar Granite, magn. 420x

- Fig. 8. Gray pear-shaped spherule with “branches” and traces after broken off protuberances or contacts with other spherules.

Explanations: a) Hnilec Granite, magn. 360x  
b) Súľ'ová Granite, magn. 600x

- Fig. 9. Irregular gray spherule, Betliar Granite, magn. 240x
- Fig. 10. Polygonally disintegrating crust of a gray spherule, Betliar Granite, magn. 2100x
- Fig. 11. Cross-sections of black spherules in a thin-section, Hnilec Granite, magn. 28x

- fig. 12. Black ferruginous spherule, Hnilec Granite, magn. 780x  
Fig. 13. Black ferruginous spherule, Hnilec Granite, magn. 1100x  
Fig. 14. Surface of a black ferruginous spherule with a visible crystalline texture, Hnilec Granite, magn. 1800x  
Fig. 15a. Cross-section of a gray spherule from Hnilec Granite. magn. 1100x  
Fig. 15b. Areal Fe, Si, Mn and Cr distributions in a gray spherule from Hnilec Granite  
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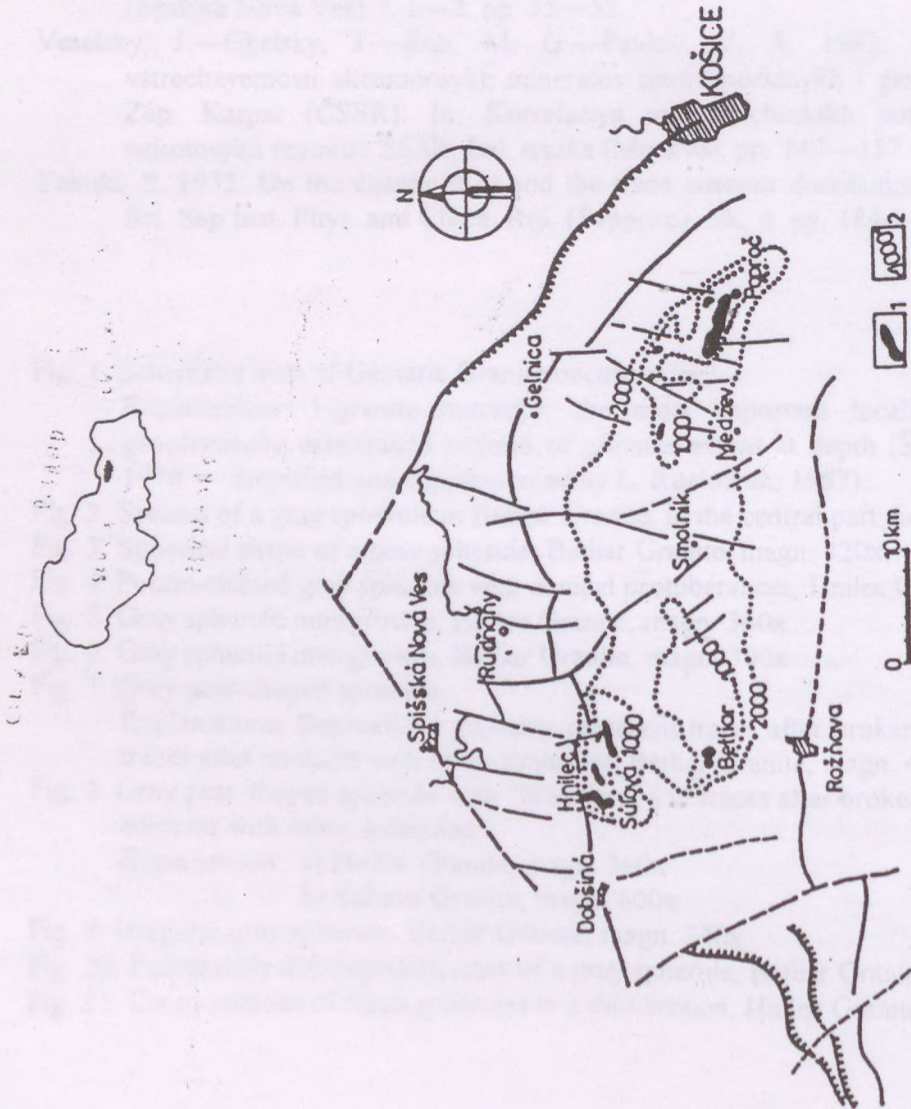




Fig. 2

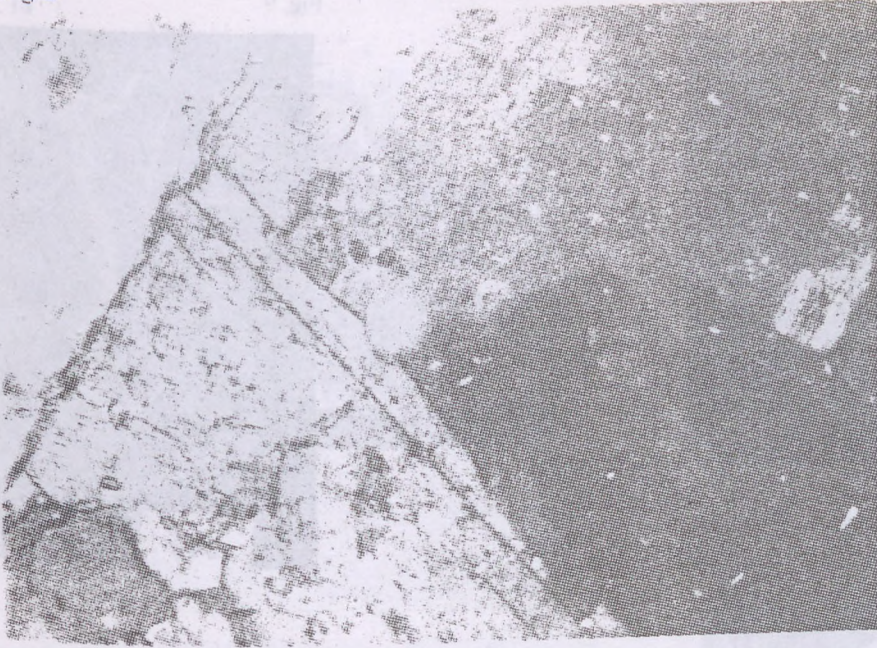


Fig. 3

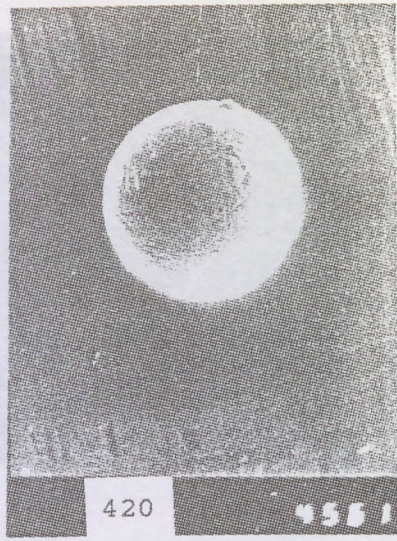


Fig. 4



Fig. 5



Fig. 6



Fig. 7



Fig. 8a



Fig. 8b



Fig. 9



Fig. 10

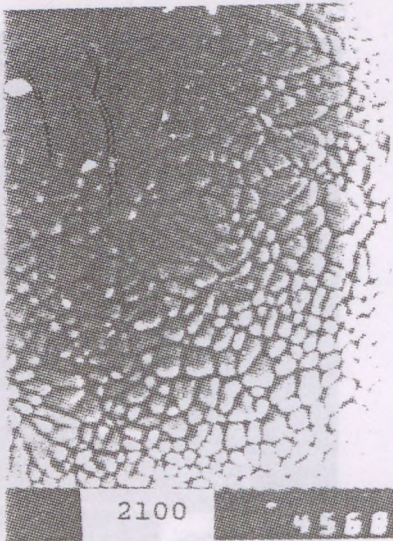


Fig. 11

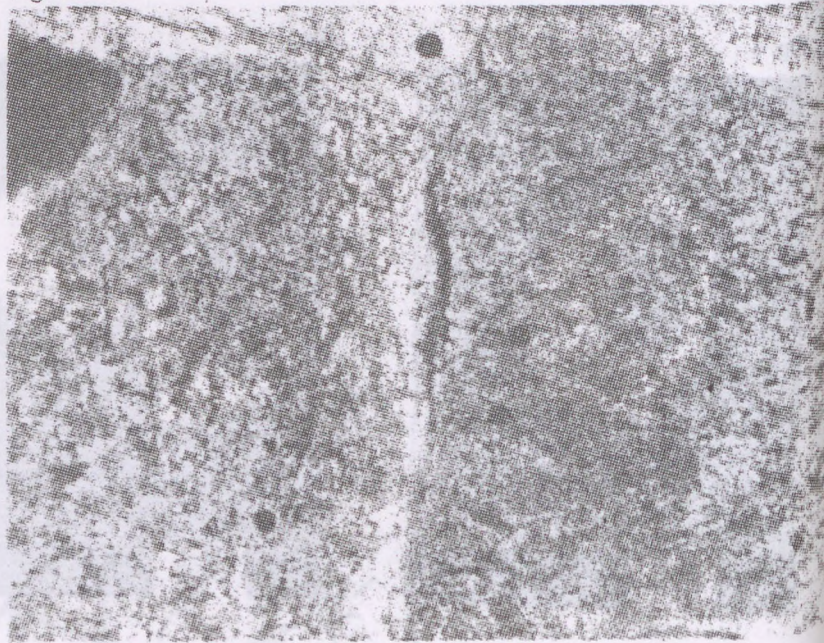


Fig. 12

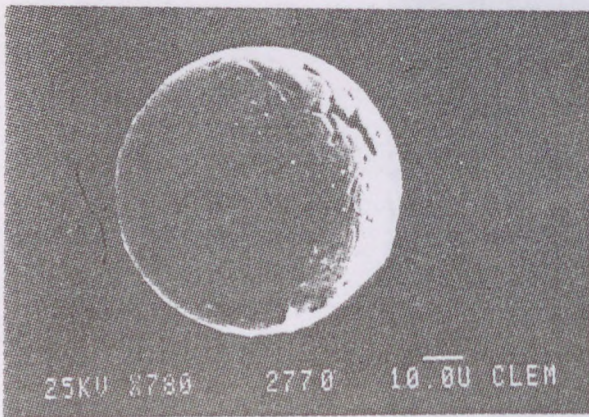


Fig. 13

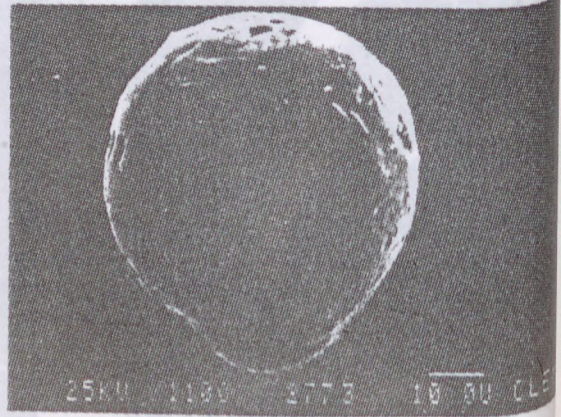


Fig. 14



Fig. 15a

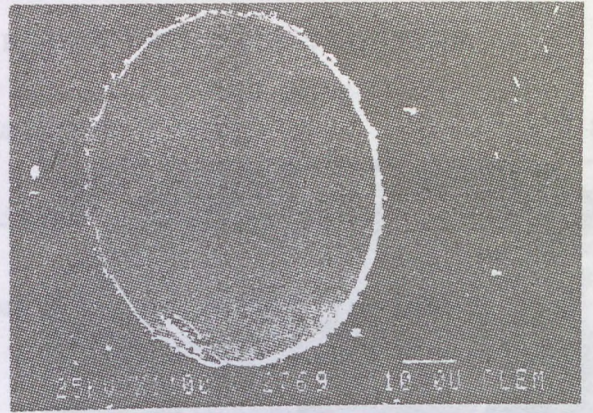


Fig. 15b

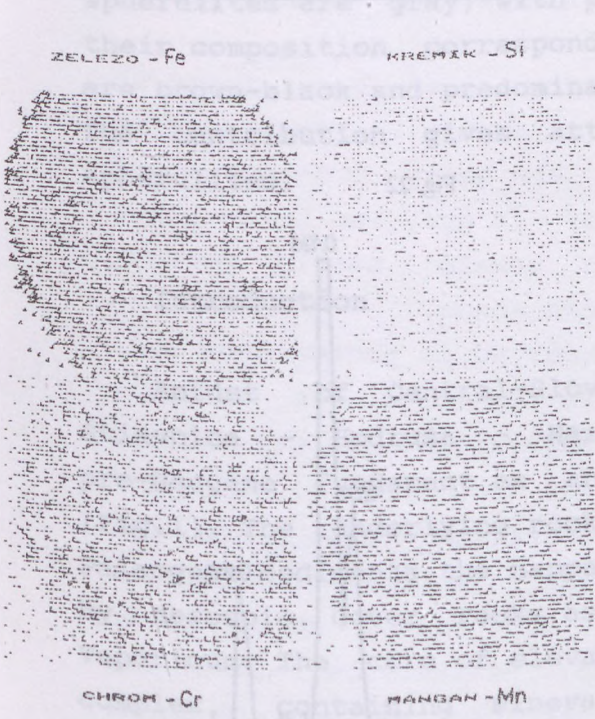


Fig. 16

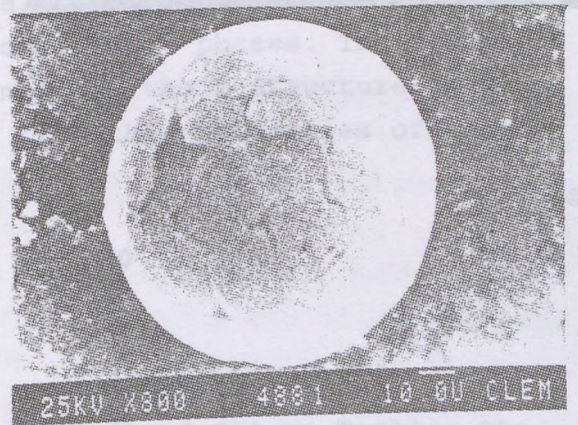


Fig. 17

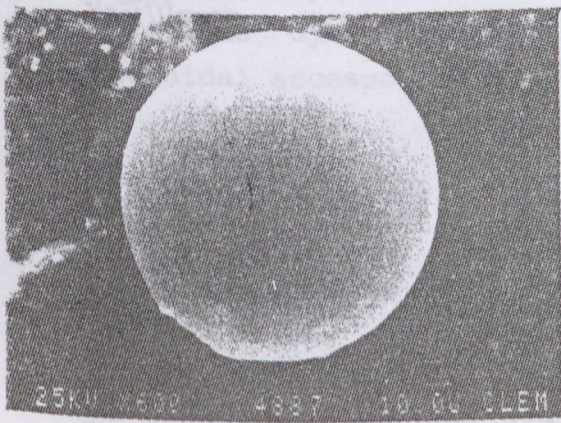


Fig. 18



Fig. 19

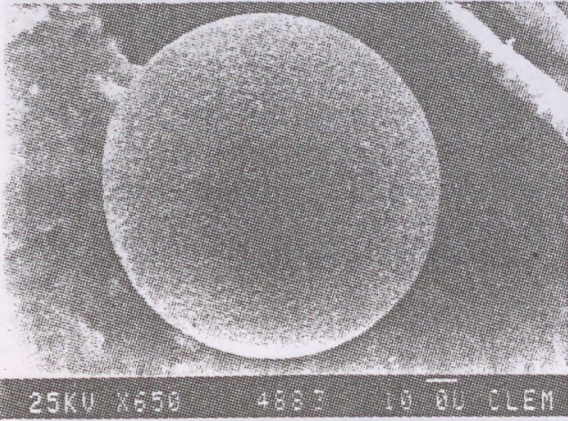


Fig. 20

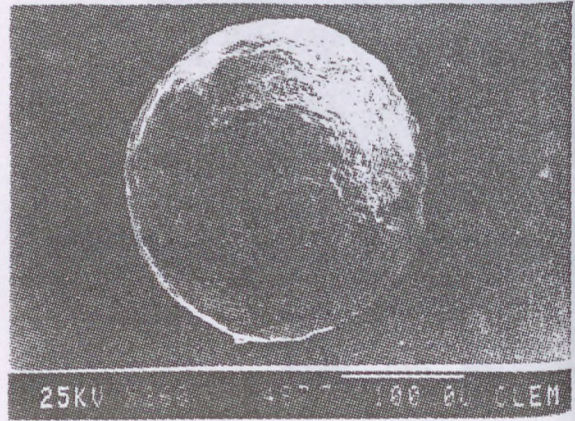


Fig. 21

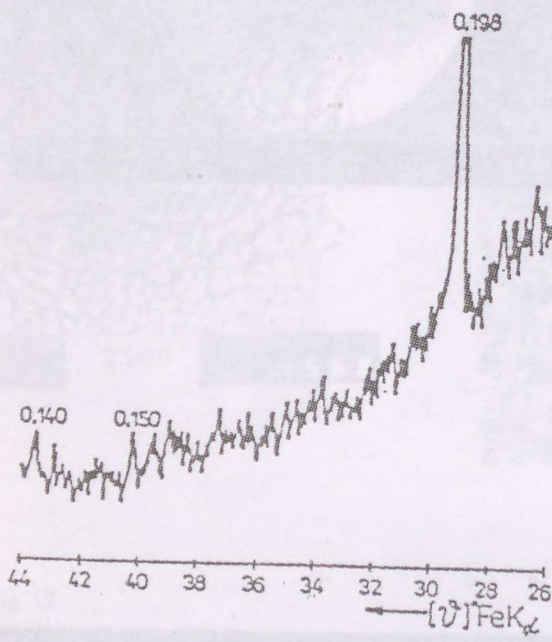
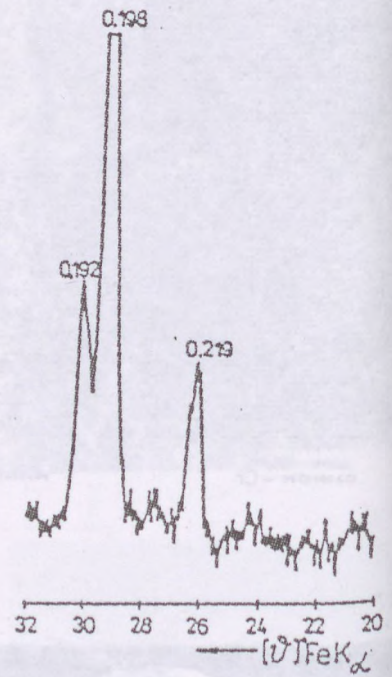


Fig. 22



## SPHERULITES IN RHYOLITE FROM BANSKÁ ŠTIAVNICA TERRITORY

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Spherulites of various size, colour and composition have been found during separation of zircons from the Banská Štiavnica rhyolite in the heavy fraction sized 0.12 to 0.09 mm. Most of spherulites are gray, with polygonal surface disintegration and their composition corresponds to some Fe-Si phases. The others are brown-black and predominant element of their structure is Fe. The contribution gives attention to the properties of these spherulites.

### Introduction

Amidst of Central-Slovakian neovolcanites among Banská Štiavnica - Hodruša - Hámre - Vyhne and Sklené Teplice the pre-Neogene fundament as an elevation emerges on the surface (Fig.1). The underlying rocks of Neogene are represented by the Paleogene sediments the nappe units of Mesozoic and Paleogene and by Mesozoic. Cover rocks as well as by crystalline complex of Veporicum. The core of elevation is formed by Hodruša intrusive complex, containing mineralized structures of the Banská Štiavnica - Hodruša Ore District become famous by the exploitation of precious and non-ferrous metals. The separation of accessory minerals from rhyolite of Banská Štiavnica - Klotilda dyke resulted in the finding of extraordinary spheroidal accessories - spherulites.

### Granite porphyry - rhyolite

In the Hodruša intrusive complex there is possible to observe the dykes of light - colour rocks such as aplite, granite porphyry and rhyolite. These rocks do not probably derive from the same magma and they are also different in geological age.

The rock which from the accessory minerals were investigated comes from the Klotilda dyke with thickness about 7 m. This dyke was discovered by workings. Its veinstuff is whitish with the visible porphyric phenocrysts of feldspate and sanidine. Matrix expressively predominates above the phenocrysts (ratio is 70:30) and it is also strongly hydrothermal altered. The rock affected especially by calcification, sericitization, adularization and silicification. Some rocks contains ore minerals above all the sulphides, which are macroscopic visible partially.

### Morphological, physical and chemical properties of spherulites

Spheroidal accessories - spherulites have been found in rhyolite of Klotilda dyke from Banská Štiavnica territory during the accessory minerals separation in the heavy fraction sized 0.12 to 0.09 mm. They occur mostly in gray and brown-black colour. The brown-black spherulites create nearly perfect spheres, while the gray ones have various shape.

Gray spherulites - as it can be observed in photographs made by means of electron microscope - are spherical but on their surface they have often fine conical elevations or depressions. The shape of spherulites can be pear-like, too (Fig. 4). Surface gray spherulites have polygonal disintegration (Fig. 3 and 5). This fact was observed at several thousandfold magnification. The polygonal disintegration is typical for rashly cooled down substances which decrease their volume, for instance: meteorites or alloys. Under binocular microscope the surface of above mentioned substances is lustrous.

Brown-black small balls create mainly regular spherical shapes (Fig. 6) and on the surface there were founded fine tumor-like elevations and depressions. The structure of spherulite surface testifies to their crystal one (Fig. 7).

Due to small dimensions of spherulites attained only several hundredth parts of millimetre and their small amount the determination of substance composition is very difficult. Qualitative analyses were realized with the application of energy-dispersion unit connected to REM-JEOL-JSM-35 CF (Metal Theory Department of Metallurgical Faculty TU Košice). On the basis of qualitative analyses obtained, the results point to the

fact that gray spherulites are composed from Fe and Si (Fig. 8), and brown-black ones are created by Fe (fig.9). Spherulites of similar composition were described in granites from Spis-Gemerian Mts. (in Jakabská - Rozložník, 1989).

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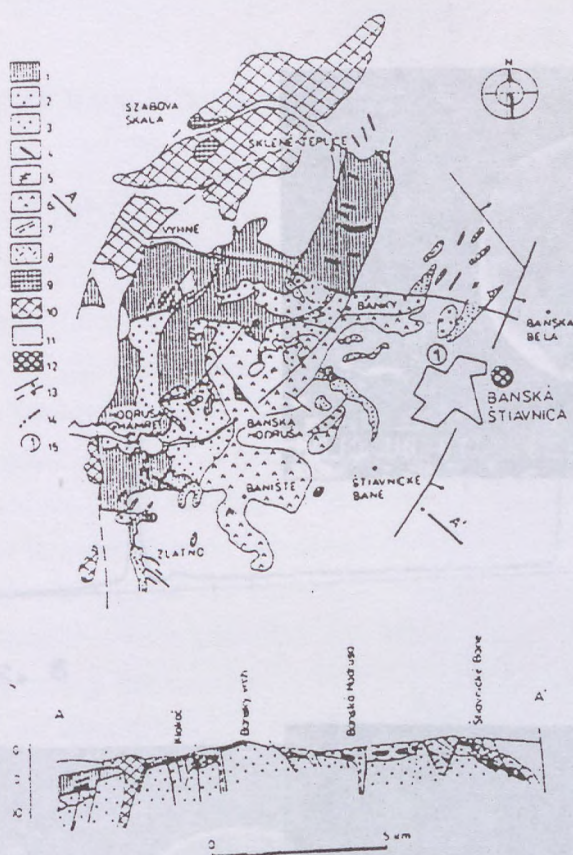


Fig.1. Schematic geological map of the Banská Štiavnica - Hodruša surroundings (compiled by Rozložník, 1988). 1- rocks of pre-Neogene fundament, 2- diorite-gabrodiorite, 3- granodiorite, 4- granodiorite porphyry, 5- skarns (only in cross section), 6- quartz diorite porphyry (sills and localities), 7- quartz diorite porphyry (dykes), 8- quartz diorite porphyry (thick dykes), 2-8- the Hodruša intrusive complex (Neogene), 9- rhyolites of the Hliník type (Sarmatian), 10- rhyolites of the Kremnica type (Sarmatian), 11- andesites - undivided (Badenian - Sarmatian), 12- basanite (Pannonian), 13- faults, 14- ore veins (only in the cross section), 15- number of sample: (1) - the Klotilda granite - porphyry - rhyolite dyke.

DESCRIPTION OF FIGURES

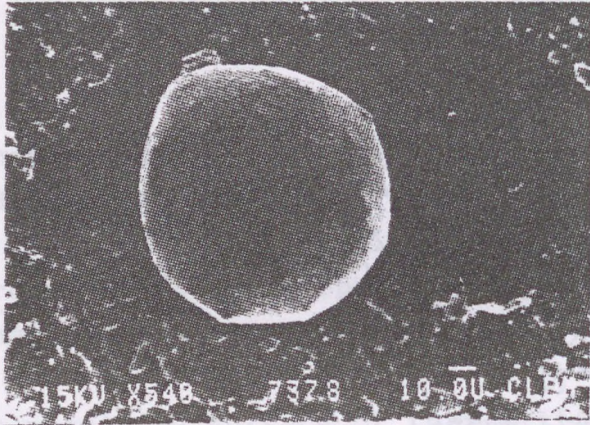


Fig. 2

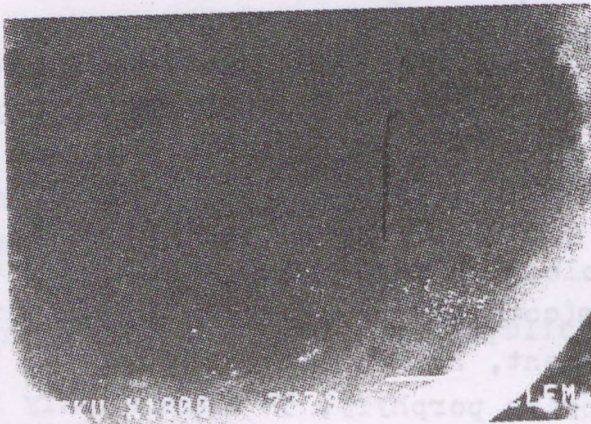


Fig. 3

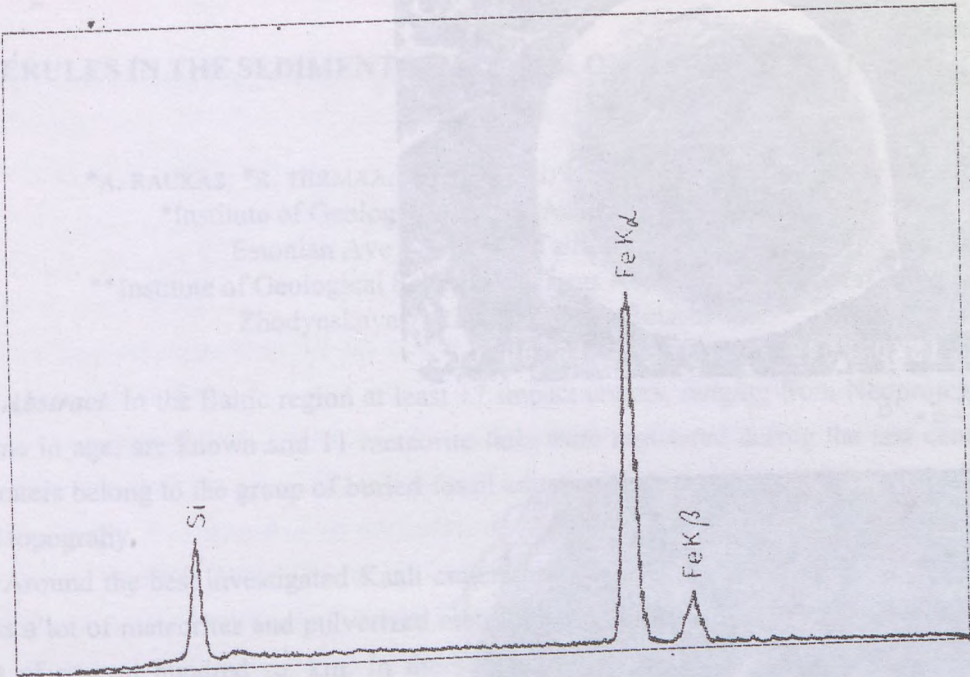


Fig. 8

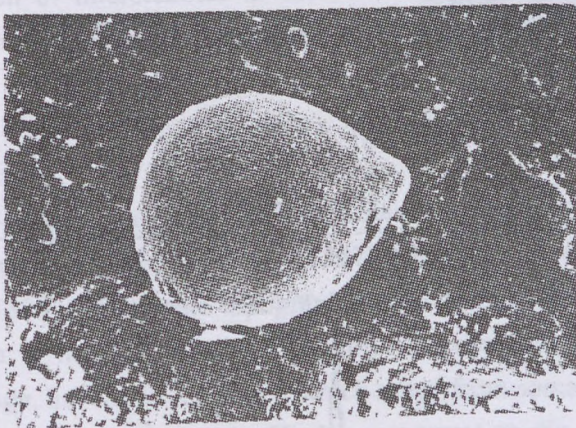


Fig. 4

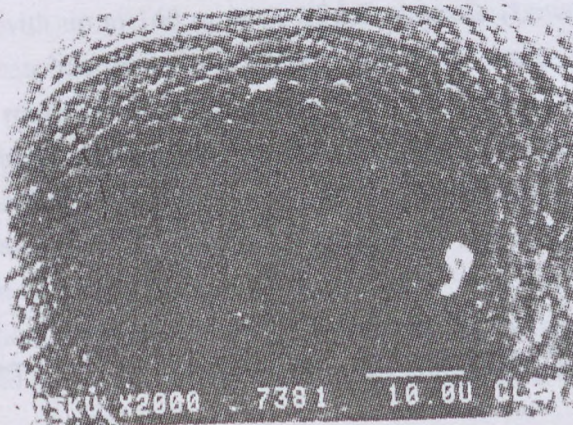


Fig. 5

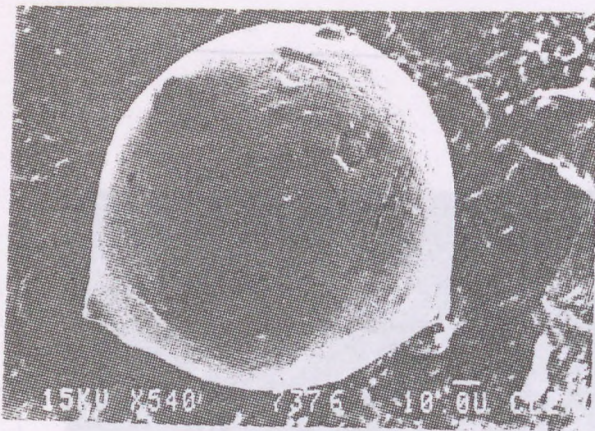


Fig. 6



Fig. 7

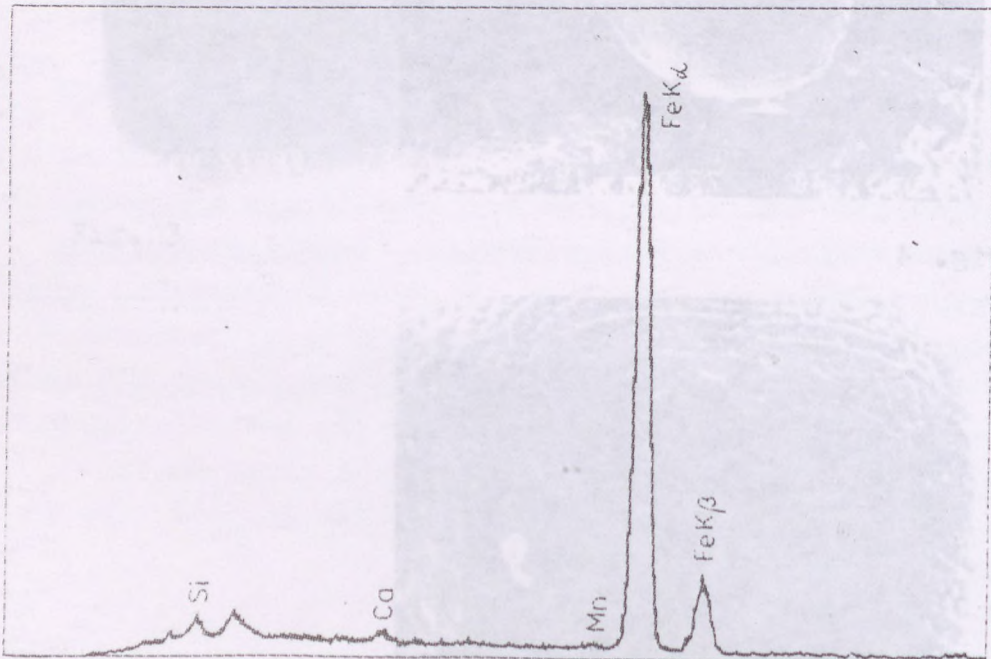


Fig. 9

# SPHERULES IN THE SEDIMENTARY COVER OF THE BALTIC STATES AND BELARUS

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**Abstract.** In the Baltic region at least 17 impact craters, ranging from Neoproterozoic to Holocene in age, are known and 11 meteorite falls were registered during the last centuries. Most craters belong to the group of buried fossil craters which are only partially visible in the present topography.

Around the best investigated Kaali craters on Saaremaa Island, West Estonia, the soil contains a lot of meteorites and pulverized meteoritic matter, established in small amounts in an area of several hundred sq. km. In the surrounding peat-bogs they are concentrated in certain stratigraphic levels. This allows to date the impact events very precisely and to find chronostratigraphical markers for the correlation of sedimentary sequences. Our preliminary investigation of microtektites in the Piila bog near Kaali shows that the Kaali meteorite explosion occurred around 7500 years ago — much earlier than it has been believed so far.

## Introduction

The impact craters in the Baltic States have attracted scientists since the first half of the past century. J. W. von Luce (1827) presented the first description of the Kaali main crater. In 1839, a foot-scale topographic map of Lake Kaali was compiled by F. K. Kruse (1842). Up to now, in the Baltic region 17 impact craters (Fig. 1) of Neoproterozoic to Holocene age, have been registered which range from some metres to more than 50 kilometres in diameter (Puura et al., 1994). The biggest crater on the area of Baltic States is the Vepriai crater in central Lithuania, which is 8 km in diameter and is situated in Palaeozoic sediments. It is filled with up to 160 m thick Middle Jurassic (Lower Callovian) lacustrine sediments, which indicate the age of the impact event. The crater is covered by 70 to 100 m thick Cretaceous rocks and Quaternary sediments.

Mizarai, another giant impact crater (5 km in diameter and up to 250–300 m deep), is located in southern Lithuania. The age of the crater is estimated according to acritarchs as Vendian–Cambrian. The covering sediments of Triassic, Cretaceous and Quaternary age are about 300 m thick.

A giant meteorite crater of Kärđla, on the Island of Hiiumaa (Estonia), 4 km in diameter and 400 m in depth, was formed approximately 455 Ma ago at the beginning of

Idavere time in the middle Ordovician and is buried under Ordovician sediments (Puura, Suuroja, 1992).

Recently, an impact structure, 3–4 km in diameter, was discovered at Dobeles, southern Latvia. For the time being very little is known about this crater.

At least three groups of impact craters of Holocene age (Kaali, Ilumetsa and Tsôõrikmäe) occur in Estonia (Fig. 1).

The first scientifically known meteorite fall in the Baltic States occurred on July 12, 1820 in the Lixna area in Latvia. A year later, on July 4, 1821 a stony meteorite of the size of a man's head fell to the earth near the village Kaiavere in Estonia (Fig. 1). On May 11, 1855, a stony meteorite shower occurred near the village of Kaanda in the north-western part of the Island of Saaremaa, referred to as Oesel in the Literature on meteoritics. A few days later, on May 17, the Iigaste meteorite fell in southern Estonia. In 1960, J. A. O'Keefe from the USA classified the Iigaste meteorite as a tektite. The Buschoff meteorite in Latvia fell on June 2, 1863 and on August 8, 1863 a stony meteorite shower occurred at Pilistvere in central Estonia. A year later, on April 12, a stony meteorite fell at Tännasilma, not far from Pilistvere (Fig. 1).

In Lithuania, the Jodzies meteorite fell on June 17, 1877. On February 9, 1929 a meteorite shower occurred near Padvarninkai and on February 2, 1933 the Zemaitkiemis meteorite fell to the earth (Fig. 1). They all belong to the group of stony meteorites.

In terms of the frequency of recent meteorite falls, Estonia seems to hold one of the first places in the world. The number of meteorite craters per one sq km is also high here than in neighbouring areas. It is because of awareness of the people in Estonia, due to the influence of Tartu University and the activity of some German estate owners — amateur naturalists. As a result, many fragments of meteorites, carefully preserved by individual collectors, found their way to the scientific institutions. The main part of the collection is curated at the Institute of Geology of the Estonian Academy of Sciences in Tallinn. About 100 specimens of this collection from all over the world are exhibited in the Museum of Geology of the Tartu University in Tartu.

Meteorites constitute only an insignificant part of the extraterrestrial material that reaches the Earth. Most of it is in the form of extraterrestrial dust (Krinov, 1976). The detailed investigations of Estonian Lower Paleozoic sequences allow us to assume that throughout geological time extraterrestrial matter reached the Earth in a pulsatory manner. After several of low rate accumulation, there were shorter periods of some years with high rate accumulation of cosmic matter (Viiding, 1984).

This paper is focused on the traces of the impact activity throughout the Pleistocene. The most complete Quaternary sequence of Belarus, several hundred metres thick were studied. Earlier (Shimanovich et al., 1993), we had studied the meteoritic matter in the surroundings of the Kaali meteorite craters with an aim of contributing to the knowledge of

extraterrestrial ferrous spherules in order to facilitate their distinction from spherules of terrestrial origin.

### Short description of sediments and spherules of the study area

#### 1. Spherules in the bedrock.

The oldest palaeometeoritic matter known in the Baltic States and Belarus so far was recovered in the Viru-Roela borehole in the central part of Estonia. It was enclosed in a Lower-Cambrian sandstone at a depth of 320 metres (Viiding, 1965). It consists of magnetite — iocite globules (diameter 20–450  $\mu$ ) or shell-like fragments (diameter up to 2–3 mm). The Ni content is below 1%. Spherules are concentrated in a layer of 2.6 m thick sand covered by clay and resting on Proterozoic migmatite granites. The strikingly high average content of magnetite globules (0.2%) and the character of spherules warrant the supposition that a giant meteor fell in this area some 570 million years ago and the products of its pulverization were deposited in the tectonically subsiding littoral sea over vast areas. According to H. Viiding (1965), every square metre in the area of the meteor fall received more than 5 kg of meteoritic matter; this encourages us to use this layer for correlational purposes.

Detailed mineralogical investigations have shown that there is more or less clear cyclicity in the accumulation of extraterrestrial spherules and they occur in different amounts everywhere (Viiding, 1984).

#### 2. Spherules in the Kaali area.

Eight meteorite craters in an area of one square kilometre occur on the Island of Saaremaa, Estonia. The initial mass of the Kaali meteorite must have been a few thousand tons. The soil both in the craters and around them contains a lot of micrometeorites and pulverized meteoritic matter. Rather high concentration of spherules has been established in the area of several hundred sq. km, mainly to the north-east of the craters, showing the projection of the supposed fall trajectory (Tiirmaa, 1994).

Various methods have been used to determine the age of the Kaali craters. Most of recent researchers maintain that the maximum age of the craters is about 4000–5000 years (Tiirmaa, 1994). Our preliminary investigation of microtektites in the Piila bog near Kaali place the age of the crater at around 7500 years ago.

According to chemical analysis, in the fragments of meteorite Fe and Ni made up 91.5% and 8.3%, respectively. The mineralogical analysis has shown that beside schreibersite, kamacite and taenite, also goethite occurs in the iron crust of the meteorite, forming there colloform textures. The iron-crust, which was formed in terrestrial conditions, mainly on account of kamacite and taenite, sometimes contains also pseudomorphoses after schreibersite. The Kaali meteorite belongs to the class of coarse octahedrites also troilite and olivine, and relying on mineralogical and chemical analyses, surmised that this meteorite

belongs to the olivine-bronzite chondrite class. This opinion is sufficiently well-founded (Yavnel, 1976). The contents of Ni, Ga, Ge and Ir indicate that the Kaali meteorite belongs to the IA group and is very similar to the well-known Odessa iron meteorite from the USA.

A. Aaloe ja R. Tiirmaa (1981) have identified in the Kaali area the following types of extraterrestrial matter (with some our modifications):

1. Meteoritic iron (micrometeorites and their microfragments);

2. Meteoritic dust

a) magnetite globules formed on the melting of meteoritic matter during the impact, or from condensed metal vapour;

b) magnetite platelets formed from heated kamacite along the cracked surfaces;

3. Microtektites (impactites)

a) magnetite-silicate tektites, formed by mixing of terrestrial silicate matter with the meteoritic iron that melted during the explosion. This type includes spherical or irregular globules and crusts;

b) silicate globules and irregular porous microtektites, formed from molten or vaporized silicate minerals during the explosion.

The same types of cosmic matter were studied later means of scanning electron microscope and microprobe equipment (Shimanovics et al., 1993). The information obtained could essentially contribute to distinguishing extraterrestrial material from magnetite particles of terrestrial origin (Industrial dust). It would also promote the research of the thick Quaternary cover of Belarus.

3. Ferriferous minerals in the Quaternary deposits of Belarus.

The area under consideration has a complicated geological setting and history, described in many papers. Both allothigenic and authigenic minerals have been encountered in the Quaternary cover of Belarus.

A large variety of iron-bearing minerals occur in lake and bog deposits and in the flood-plain facies of alluvium. More than 3000 photos of magnetite, pyrite and siderite grains have revealed a high variability in grain morphology. Most particles have undergone phase transformation in the zone of hypergenesis. This hampers their genetical interpretation.

The investigated iron-bearing minerals can be subdivided into four main groups: 1) crystals; 2) microconcretions, irregular crystalline aggregates and growths with have developed on plant and animal remains; 4) spherical, (globe-shaped) grains (Shimanovich et al., 1995) Attention was focused on the fourth group. The surface of the grains is usually smooth and often consists of microblocks which account for the scaly pattern of the surface. As a result of weathering, caverns and fissures, occasionally in the form of growth have developed. Morphologically and genetically, this is a complicated group which associates formations similar in outlook but different in genesis: microbiological, hypergene, technogenic and cosmogenic.



In terms of their shape and dimensions, the globules of microbiological origin resemble the so-called framboids of sulphides in coal-bearing deposits of different age, which have been formed as a result of oxidation and substitution of initial colloidal drops or coacervates. It is not excluded that part of the spherules consists of impregnated bacteria.

When together with the newly-formed aqueous ferric oxide also ferrous ions occur hypergenic ferriferous minerals will form. The slower the rate of oxidation, the more complex and effective is the process of crystallization. Occasionally, the formation of magnetite is preceded by metastable phases of "green rust" which may arise either during the slow oxidation of ferrous oxide (solide phase) or precipitation of the corresponding carrier compounds the further transformation of which is controlled by the particular environmental conditions.

Technogenic globules formed by industrial emissions are common in surface sediments. It seems that the formation and development of  $Fe_3O_4$  takes place when ions of  $Fe(OH)_3$  with a hydrocomplex of trivalent iron precipitate slowly within the thin layer outlining the embryonic nuclei. However, some globules containing Fe, Ni, Cr and Mn may correspond to the orbital debris originating from spacecraft or their engines (Zbik, Gostin, 1994).

Some globules have a specific Widmanstätten surface pattern indicative of their extraterrestrial origin. Unfortunately, the spherules of fine sand and silt size have often undergone corrosion and the specific features of extraterrestrial matter have been effaced. Our preliminary results have shown that extraterrestrial particles are rather rare in the Byelorussian sequence and clear stratigraphical markers of the pulverized matter are absent there. This hampers the establishment of cyclicity in the meteoritic activity.

Specific features of extraterrestrial spherules have partially been altered in active dynamic conditions, as for example, in river flows. It means that in the future much more attention should be paid to interglacial lake and bog sediments, where especially microtektites must have been well preserved.

#### 4. Main types of magnetite globules.

Magnetite globules both in the surroundings of the Kaali meteorite crater in Estonia and in the Quaternary deposits of Belarus vary in roundness, internal structure and microsculpture of the surface. The following varieties can be distinguished: spherical, rounded, elliptic, ovate-tubercular, oolitic or drop-like. Occasionally, strongly crusted forms are encountered (Photos 1 and 2). In terms of internal structure, the following forms can be distinguished:

— compact globules with metallic nucleus (Photo 1). Their uneven surfaces have small irregular kidney-shaped or drop-like projections. The nuclei vary in appearance. They may show Widmanstätten pattern (Photo 5) or be rod-like (elongated), resembling a prism structure;

—hypothetically rounded forms, the surfaces of which are covered with a pattern of regular moonlets resembling craters (Photo 6). Probably these have resulted from the burst of gas-filled vesicles inside melted meteoritic matter. Some spherules are extremely rich in vesicles (Photo 7).

Polyhedron decorated spherules (Photo 8) have most frequently octahedral, hexahedral, tetrahedral or trihedral plates, sometimes the spherules are covered with rather deep irregular hollows (Photo 9). Dendrite decorated (Photo 10) spherules have a regular rounded or a bit oval shape and a rough surface covered by needle-like magnetite dendrite crystals (Zbik, Gostin, 1994).

Occasionally, complicated conglomerate-like magnetite spherules (Photo 11 and 12) of unknown genesis can also be found.

### Conclusions

Research into ferriferous minerals has revealed a great diversity of forms due to the different facial and physico-chemical conditions of their formation. The morphotypes of grains and the character of changes on their surfaces are due to mechanical and chemical processes. Comparison of ferriferous minerals from different regions on the basis of their micromorphological indices has enabled us to elucidate similar features, particularly in magnetites, spherical in outline. One has to accentuate that if Widmanstätten structures are absent then it is difficult to differentiate extraterrestrial ferriferous spherules from those of some other origin. In that case microprobe studies are needed.

### Acknowledgement

This research was financially supported by the PACT-project "Environmental History of the Baltic Region". The authors thank Mr. Rein Vaher for producing the drawing and Mrs. Helle Kukk for improvements of the English text.

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### Captions

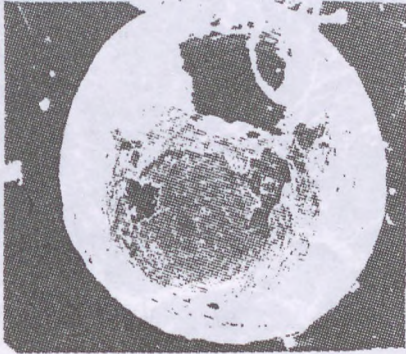
Fig. 1. Distribution of impact craters and meteorite falls in the Baltic States. I — Craters: E1 — Kaali; E2 — Kärddla; E3 — Ilumetsa; E4 — Tsõõrikmäe; Lal — Dobeles; L1 — Mizarai; L2 — Vepriai. II — Meteorite falls: 1 — Kaande (Oesel); 2 — Tännasilma; 3 — Pilistvere; 4 — Kaiavere; 5 — Iigaste; 6 — Nerft; 7 — Buschoff; 8 — Lixna; 9 — Padavarninkai; 10 — Jodzie; 11 — Zemaitkiemis.

Table I and II Varieties of spherules and their microtopography.

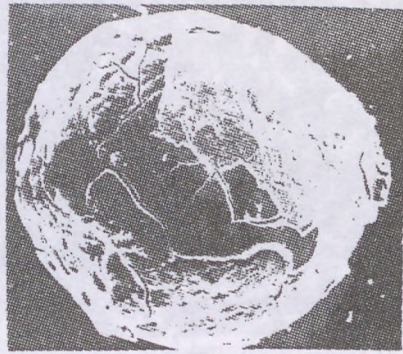
1. Rounded magnetite globule with weathered crust and metal core from the Kaali area, Estonia (94x).
2. A loss of conchoidal mantle from the Vedrich area, Belarus (440x).
4. Spherule from the Pinsk area, Belarus, showing lost polygonal fragments (540x).
5. Magnetite spherule from the Kaali area, Estonia. The metal core surface has a Widmanstätten structure (1800x).
6. Spherule from the Kaali area, Estonia. The pitted surface has been caused by collapse of vesicles (540x).
7. Vesicle-rich surface of a magnetite spherule from the Kaali area, Estonia (780x).
8. Polyhedron decorated spherule from the Kaali area, Estonia (860x).
9. Surface of the magnetite spherule from the Kaali area, Estonia, is covered with irregular holes (1000x).
10. Dendrite decorated spherula from the Pinsk area, Belarus (760x).
11. Conglomerate-like magnetite spherule from the Pinsk area, Belarus (360x).
12. The same spherule with 1300x blow up.



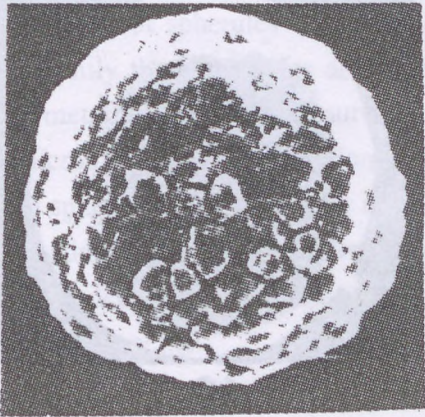
WITHIN THE BORDERS OF CYCLOGENIC RING STRUCTURES OF THE MOST DIVERSE SCALES ON THE TERRITORY OF RUSSIA AND KAZAKHSTAN



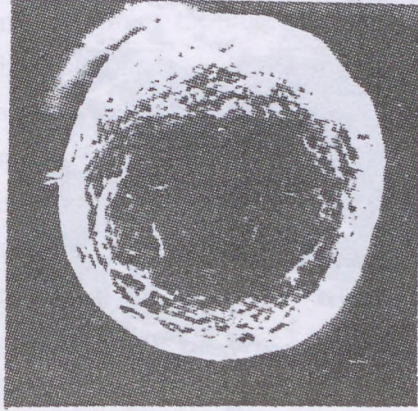
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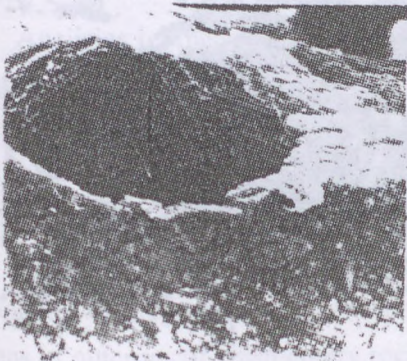
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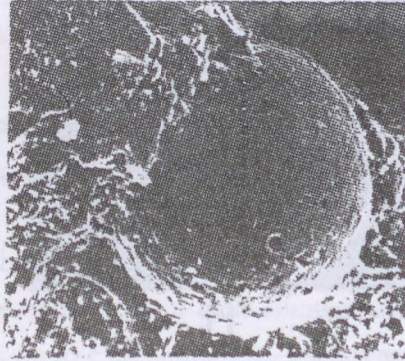
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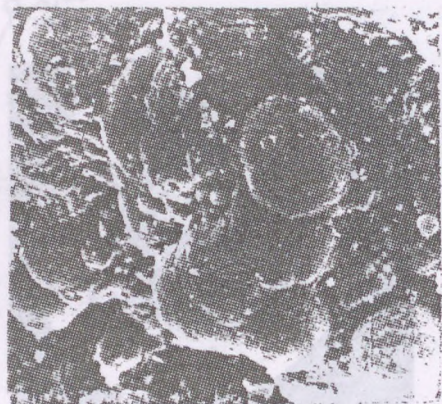
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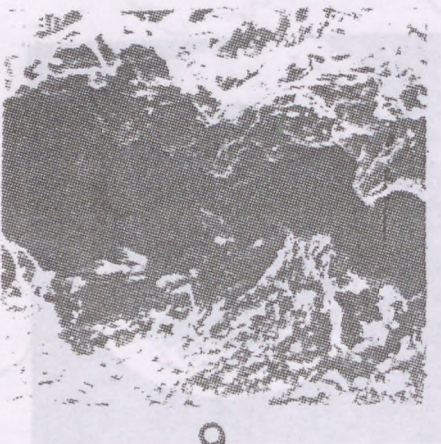
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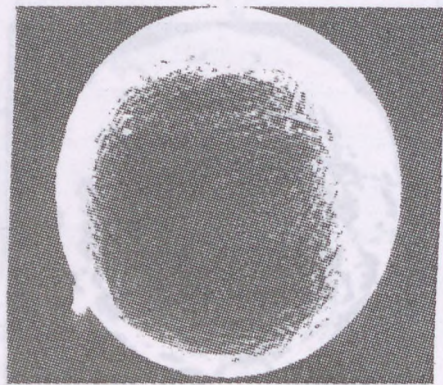
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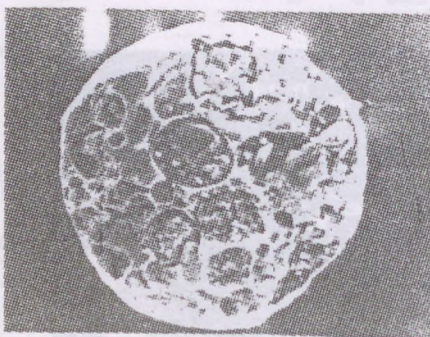
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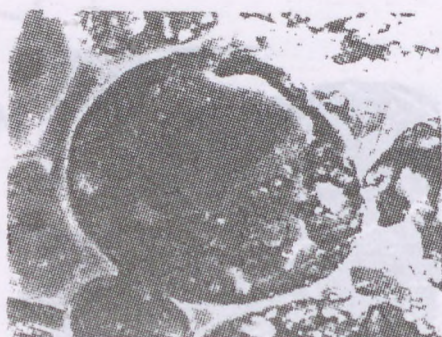
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10



11



12

FINLAND

BALTIC SEA

RUSSIA

POLAND

RUSSIA

BELARUS

100 km

## WITHIN THE BORDERS OF COSMOGENIC RING STRUCTURES OF THE MOST DIVERSE SCALES ON THE TERRITORY OF RUSSIA AND KAZAKHSTAN

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In the north-east of Russia there is a gigantic presumably impact Verchojansko-Kolimskaja ring structure, which has section 2100 x 1700 km. This structure was identified in 1976 (1). It is the biggest ring structure in the area of the former USSR. Within this structure in alluvial deposits of many streams between the rivers Kolima-Alazeja, Alazeja-Indigirka, Indigirka-Jana and Jana-Omoloi there are frequently magnetical and nonmagnetical spherules.

Halo of their dissipation has breadth 150–200 km and length about 1000 km. Halo has latitudly orientation. This spherules called janits, because maximum their spreading is alluvion of Jana river. Their amounts here reach  $100 \text{ g/m}^3$  of alluvion.

The size of spherules 0,01–0,05 mm, sometimes the biggest spherules go up to 0,2 mm. Frequently these spherules are zoned but sometimes — hollow. Zoned balls consist of forgeable metal kernels and frail outward cover.

Microdiffraction investigations by means of electronic microscope show that the kernels of spherules are made up by alfa-iron.

In addition with said above there are microcrystallites with sizes 50–100 angstrom which have definite orientation in matrica alfa-iron. These microcrystallites are simply identified as magnetite.

Shell of spherules which has thickness from  $1/3$  to  $1/2$  of their radius is made up mainly by magnetite.

However apart from magnetite in this shell there are mono and polycrystalline particles of jozite.

Analises by means of microzond do not establish Ti, Mn, Ni, Co and other elements in magnetite and jozite.

The surface of growing together jozite–magnetite shell of spherules with a metal kernel has features which are typical for many of intermetallic combinations, getting crystalized in conditions of quick hardening.

The halo of spreading of described above magnetical balls does not coincide with area of spreading of magmatic rocks. In opposite, maximum of their amounts are connected with the areas of outcrops of sedimentary rocks of middle carboniferous–latejurenious age.

All facts lead to conclusion that the mentioned magnetical spherules don't have a magmatic nature. There are basises to say that these spherules were originated in result of desintegration of large space body what stipulated the rise of Verchojansko–Kolimskaja ring structure.

It should be emphasized that the area of spreading of described magnetical balls coincides with periphery of Kolimski median massif which can be regarded as central uplift of this cosmogenic structure.

Analysis of geological data shows that the structure under consideration appeared at the end of the early cretaceous period.

Unlike of janites in Kazakhstan not infrequently metal magnetical small balls are revealed in magmatic rocks of diverse composition and age. For example may be bring the porphyry-copper deposits Aktogai and Aidarly.

For the confirmation of the cosmogeneous nature of explosions forming craters to which the Aktogai and Aidarly deposits are confined, a meteorite substance found in their ores may be used. E. M. Poplavko found tear-shaped isolations of troilite contained in pyrite crystals. Earlier these tear-shaped isolations were taken as pyrrhotine or cubanite. The size of the prolate-oval microinclusions of troilite is from 0,002 to 0,01 mm. E. M. Poplavko discovered also tear-shaped isolations of native iron of nearly the same dimensions.

The check showed that the mineral previously defined as pyrrhotine in the ores of the Aktogai and Aidarly deposits turned out to be troilite. Reflectivity,\* microhardness and crystalline mineral texture by microdiffraction and the dimensions of its crystal lattice were defined. All determined parameters undoubtedly indicate troilite. The mineral microhardness measured by S. I. Lebedeva (Institute of Mineralogy, Geochemistry and Crystal Chemistry of Rare Elements, IMGRE) made up 186, 249, 229, 187, 205 kg/mm<sup>2</sup>. The mineral microhardness according to reference books varies from 157 to 286 kg/mm<sup>2</sup>, that of pyrrhotine — 210–367 kg/mm<sup>2</sup>. Hardness according to the Moos scale is No 4. Anisotropy of the first and second kind of hardness is ascertained. Coefficients of anisotropy: K1 = 1.10; K2 = 1.28. Microfragility of the investigated troilite (definite by S. I. Lebedeva) according to the coefficient measured and calculated by S. I. Lebedeva by the formula is 2.8–2.9. Sizes of elementary cells are determined by the data of microdiffraction by V. T. Dubinchuk (VIMS): a0 = 5.99+0.03 angstrom, c0 = 11.82+0.03 angstrom. The dimensions of pyrrhotine are substantially different: a0 = 3.44, c0 = 5.69 angstrom.

It is well-known that troilite is an ordinary component of all classes of meteorites, in particular-stone, constituting not less than 99% of all meteorites, colliding with Earth. About 6% of troilite is contained in them exactly.

Tear-shaped very small isolations apparently represent solidified particles of the meteorite substance melt initiated at the moment of explosion injected into the deep-penetrating cracks in target rocks exposed to intensive fragmentation during this explosion. It is essential that troilite s revealed principally in the upper horizons of ore bodies. Permanent presence of troilite in pyrite as tear-shaped isolations comes about as a result that the

\* Troilite reflection was investigated on a double-beam microspectroreflectometer BLESK designed by the State Optical Institute. objective 21X; A=0.65; reference – silicon; analyst T. N. Chvileva, IMGRE.



composed by its isolations served as a seed during ore deposition, i.e. during the hydrothermal process manifestation provoked or intensified by a cosmogeneous explosion. The possible activation of not only hydrothermal but also magmatic process as a result of cosmogeneous explosion is indicated by dykes and subvolcanic stocks composing the ring belt according to the data of A. I. Poletayev and coauthors.

The age of these magmatic bodies determines the time of cosmogeneous bombardment as Late Carbonaceous.

The picture observed at deposits appeared as a result of simultaneous powerful explosion of two fragments of one meteorite deepened in target rocks.

An irrefutable and reliable proof of the meteorite origin of ring structure is the location of proper meteoritic substance. Meteoritic debris are found in regions of a comparatively small number of craters. Very often a meteoritic substance is found in the form of metal spherules or balls: hollow, elongated, in the form of flat moulds, wires, drops. The above-mentioned particles of meteoritic substance are usually distinguished in testing technological samples at different deposits of useful minerals confined to cosmogenic structures. A vivid example of meteoritic substance in a similar form are numerous metal particles recovered during processing of technological samples, from deposits of small dust-like technical diamonds in Northern Kazakhstan.

Apart from numerous balls other forms have been discovered in these samples: drops, wires, fissure casts. If balls may represent themselves the meteorites residues burnt in the atmosphere and having not reached the Earth then these forms could be formed only on the Earth. Their formation can be ensured only in the case if a melted substance of cosmogenic body is injected in deeply penetrating fissures of the rocks of the target.

Composition of these particles is as follows: alfa-iron, jozite –  $\text{FeO}$ , magnetite –  $\text{Fe}_3\text{O}_4$ , hematite –  $\text{Fe}_2\text{O}_3$  and combination of these components. Particles of plessite are encountered comparatively seldom. Silicate glass particles may be encountered. Sometimes silicate glass obtains the form of metal balls. The evidence to consider these particles a meteoritic substance is the discovery of some of them which have the form of drops flowing down the fissures of the walls. Similar form is possible only if the melt is seeping from above. Other is excluded.

Outward appearance of these particles is clearly seen when magnified by electronic microscope. When studied under binocular lens they seem to be amorphous. But in fact they have a fine crystalline structure. The dimensions of these particles are from 100–300  $\mu\text{m}$  up to 1 mm.

Among these particles drop-like segregations of troilite are found. Earth troilite is extremely rare, and in chondrite, mostly occurred class of meteorites, it amounts to 6% on the average.

In a porphyry copper deposit Aktogai the drop-like formations of troilite (2) are abundance in pyrite crystals. Here also occur "drops" of alfa-iron (pure iron). Meteoritic substance (troilite and alfa-iron) in this deposit for the first time was identified by A. M. Poplawko, a mineralogist from IMGRE (Moscow). Later on troilite was confirmed by the electronic microscope study at VIMS by V. T. Dubinchuk. In Aktogai other features of cosmogenic influence are discovered, planar structures in quartz breccia, regular orientation of multiple structural elements of ore stockwerk (2) in particular.

In connection with said above — the mentioned spherules have very great importance for global correlation (3).

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# SPHERULES AND MICROTEKTITES IN THE ?CONIACIAN--MIDDLE CAMPANIAN SEDIMENTS OF THE BAKONY MTS (HUNGARY) A PRELIMINARY REPORT

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<sup>3</sup>Üröm, Rákóczi u.53, H-2096

## Abstract

The Senonian sequence of the borehole Magyarpolány Mp. 42 in the Northern Bakony Mts. was investigated. Spherules of microscopic size and glassy particles of cosmic origin were observed in the preparations made of washing residue. In this borehole spherules were first found by A. Szarka (1991), however, only in a single sample (626.5 m, Csehbánya Formation).

In addition to A. Szarka, the author was able to isolate spherules from all formations (Csehbánya, Ajka, Jákó and Polány Formations), in a decreasing number upwards. The first author was the first to find glassy meteorite particles in the marine formations and in the upper part of the Ajka Coal Formation.

According to the investigations carried out so far, spherules are abundant in the lower part (?Coniacian to Santonian) of the borehole. On the other hand, particles of glassy micrometeorites are more frequent in the upper part of the Polány Marl Fm. (Campanian).

## Introduction

During the study of the wash residues of the ?Coniacian--Middle Campanian core materials furnished by the borehole Magyarpolány Mp 42 strongly refractive, metallic spherules and slightly refractive, glassy microtektites, both of microscopical size were observed. The spherules were firstly described from one sample of the above mentioned core material in the MSc thesis of A. Szarka (1991). He found they consist of iron and considered them as being of interplanetary origin.

This note is to present a short morphological, textural and compositional characterization of the above microscopical bodies together with data about their stratigraphic position.

## Geographic location and geological setting

The borehole Magyarpolány Mp 42 was drilled in 1980 for coal exploration 2 km NE from the church of Magyarpolány village, in the northern part of the Bakony Mts (Fig. 1).

It penetrated a 734 m thick ?Coniacian-Middle Campanian sequence, which discordantly covers the Upper Jurassic-Lower Cretaceous Mogyorósdomb Limestone Formation of Biancone facies and is discordantly overlain by the shallow marine Nummulitic Limestone of Middle Eocene age (Fig. 2). The ?Coniacian--Middle Campanian is divided to 4 formations by Mészáros (1980) as follows:

- 19.0 m - 455.0 m Polány Marl of marly lithology with Inoceramus
- 455.0 m - 517.3 m Jákó Marl, marls, clay marls with Gryphaeas, other molluscs and corals
- 517.3 m - 637.3 m Csehbánya Formation, terrestrial variegated clay, clay marl, silt, sand, gravel with freshwater intercalations
- 637.3 m - 727.6 m Ajka Coal Formation
- 727.6 m - 734.0 m Csehbánya Formation

### Results

Spherules and microtektites are present throughout the ?Coniacian--Middle Campanian section, in marine as well as in continental facies (see Table 1).

Wash residues of 67 samples were selected for analysis. 20 samples contained microscopic cosmic particles. Spherules were identified in 10 samples and microtektites in 8 samples. Both spherules and microtektites were found in 3 samples (see Table I). Four spherule frequency maxima were identified at 700.0 m, 701.4 m, 655.0 m and 626.5 m, a significant microtektite frequency maximum was found at 372.0 m with 155 microtektite grains. In addition, 2 lesser microtektite maxima were observed at 707.0 m and 701.4 m which were parallel to the first two spherule peaks.

Occurrence of cosmic particles in the borehole Mp 42 (Table I):

Mp 42 (m)	Spherules (pieces)	Microtektites (pieces)
100		2
115		1
175		1
185		1
372		155
450	1	3
500		
527.3		1
583.6		1
626.5	32	
627.4	1	
638.5	1	
655	15	
700	5	
701.4	30	18
707	35	8

## **Microspherules**

Bluish black, brownish black, strongly refractive, hollow spheres (diameter 0.063-0.25 mm) or more seldom half spheres, cavernous spheric bodies and flattened bodies with characteristic surface pattern (Fig. 3). A grain found in the Polány Marl at 450 m depth shows polygonal plus pentagonal and elongated hexagonal pattern and fluidal texture on SEM images.

Spherules have been found in the Polány Marl (SEM fotos see on Plate I), in the silty intercalations of the Ajka Coal Measures and in the freshwater intercalations of the Csehbánya Formation. In these latter spherules are accompanied by the calcareous alga *Munieria grambasti* (Bystrický) ssp. *sarda* Cerchi et al. The spherules studied by Szarka consist of almost pure iron with very low amount of Ti, Mn, Ca, Si and Al based on microprobe analysis (Szarka, 1991). The spherules of the Csehbánya Formation were found by EDAX study to consist of  $\alpha$ -Fe (Kákay Szabó O. oral comm.).

## **Microtektites**

Slightly refractive, yellowish, more or less translucent, inclusion containing, glassy bodies with diameter varying between 0.63 mm to 0.30 mm. They are characterized by fibrous surface pattern and mostly show spheric, ellipsoidal, spindle-like, tongue-like, more seldom taped slick-like forms. The spheric forms bear 1-3 curved, twisted appendices (Plate I).

A further study of the subject is of importance in view of the terminal Cretaceous extinction.

## **Conclusions**

1. Spherules and microtektites have been found in the ?Coniacian--Middle Campanian section penetrated by the drilling Mp 42.
2. Spherules occur with the exception of a single sample (450 m) only in the Ajka Coal and Csehbánya Formations whereas microtektites occur in every formations studied. Their maximum is in the lower member of the Polány Marl Formation (372 m).
3. Two types of cosmic material reached the Earth surface at Magyarpolány during the impact: an  $\alpha$ -Fe containing metalliferous and a glassy material.
4. Few analyses show the spherules consist almost entirely of iron in form of  $\alpha$ -Fe.

## **Aknowledgements**

This work was supported by the grant of the National Science Fond OTKA No. T 015783 (Bakony-Gubbio Correlation). The studies are included into the IGCP Project No. 384: "Impact and Extraterrestrial Spherules: New Tools for Global Correlation." K. Brezsnýánszky, L. Stegena, I. Vető and I. Viczián are thanked for their technical help and O. Kákay Szabó for consultation.

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Manuscript received: 18th November 1995.  
 Accepted: 15th April 1996.

Occurrence of cosmic particles in the borehole Mp 42 (Table 1).

Sample No.	Depth (m)	Particle Type	Notes
1	10.5	Spherule	Small, spherical, glassy
2	10.5	Microtektite	Small, angular, glassy
3	10.5	Spherule	Small, spherical, glassy
4	10.5	Microtektite	Small, angular, glassy
5	10.5	Spherule	Small, spherical, glassy
6	10.5	Microtektite	Small, angular, glassy
7	10.5	Spherule	Small, spherical, glassy
8	10.5	Microtektite	Small, angular, glassy
9	10.5	Spherule	Small, spherical, glassy
10	10.5	Microtektite	Small, angular, glassy
11	10.5	Spherule	Small, spherical, glassy
12	10.5	Microtektite	Small, angular, glassy
13	10.5	Spherule	Small, spherical, glassy
14	10.5	Microtektite	Small, angular, glassy
15	10.5	Spherule	Small, spherical, glassy
16	10.5	Microtektite	Small, angular, glassy
17	10.5	Spherule	Small, spherical, glassy
18	10.5	Microtektite	Small, angular, glassy
19	10.5	Spherule	Small, spherical, glassy
20	10.5	Microtektite	Small, angular, glassy

This work was supported by the grant of the National Science Foundation (OTKA No. 018783/Bakony-Gubis-Geology). The studies included into the IGP Project No. 364: Impact and Extraterrestrial Spherules. New tools for cosmic spherule identification were developed for their recognition and O. Káráy Szabó for consultation.

## Figures

Fig. 1. Geographic setting of Magyarpolány Mp 42 borehole.

Fig. 2. Geology of Magyarpolány Mp 42 borehole.

Fig. 3. Microtektites and microspherules from Magyarpolány Mp 42 borehole.

## Plate I.

Figs 1-6. Spherules, SEM photos.

Borehole Magyarpolány Mp 42, 450 m, Polány Marl Formation, Upper Cretaceous.

1. 130x, 2.-5. 150x, 6. 300x

## Plate II.

Figs 1.-6. Microtektites (with pores in Figs. 1-4), Fig. 7. Spherule, SEM photos, borehole Magyarpolány Mp 42.

Fig. 1. 115 m, 130x, Ganna Member of Polány Marl Formation, Middle Campanian.

Fig. 2. Part of Fig. 1, 520x.

Fig. 3. 175 m, 130x, Ganna Member of Polány Marl Formation.

Fig. 4. 527,3 m, 78x, Csehbánya Formation, ?Coniacian-Santonian.

Fig. 5.-6. 100 m, 130x Ganna Member of Polány Marl Formation.

Fig. 7. 701,4 m, 260x, Ajka Coal Formation.

Photos by I. Bodrogi - V. Takács - M. Pellérdy

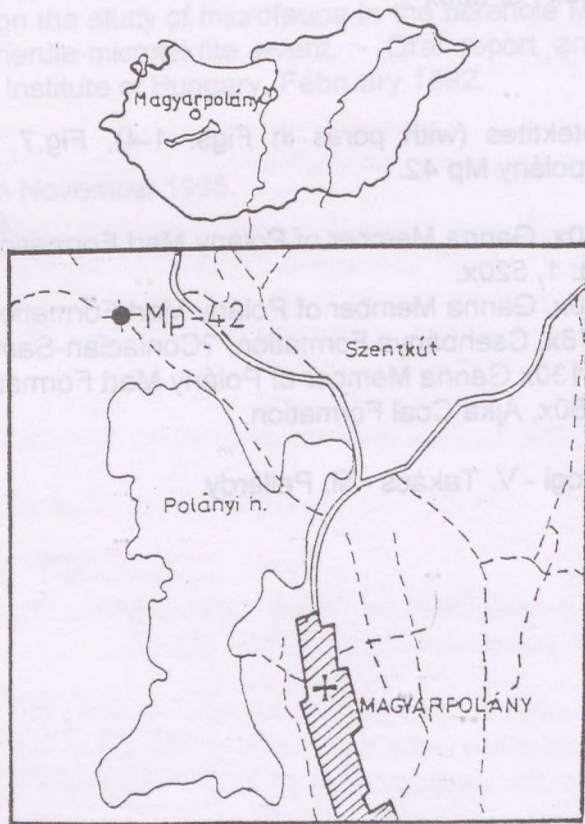
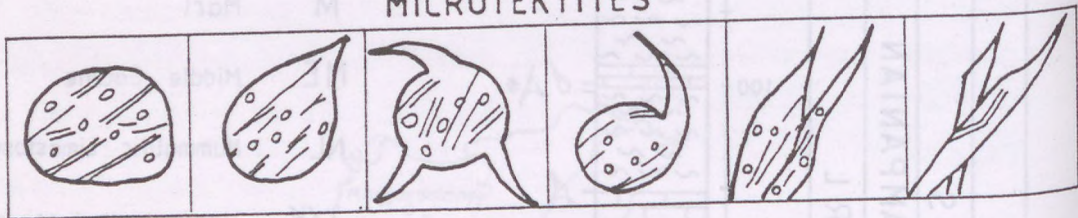


Fig. 1.





MICROTEKTITES



MICROSPHERULES

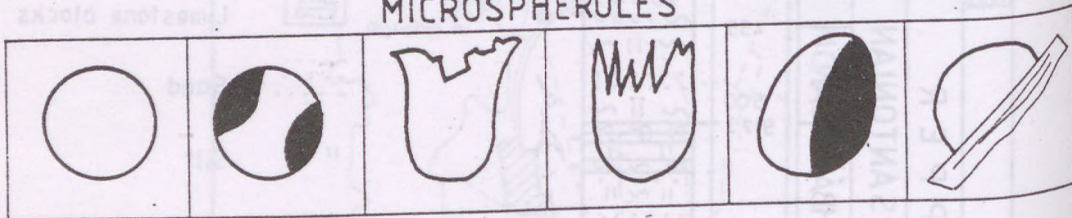
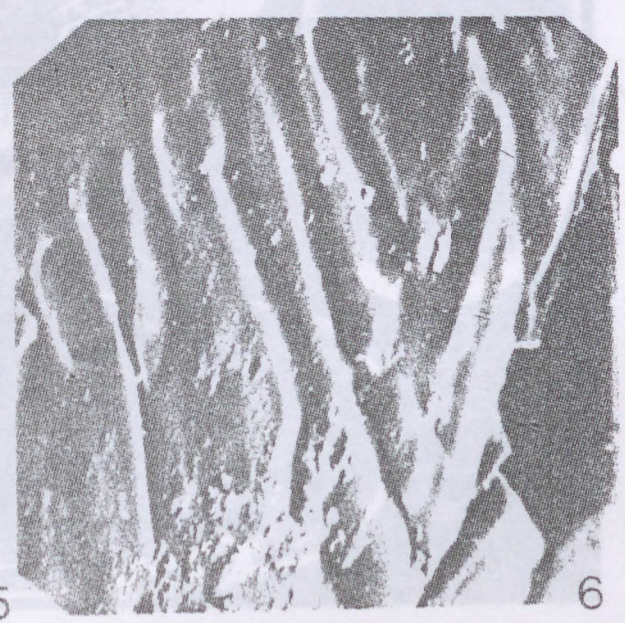
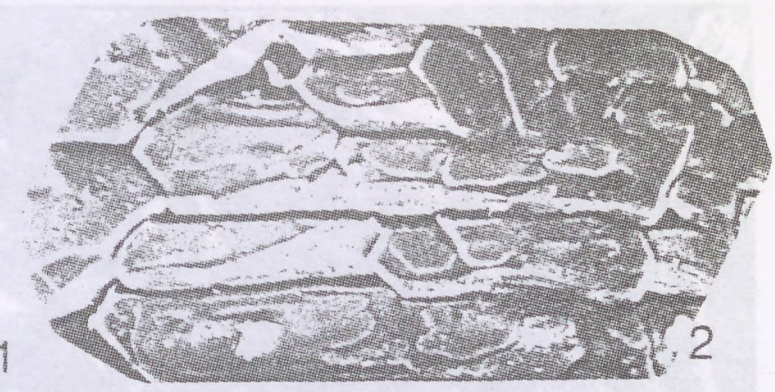
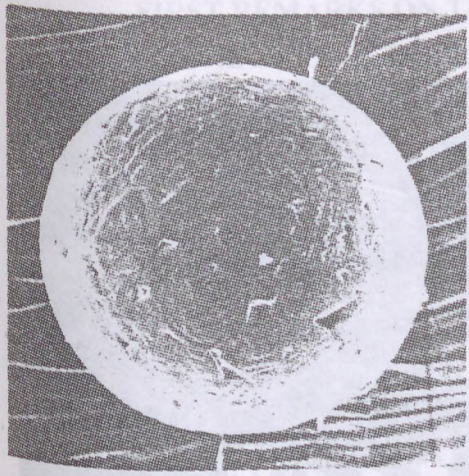
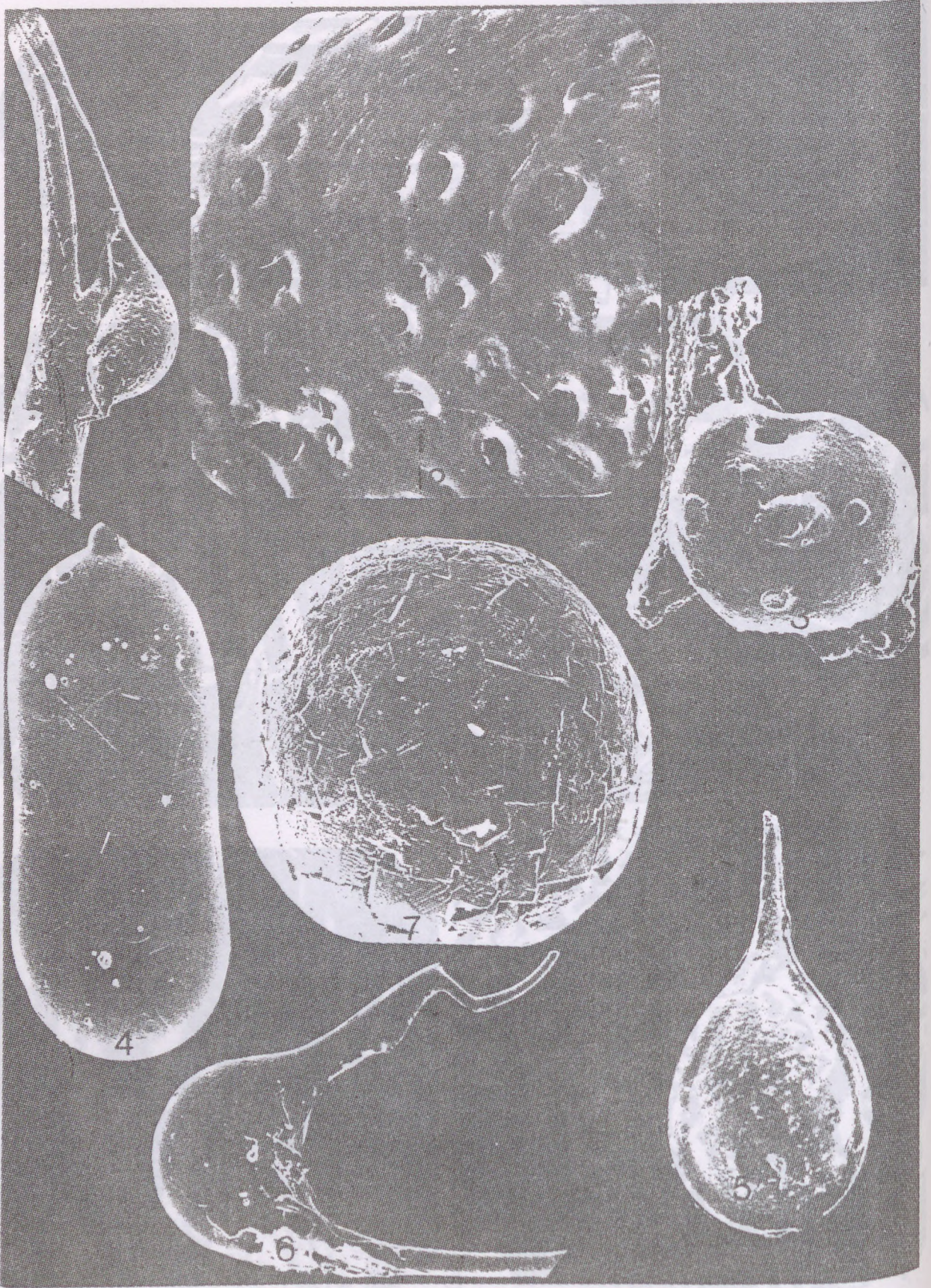


Fig. 3

THE SIGNIFICANCE OF THE OCCURRENCE OF EXTRATERRESTRIAL  
IN THE SENONIAN ALLUVIAL SEDIMENTS





# FIRST REMARKS ON THE OCCURRENCE OF EXTRATERRESTRIAL MAGNETIC SPHERULES IN THE SENONIAN ALLUVIAL SEDIMENTS OF THE SOUTHERN BAKONY MOUNTAINS, HUNGARY

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**Abstract.** Magnetic spherules were found during the micromineralogical study of the Upper Cretaceous Csehbánya Formation. Chemical analysis of the samples showed that they consist of Fe and small amounts of Ca, Al, Si, Mn and Ti. Since they contain only traces of titanium, their volcanic origin is questionable. It is suggested that they are rather of impact origin.

## Introduction

The subject of my M.Sc. thesis was the micromineralogical study of the Senonian alluvial clastic sedimentary deposits of the Csehbánya Formation, in the Southern Bakony Mts. The results will be published in an other, more detailed paper (SZARKA in prep). Samples were collected from borehole Mp-42, Magyarpolány (Fig.1.) For the exact location the reader is referred to Bohn, 1982.

In one of the samples prepared for micromineralogy (Fig.2.), strange shiny black spheroidal grains were found. They amounted to 7.72 weight percent of the heavy fraction of the sample. Their occurrence having been limited to one single sample only, the contamination origin could be safely discarded.

The grains were studied by scanning electron microscopy and electron microprobe equipped with an Energy Dispersive Analyser. The device used was the AMRAY-1830-I electron microprobe of the Department of Petrology and Geochemistry, Eötvös University. The technical parameters were as follows: acceleration voltage 20 kV, measuring time of the spectra 60 sec. The analysis was made with the help of Miss M. Jánosi and Dr. K. Török.

## Results

By their shape, the spherules can be divided into three groups: *spherical* (Photo 1.), *droplet-shaped* (Photo 3.) and *irregular* (Photo 4.). Their surface is variegated. In some cases the surface pattern exhibits linear features that look like those of the octahedrite-type

meteorites (Photo 2.). In other cases, there are "chain"-like features on their surface, (Photo 5., 6.) which may be intergrown individual octahedral crystals (Photo 7.,8.). Again others show a "mosaic" pattern made up of polygonal (pentagonal-hexagonal) sheets (Photo 9.). Some grains seem to be etched or built up by skeleton crystals (Photo 10., 11.).

Some of the grains were fractured, thus their internal structure could be observed. The fracture surface is irregular, splintery. Most spherules have a central. Its shape is more or less elongate spherical (Photo 12.). In some cases this hole has reached the surface of the grain (Photo 13.) suggesting the strength of expanding internal gas(?) having surpassed that of the material of the spherule. The wall of the internal hole shows the same type of pattern as the surface of the spherule, only the phenomenon is much more "pronounced" in the hole than on the surface (Photo 14.). The "chains" seen on the internal surface may have been melted and then suddenly frozen again. In also some samples also drusy crystals attached to the internal surface were observed (Photo 15.). The shape of the drusy crystals is similar to the "cockscomb aggregates" of marcasite (Photo 16.).

The chemical composition of the spherules is rather constant: Their main constituent is iron-oxide (Fig.3.). No enrichment of Ni or Co was found, not even in the "octahedrite"-type grains. — Some samples contained Ti, Mn, Ca, Si and Al (Fig.4.). (The Au, Cl and Br of the spectra arise from sample preparation: dissolving in hydrochloric acid, separation by bromoform, coating by gold.) Only one grain was different: its colour was orange (instead of black), and it contained only Ca, Al and Si (Fig.5.).

## Discussion

Magnetic spherules are common on the surface of the Earth. They may develop in two main ways (JAKABSKA & ROZLOZNIK, 1989; MARVIN & EINAUDI, 1967) either **extraterrestrial** or **terrestrial**.

The extraterrestrial ones may be *ablation particles* of meteorites, or *vaporized particles of large bolides* hitting the earth and creating large craters. They may be also *tektites*: likewise formed in association with impacts but originating as the molten fraction of terrestrial rocks heated up by the impact. They may be also grains of *asteroidal or cometary origin* (interplanetary material).

Terrestrial spherules may be the results of *volcanic activity* or of human *industrial activity*. In addition, spherules may be formed also in *other natural ways*: like in case of forest fires, lightning discharges or biogenically by marine organisms (MARVIN & EINAUDI, 1967).

Most magnetic iron-rich spherules mentioned in the literature have a nickel- and cobalt-rich core (see e.g. FECHTIG & UTECH, 1964; BLANCHARD et al., 1980; BROWNLEE, 1981). Neither of the spherules found in the Csehánya Formation had nuclei of this type. FECHTIG & UTECH (1964) showed that during the ablation of the Ni-Fe-rich part of meteorites

nickel is usually normally concentrated rather in the metal phase and is only partially incorporated into the oxide phase. This would comply with the probably oxidic mineralogical composition of the "Csehbánya" spherules. (Because of the scarcity of the grains in the micromineralogical separate I could not perform on X-ray analysis of the spherules but their highly magnetic behaviour suggests that they are probably of magnetitic composition.) Magnetite spherules may be formed by volcanic activity, too. Volcanic magnetites are, however, generally rich in titanium (MARVIN & EINAUDI, 1967) which is not of the case of the "Csehbánya" iron-oxide spherules.

Magnetic grains similar to some of the spherules found in the Csehbánya Formation were described by FUNICELLO & FULCHIGNONI (1969). Their samples were collected from Quaternary (early Sicilian) sedimentary rocks of the Pontina plain and the surroundings of Ascoli Piceno. They concluded that the grains were of extraterrestrial origin. They were very small, with a maximum diameter of 0.015 mm. As to shape, they were either spherical or droplet-shaped, and their surface showed the characteristic "chain" pattern. They also had fractured grains exhibiting the internal hole with the same pattern on their wall.

### Conclusion

Because of their size, shape, surface and interior pattern and chemical composition, I assume that the spherules of the Csehbánya Formation are of extraterrestrial origin. Though most of the grains are spherical, droplet-shaped ones with radial lines on their surface (striking backwards from the apex) have also been found. According to Dr. C. Koeberl (University of Vienna, Department of Geochemistry, pers. comm.) the shape and pattern of the described spherules are similar to those of extraterrestrial spherules found in polar ice.

The structure of the wall and the shape of the internal holes (elongate spherical with signs of expansion of entrapped gas) shows that the material was melted at extremely high temperatures (MARVIN & EINAUDI, 1967) and was cooled down instantaneously. The chemical composition is rather constant, i.e. with the exception of one single Ca-Al-Si grain they were rich in iron. They contain neither Ni nor Co. Their very low Ti-content suggests that they are definitely of non-volcanic origin. Since lithophilic elements are present in them, it is highly possible that they are fragments formed during a meteorite impact. Classical tektites being mostly of glassy composition, I think that these magnetic spherules may not be tektites *sensu stricto* but rather impact-related fragments or droplets formed from a mixture of extraterrestrial and terrestrial material when some highly metallic meteorite hit the earth surface.

Since for the time being their occurrence is limited to one particular sample, they may be considered as the result of an event and may therefore potentially be used as an index horizon in the correlation of the Csehbánya Formation.

## Acknowledgements

The author wishes to express his thanks to C. Koeberl (University of Vienna) for his useful advice and comments on the material. Special thanks are due to M. Jánosi and K. Török for their patience in preparing the SEM photographs.

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Fig. 1

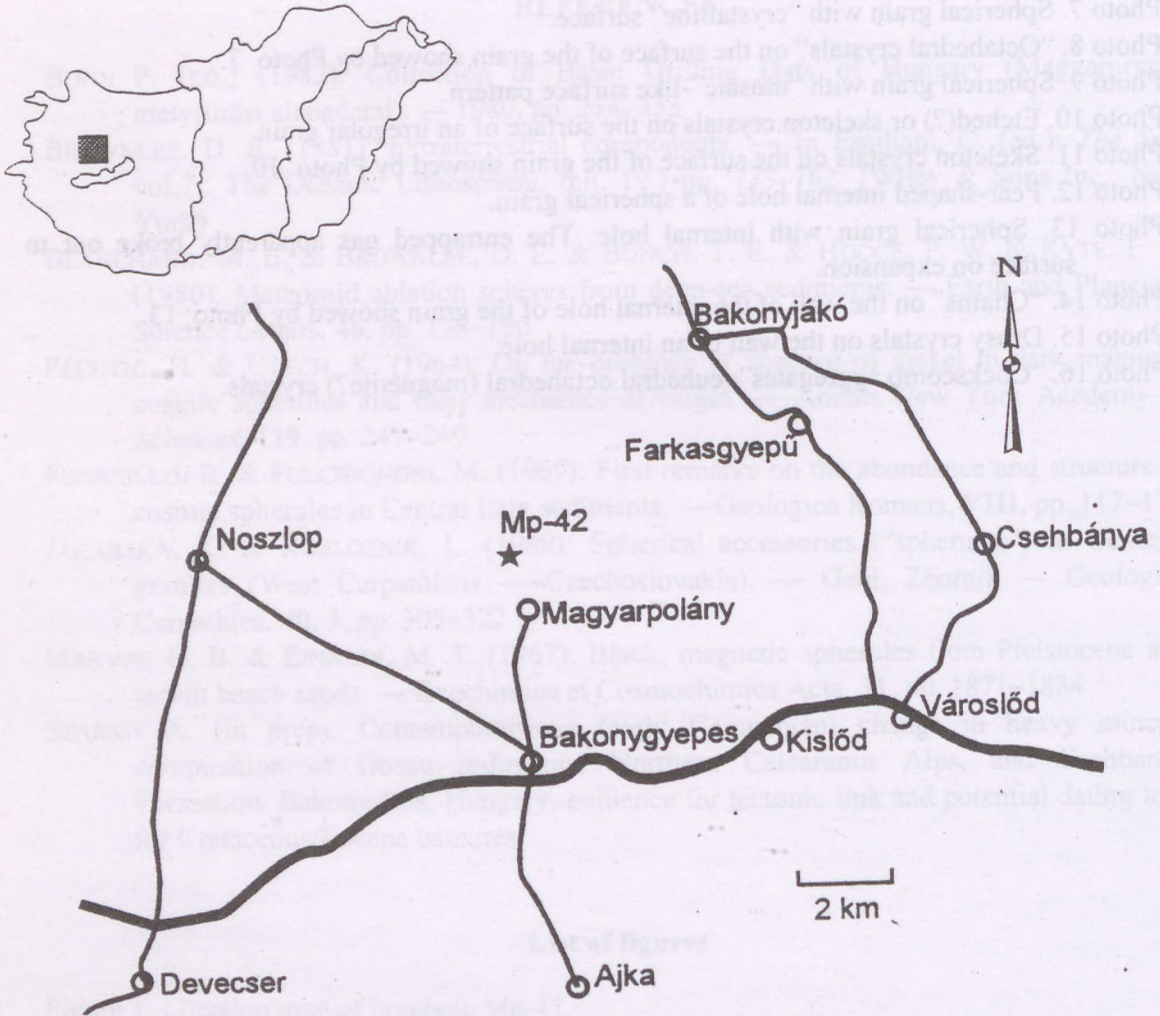


Fig. 2

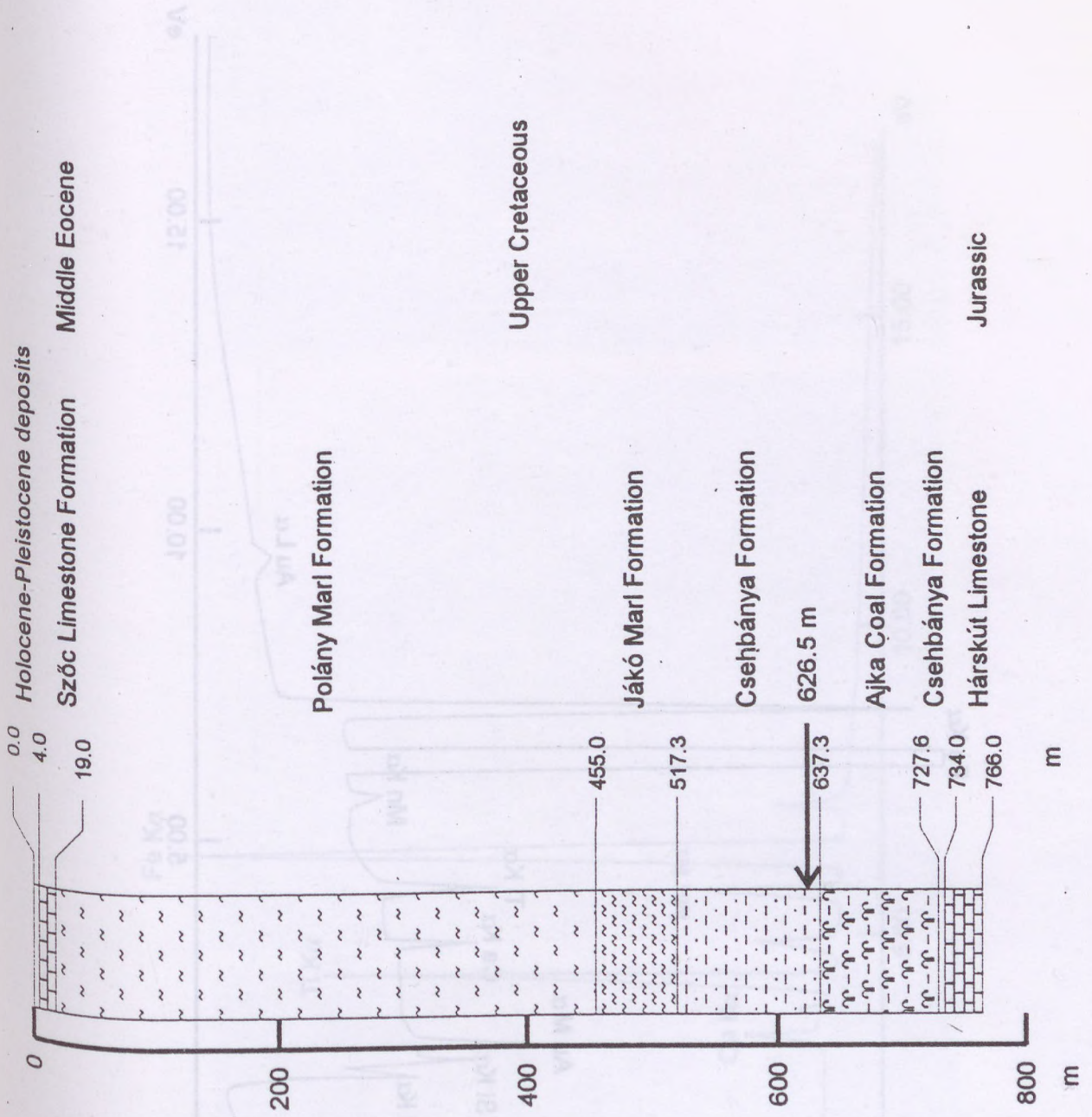


Fig. 3

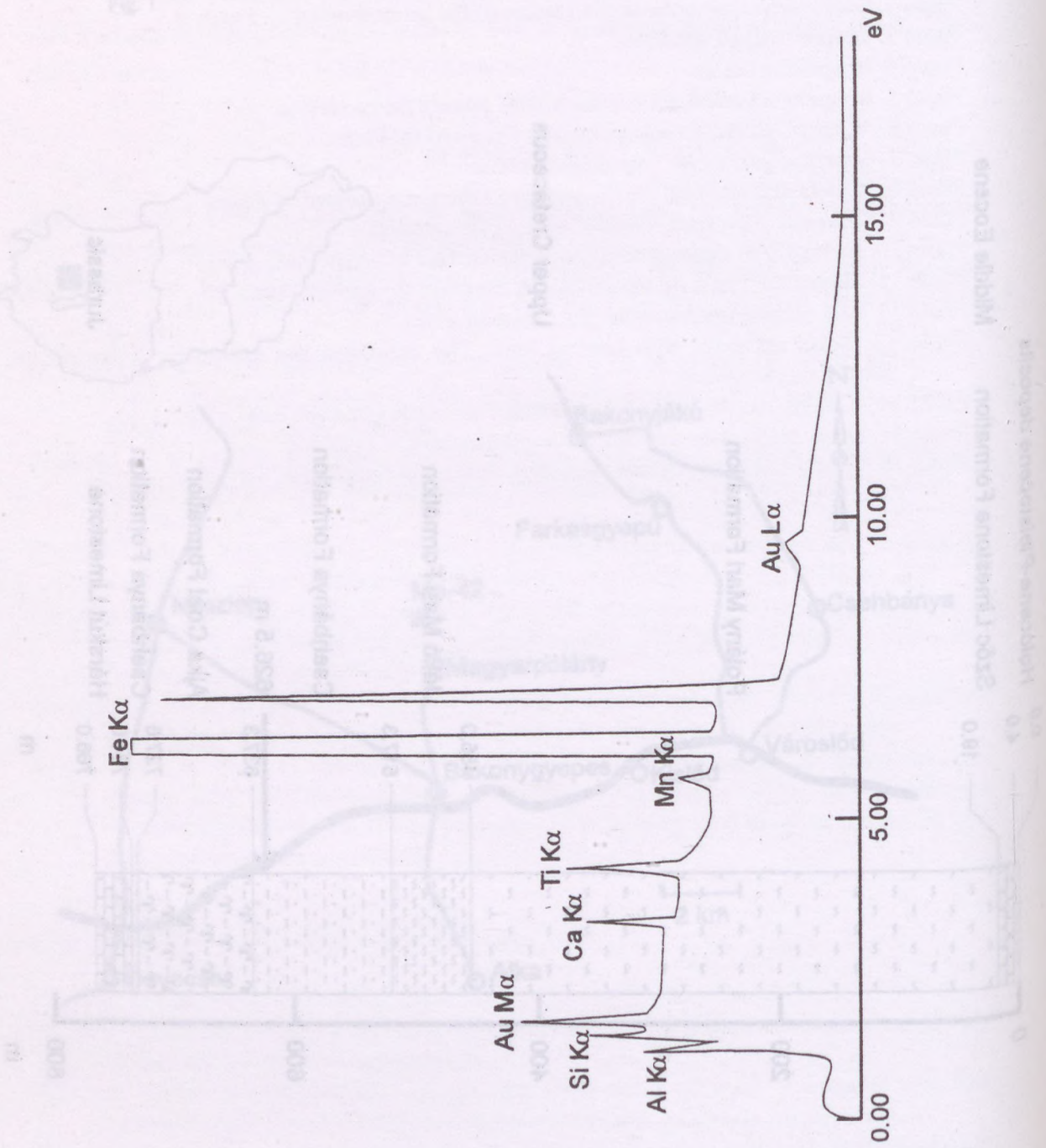


Fig. 4

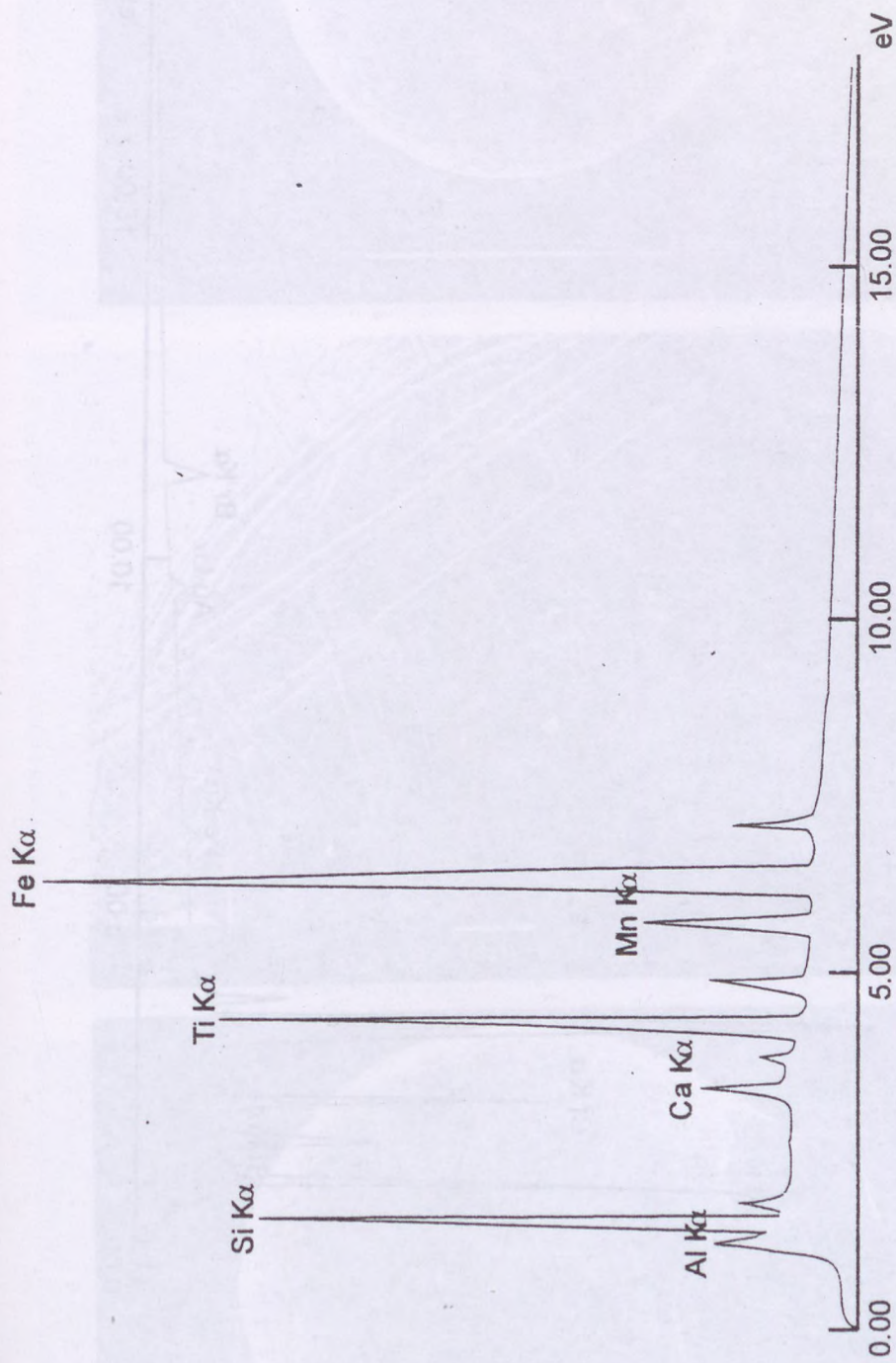
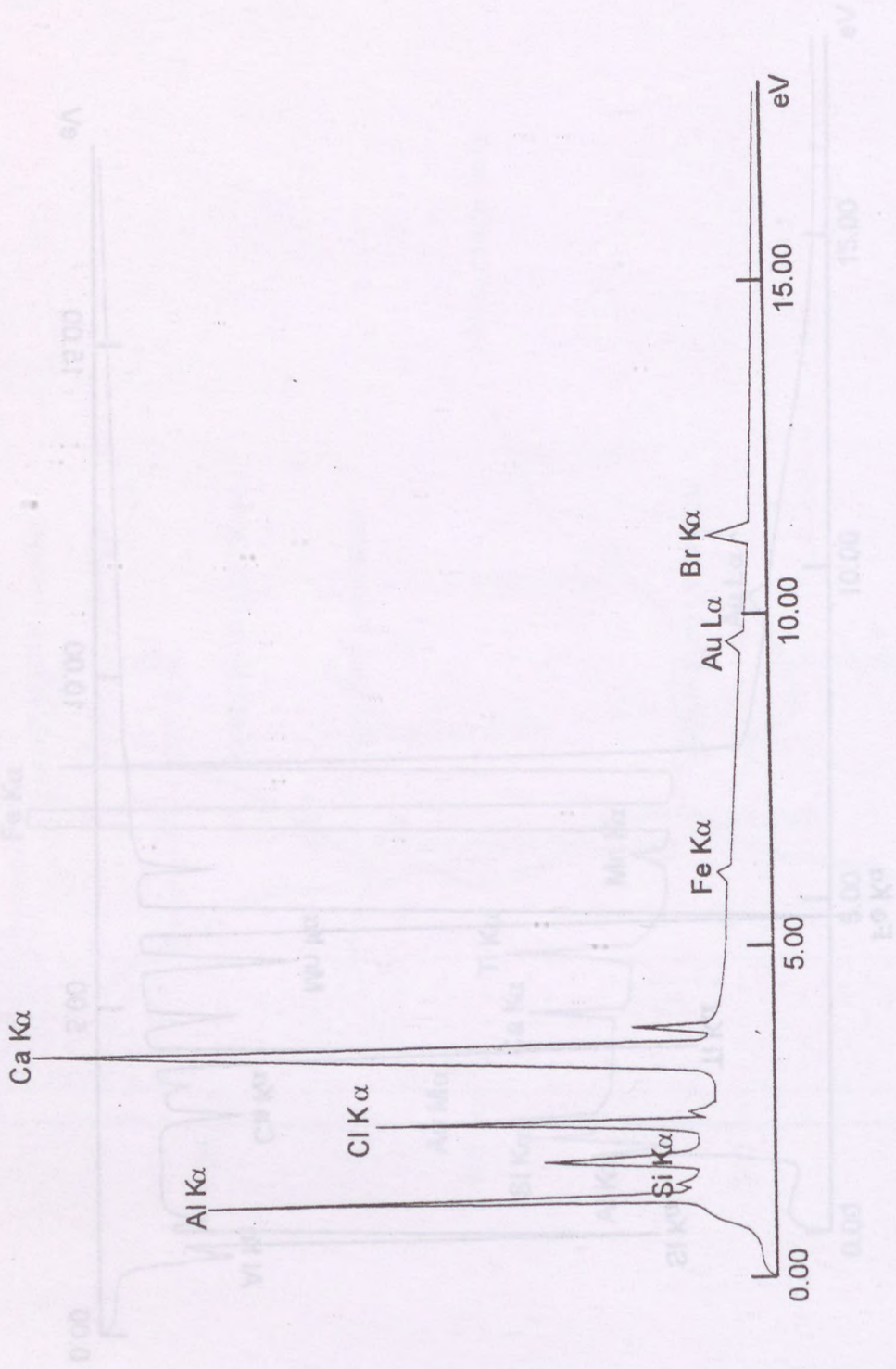


Fig. 5



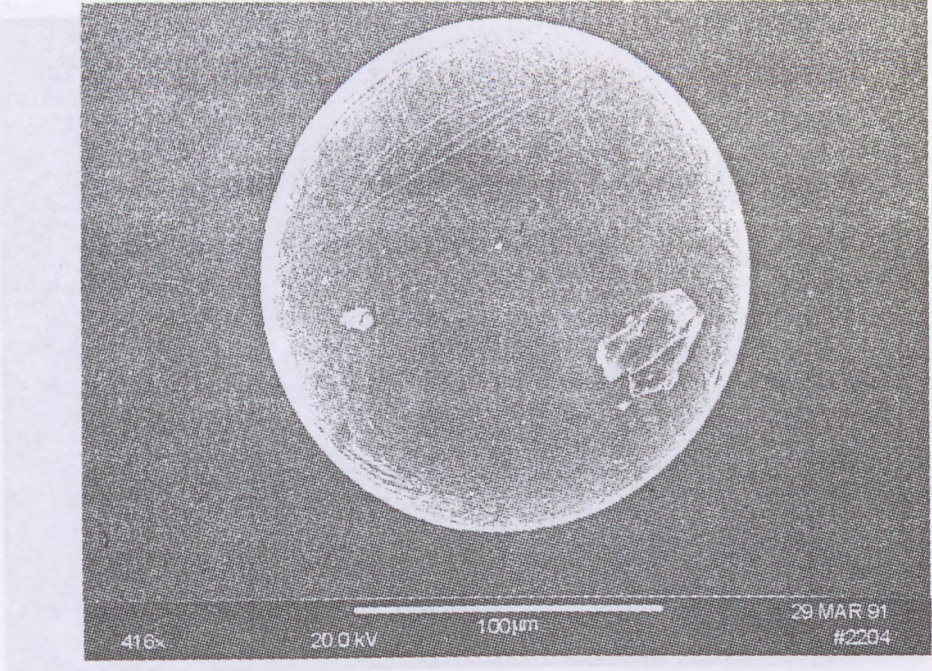


Photo 1

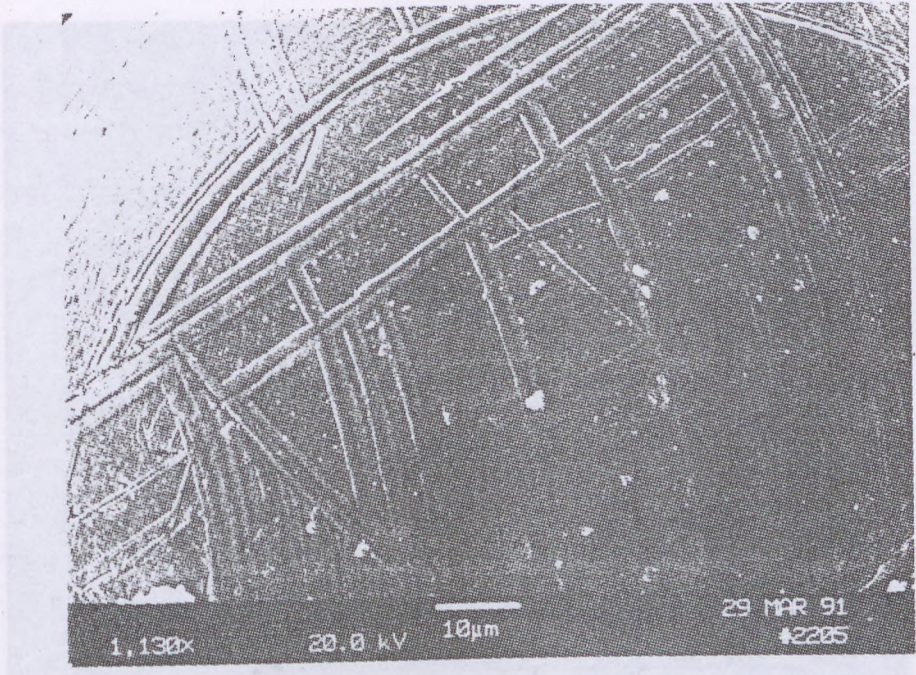


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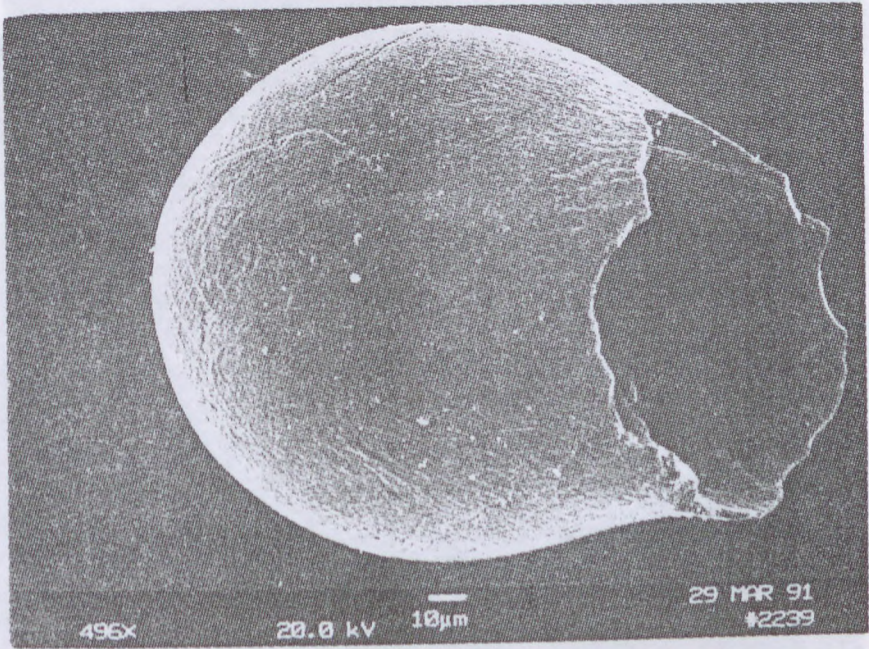


Photo 3

Fig. 3

Photo 4

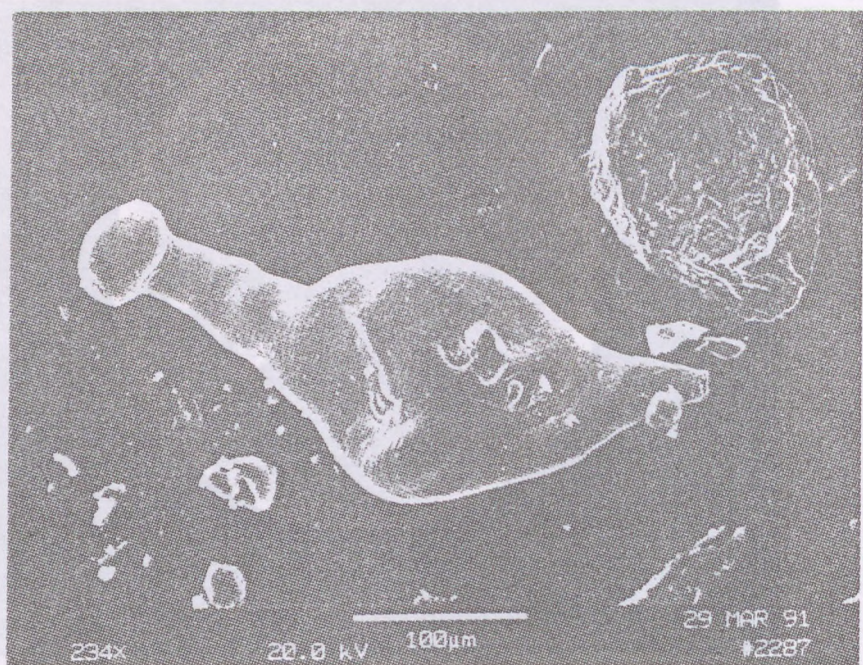


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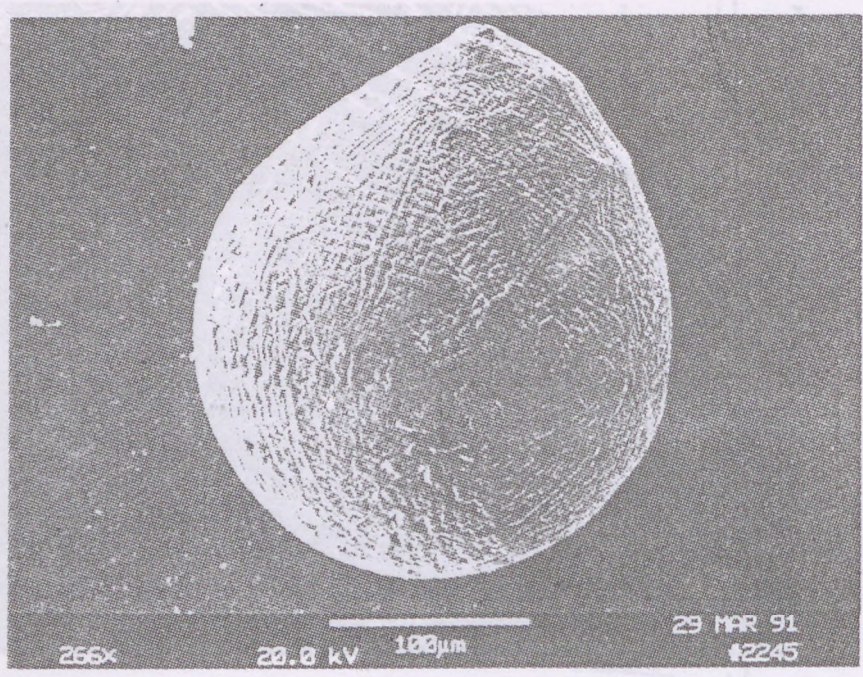
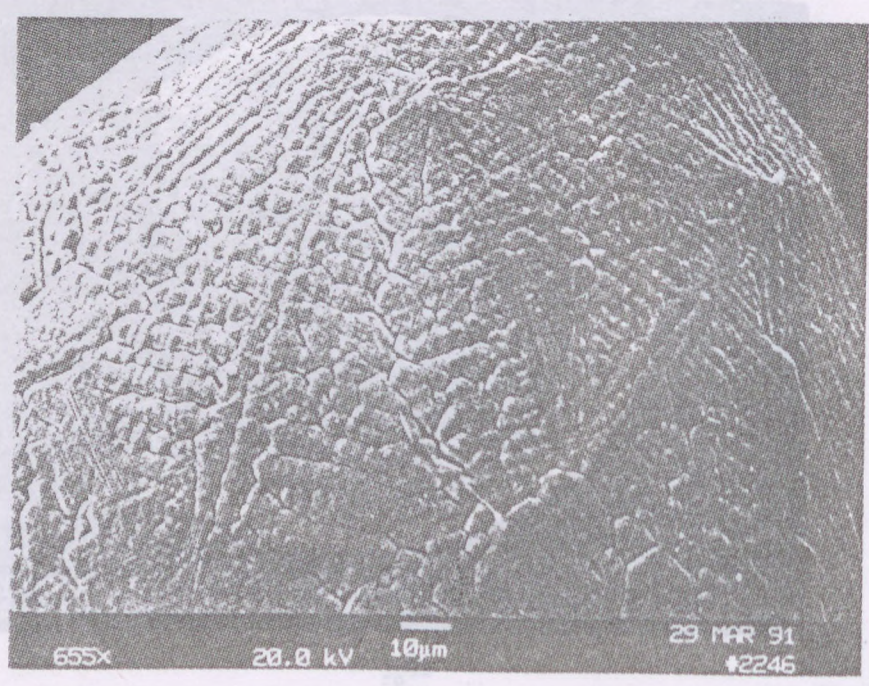


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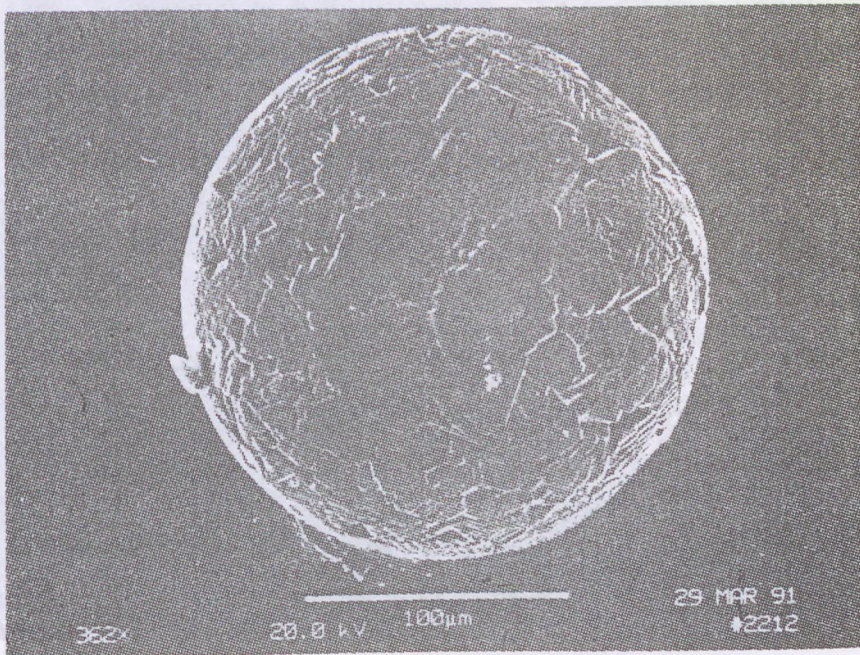


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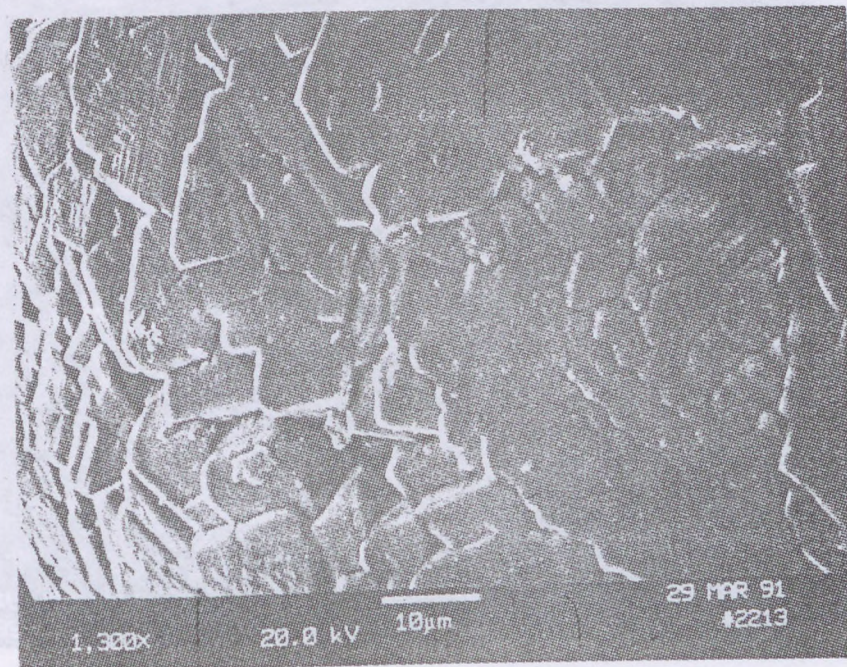


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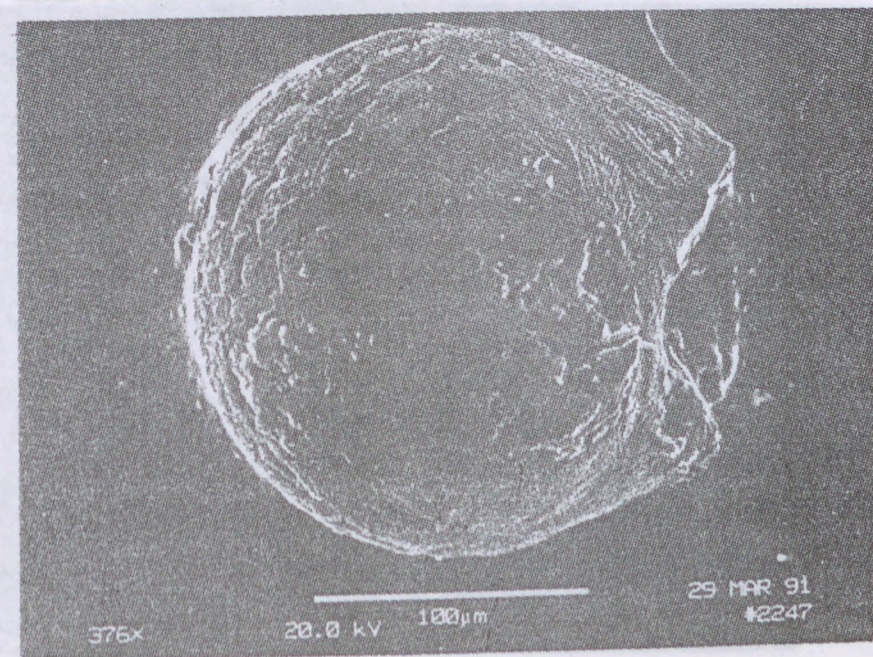


Photo 9

Photo 10

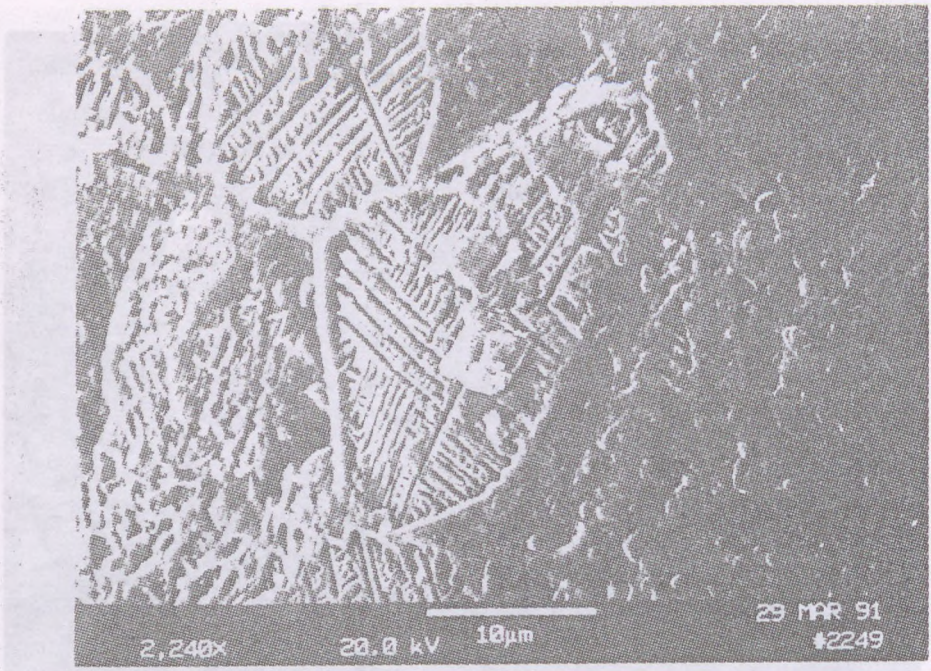


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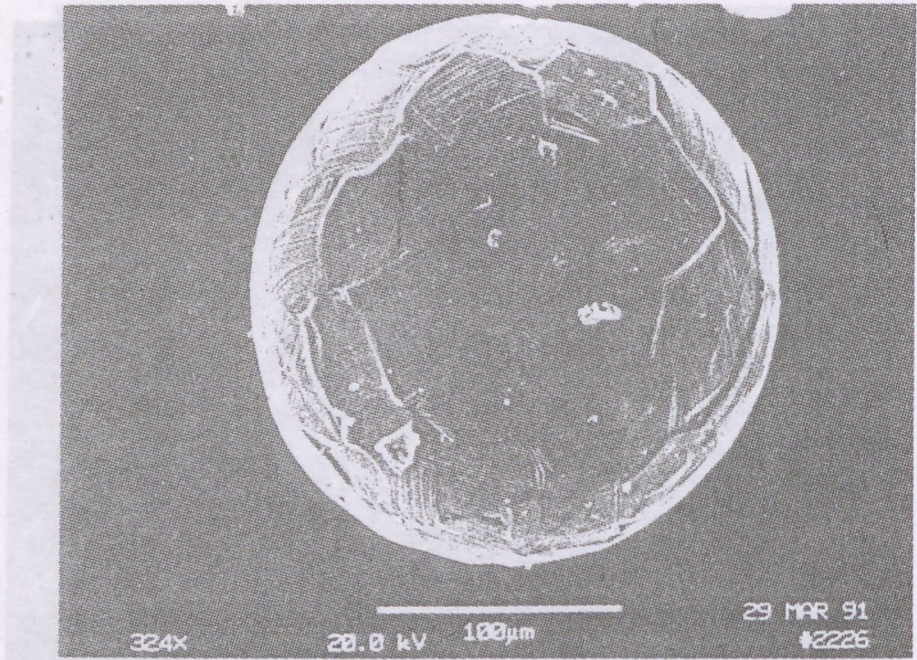
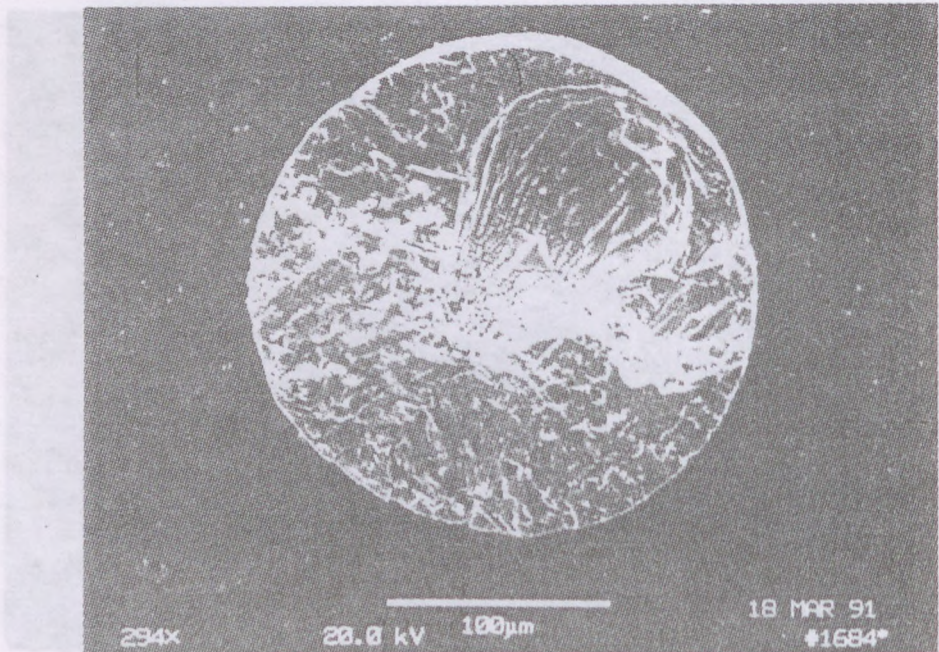


Photo 12



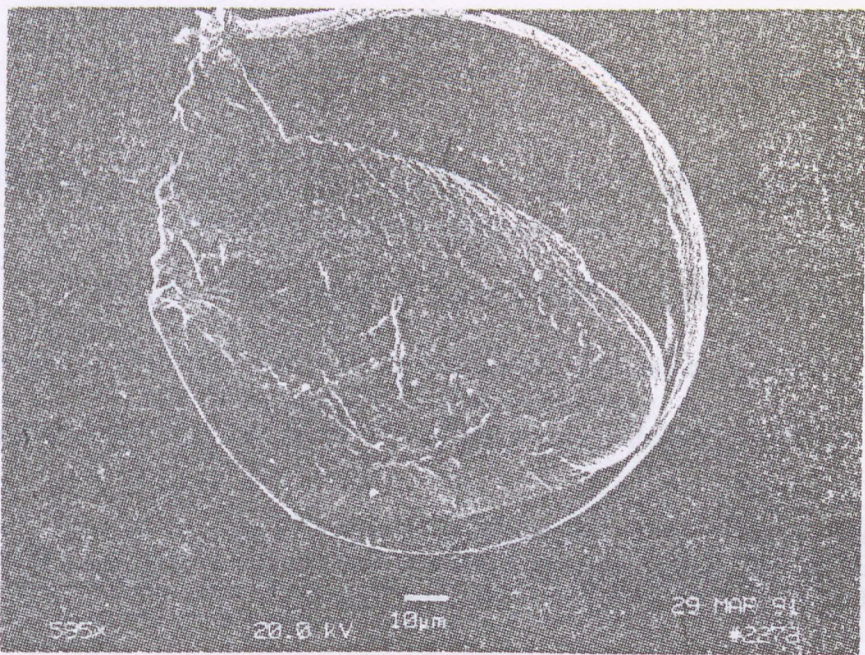


Photo 13

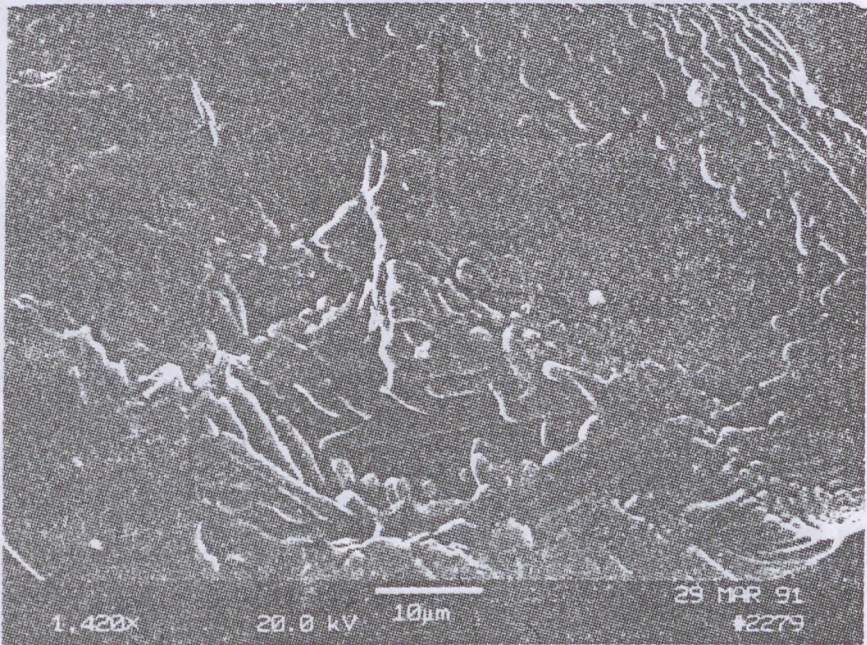


Photo 14

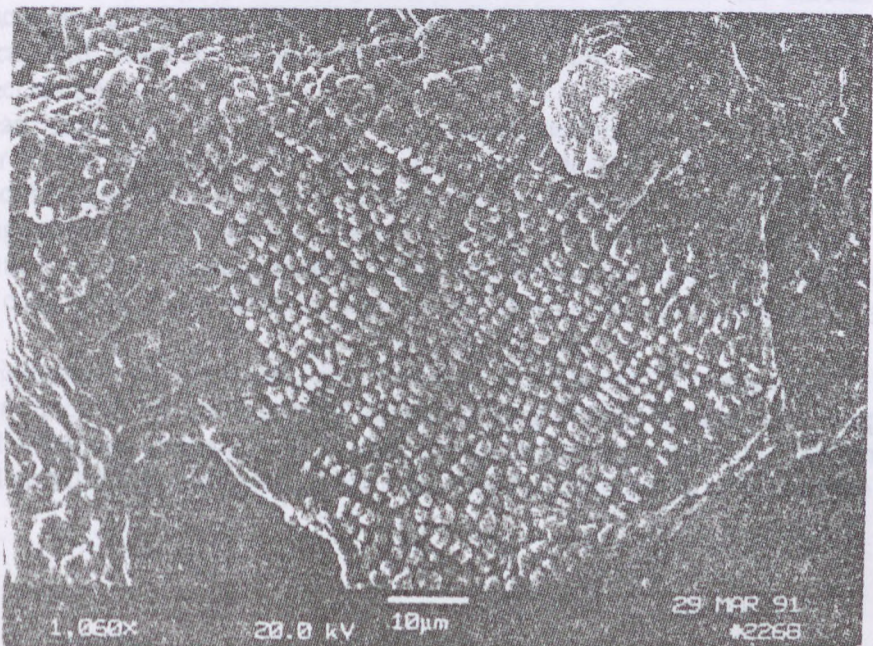


Photo 15

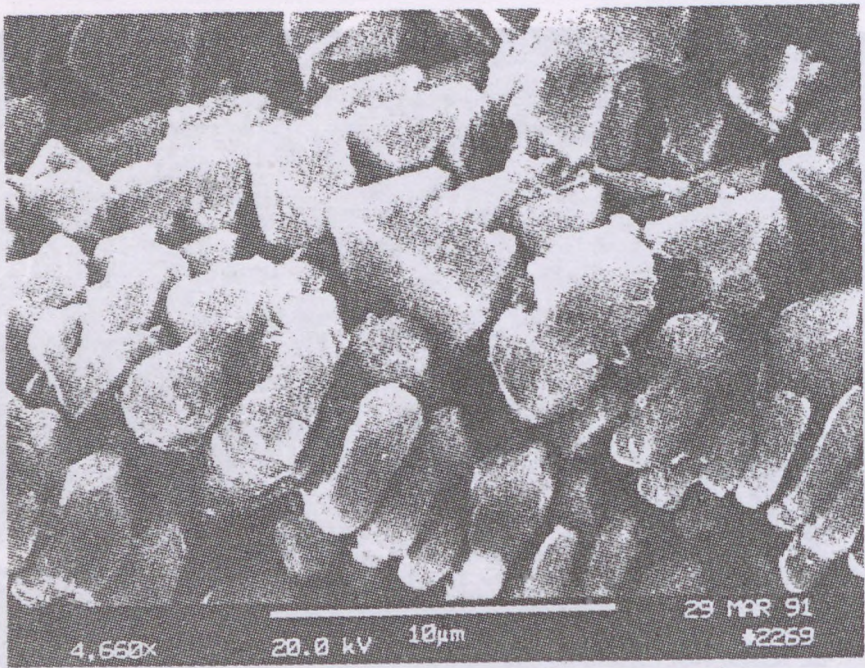


Photo 16



# Analysis of various types of extraterrestrial spherules by scanning proton microprobe

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## Abstract

The determination of elemental composition of the spherules collected from the Fekete Körös (Crisu Negru) area, Romania and the Kaba meteorite fall area, Hungary analysed by EPMA (Electron Probe Microanalysis) are extended by the use of microbeam PIXE technique (Proton Induced X-ray Emission) at the Debrecen scanning proton microprobe with special attention to trace elements.

## 1. Introduction

Electron Probe Microanalysis is a widely used analytical technique in the study of geological samples. However its sensitivity is not satisfactory in many cases. MicroPIXE, with its lower detection limits, can be a complementary method to EDAX, especially in the determination of trace elements heavier than Sodium.

MicroPIXE measurements have been carried out on samples of which the major elemental composition was already known from EDAX investigations. The examined spherules were collected by Á. Hadnagy (Romania) and P. Solt (Hungary) from the Fekete Körös (Crisu Negru) area, Romania, and the Kaba meteorite fall area, Hungary, respectively. The major and trace elemental composition of various types of spherules are presented.

## 2. Experimental

The scanning proton microprobe [1] of the Institute of Nuclear Research (Atomki) in Debrecen is built up to a 5 MV Van de Graaff particle accelerator. The accelerated ions (usually protons) are focussed to the surface of the sample by means of an Oxford-type quadrupole magnetic doublet lens system. The minimum beam spot size is  $1 \times 1 \mu\text{m}^2$ . The beam can be scanned to a maximum size of  $2.5 \times 2.5 \text{ mm}^2$ , thus elemental maps of the sample can be generated.

When accelerated ions hit the target, different reactions can happen. The atoms of the target material can be ionised, secondary electrons can escape from the surface of the sample, characteristic and bremsstrahlung x-ray radiation can occur, the irradiating particles can be backscattered, nuclear reactions can take place, while the energy of the incoming particles is decreasing.

One can detect any of the above mentioned photons or particles with suitable detectors. For PIXE (Proton Induced X-ray Emission) elemental analysis, characteristic x-rays have to be detected. The microprobe facility is equipped with a Canberra Si(Li) detector having an energy resolution of 170 eV at 5.895 keV (Mn  $K\alpha$  line). Signals are fed via a preamplifier, a signal processor and an ADC unit to the data acquisition computer. The computer can control the beam position, thus capable to scan the beam only on the areas of interest.

The microprobe facility is also equipped with a colour CCD camera and a 14 inch colour monitor which shows the irradiated sample looking through a zoom microscope. The maximum and minimum magnification of this sample observing system is 500x and 50x, respectively. This allows an easy method for localizing the desired area of the sample for the analysis.

The sensitivity of PIXE is in the range of 0.1-10 ppm depending on the accumulated charge, the matrix composition of the sample, the energy of the proton beam and many other experimental options. Thus EPMA and microPIXE are complementary methods.

### 3. Samples

The investigated samples were spherules [2], i.e. small spheroid objects either terrestrial or in more cases extraterrestrial of origin. It is possible to determine their origin using the results of the major and trace elemental composition measurements.

#### 3.1. Tektites

Honey-yellow spherules from the placer of the western part of the Királyerdő mountains (Piatra Craiului) have mainly Ca and almost the same amount of Cu and Zn trace elements. The sample No. 407B/5 have more Si and Ca than the sample No. 339B/3/2 (Fig. 1 and 2). The latter has some Mg and Sr as well.

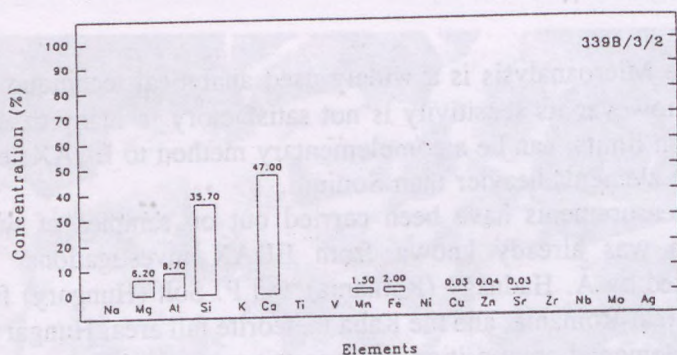


Figure 1. Elemental composition of the sample No. 339B/3/2

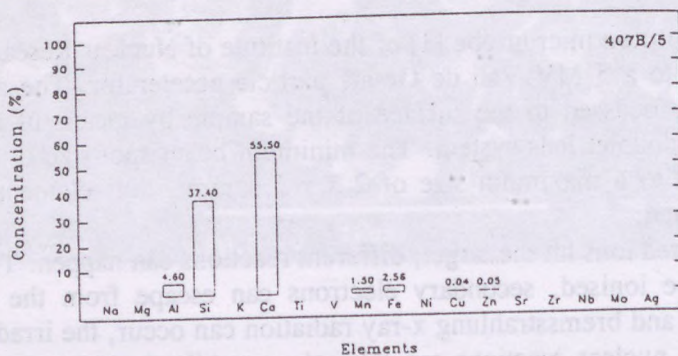


Figure 2. Elemental composition of the sample No. 407B/5

#### 3.2. Magnetospherules

Magnetospherules have been found in the placers of the western part of the Kodru mountains (Codru). These spherules contain both Cu and Zn trace elements approximately, in the same amount (Fig. 3 - 6).

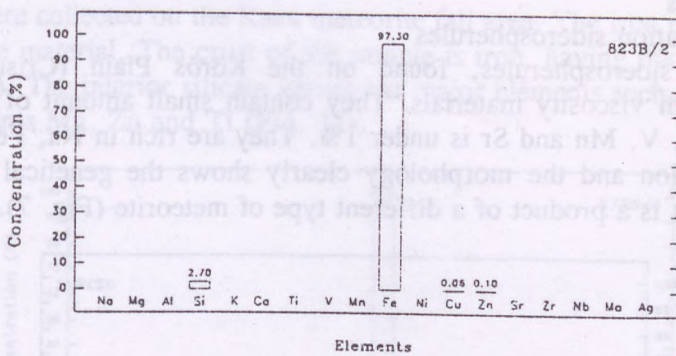


Figure 3. Elemental composition of the sample No. 823B/2

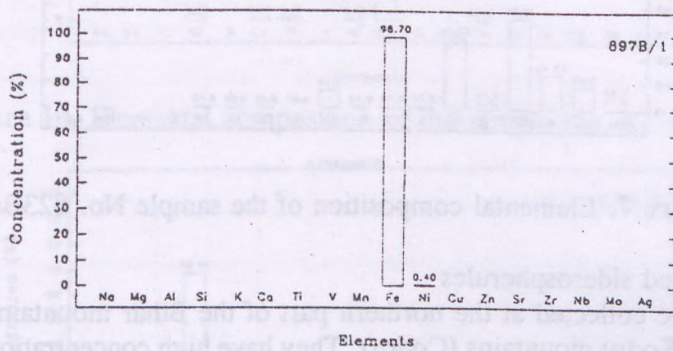


Figure 4. Elemental composition of the sample No. 897B/1

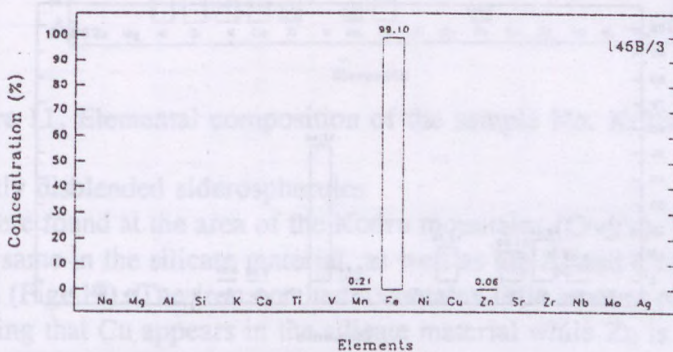


Figure 5. Elemental composition of the sample No. 145B/3

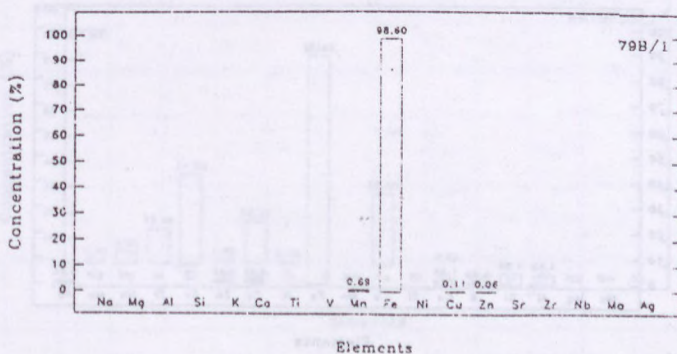


Figure 6. Elemental composition of the sample No. 79B/1

### 3.3. Siderospherules

#### 3.3.1. Tektite formation siderospherules

Tektite formation siderospherules, found on the Körös Plain (Crisului), are black, blendclothfibre, high viscosity materials. They contain small amount of Cu and Zn. The concentration of Ti, V, Mn and Sr is under 1%. They are rich in Na, Fe, Ca and Si. The elemental composition and the morphology clearly shows the genetical similarity to the tektites, however, it is a product of a different type of meteorite (Fig. 7).

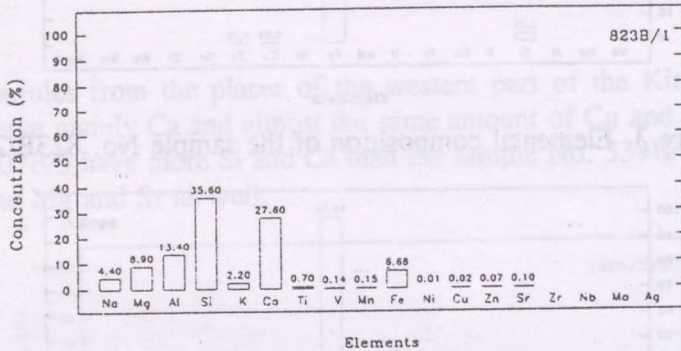


Figure 7. Elemental composition of the sample No. 823B/1

#### 3.3.2. Non disblended siderospherules

These spherules were collected at the northern part of the Bihar mountains (Bihar) and the southern part of the Kodru mountains (Codru). They have high concentration of Fe and traces of Cu and Zn (Fig. 8 and 9).

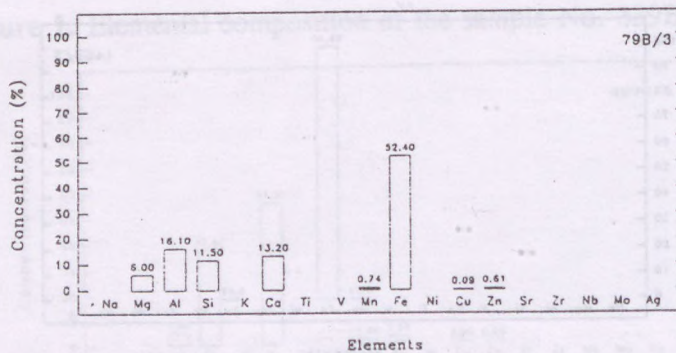


Figure 8. Elemental composition of the sample No. 79B/3

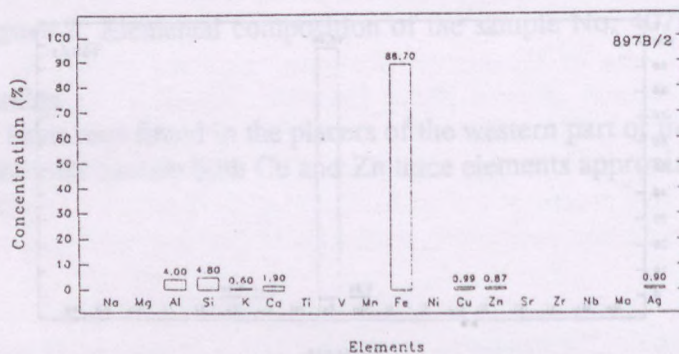


Figure 9. Elemental composition of the sample No. 897B/2



### 3.3.3. Perfectly disblended siderospherules

The samples were collected on the Kaba meteorite fall area. The iron is perfectly disblended from the silicate material. The crust of the sample is iron, having traces of Si, Ca, Ti, Mn and Zn (Fig. 10). The interior silicate kernel has major elements such as Si, Al, Ca and Fe, and trace elements Mn, Zn and Ti (Fig. 11).

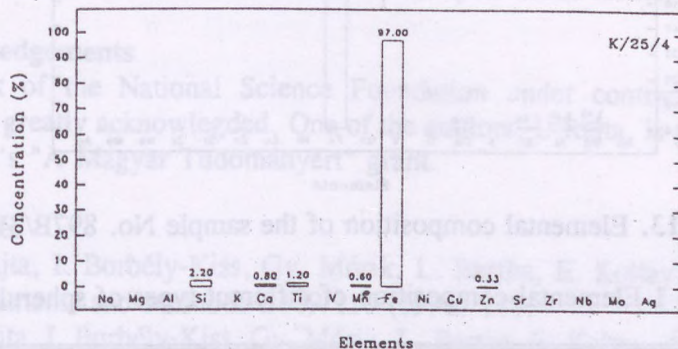


Figure 10. Elemental composition of the sample No. K/25/4 (crust)

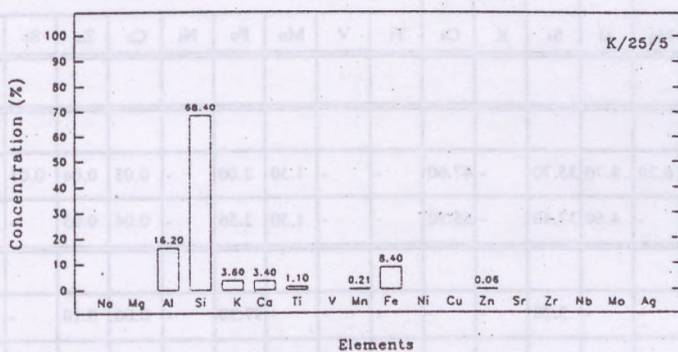


Figure 11. Elemental composition of the sample No. K/25/5 (kernel)

### 3.3.4. Imperfectly disblended siderospherules

The spherules were found at the area of the Kodru mountains (Codru). The amount of Si and Fe is almost the same in the silicate material, as well as the Al and Ca. Traces of Cu and Ti have been found (Fig. 12). The iron octahedra contains little amount of Mg, Al, Si, Ca and Zn. It is interesting that Cu appears in the silicate material while Zn is in the iron octahedra (Fig. 13).

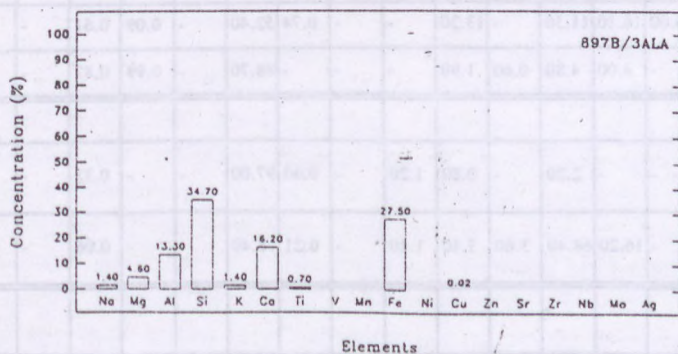


Figure 12. Elemental composition of the sample No. 897B/3ALA

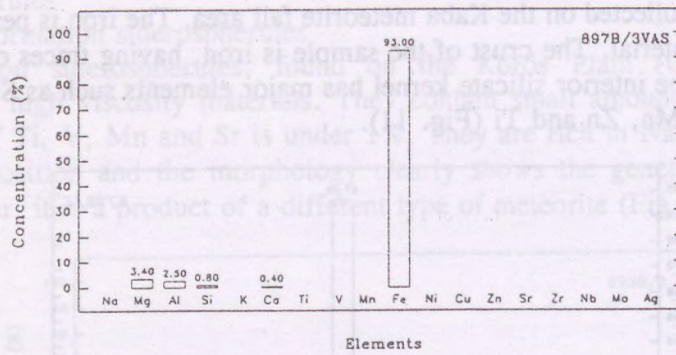


Figure 13. Elemental composition of the sample No. 897B/3VAS

Table 1 Elemental composition of different types of spherules

Elements in %	EDAX results																-	-	-	-	-	-	-	-
	EDAX results						PIXE results										-	-	-	-	-	-	-	
	Na	Mg	Al	Si	K	Ca	Ti	V	Mn	Fe	Ni	Cu	Zn	Sr	Zr	Nb	Mo	Ag						
Spherule types																								
I. Tektites																								
339B/3/2	-	6.20	8.70	35.70	-	47.00	-	-	1.50	2.00	-	0.03	0.04	0.03	-	-	-	-						
407B/5	-	-	4.60	37.40	-	55.50	-	-	1.50	2.56	-	0.04	0.05	-	-	-	-	-						
II. Magnetospherules																								
823B/2	-	-	-	2.70	-	-	-	-	-	97.30	-	0.06	0.10	-	-	-	-	-						
897B/1	-	-	-	-	-	-	-	-	-	98.70	0.40	-	-	-	-	-	-	-						
145B/3	-	-	-	-	-	-	-	-	0.21	99.10	-	-	0.06	-	-	-	-	-						
79B/1	-	-	-	-	-	-	-	-	0.69	98.60	-	0.11	0.06	-	-	-	-	-						
III. Siderospherules																								
Tektite Formation																								
823B/1	4.40	8.90	13.40	35.60	2.20	27.60	0.70	0.14	0.15	6.68	0.01	0.02	0.07	0.10	-	-	-	-						
Non Disblended																								
79B/3	-	6.00	16.10	11.50	-	13.20	-	-	0.74	52.40	-	0.09	0.61	-	-	-	-	-						
897B/2	-	-	4.00	4.80	0.60	1.90	-	-	-	88.70	-	0.99	0.87	-	-	-	-	-0.90						
Perfectly Disblended																								
a. crust K/25/4	-	-	-	2.20	-	0.80	1.20	-	0.60	97.00	-	-	0.33	-	-	-	-	-						
b. interior silicate kernel K/25/5	-	-	16.20	68.40	3.60	3.40	1.10	-	0.21	8.40	-	-	0.06	-	-	-	-	-						
Imperfectly Disblended																								
a. silicate material 897B/3ALA	1.40	4.60	13.30	34.70	1.40	16.20	0.70	-	-	27.50	-	0.02	-	-	-	-	-	-						
b. iron octahedra 897B/3VAS	-	3.40	2.50	0.80	-	0.40	-	-	-	93.00	-	-	-	-	-	-	-	-						

#### 4. Conclusions

EDAX and PIXE are complementary methods and their combined application proved to be very useful for the investigations of spherules. It is surprising that the investigated spherules, collected in different sites and having different genetics, contain either Cu or Zn or both. In the disblended spherules Zn linked to the iron material, while Cu stays in the silicate environment. The Cu and Zn content of the samples needs further investigations.

#### 6. Acknowledgements

The support of the National Science Foundation under contracts A080, T017040 and T014058 is greatly acknowledged. One of the authors, I. Rajta, is grateful to the Hungarian Credit Bank's "A Magyar Tudományért" grant.

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#### Introduction

The measurement of the composition of spherules is one of the most important steps in the process of determination of their origin. Therefore, the spherules are widely investigated by different analytical methods such as nuclear absorption analysis (NAA) [1,2], electron microprobe with X-ray spectroscopy [3] and proton microprobe techniques (PIXE) [4,5]. Mass spectrometry has also been used (e.g. with thermal ionization [6,7] or with ion microprobe [8]), especially for studying isotopic compositions.

The aim of our investigations is to apply time-of-flight mass spectrometry with laser ionization to the elemental analysis of different spherule samples. This method has several very advantageous characteristics. In principle the laser ionization can evaporate and ionize any solid material. Therefore, the spherules can be directly analysed without any special preparation except fixing them on a substrate. The TOF MS provides a complete mass spectrum detecting all the elements in each laser shot. The tight focusing of the laser beam enables the analysis of the samples with high spatial resolution. Moreover, by shooting onto the same spot many times the material can be sputtered, so depth profiling can be carried out. However, the intensity, energy density and spot size of the laser radiation are of crucial importance which must be well controlled so as to be able to obtain linear and non-selective ion detection. This method can be extended with the use of tunable laser radiation, when extremely high sensitivity can be reached by resonance ionization for certain selected elements.

4. Conclusions

EDAX and PIXE are complementary methods and their combined application proved to be very useful for the investigations of speckles. It is surprising that the investigated speckles, collected in different sites and having different genetic, contain either Cu or Zn or both. In the disintegrated speckles Zn linked to the iron material, while Cu stays in the silicate environment. The Cu and Zn content of the samples needs further investigations.

6. Acknowledgements

The support of the National Science Foundation under contracts A080, T017040 and T014028 is gratefully acknowledged. One of the authors, F. Rajta, is grateful to the Hungarian Credit Bank's "A Magyar Tudományos" grant.

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Sample	EDAX													
	Si	Al	Fe	Cu	Zn	Ca	Mg	S	P	K	Na	Cl	Br	I
1	10.5	1.2	15.8	0.5	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
2	12.1	1.5	18.2	0.6	0.3	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
3	11.8	1.4	17.5	0.5	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
4	13.2	1.6	19.1	0.7	0.4	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
5	12.5	1.5	18.5	0.6	0.3	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
6	11.9	1.4	17.8	0.5	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
7	13.5	1.7	19.5	0.8	0.5	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
8	12.8	1.6	18.8	0.7	0.4	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
9	11.7	1.3	17.2	0.4	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
10	13.1	1.6	19.0	0.7	0.4	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3

# Investigation of the elemental composition of microspherules by laser ionization mass spectrometry – preliminary report

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## Abstract

We have applied Laser Ionization Mass Spectrometry (LIMS) to analyse magnetic and non-magnetic microspherules found in geological samples which had been collected in several locations in Hungary. We used a home-built Time-Of-Flight Mass Spectrometer (TOF MS), together with an excimer laser for the evaporation and ionization of the solid materials. We have tested two methods for fixing the spherules on Si substrates so as to manipulate them in the MS. Firstly polystyrene dissolved in benzene was used as a glue, and secondly the spherules were embedded in an indium film. Both methods proved to be applicable to fasten the spherules to the substrate. Moreover, the background levels originating from the fixing layers were reasonably low. In the first experiments of this kind we have detected the basic components of several spherules belonging to both the magnetic and the glassy types, and some qualitative information has also been obtained on certain impurity constituents of the samples.

## Introduction

The measurement of the composition of spherules is one of the most important steps in the process of determination of their origin. Therefore, the spherules are widely investigated by different analytical methods such as nuclear activation analysis (NAA) [1,2], electron microprobes with X-ray spectroscopy [3] and proton microprobe techniques (PIXE) [4,5]. Mass spectrometry has also been used (e.g. with thermal ionization [6,7] or with ion microprobe [8]), especially for studying isotopic compositions.

The aim of our investigations is to apply time-of-flight mass spectrometry with laser ionization to the elemental analysis of different spherule samples. This method has several very advantageous characteristics. In principle the intensive laser radiation can evaporate and ionize any solid material. Therefore, the spherules can be directly analysed without any special preparation except fixing them on a substrate. The TOF MS provides a complete mass spectrum detecting all the elements in each laser shot. The tight focusing of the laser beam enables the analysis of the samples with high spatial resolution. Moreover, by shooting onto the same spot many times the material can be sputtered, so depth profiling can be carried out. However, the intensity, energy density and spot size of the laser radiation are of crucial importance which must be well controlled so as to be able to obtain linear and non-selective ion detection. This method can be extended with the use of tuneable laser radiation, when extremely high sensitivity can be reached by resonance ionization for certain selected elements.

### Experimental arrangement

The experimental arrangement [9] (Fig. 1) is based on a TOF MS equipped with an ion reflector, which was designed and built in the KFKI Research Institute for Atomic Energy. The spherules mounted on a silicon wafer slice are placed into the ion source section of the MS, and an excimer laser is used for the ablation and ionization of the solid material. The sample is manipulated either by a manual manipulator or by a stepper-motor-driven XY-table. The solid surface is illuminated at an incidence angle of  $45^\circ$  by the focused beam of the pulsed XeCl ultraviolet excimer laser (pulse duration 20 ns, wavelength 308 nm). The laser beam is focused to a small spot of about 30-50  $\mu\text{m}$  diameter on the sample. This is achieved by focusing the beam onto an aperture, which is imaged by a lens of short focal length onto the sample surface.

The ions are accelerated by an extractor electrode into the drift tube of the TOF MS. The ion beam is collimated by an electrostatic lens along the flight path. The ions are turned back by an ion reflector, which consists of a series of electrodes generating a uniform potential increase from -3.5 kV to +0.5 kV. The faster ions, penetrating deeper into the ion reflector, suffer a longer time delay than the slower ions. Therefore, the ion reflector compensates the broadening of the ion packets of certain masses, which is due to the initial velocity spread in the laser plasma. Consequently, the mass resolution of the MS is good ( $m/\Delta m > 300$ ) despite the relatively high energy spread of the ions in the laser plasma. The ions are detected by an electron multiplier; the signal is measured by a transient recorder and a fast digital oscilloscope; and the data are processed by a personal computer.

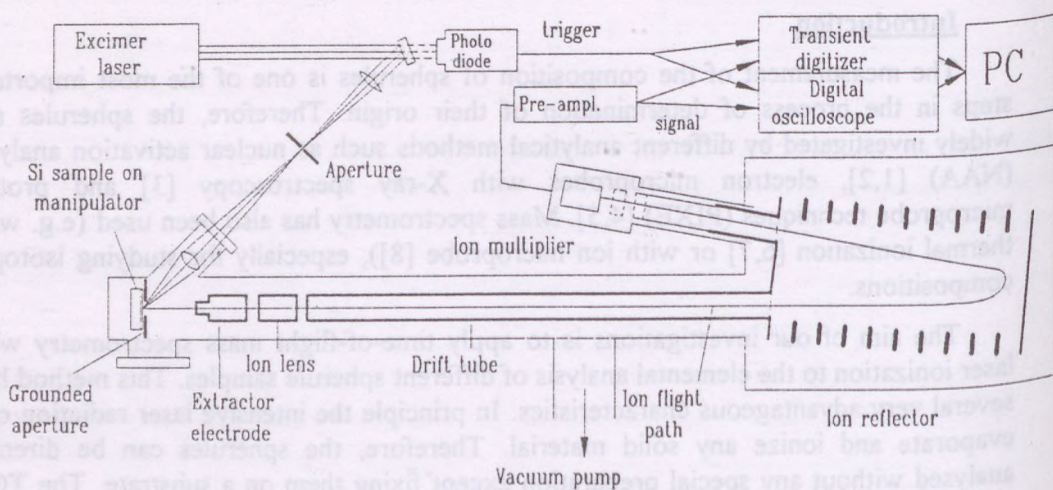


Figure 1. Scheme of the laser ionization mass spectrometer set-up [9].

### Results of the experiments

In our experiments we aimed to analyse a number of spherules, which were found in several locations in Hungary and belonged to the following types:

1. Magnetic spherules collected in the area of the 1857 meteorite fall near Kaba (from the point of view of the geological chronology: Recent). The studied species were black, spherical, and their diameters were between 0.2-0.4 mm.
2. Magnetic spherules found in fire clay samples from the Upper Oligocene quarry near Felsőpetény (age 30 million years). The analysed spherules were black with brownish stains (which seemed to be oxidized parts), one of them had a crater; and their dimensions were between 0.2-0.3 mm.
3. Glassy spherules found in gray limestone samples near Aszófő; age: Middle Triassic (220 million years), Anisian stage, Pelsonian substage, Balatonites balatonicus zone. The investigated ones were light or dark yellow, perfect spheres of diameters between 0.2-0.3 (max. up to 0.4) mm.

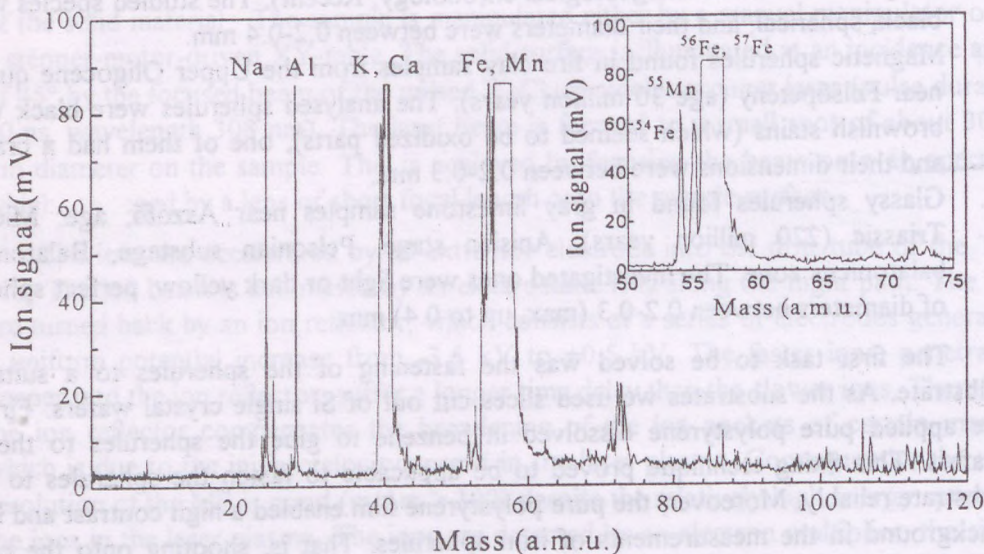
The first task to be solved was the fastening of the spherules to a suitable substrate. As the substrates we used slices cut out of Si single crystal wafers. Firstly we applied pure polystyrene dissolved in benzene to glue the spherules to the Si wafers. This fixing technique proved to be applicable to fasten the spherules to the substrate reliably. Moreover, the pure polystyrene film enabled a high contrast and low background in the measurements of iron spherules. That is, shooting onto the pure polystyrene surface with moderate laser intensity we obtained no ion signals, while measuring with the same settings on the magnetic spherules provided measurable mass spectra. However, for the ablation of the glassy spherules we had to apply higher laser intensity. In this case the mass peaks originating from the polystyrene material near the spherule were sometimes not negligible, or at least not unambiguously distinguishable.

In order to discriminate between the mass peaks originating from the spherule and from the "glue", we tried to use another material for fixing the spherules. We chose indium, which is soft enough to embed the spherules. Furthermore, indium, being an element rather than a compound, provides a distinct mass spectrum, unlike polystyrene, which gives rise to a number of mass peaks belonging to different molecules, which are very difficult to identify.

Our experiments with indium were very promising, because indium was also applicable to fixing the spherules reliably, and it provided an excellent contrast for the spherules in the measurements. Having a low ionization potential of 5.8 eV, indium was very easy to ionize by the laser radiation. Therefore, if the laser radiation hit the In layer, the In signal was always predominant in the mass spectra. In most of these cases the mass spectra also contained other peaks originating from impurities of the In layer. However, the In peaks were always the most intensive. Consequently, if no In peaks could be seen in the mass spectra, it proved unambiguously that there was no signal originating from the fixing layer at all.

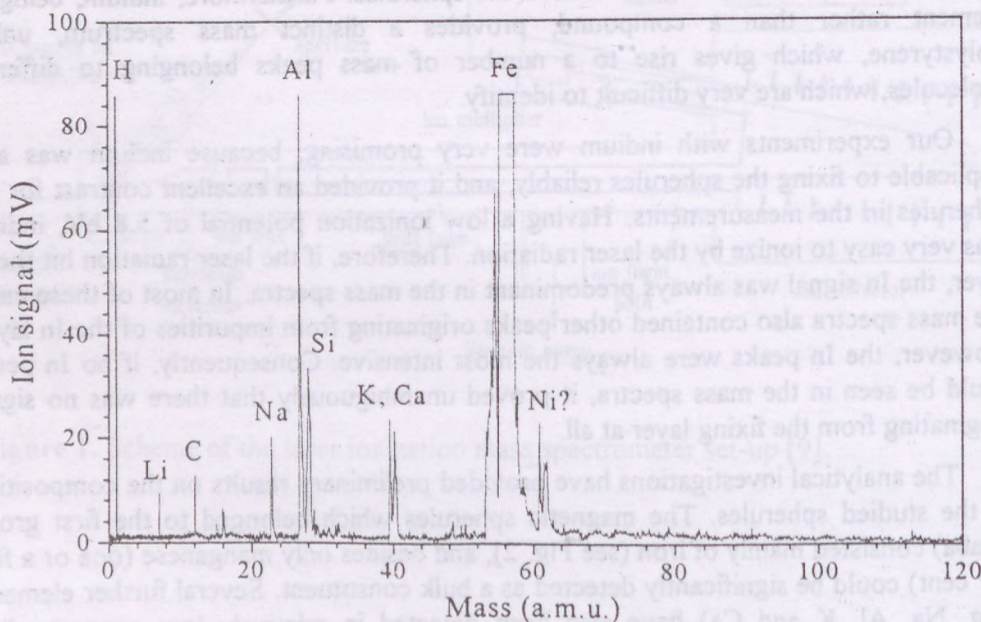
The analytical investigations have provided preliminary results on the composition of the studied spherules. The magnetic spherules which belonged to the first group (Kaba) consisted mainly of iron (see Fig. 2), and besides only manganese (one or a few per cent) could be significantly detected as a bulk constituent. Several further elements (e.g. Na, Al, K and Ca) have also been detected in relatively low concentration, predominantly in the first few shots onto a certain spot on the spherule, which can be

the consequence of their presence on or near the surface of the spherules, probably as surface impurities. In addition, several peaks of relatively higher mass values have also been detected with variable reproducibility, which could perhaps be attributed to the appearance of different iron-oxide molecules or clusters.



**Figure 2.** Mass spectrum obtained on a magnetic spherule of the first type (Kaba) in one of the first few shots onto a spot. The inset shows the non-zero part of another mass spectrum originating from a deeper layer of the spherule.

The magnetic spherules that belonged to the second group (Felsőpetény) also consisted mainly of iron, but some characteristic differences could be observed (Fig. 3). Firstly, the appearance of the mass peak of hydrogen seemed significant in these

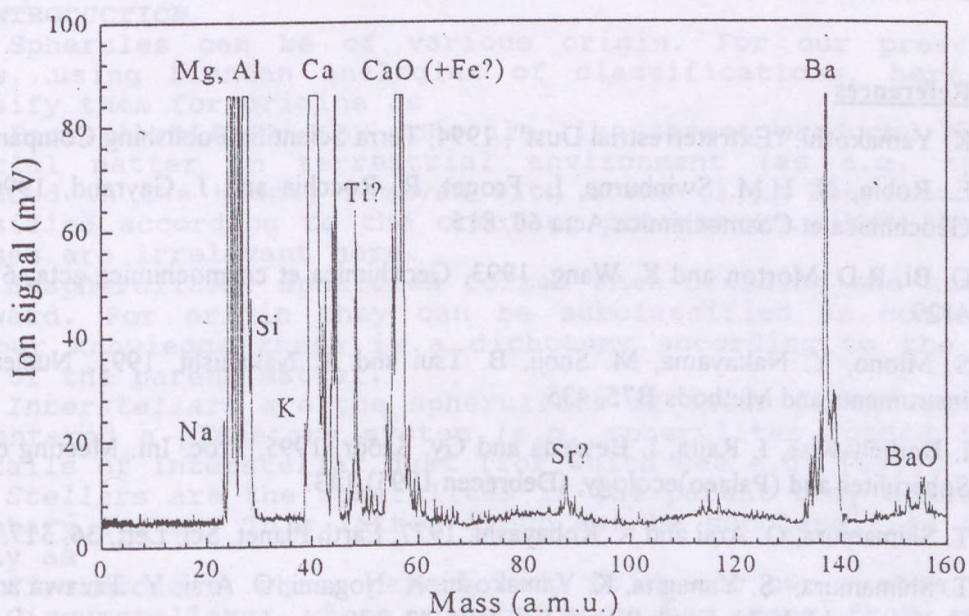


**Figure 3.** Mass spectrum measured on an iron spherule originating from Felsőpetény.



spherules. This feature proved to be distinctly different from the characteristics of the spherules belonging to the other groups. Besides, Li, Na and Al have been detected with reasonable reproducibility. In addition, the appearance of several further elements (e.g. C, Si, K, Ca, Mn, Ni) could be assumed in certain laser shots, but their reproducibility was rather poor and the identification should also be checked.

The glassy spherules of the third group (Aszófő) seemed to consist mainly of calcium (see Fig. 4), and significant amounts of Na, Mg, Al, K, CaO, Ba and BaO constituents could be detected. The mass peak of CaO coincides with that of the dominant isotope of iron, so it is difficult to decide whether there is iron in the spherules, but the presence of a low concentration of Fe can be assumed on the basis of the appearance of its other isotopes. In some laser shots the mass peaks of Li and Si appeared. The presence of low amount of Ti and Sr could also be assumed, but their identification should yet be confirmed.



**Figure 4.** Mass spectrum obtained on a glassy spherule of the Aszófő-type.

It should be noted that these glassy spherules probably have lower absorption for the laser radiation and also higher binding energy than the iron spherules. Therefore, they had to be measured with increased laser intensity. In this case it is more difficult to retain the linearity of the ion detection. Consequently, the favourable laser intensity with tolerably small spot size could hardly be achieved. Therefore, the reproducibility was somewhat poorer in this case. Moreover, we cannot exclude the possibility that some constituents having high ionization potential could not be significantly detected.

Our main purpose in the further experiments is to increase the laser intensity while retaining the pulse energy, so as to detect the components of different ionization potential values with uniform sensitivity. One possible solution to this problem is the tighter focusing of the laser beam, which raises considerable experimental difficulties, especially in case of the excimer laser. A more promising development can be the application of extremely short laser pulses (under 1 ps of pulse length), in which case

the intensity can be very high while the overall pulse energy can be kept reasonably low. Such an experimental arrangement would result in significant improvement and would provide new possibilities in the measurements.

### Acknowledgements

The spherule samples were provided by the researchers of the Geological Institute of Hungary, whose substantial contribution is gratefully acknowledged. The field reworking of the area of the meteorite fall near Kaba was carried out by Gy. Don and P. Solt. The magnetic spherules originating from Felsőpetény were collected by P. Solt, Gy. Don and Cs. Detre. The glassy spherules found near Aszófő were collected by L. Dosztály. The work was financially supported by the Hungarian Space Office, and the Hungarian Scientific Research Fund ("OTKA") under the contracts T-014958 and T-016472.

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## ON INTERPLANETARY AND INTERSTELLAR SPHERULES

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### ABSTRACT

According to statistical analyses based on the new Catalog of the Antarctic Meteorites compiled by Yanai, Kojima & Haramura detailed maps of compositios can be given for meteorites of the Solar System. Spherulites falling to the unpopulated regions can be suspected as of planetary, interplanetary or interstellar origin.

### 1. INTRODUCTION

Spherules can be of various origin. For our present purposes, using Linnéan analogies of classifications, here we can classify them for origins as

*Paraspherulites*: any spherule-like object produced from terrestrial matter in terrestrial environment (as e.g. spherules produced in the kidney removed with urine [1]). They can be subclassified according to the creating process &c., but these subclasses are irrelevant here.

*Euspherulites*: spherules formed when crossing the atmosphere downward. For origin they can be subclassified as comes below. For our knowledge there is a dichotomy according to the prehistory of the parent matter.

*Interstellars* are the spherulites if their parent matter has not entered a planetary system (e.g. spherulites formed from the crystals of interstellar dust (for which see e.g. [2])).

*Stellars* are the spherulites if the parent body comes from a planetary system. This latter group can be subclassified dichotomously as

*Circumsolars*: whose parent body is in our own system.

*Circumstellars*: whose parent body has come from an alien system.

In addition both groups can be subclassified, but now we show only the Circumsolar group. Namely it divides into 3 groups as

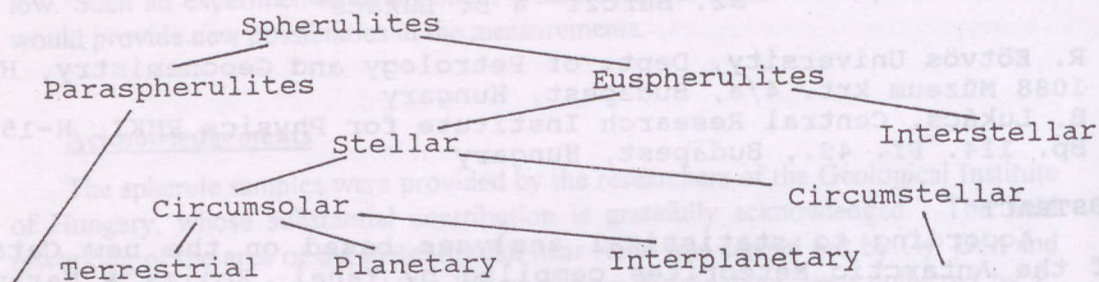
*Terrestrials*: produced from terrestrial matter but under cosmic conditions. E.g. if tectites reenter the terrestrial atmosphere, spherules are formed in the same way as at meteorite entries.

*Planetaries*: from the planetary bodies of our system.

*Interplanetaries*: from meteorites, dust &c. of the Solar System produce spherulites in the usual way.

Hopefully this classification is complete. Now, if spherulites are studies for astronomical information, then these classes must be distinguished from each other.

The scheme reads as follows



The structure of the scheme reflects the fact that circumstellar matter composition is more similar to the planetary and interplanetary ones than to that of interstellar dust, because both the Solar System matter and the circumstellar one have undergone condensation and gravitational selection, but interstellar dust has not.

It is, perhaps, easy to recognise some types of Class 1 (Paraspherulites), since the way of creation is different from all others (Euspherulites). However the shapes and surfaces of all Euspherulites are similar, because they have suffered the same mechanism when crossing the terrestrial atmosphere. So they may differ only for composition.

In this paper we give some criteria to distinguish among Euspherulites for composition.

## 2. PRIMORDIAL AND DIFFERENTIATED COMPOSITIONS

The primordial or cosmic matter has a characteristic abundance of atoms or nuclei, which is shown in Fig. 1. The abundance shows two roughly exponentially decreasing curves, the higher for even nuclear charges, the lower for odd ones. At some nuclei peaks are seen. All these characteristics have simple explanations from cosmology and nuclear physics. Without unnecessary details, the stellar nucleosynthesis started with more than 90 % H, some % He and anything else below 0.1 % (number %'s). Then, as textbooks state [3]:

The first step is  $4\text{H} \rightarrow \text{He}$ , possible in any new stars above cca. 0.05 solar mass. Then He concentration is growing. When H is sufficiently depleted, this reaction stops.

The second step is  $3\text{He} \rightarrow \text{C}$ , with parallel reactions into N and O. Because of high Coulomb barrier this reaction is possible only in stars of substantial mass. Between He and C there is practically no possible endproduct. To understand the reason let us use the notations: Z for proton number and N for neutron number in a nucleus. Then quantum statistics would prefer  $N/Z=1$ , but the Coulomb energy of protons weaken the binding, so

$$d(N/Z)/dN > 1$$

So from He upwards stable nuclei with  $Z < N$  are rare. Now, actually, after the first step one has only He ( $N/Z=1$ ) and H ( $N/Z=0$ ) in relevant quantities. So, by using only He nuclei, one can get  $Z=N$  nuclei of Be and C, and the first one is unstable, by using H and He one would get  $Z > N$  nuclei, all unstable. The only alternative is the inverse  $\beta$  decay of a proton, which is a very unprobable reaction. So at the end of the second step, possible only in the minority of the steps, the product is (C,N,O). (2.1)

The third step starts from (C,N,O), with some (H,He). The most probable endproduct is (Mg,Al,Si), with the neighbours.

Henceforth the further steps are somewhat mixed with each other. However the final step of normal nuclear reactions is the ferrous triad (Fe,Co,Ni) since there the binding energy/nucleon is maximal.

So one expects an exponentially descending curve with plateaus at (C,N,O), (Mg,Al,Si) and (Fe,Co,Ni). In addition some nuclei are exceptionally strongly bound (magic numbers) and then the abundance is enhanced there. Such nuclei are, e.g. Ca and Pb. Finally, more bound clusters are possible in nuclei with even Z or N than for odd ones, and this is the reason for the two parallel curves. Then

Fig. 1 is understood, and, indeed, is characteristic for the whole Universe at large scales today. In earlier times the curve was structurally similar, but descended more rapidly.

Now, in solid matter He is practically absent and H is depleted. It seems that C and N are depleted too, because they are moderate to form crystallizing compounds, but O is not depleted at all, and the next triad + Fe are proper atoms to participate in crystals. But the actual abundance ratios depend very much on the condensation conditions.

Fig. 2 is a comparison between abundances of H, (primordial), C, N & O (second fusion step), Mg, Al & Si (third step) and Fe (final step) in the Universe and in the terrestrial crust. Paraspherulites will have very peculiar composition, and Terrestrials will have more or less that of the crust or of special rocks as basalt &c. If a spherulite composition is not very peculiar or complicated, but is far from terrestrial, then it is probably Interstellar, and if it is not far from terrestrial but definitely different, then it may be Interplanetary or Circumstellar.

Meteorites are either byproducts of planet formation or fragments of planets. So generally Interplanetaries and Circumstellars must resemble more or less meteorite compositions. Since Circumstellars must be rare at Earth, at first anything resembling meteorite compositions must be regarded as Interplanetary, until the opposite is not proven. Characteristic meteorite compositions will be shown in Sect. 4.

Practically nothing is known about chemical compositions in alien planetary systems. We can rely only on common sense, on the fact that chemistry is the same anywhere, and that we do know something about the protostellar steps of contraction from calculations of stellar evolution [4]. We tried with a coherent picture elsewhere [5], but here we can omit the details because spherulites cannot be formed from ices, so anyway some silicates are expected. Isotope dates may help, of course, anything beyond 4.5 My must come from outside the Solar System. However for composition we can mention a clear signal: in any system not contemporary with ours the ratio Fe/(Mg,Si,Al) must be different from the Solar System ratio because this ratio depends on time.

### 3. ON THE CONDENSATION AND METEORITE CLASSIFICATIONS

In this Section we concentrate on Solar System bodies. In our system there was originally a tendential relation between solar distances and chemical compositions, whose explanation was first given by Barshay and Lewis [6]. Omitting all the details, in a mixture of atoms and molecules the composition of condensing

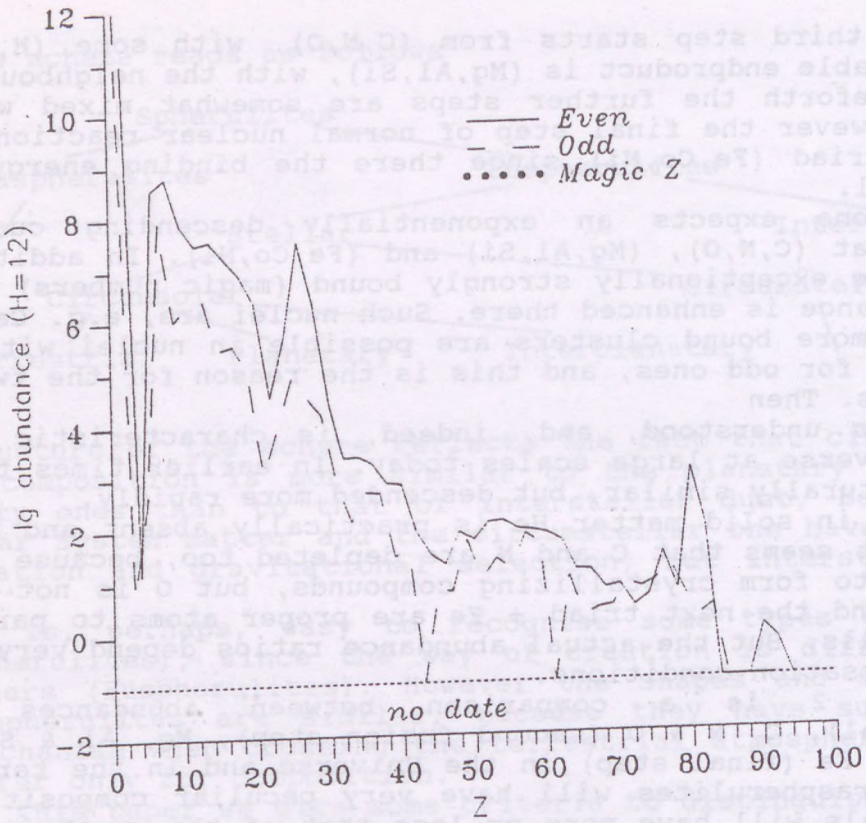


Fig. 1: Cosmic abundances. ..: Magic Z's. Fe is not magic.

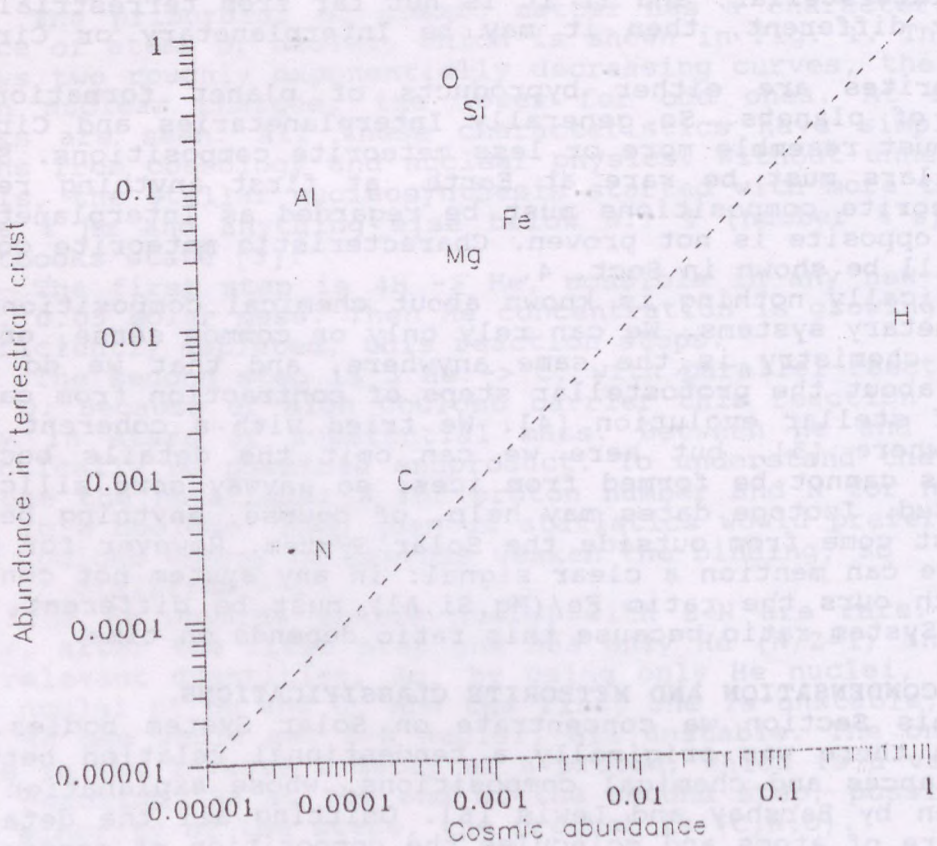


Fig. 2: Cosmic vs. crust abundances of H, C, O and N.

molecules depends on temperature, so on distance. Ref. 6 gave a Table for temperatures above which a molecule cannot condense; it is recapitulated here as

Temp. (K)	Mineral
1600	Refractory oxides
1300	Iron
1200	Enstatite (pyroxene)
1000	Feldspar
1200-490	Olivine
680	Troilite
550	Tremolite
425	Serpentine
<hr/>	
175	water-ice
150	Ammonia-hydrate ice
120	Methane-hydrate ice
65	Methane, Argone ice

As for spherulites, we may forget about matter below the horizontal line. All minerals above the line exist in meteorites so may be important for spherulites too. These temperatures are palaeotemperatures from the time of condensation and it seems that they are rather doubles of the present temperatures in the Solar System [5], [6].

However these compositions are average and primordial ones; for differentiated bodies the surface is different, made of minerals of mass density below the average, and chemical processes may have gone further in the bodies. The first remark is important, but imposes no great difficulty: very probably astronomers know the surfaces of all bodies large enough for being differentiated in the System. (Exceptions are Venus and the giant planets, but thence no spherulites are expected.) As for the chemical processes after condensation, one can learn something from meteorites.

Classifications of chondrites, the main group of meteorites, have been developed in the idea that these meteorites have Solar System origin. This idea was confirmed with radiometric age measurements. The chemical composition of chondrites are in strong relations with each other. Their mineral compositions consist of minerals forming sequences, if slow thermal metamorphosis is imagined as a main process changing the actual mineral composition from a suspected initial, accretional one. Surviving classifications of chondrites are based on thermal evolutionary processes. Two of the classifications played important role: the Prior-Rose-Wiik-Mason-Anders classification and the Van Schmus - Wood classification.

The PRWMA classification (which we call metallurgic one, we shall see, why) first used pyroxenes, but together with Fe contents in oxide, sulphide and metallic phases in order to span a field for their compositions. Fe is the main actor in this play, because this element could change its phase according to thermal evolutionary processes. In 1962 Mason suggested that chondrules had been formed from the groundmass of C1 type carbonaceous chondrites [7]. During this process some of the FeO content of the mainly serpentine groundmass had been transformed. One part of

FeO went to olivine and pyroxenes, the other part of it had been reduced by carbon. Wiik [8] has found (on a restricted amount of data, about 30 selected measurements) that two different total iron contents are characteristic for chondrites: one with cca. 22 weight %, the other with 27 %. Although E, H, L, LL, and C groups of chondrites preserved the original pyroxene-type groups of chondrites in the metal plus sulphide Fe versus oxidized Fe diagram, an evolutionary process of FeO reduction was also involved in the "metallurgic" classification [7], [9], [10].

The second great step in chondrite classification focused efforts on the summarizing of textural characteristics. The idea was recognized in such studies that textures can be arranged gradually, if distinctive features of chondrule obscuring, and the content of some elements or compounds were selected as definitive characteristics of petrologic classes. The six petrologic classes [11] formed textural groups almost "perpendicular" to the "metallurgic" groups. The two systems were different, therefore some combination (i.e. a cross-multiplication) of the earlier and the later one (groups with figure and classes with numbers) gave large number of "boxes" where to collect observational data on textural features of new meteorites. The Van Schmus - Wood table was very useful in focusing attention to the gradual changes, which could have been caused by processes. If a collection of chondrites with representatives of all Van Schmus-Wood boxes were in hands, some processes could become alive for spectators of thin sections in the microscope. This is the necessary condition to be able to see that gradual changes in the texture can be related to the inner processes of a larger body, the parent body. With the presumption that thermal history of a larger parent body is in the background of the transformational processes most of the textural characteristics and changes might be understood as results of partial processes in this thermal history: diffusion, reduction, melting, flowing out of melt, etc. All these happened by the effect of heat inside the parent body during its early history [12].

#### 4. PATHS OF EVOLUTION FOR CHONDRITE GROUPS

In our earlier works we projected the results of suggested metallurgic transformational process of FeO reduction or/and oxidation (i.e. the compositional position in the metallurgic field) onto the Van Schmus - Wood boxes. In the next step we projected the average data of the measurements on Fe related compounds [13] for Van Schmus - Wood boxes onto the metallurgic field, and we had received PATHS of METALLURGIC TYPE TRANSFORMATIONS for the box members inside the metallurgic field. For the present purposes the details of these transformations (mainly suspected only at this moment) are irrelevant; it is enough to know that there are rules for them, i.e. compositions of existing meteorites are not random. This paper gives some ideas how to try to identify extrasolar chondrites according to the Fe/Si and similar basic elementary ratios, initial conditions not only for Solar System materials, but for alien planetary system materials too.

The two great collections of (meteorite and partly planetary) data have been come to existence by the work of NIPR in Tokyo. First 20 copies of a nice collection of Antarctic meteorites have been made, all containing 30 thin sections of different meteorites, forming a small set of meteorites. Then a data set of chemical compositions of more than 550 meteorites, has



been collected and published at NIPR [13]. R. Eötvös University and our Academic Subcommittee on Matter Evolution received these extraordinary valuable data sets and we are working on them.

First a model was established how to arrange all the thin sections into a system, where thermal metamorphic events touched and affected all parent bodies, of which these samples had been originated [14]. In another approach we looked for data about the carbon content of the meteorites, because this abundance, unfortunately, is missing from the NIPR data set. With data of Otting and Zähringer [15] we could project carbon data to the Van Schmus-Wood table [11]. Taking FeO and Fe data from the NIPR set one gets Fe-C-FeO triple-data to all van Schmus-Wood boxes. This way one can project the results of the suggested metallurgic transformational processes of FeO reduction or/and oxidation (i.e. the compositional position in the metallurgic field) onto the Van Schmus - Wood table. By projecting the average values of the Fe containing compounds from Van Schmus - Wood boxes onto the metallurgic field PATHS OF METALLURGIC TYPE TRANSFORMATIONS are found for the box members inside the metallurgic field [14].

This projection gave a great impetus to study the thermal transformational processes affecting meteorite parent bodies. It turned out that chondrule obscuring and final vanishing gave a rather smooth sequence of thermal effects, (the larger Van Schmus-Wood class numbers, the larger heat impact on the parent body. So this sequence serves as a good background (a smooth frame of reference for events) to exhibit detailed steps in the metallurgic process in different chondrite parent bodies. The COMPOSITIONAL PATHS of H, L, and LL chondrites showed first reductions, then oxidation, finally reduction again and flowing out of melt (iron) with the increasing petrologic class numbers. In E chondrites reduction, in C's oxidation seems to ran away. Fe/Si and Mg/Si initial condition parameters suggested different places of origin for the different groups of meteorites' parent bodies.

This work may give aspects and facts to find extrasolar chondrites according to the Fe/Si and similar basic elementary ratios, as initial conditions not only for Solar System materials, but for alien planetary system materials, too.

As told above, all planetary surfaces are more or less known. Mercury is too deep inside Sun's gravity well to expect Mercurian fragments on Earth. Similarly, Venus' thick atmosphere retains the fragments. Lunar rocks are well known for details. Fragments of Mars are found in meteorite collections and the Martian moons are similar to C asteroids. Asteroid fragments are among the meteorites, and beyond the asteroid belt surfaces are generally icy, so the fragments do not survive as spherulites. Therefore if we see the meteorites, we know almost all possible Solar System compositions and surely the possible Interplanetary spherulite compositions.

##### 5. POPULATED AND EMPTY REGIONS IN METEORITE COMPOSITIONS

By the tremendous work of Yanai, Kojima and Haramura [13] now the compositions of 568 meteorites, measured in a homogeneous way, are available. The overwhelming majority of these meteorites has been collected from the Antarctic permanent ice, so preservation was as good as possible and the abundances reflect the original ones as well as possible on Earth. So we cannot do anything else than to regard the sample as representative. Here we give 4 Hertzprung-Russel-type diagrams for the existing compositions.

On the Figures we distinguish among chondrites (the 5 main groups), basalts, other achondrites, other special meteorites (as e.g. the lunar and Martian ones, irons, stony-irons, &c.); for these classes see [13]. 4 "unique" meteorites are omitted, because no statement is known for their origins and so there is a slight possibility that they may not be Interplanetaries.

Fig. 3 is the usual plot: percents of Fe not in oxides (i.e. metallic and sulphide Fe) vs. oxidized Fe. Only now the Fe quantity is normalised to Si, in numbers of atoms. Two branches are seen. The "main branch" runs roughly between 1.6 oxidized Fe/Si and nothing else, and 1.6 not oxidized Fe/Si and no oxide. This suggests total Fe/Si (atomic)  $\approx$  1.6 as initial condition. At the upper end the branch is poorly populated. The "parallel branch" runs at lower total Fe content but not too far. These two branches contain practically all chondritic (i.e. not too much differentiated) meteorites, so they reflect the original meteorite compositions. There is a well-populated area around 0.65 oxidized Fe/Si with almost no other Fe. Otherwise achondrites and "others" thinly populate the strip where oxidized Fe/Si  $<$  0.6 and non-oxidized Fe/Si is a few %. The usual explanation is melting and gravitational differentiation in the parent body: metallic iron has gone down into the center. This would result in Fe meteorites high up along the vertical axis. But other regions of the plot are practically empty.

This fact gives serious constraints on theories of origins of meteorites; but that is not the topic of our paper. (For possible evolutionary paths see [14].) If spherulites appear in the empty regions, their composition does not seem interplanetary, and they may be suspected to be of other origins.

Fig. 4 is the same, but now metallic iron is opposed to Fe compounds. Compared to Fig. 3, the biggest difference is the separation of C chondrites.

Again the two parallel branches are clearly seen. There is a small mixing within the groups L and LL or within C, H and E, but there is practically no mixing between (C,H,E) (main branch) and (L,LL) (parallel branch), achondrites clearly separate, and there is a well-expressed diagonal void between the branches.

Fig. 5 is total Fe vs. Mg, both normalised to Si. Such a plot reflects the initial conditions of meteoritic bodies, at least for chondrites. A chondrite body was never in molten state, because melting would have obliterated the chondrules. Now below the melting point of Fe-C eutectic total Fe, MgO and SiO<sub>2</sub> are invariant.

Then let us see first chondrites. Their region is rather compact. LL's, L's and H's occupy an almost vertical compact region, whence some E's deviate leftward, some C's rightward. Most achondrites form another, horizontal line, while a few achondrites are at the top of the diagram. Here obviously differentiation is seen: liquid iron is lost from some fragments and is accumulated into some others. The large spread in Mg/Si seems to need more complicated differentiation processes. Here we do not go into details; these Figures anyway collect some facts.

Fig. 6 is similar to Fig. 5, but now with only the oxidised Fe. Here the orthogonal structure is obliterated on the right hand side of the chondrites.

There is loss of information if we verbalise the 4 Figures. However one can tell that large areas are empty on the diagrams. For example spherulites at the upper right direction of the main

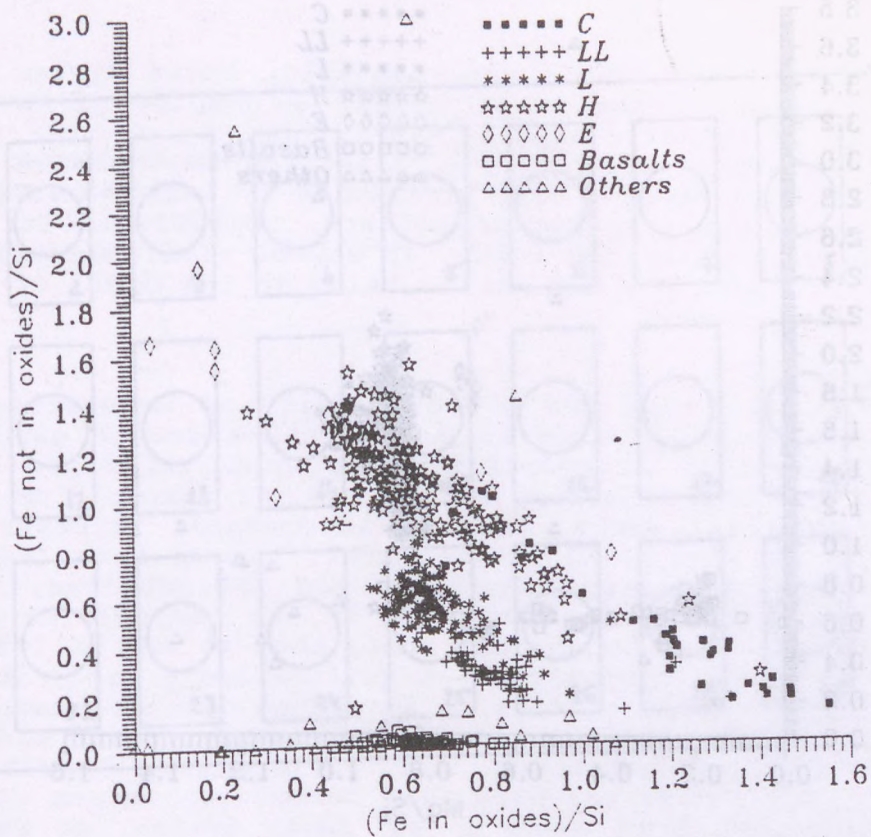


Fig. 3: Oxidised iron vs. other iron.

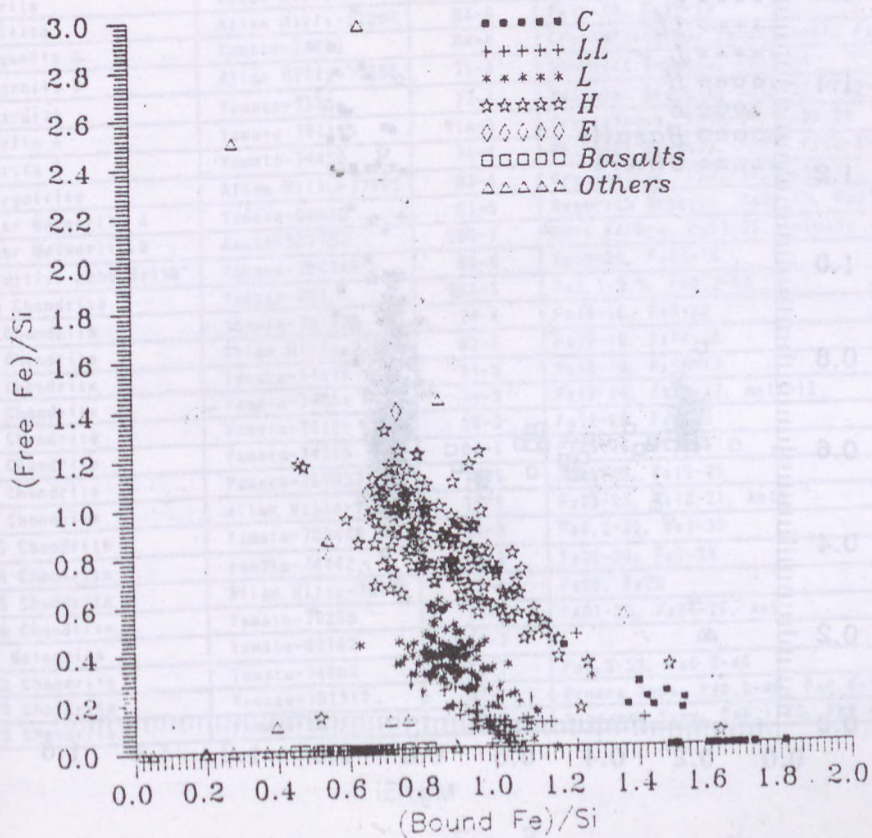


Fig. 4: Metallic iron vs. bound iron.

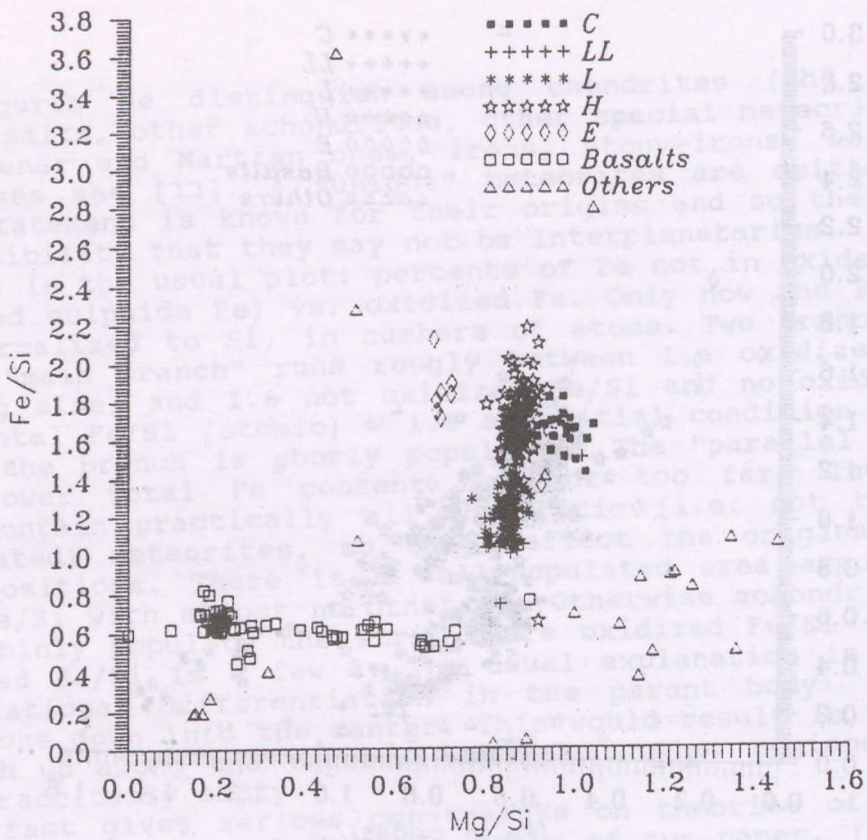


Fig. 5: Total iron vs. magnesium.

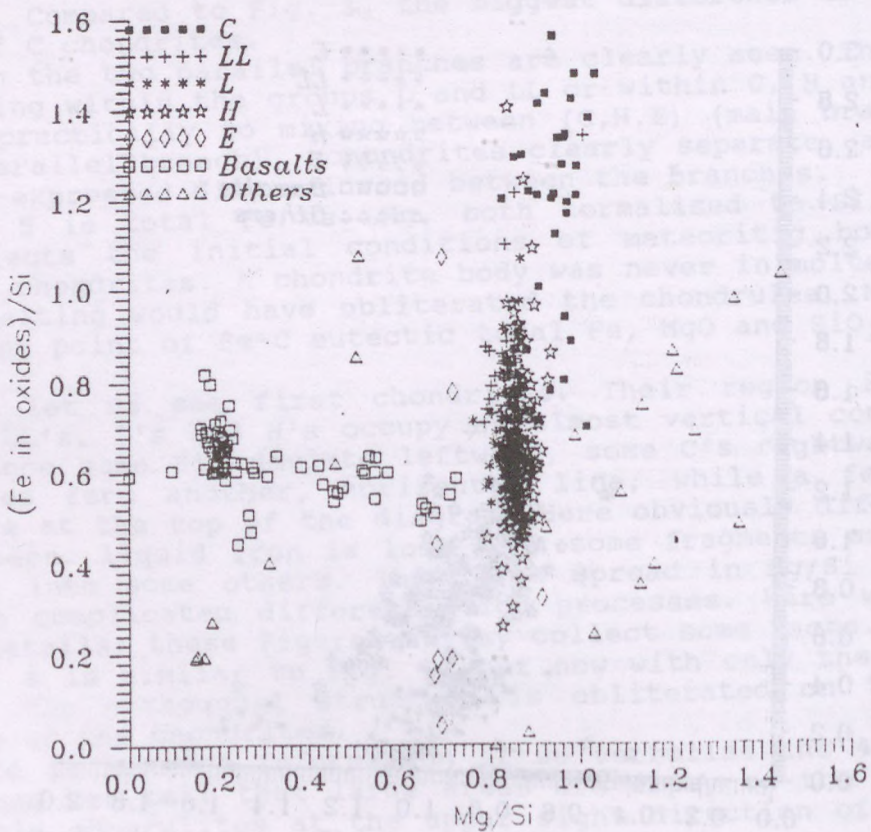
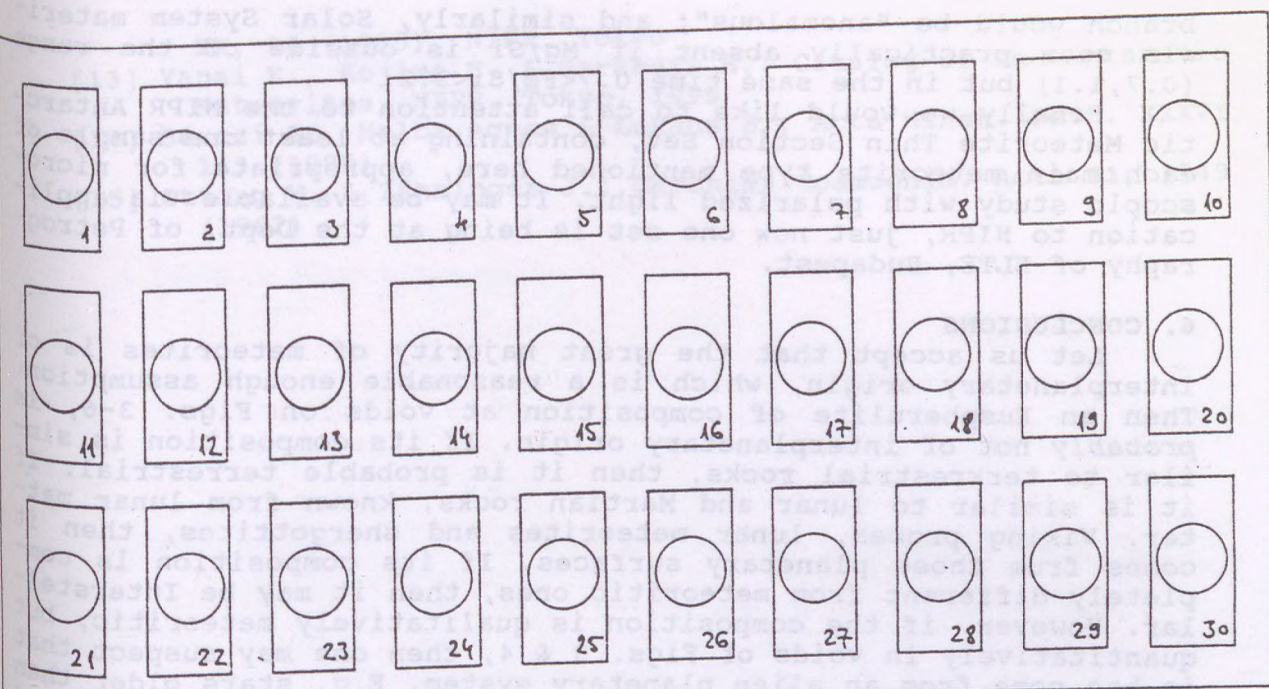


Fig. 6: Oxidised iron vs. magnesium.



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1993.6

No.	Type	Meteorite Name	Sub Number	Remarks
1	Pallasite	Yamato-8451	50A-3	Pyroxene Bearing, Fa9-11, Fs8-9
2	Mesosiderite	Allan Hills-77219	74-5	Fs19-31, An90-96
3	Aubrite	Allan Hills-78113	82-1	Monomict Breccia, Fs0-0.1, An25
4	Ureilite	Allan Hills-77257	64-5	Fa1-14, Fs11-13
5	Diogenite A	Yamato-74097	64-4	Crystalline(Recrystallized), Fs23-26
6	Diogenite B	Allan Hills-77256	71-4	Monomict Breccia, Fs22-25
7	Howardite	Yamato-7308	77-3	Polymict Breccia, Fa15-33, Fs21-57, An85-96
8	Eucrite A	Yamato-791195	61A-3	Crystalline, Fs54-57, An89-92
9	Eucrite B	Yamato-74450	74-4	Polymict Breccia, Fa72, Fs26-31, An78-93
10	Shergottite	Allan Hills-77005	93-4	Crystalline, Fa25-31, Fs20-21, An49-56
11	Lunar Meteorite A	Yamato-86032	51-6	Regolith Breccia, Fa63-93, Fs17-41, An91-97
12	Lunar Meteorite B	Asuka-801757	22B-2	Mare Gabbro, Fa87-95, Px(wide range), An74-96
13	"Primitive Achondrite"	Yamato-794046	53-6	Fa18-20, Fs13-16
14	EH3 Chondrite	Yamato-691	53A-3	Fa0.1-2.5, Fs0.3-20
15	H3 Chondrite	Yamato-791428	74-4	Fa16-18, Fs5-26
16	H4 Chondrite	Allan Hills-77233	82-4	Fa16-18, Fs14-16
17	H5 Chondrite	Yamato-74079	51-3	Fa16-19, Fs15-17
18	H6 Chondrite	Yamato-74014	70-3	Fa18-20, Fs16-17, An11-12
19	L3 Chondrite	Yamato-74191	50-3	Fa12-25, Fs4-25
20	L4 Chondrite	Yamato-74355	84-1	Fa23-26, Fs20-21
21	L5 Chondrite	Yamato-790957	72-5	Fa23-25, Fs19-21
22	L6 Chondrite	Allan Hills-769	75-6	Fa23-25, Fs18-21, An11
23	LL3 Chondrite	Yamato-790448	64-3	Fa0.2-22, Fs1-30
24	LL4 Chondrite	Yamato-74442	62-3	Fa28-30, Fs7-24
25	LL5 Chondrite	Allan Hills-78109	83-4	Fa28, Fs23
26	LL6 Chondrite	Yamato-75258	74-3	Fa31-33, Fs24-25, An9
27	CI Meteorite	Yamato-82162	4-7	
28	CM2 Chondrite	Yamato-74662	50-7	Fa0.2-53, Fs0.5-45
29	C03 Chondrite	Yamato-791717	66-1	Ornans Type, Fa0.2-66, Fs0.6-14, An79
30	CV3 Chondrite	Yamato-86751	52-3	Vigarano Type, Fa0.1-45, Fs0.4-10, An46

Fig. 7.

branch would be "anomalous"; and similarly, Solar System materials seem practically absent if Mg/Si is outside of the range (0.7,1.1) but in the same time  $0.7 < \text{Fe/Si} < 2.2$ .

Finally we would like to call attention to the NIPR Antarctic Meteorite Thin Section Set, containing at least one sample of each main meteorite type mentioned here, appropriate for microscopic study with polarized light. It may be available via application to NIPR, just now one set is being at the Dept. of Petrography of ELTE, Budapest.

## 6. CONCLUSIONS

Let us accept that the great majority of meteorites is of interplanetary origin, which is a reasonable enough assumption. Then an Euspherulite of composition at voids on Figs. 3-6, is probably not of interplanetary origin. If its composition is similar to terrestrial rocks, then it is probable terrestrial. If it is similar to lunar and Martian rocks, known from lunar matter, Viking probes, lunar meteorites and shergottites, then it comes from those planetary surfaces. If its composition is completely different from meteoritic ones, then it may be Interstellar. However, if the composition is qualitatively meteoritic, but quantitatively in voids of Figs. 3 & 4, then one may suspect that it has come from an alien planetary system. E.g. stars older than Sun formed in a younger Galaxy with different Fe/Si ratio, and meteorite parent bodies may have been formed at regions with temperatures different from those in our Solar System, so Fe/Si may be such, which is unpopulated on our Figures. Such bodies are very probably rather rare here, but in the neighbour of Sun there is an abundance of red dwarfs, of which some may be older than Sun.

## ACKNOWLEDGEMENTS

Illuminating discussions with Drs. K. Yanai, H. Kojima & A. K. Pedersen are acknowledged. Partly supported by OTKA N°'s 14958 & 21209.

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**Keywords:** Városmajor - 1 borehole, Eocene-Oligocene boundary, spherules, Bakonyjako - 528 borehole, Upper-Cretaceous, glaucofan spherules

**Abstract:** Spherule content of sequence of Városmajor - 1 borehole and Bakonyjako - 528 borehole was examined.

In the case of Városmajor - borehole 46 pieces of spherules have been found. According to their magnetic properties, shape and colour they can be arranged into four groups. All the spherules have been found in the sediments of Bakonyjako - 528 borehole are glaucofan. Their number is 742.

## Introduction

Purpose of investigating the sequence of two boreholes was to examine if their sediments contain spherules. Formations of Eocene-Oligocene boundary are exposed by the Városmajor - 1 borehole (Fig. 1).

In the case of Bakonyjako - 528 borehole Upper-Cretaceous sediments can be studied (Fig. 2).

There is only one common thing can be found concerning the material of the above mentioned two boreholes: both of them contain spherules.

## Description

### *Spherules from the sediments of Városmajor - 1 borehole*

The number of spherules found here is 44.

According to their magnetic properties, shape and colour they belong into four types (Table 1).





Spherules from the  
Sediments of Borehole Városmajor - 1  
and Borehole Bakonyjákó - 528,  
Hungary

Á. Dávid - A. Rác - É. Koleszár

**Keywords:** Városmajor - 1 borehole, Eocene-Oligocene boundary, magnetic spherules, Bakonyjákó - 528 borehole, Upper-Cretaceous, glassy spherules

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### Description

#### *Spherules from the sediments of Városmajor - 1 borehole*

The number of spherules found here is 44.

According to their magnetic properties, shape and colour they belong into four types (Table 1).

Their size is between 200-400  $\mu\text{m}$  in diameter. The biggest ones belong into 1.1 type. Most of the spherules have been found in sediments from 62,5-120 m (16) and 144,5-163,6 m (11) depths. Glassy spherules occur in the upper ninety meter of the sequence. The deepest part of the borehole where spherule have been found is between 220,8-225,8 m.

### *Spherules from the sediments of Bakonyjako - 528 borehole*

There are 742 spherulites have been found here (Fig. 2). All of them seem to be glassy. Their shape shows great variety: rounded, tear-drop shaped, elongated. They are extremely large (200-1000  $\mu\text{m}$ ). More than 60 percent of them is colourless. The others are yellow or light-brown. Levels between 58,5-58,6 m and 80,5-80,6 m depths are the richest in spherules. There are 248 pieces and 292 pieces have been found here.

In the case of both localities further investigations need to examine the chemical composition and origin of these small particles.

**Table 1**

Classification of spherulites found in the sediments of Városmajor 1 borehole according to their colour and shape

MAGNETIC		NON-MAGNETIC	
BROWNISH-BLACK ROUNDED ①	SHINY, BLACK ROUNDED ②	COLOURLESS ROUNDED ③	YELLOW ROUNDED ④
29,0-35,1 : 5 pc 35,1-41,7 : 2 pc 49,5-53,6 : 1 pc 58,0-62,5 : 2 pc 90,0-95,2 : 3 pc 95,6-96,0 : 4 pc 116,4-120,0 : 3 pc 132,0-135,1 : 1 pc 144,5-146,8 : 5 pc 146,8-147,1 : 3 pc 163,4-163,6 : 3 pc 171,0-173,9 : 1 pc 217,2-218,3 : 1 pc 220,8-221,2 : 3 pc	53,60-57,3 : 2 pc 62,5-68,4 : 2 pc	90,00-95,2 : 1 pc 49,5-53,6 : 1 pc 62,5-68,4 : 1 pc 95,6-96,0 : 1 pc	7,0-9,0 : 1 db
össz: 37 pc	4 pc	4 pc	1 pc

Fig. 1

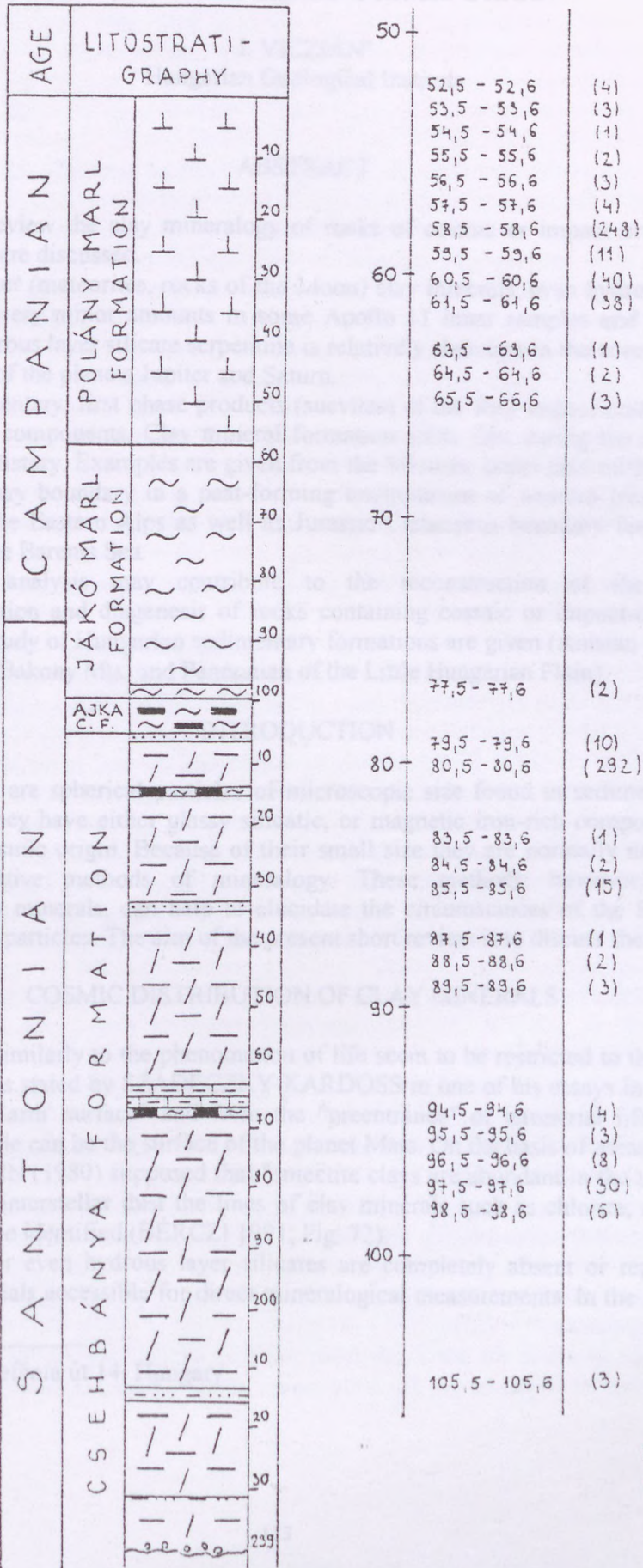
Sequence of the Városmajor 1 borehole

age	for- mation		
Oligocene	KISCELL CLAY	7,0 - 9,0	(1)
		29,0 - 35,1	(5)
		35,1 - 41,7	(2)
		49,5 - 55,6	(2)
		53,6 - 57,3	(2)
	TARD CLAY	58,0 - 62,5	(2)
		62,5 - 68,4	(3)
		90,0 - 95,2	(4)
		95,60 - 96,00	(5)
		116,4 - 120,0	(3)
Eocene	BUDA MARL	132,0 - 135,1	(1)
		144,5 - 146,8	(5)
		146,8 - 147,1	(3)
	163,4 - 163,6	(3)	
	171,0 - 173,9	(1)	
	217,2 - 248,3	(1)	
220,8 - 225,8	(1)		

(5) - number of  
spherulites

Fig. 2

SEQUENCE OF BAKONYJÁKÓ - 528 BOREHOLE



(1), (2) .. - number of spherulites



# THE POSSIBLE ROLE OF CLAY MINERALOGY IN THE STUDY OF MICROSPHERULES OF COSMIC ORIGIN

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## ABSTRACT

In this short review the clay mineralogy of rocks of cosmic or impact origin and of their enclosing sediments are discussed.

In cosmic matter (meteorites, rocks of the Moon) clay minerals, even hydrous layer silicates are extremely rare (very minor amounts in some Apollo 11 lunar samples and in carbonaceous chondrites). The hydrous layer silicate serpentine is relatively abundant in the zone of asteroids and among the satellites of the planets Jupiter and Saturn.

The non-sedimentary, first phase products (suevites) of the Ries impact crater at Nördlingen contain fresh glassy components. Clay mineral formation starts first during the subsequent post-impact sedimentary history. Examples are given from the Miocene crater lake of the Ries structure, the Cretaceous/Tertiary boundary in a peat-forming environment of western North America and marine deposits of the Eastern Alps as well as Jurassic/Cretaceous boundary formations near an impact structure in the Barents Sea.

Mineralogical analysis may contribute to the reconstruction of the conditions of sedimentation, alteration and diagenesis of rocks containing cosmic or impact-derived material. Examples from the study of Hungarian sedimentary formations are given (Anisian of Mecsek Mts., Upper Cretaceous of Bakony Mts. and Pannonian of the Little Hungarian Plain).

## INTRODUCTION

Microspherules are spherical particles of microscopic size found in sedimentary rocks and Recent sediments. They have either glassy silicatic, or magnetic iron-rich composition. They are supposed to be of cosmic origin. Because of their small size they are normally not accessible for traditional determinative methods of mineralogy. These methods, however, including the determination of clay minerals, can help to elucidate the circumstances of the formation of the rocks that contain the particles. The aim of the present short review is to discuss these possibilities.

## COSMIC DISTRIBUTION OF CLAY MINERALS

Clay minerals, similarly to the phenomenon of life seem to be restricted to the surface of the planet Earth. As it was stated by SZÁDECZKY-KARDOSS in one of his essays in 1975, "they are known only on the Earth's surface" and form the "preentrance" of terrestrial life (p. 163). One exception from this rule can be the surface of the planet Mars. On the basis of measurements of the Viking program BANIN (1980) supposed that "smectite clays are abundant in the soil of Mars". In the IR spectra of the interstellar dust the lines of clay minerals such as chlorite, montmorillonite and serpentine could be identified (BÉRCZI 1991, Fig. 72).

Clay minerals or even hydrous layer-silicates are completely absent or represent extreme rarity in cosmic materials accessible for direct mineralogical measurements. In the Apollo 11 lunar

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samples only extremely rare di- and trioctahedral phyllosilicates could be detected by electron diffraction study (DREVER et al. 1970) which were not more closely specified. Among the meteorites the carbonaceous chondrites may contain clay mineral-line phases. In the Orgueil meteorite minor amounts of "pseudo-chlorite" (ORCEL et al. 1972), in the Allende carbonaceous chondrite intergrown mica and montmorillonite and a serpentine-like phase were found (TOMEOKA, BUSECK 1982a,b).

Contrary to true clay minerals the hydrous layer silicate serpentine is relatively abundant in the Solar System in the zone of asteroids and among the satellites of Jupiter and Saturn. Serpentine constitutes e. g. essential part of the 4 Galilean satellites of Jupiter. Its occurrence is typical in a zone which occupies intermediate position between planets constituted mainly by silicates, e. g. Earth, Moon, Mars and those containing much water like Jupiter and Saturn. Serpentine may be a constituent of carbonaceous chondrites and is supposed to occur in comets (BÉRCZI 1991).

Microspherules of cosmic origin can remain fresh over very long periods of time. In glassy microspherules found in Carboniferous of Upper Silesia no water contents could be detected by IR method (MANECKI and SKOWRONSKI 1970). On the other hand, glauconite-bearing microspherules proved to be of diagenetic, not of impact origin in the layers near the Cretaceous/Tertiary boundary at Gubbio, Italy (NASLUND et al. 1986).

#### THE LACK OF CLAY MINERALS IN IMPACT PRODUCTS

Clay minerals are practically absent in particles and rocks formed from terrestrial material by the effect of impact of cosmic bodies.

Tektites and micro-tektites consist of glassy silicate material which may contain also particles of pure  $\text{SiO}_2$  glass (lechatelierite) but are devoid of primary crystallites (GLASS 1990). No hydrous devitrification and transformation of the glass into clay minerals could be observed.

The infilling of the impact crater of Ries at Nördlingen, Germany, was extensively studied (FÜCHTBAUER et al. 1977, LEMCKE 1981). The rocks on the basis of the sequence, called suevite, show effects of shock and melting phenomena but no evidence of subsequent clay mineral formation was found. The same is true for the majority of the overlying so called Graded Unit which is most probably the product of the subaeric fall of the cloud of suevite debris produced by the impact. Alteration products of glass such as montmorillonite, zeolites and calcite appear first in the fine-grained groundmass of the upper part of the Graded Unit (JANKOWSKI 1977b, FÜCHTBAUER et al. 1977) introducing a subsequent lacustrine sedimentation in the crater basin.

#### CLAY MINERALS IN SEDIMENTARY ROCKS CONTAINING IMPACT OR COSMIC MATERIAL

There is a great variety of composition of rocks hosting impact-derived material or particles of cosmic origin.

In the lacustrine sequence of the *Ries impact crater* near Nördlingen, Germany, various sediments are present (SALGER 1977, JANKOWSKI 1977a, 1980, 1981). In the lower part of the sequence glassy detrital material derived from the crater walls predominates in the sediments which has been altered into montmorillonite and zeolites. Diagenetic transformation produced illite from montmorillonite in hypersaline periods of the lake. Detrital clay minerals such as micas, chlorite, kaolinite and montmorillonite derived by weathering of micas, appear only in the upper portion of the sedimentary sequence.

In a distance of about 80 km south from the Ries crater sporadic debris derived from the impact products can be found in the fluvial layers of the Upper Freshwater Molasse horizon of



the Molasse Basin. In the molasse sediments of this zone normal terrigenous detrital clay minerals were found (e. g. near Augsburg, see VICZIÁN 1984), no traces of the impact material can be detected. There are, however, restricted bentonite layers in the molasse sequence of the area, the glassy components of which are remarkably synchronous with the Ries event (Ries:  $14.7 \pm 0.4$  Ma, bentonite: 14.4-14.6 Ma). They were connected with the impact event by several authors (GENTNER and WAGNER 1969, see HEROLD 1970 and VOGT 1980). No direct mineralogical evidence supporting this theory was found except the very sporadic occurrence of diaplectic plagioclase glass (?) in the sediments (HARR 1976). Recently, however, the connection of the Bavarian bentonites with the Ries event was questioned and the source area was located to the Carpathian volcanic region (UNGER, NIEMEYER 1985).

The *Cretaceous Tertiary boundary* is being extensively studied because the theory of a catastrophic impact event on this boundary is widely accepted.

In the *western Interior of North America* a few cm thick clay layer represents this boundary (POLLASTRO, BOHOR 1993). It was deposited in a peat-forming environment. The clay minerals differ from those found in marine K/T boundary sequences due to special circumstances of deposition and alteration which prevailed in this region. The layer consists of two subunits depending from the mechanism of the impact: the lower one, called the "melt ejecta layer" underwent kaolinitic alteration of glassy fragments including hollow spherules (microtektites). The upper one, called "fireball layer" has been altered into smectite. These impact-derived layers differ significantly from other clays found in the sequence, namely from tonsteins and detrital shales.

In a marine K/T transition sequence of the *Northern Calcareous Alps* the boundary clay differs from other clay layers in the sequence. It contains remarkably little quartz and detrital minerals such as plagioclase, micas and chlorite while it is rich in "expandable clays" and kaolinite which are the devitrification products of fine-grained vitric material derived by the impact (PREISINGER et al. 1986, LAHODYNSKY 1994).

The *Jurassic Cretaceous* transition beds were studied in a borehole drilled in the *Barents Sea* which penetrated a marine shelf succession of clay- and siltstones (DYPVIK et al. 1995). The upper part of this sequence above the Early Volgian (=Tithonian) beds contains enhanced quantities of smectite. The authors think that smectite may be the devitrification product of glassy particles derived from the closely located Mjølneir Structure, a possible extraterrestrial impact crater.

In *Hungary* glassy and magnetic microspherules were found in several stratigraphic horizons ranging from Triassic to Recent sediments (DETRE et al. 1995). The clay minerals of the particular host rocks are not yet known. In some instances, however, clay minerals were investigated from the same stratigraphic formation but from another localities. Even so, some conclusions can be drawn concerning the mode of formation of these samples. A few examples are given here:

One of them is the *Lower Anisian Vöröshegy Dolomite Member in the Mecsek Mts.* (RÁLISCH-FELGENHAUER 1995). In this member there is a transition between two clay mineral associations (VICZIÁN 1993). The lower one is illite+Mg-chlorite+corrensit formed in a restricted basin environment, the upper one is detrital illite typical of shallow marine sedimentation.

In the *Upper Cretaceous of the Bakony Mts.* microspherules were found in alluvial sediments of the non-marine Csehbánya Formation as well as in the Ajka Coal Formation and in the marine Polány Marl Formation (SZARKA 1994, BODROGI 1994, 1995, SIEGL-FARKAS and WAGREICH 1994). According to clay mineral analyses Csehbánya Formation and clastic intercalations of the Ajka Coal Formation are characterised by the detrital association of illite-chlorite (VICZIÁN 1988). The Polány Marl is similar but contains more smectite which is a sign of the open marine conditions of sedimentation (VICZIÁN 1987). The possible contribution of a volcanogenic component (VASKÓ-DÁVID 1994) is not yet clear.

Spherules were found in the *Upper Pannonian* deposits of the borehole Nagylózs 1 in the *Little Hungarian Plain* (SZÖÖR and RÓZSA 1995). No clay mineral analysis was performed from this well. Stratigraphically equivalent sediments from another borehole contain detrital terrigenous polymineralic clay mineral association of illite+chlorite in the bulk rock and highly expandable illite/smectite in the  $<2 \mu\text{m}$  fraction (borehole Szombathely II, VICZIÁN 1990).

It is planned to investigate the mineralogy of the samples themselves which contain spherules in order to study the particular conditions of their formation.

## CONCLUSIONS

1. Meteorites and micrometeorites of cosmic origin usually contain no clay minerals, even hydrous layer silicates are rarity. Ejecta produced from terrestrial material by impact of a cosmic body are in most cases of fresh unaltered glassy composition.

2. Diagnostic features of cosmic or impact origin can be detected mainly by morphological or geochemical studies rather than by bulk mineralogical methods.

3. The clay minerals formed later of the cosmic or impact-produced particles reflect the circumstances of sedimentation, alteration and diagenesis of this material. The same is true for sedimentary rocks hosting microspherules or other particles produced by a cosmic event. Bulk mineralogical methods such as X-ray analysis may contribute to the reconstruction of the geological history of these formations.

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## Exploration of spherules in the Kaba meteorite fall area

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### 1. Preface:

In the beginning of investigation of spherules in Hungary in 1994 we suggested field work reviewing some meteorite fall areas. In every reports of meteorite fall observations we can hear or read about detonations, crackings, smoke, etc., which means that there are a lot of little, microscopic particles in the area around the fall. So we had real hope to find some micrometeorites and spherules by collecting soil samples in the area of some well documented meteorite falls.

### 2. The Kaba meteorite fall:

Kaba is a village in the Great Hungarian Plain about 200 km east of Budapest. April 15 of 1857 at ten oclock in the evening some inhabitants noticed a fireball coming from south. Mr. Gabor Szilagyí, who also had seen the spectacular event and heard the detonations too, next morning just riding to his farm north of the village, suddenly noticed a black rock flattened into the sandy road. His horse sprang back from the stone. In the evening on the way home Szilagyí digged out the black stone with his friends and carried it to the magistrate of the village. The principal gave the meteorite to the famous Reform College of Debrecen, which town is near to Kaba. The fall is documented by J. Török/1858/, who was the geography teacher of the College and the first analyses were made by F. Wöhler/1858/. Since then a lot of investigations were made and the meteorite became famous as Kaba III /CV3/ carbonaceous chondrite /A. Hoffer 1928, K.I. Sztrókay 1960, 1961, W.R. Van Schmus and J.M. Hayes 1974, L.P. Keller, P.B. Buseck 1990, etc./

### 3. Field work ;

As the cottage of Szilagyí is already destroyed and the roads and tracks around Kaba changed in the past 140 years, first we had to look for some old maps. In the Library of the Military History Museum in Budapest we could find some excellent documents of military mapping of Hungary 1826-66 and 1861-68., so we could compare the place of the cottage and the roads north from Kaba on modern topographical maps. The possible meteorite fall area, which we have to investigate is about 40km<sup>2</sup>. Our first field work plan consisted of three parts / look the map / :

1. Cross section of the fall direction /1.-2. and 21/27.
2. The area north of Kaba to the cottage of Szilagyí 3.-20.
3. Round the village /28.-52.

The land around Kaba is flat plain with some little lakes, streams and water channels, everywhere sugar beat fields, somewhere fruit gardens. The speed of accumulation of soil is about 10 cm in hundred years, so we tried to collect the samples from 10-15 cm deep. Bellow the humusz and soil there are sand and clay from holocene and upper pleistocene. Up to now we collected 52 samples from different places, each sample 1-1,5 kg.

### 4. Laboratory work:

In the Hungarian Geological Institute from all the samples first the dokumentation samples were separated. After drying, the material was washed with wtaer and then we used hydrogeneperoxide

and hydrochloric acid too for additional cleaning the samples. During sluicing different sieves were used, the finest 60 micron, but we also tried to save the finest, smallest fraction too. We used magnet for to collect the magnetic particles. The sorting and separation was made under microscopes.

#### 5. Results:

After picking we collected up to now more than 400 spherules from 20 to 500 micron / most of them 100-150. micron in diameter/. The particles are rounded, but also droplets, tear drops, dumb-bell and pear forms occur. There are spherules in matrix too. About one third of the material is magnetic and metallic, one third is glassy and one third is scoriaceous.

The distribution of different kinds of spherules in different samples is rather great, so already we could distinguish the probably fall area / see the map /. To have an exact investigation for the future reconstruction of the Kaba meteorite fall area, sample collection by quadratic method is planned by considering the results of our work. First we have to make lot of chemical analyses of the spherules to distinguish; which are from the Kaba meteorite fall, which are other extraterrestrial ones, which are terrestrials and if there are industrials too. The mineralogical investigations are just made by O.Kákay-Szabó, the mineralogist of the Hungarian Geological Institute.

#### 6. Proposal:

By the results of our investigations we can suggest to other meteorite researchers, and research groups for meteorite fall reconstructions.

-Supported by the Hungarian Space Office and the OTKA grant T 014956

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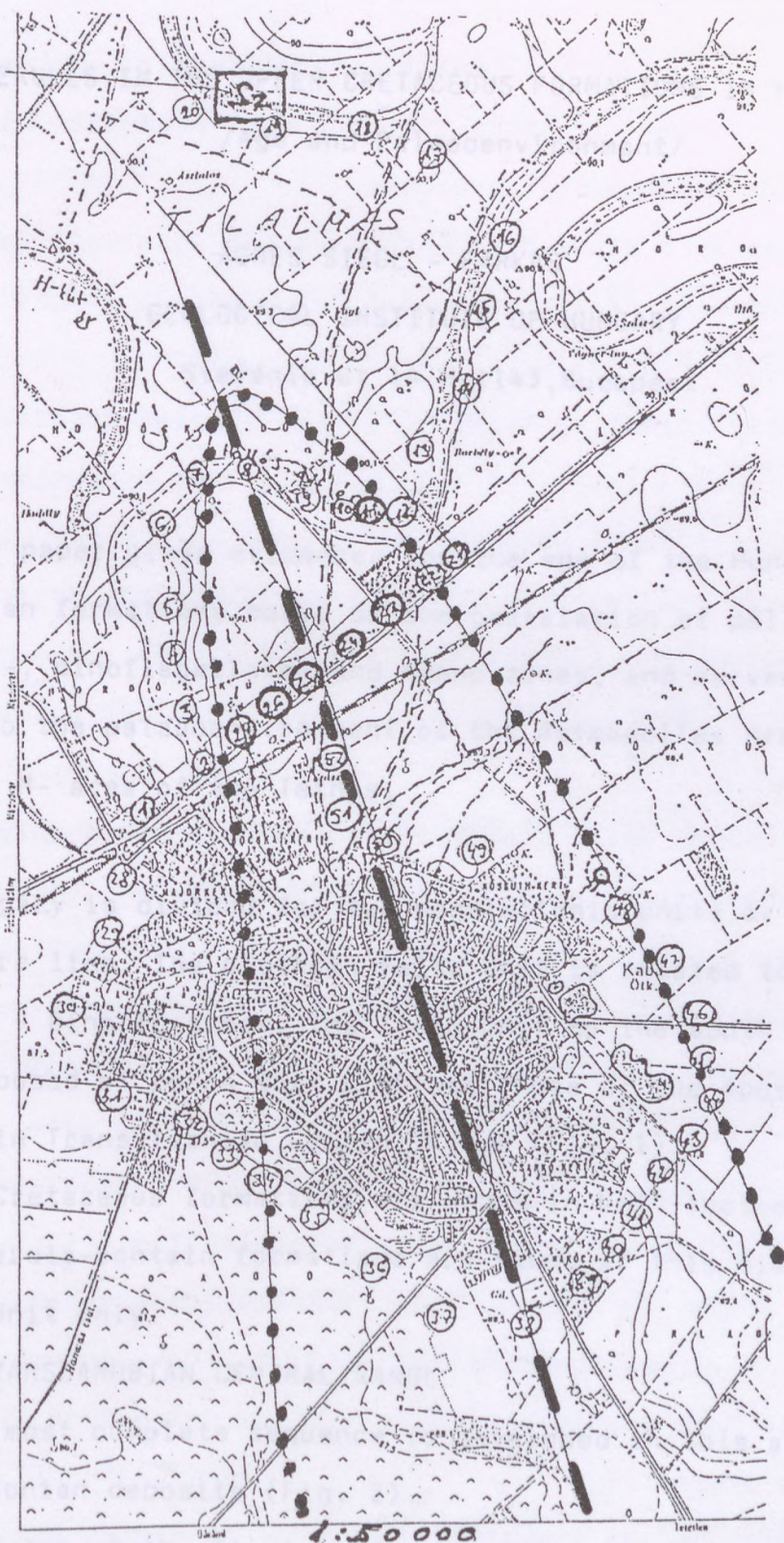


Fig. 1. Map of the Kaba meteorite fall area.

Sz - the site of the Szilágyi cottage, North of Kaba village

--- tracks in 1857

1.-52. Numbers of the sample collecting sites

— — — vector of the fall from  $170^{\circ}$  to  $350^{\circ}$





# SPHERULES IN THE UPPER CRETACEOUS FORMATIONS IN HUNGARY

/Age and Palaeoenvironment/

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The paper gives evidences for the age of the Hungarian Senonian formations, based on the correlation of pollen and spores-, dinoflagellata- and nanno zones, and serves some data to the palaeoenvironment of the Normapolles province on the N- area of the Tethys.

Hungary is divided for two megatectonic units by a SW-NE fracture line. The northern Pelso Unit is related to the Alpine - Dinaric areas, the Tisza Unit to the south of it, is composed of rocks that similare those of the Apuseni Mountains in Transylvanian Central Range (Fig. 1).

Upper Cretaceous formations are known in both tectonic units.

Spherula-contain formations are found at this time in the Pelso Unit only.

## 1.1. TRANSDANUBIAN CENTRAL RANGE

The most complete sequence is developed in this area of the Senonian deposits (Fig. 2).

Here on top of the fluvial (Csehbánya Formation) and/or on

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These investigations are supported by the OTKA grant T 014958

the lacustrine (Ajka Coal Formation) sediments, the marine (Jákó Marl-, Ugod Limestone-, Polány Marl Formation) formations are continuously developed.

Spherules were studied in Csehbánya and Ajka Coal Formation in the area of Magyarpolány (Bakony Mts), in bh. Mp. 42 (Szarka 1991, Bodrogi pers. comm.).

In Csehbánya Formation magnetic spherules were found. Because they contain traces of titanium only, it suggests rather impact than volcanic origin (Szarka 1994).

Spherules studied from the Polány Marl Formation in territory of Bakonyjákó (Bakony Mts.) in bh. Bj. 528, and Nagygörbő (Keszthely Mts.) in bh. Ng 1 were determined as magnetic and glassy.

At Ganna in bh. Gat. 1 glassy spherules were found only.

The spherula contained parts of Csehbány- and Ajka Coal Formation is ranged to the Late Santonian: *Oculopollis-Complexiopollis* -, *O. zaklinskaiae* - *B. globosus* -; *Hungaropollis Dominancia*-Zones and to the CC16 - CC17a Nanno Zones (Siegl-Farkas 1995), (Fig. 3).

The youngest marine Polány Marl Formation is ranged to the Campanian and to the early Early Maasrichtian.

The part of bh. Bj 528. of Polány Marl belongs to the *L. bajtai* - *L. lenneri* Assemblage Zone, to the *Odontochitina operculata* Zone and to the CC17b - CC18a Nanno Zone (Lantos et al. in press).

Sediments of bh. Ng. 1 was developed during the *P. bakonyensis*-

P. subhercynicus Assemblage Zone, the Pyxidinoopsis bakonyensis Zone and the CC21 - CC22ab Nanno Zone (Siegl-Farkas and Wagreich 1994).

The youngest part of the Polány Marl in bh. Gat. 1 was sedimented in time of Plicapollis - Subtriporopollenites Assemblage Zone and the CC22c Nanno Zone (border of the Campanian - Maastrichtian, Siegl-Farkas 1995).

Based on this data the Spherulite - contain sediments were developed during the Late Santonian - Early Maastrichtian in territory of Transdanubian Central Range.

### 1.2. UPPONY MOUNTAINS

In the Uppony Mountains (N Hungary) the Senonian is represented by about 60 m of Gosau type sediments. The Nekézseny Conglomerate Formation made up of conglomerate bands with some thin marly layers (Fig. 4).

This formation is a redeposited sequence (Brezsnyánszky et Haas, 1983).

On the basis of palynological evidences it is considered to have been deposited during the Late Santonian - Campanian (Siegl-Farkas, 1984).

Magnetic and glassy spherules were appeared in the marly layers of the railway-cut at Nekézseny.

### 3. PALAEOENVIRONMENT

According to the newest palaeogeographic maps (Hay et al. 1994 )

the boundary of the Tethys and the Boreal region developed at the border of the subtropical and the polar oceanic fronts during the Upper Cretaceous.

This boundary were situated in the territory of the present-day Central Europe.

The extension of the Tethys was about three times greater than that of the Mediterranean Sea. It was warm and shallow. The Hungarian Upper Cretaceous formations were deposited on the northern, richer in archipelagos areas of the Tethys.

On the dry lands, abundant fern, pine and diversified dominantly angiospermous (Normapolles) vegetation grew.

On the surrounding elevated relief, erosion of Carboniferous and Triassic formations took place.

In the water of the sea, abundant phytoplankton associations vegetated (dinoflagellata and nannoplankton).

On the basis of the occurrence of the rich sporomorph remnants occurring frequently in massulae, the sedimentation took place in the naershore region.

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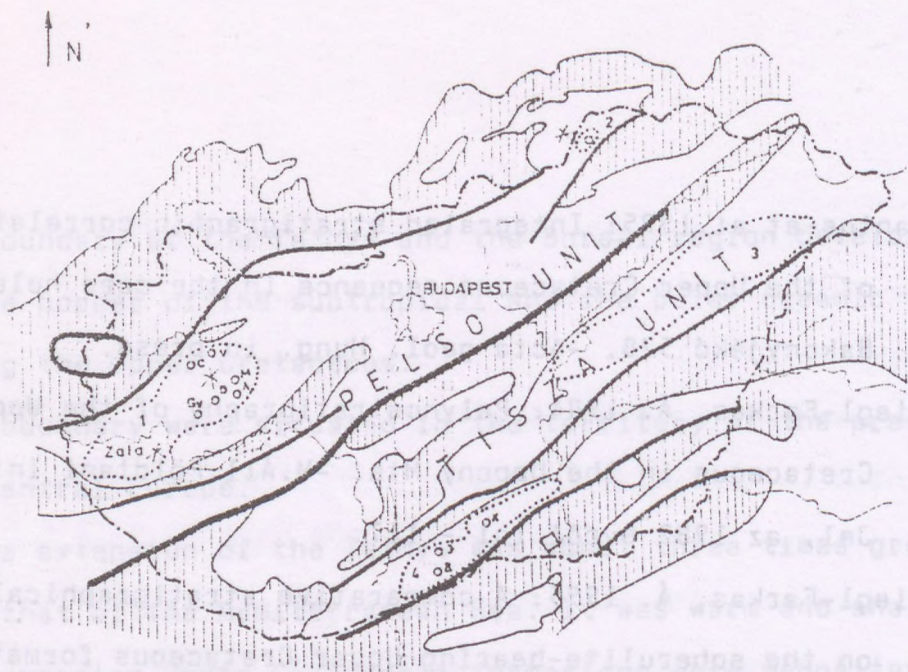
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



-  Unmetamorphosed Mesozoic formations
-  Metamorphic Mesozoic formations
-  Palaeozoic and Precambrian formations
-  Upper Cretaceous formations:

Figure 1. Upper Cretaceous localities. 1. Transdanubian Central Range (A, Ajka; C, Celldömök; D, Devecser; Gy, Gyepükaián; M, Magyarpolány; V, Vinár); 2. Gosau (N, Nekézseny); 3. Flysch; 4. S. Great Hungarian Plain (B, Bácsalmás; J, Jánoshalma; K, Kiskunmajsa).

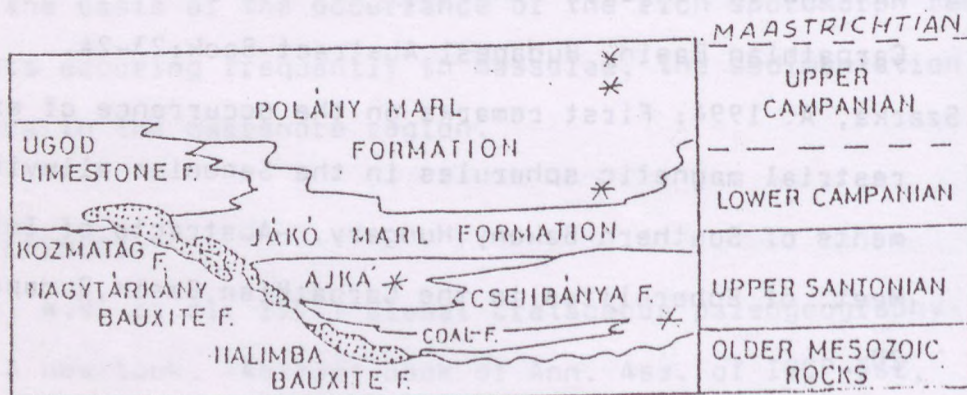


Fig.2 Simplified section to illustrate relationships Senonian formations

\* appearance of spherules

Siegl 1995 August

LATE SANTONIAN		CAMPAANIAN		STAGE	
CC16		CC24-CC29		NAINDO	
C		DINOFLA - GELATA		DINOFLA - GELATA	
D		ADOLITO PYRIDINOP		ADOLITO PYRIDINOP	
E		SILITWA SIS. BAK.		SILITWA SIS. BAK.	
F		2		2	
G		I		I	
H		I		I	
POLLEN		1984 1984 1984 1984		1984 1984 1984 1984	

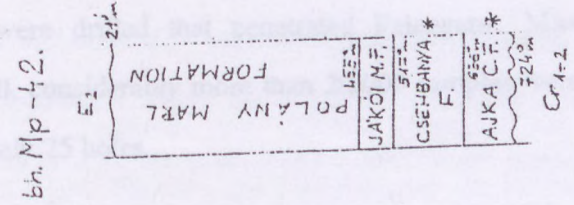
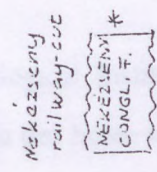
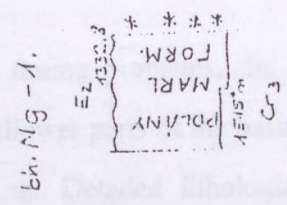
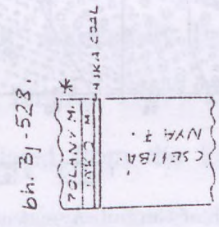
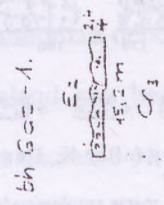


Fig. 3. Comparative biostratigraphical study on spherulite-bearing Upper Cretaceous formations of Hungary

\* spherulite-contain layers





## PROSPECTS FOR INVESTIGATION OF THE MAGNETIC SPHERULES

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The Pannonian Basin, a typical back-arc depression, subsided rapidly during late Miocene and Pliocene times to receive several km thick, dominantly fine-grained, brackish-water sediments, the so called Pannonian deposits (e.g. Jám bor, 1990). By the end of Sarmatian (~12 Ma) the Pannonian inland sea became isolated from the Eastern Paratethys and the Mediterranean. Subsequent decrease of salinity resulted in the evolution of a special endemic mollusc and ostracod fauna which are indicative mostly of depositional environments rather than age.

To solve correlation and timing problems, the Geological Institute of Hungary started a project in 1982. In the shallower parts of the basin, ten test holes were cored continuously to depths of 500 to 2000 m. Detailed lithological, sedimentological, paleontological and paleomagnetic studies were carried out on the cores. During and following this project further stratigraphic test holes were drilled that penetrated Paleogene, Miocene, Pliocene and Pleistocene deposits. In all, considerably more than 20,000 samples were collected on 0.5 m intervals from approximately 25 holes.

Samples were collected at the drill sites and immediately placed in cubic plastic boxes. They were measured in a cryogenic magnetometer at the joint laboratory of the Geological Institute of Hungary and the Eötvös Loránd Geophysical Institute. Zones of normal and reversed polarities were developed from inclinations, then were correlated among the drill core sections and with the geomagnetic polarity time scale. These correlations led to an improved understanding of the accumulation history of the basin (e.g. Elston et al. 1990, Lantos et al. 1992).

During the evaluation of data it was noted that several samples exhibited extremely high intensity of magnetization. The magnetization of these samples was 10-100 times stronger than that of the adjacent samples and ranged from  $10^{-1}$  to  $10^{-3}$  A/m. Additionally, magnetic directions in some of the anomalous samples also differed significantly from the adjacent ones. Figure 1 shows a relatively short part of the paleomagnetic record in the Szombathely test hole, drilled in NW Hungary. Magnetic intensities commonly range from  $10^{-4}$  to  $10^{-3}$  A/m for the fluvial Upper Pannonian samples that consist of sand, silt and clay. Anomalous samples with high intensities can be seen at 300, 303 and 337 m in Figure 1. High positive inclination of the sample at a depth of 303 m does not fit with adjacent directions. Such anomalous inclinations were discarded from the dataset used for stratigraphic interpretation.

The very high intensity of magnetization in rock samples can be explained in three different ways. Firstly, increase of intensity can be related to increase in the concentration of the magnetic minerals. This is only a theoretical explanation. Abrupt increase in the concentration of magnetic minerals is unlikely in the Hungarian sections because of the depositional history of the basin. Secondly, high intensities can be produced by secondary magnetizations due to subsequent chemical alterations. Particularly important secondary mineral may be the greigite ( $\text{Fe}_3\text{S}_4$ ). The intensities of greigite-bearing sediments are at least an order of magnitude higher than the intensities recorded in the Upper Pannonian strata (Reynolds et al., 1994). Chemical alterations might have occurred in some parts of the Neogene sections in Hungary, and secondary greigite may occasionally be the main carrier of magnetization.

Finally, very high intensities may be caused by any materials that are independent of the accumulation of sediments. Extremely high magnetic intensities therefore may mark horizons where foreign magnetic particles are present, and the origin of foreign grains may be extraterrestrial. Magnetic particles deriving from the drilling process itself are very unlikely, because the samples were collected from the central parts of the cores, away from surfaces that had been in contact with the core barrel. In addition, samples were cut from the rock with brass or bronze tools.

Numerous samples have been recognized in the magnetostratigraphic records that display extremely strong magnetizations relative to the adjacent samples. None of the extreme samples have been either reviewed or analysed yet. The selection and study of these samples can be useful in the investigation of extraterrestrial magnetic materials. Following selection of anomalous samples, the paleomagnetic records should be compared with lithological and sedimentological logs. Then a study of the selected samples under microscope may result in the finding of extraterrestrial magnetic minerals.

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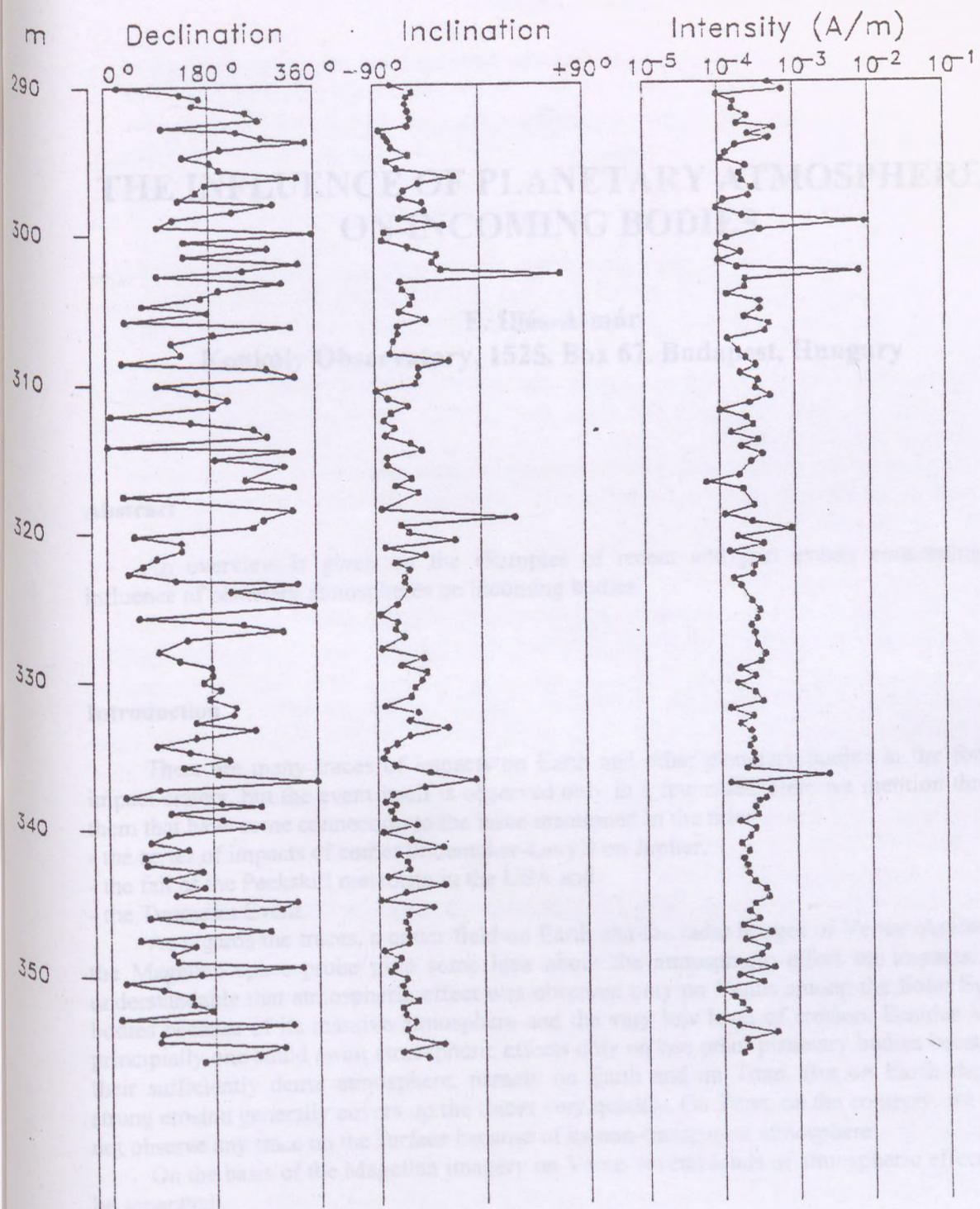
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Figure caption

Figure 1. A part of the arbitrary declination, inclination and magnetic intensity records from the Szombathely II. core hole.





# THE INFLUENCE OF PLANETARY ATMOSPHERES ON INCOMING BODIES

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## Abstract

An overview is given on the examples of recent and past events concerning the influence of planetary atmospheres on incoming bodies.

## Introduction

There are many traces of impacts on Earth and other planetary bodies in the form of impact craters, but the event itself is observed only in a few cases. Here we mention three of them that have some connection to the issue mentioned in the title:

- the series of impacts of comet Shoemaker-Levy 9 on Jupiter,
- the fall of the Peekskill meteorite in the USA and
- the Tunguska Event.

As regards the traces, a crater field on Earth and the radar images of Venus obtained by the Magellan space probe give some idea about the atmospheric effect on impacts. It is understandable that atmospheric effect was observed only on Venus among the Solar System bodies because of its massive atmosphere and the very low level of erosion. Besides Venus principally one could await atmospheric effects only on two other planetary bodies because of their sufficiently dense atmosphere, namely on Earth and on Titan. But on Earth the very strong erosion generally covers up the traces very quickly. On Titan, on the contrary, we could not observe any trace on the surface because of its non-transparent atmosphere.

On the basis of the Magellan imagery on Venus several kinds of atmospheric effects can be separated:

- lobate ejecta blanket, and/or missing of some sectors of the ejecta blanket,
- synchronous impacts of chunks of exploding bodies.
- cut off of crater diameters.
- traces of "tunguska events".

## Impact of Comet Shoemaker-Levy 9 (SL-9) on Jupiter

The only impact event of a natural body that was forecasted and consequently observed so far in the history of mankind is the impact of comet Shoemaker-Levy 9 into Jupiter [1]. The comet was fragmented at least into 22 pieces beforehand because of Jupiter's tidal effect. The series of impacts spread over 6 days between 16-22 July 1994. In spite of the fact, that some people doubted that any effect could be observed from the Earth, many telescopes – almost every in orbit – followed the events. The effect in some cases was extremely violent. The scenario observed was as follows:

- The meteor flash phase: when the body arrived into the atmosphere it was heated up and glared because of the air drag. The temperature reached 10,000 K at this first flash.
- Breaking apart: after a long path in the atmosphere the incoming body was heated up to such a degree that it exploded.
- The explosion was followed by the fireball phase: the material of the impactor was launched (Fig. 1) as high as 3000 km above the cloud deck into the upper atmosphere and the temperature reached 30,000 K.
- Some 5-10 minutes later a splash down of the launched material occurred, it fell back on the lower layers of the atmosphere with a velocity of 5-12 km/sec, that caused a heating of the atmosphere by cca 2000 K.
- After 90 minutes a dark spot appeared on the impact site (Fig. 2) the material of which is unknown. Around a central dark spot that spread out with smaller velocity, another almost concentric dark egg shaped ring appeared and spread out with the sound velocity of the minimum temperature layer like a wavelike phenomenon. It reached about 20,000 km size in some cases.

The molecules that were detected: in the first 2 minutes strong emission of  $\text{NH}_3$  and strong absorption of  $\text{CH}_4$ , in the 4th minute strong emission of  $\text{CO}$  (2000-3000 K is needed), in the 6th minute ultraviolet emission of  $\text{H}_3^+$  (1400 K is needed), in the 12th minute  $\text{H}_2\text{O}$  was detected. In the 16th minute  $\text{H}_2$ , in the 30th minute Li, Na, K, Ca, Fe,  $\text{H}\alpha$ , after the 45th minute S and neutral and ionized metal ( $\text{MgI}$ ,  $\text{MgII}$ ,  $\text{FeI}$ ,  $\text{FeII}$ ,  $\text{SiI}$ ) emerged. After 8 days  $\text{SiO}_2$ ,  $\text{C}_2\text{H}$ ,  $\text{H}_2\text{O}$  then,  $\text{CO}$ ,  $\text{HCN}$ ,  $\text{CS}$  appeared in emission that went into absorption by September.  $\text{CS}_2$  could be detected even in April 1995.

On the basis of the observations (with  $10^{15}$  g mass coming in) a thermochemical model indicates that everything has been vaporized, the material of the impactor and the neighbouring atmospheric envelop as well. Then photolysis, chemical reactions, condensation took place as well as interaction among participants. Consequently, there is no chance for the original molecules of the impactor to be observed.

### The Peekskill meteorite

The fall of a larger chunk of meteoroid occurred on 9 Oct. 1992 at 23 hour  $48 \pm 1$  min UT over the Eastern part of USA [2, 3]. The 700 km path of the falling body between 46.4-33.6 km altitudes was recorded by 14 videocameras. This is the first case when moving pictures stay at disposal on a bolide. The orbit could have been reconstructed. This is only the fourth case when the orbit of a meteorit is known up till now. The inclination was  $3.4^\circ$  to horizontal. At 41.5 km height break up occurred and afterwards more than 70 pieces could be observed (Fig. 3). The bolide's velocity outside the atmosphere was  $14.72 \pm 0.05$  km/sec. and 12.4 kg ordinary chondrite has been found.



## The "Tunguska Event"

The so called Tunguska meteoroid earlier was thought to be a comet that blew up at about 6-9 km height over the Tunguska River in Siberia on 30. June 1908, 0<sup>h</sup> 14<sup>m</sup> UT with energy about 20-50 Mtn TNT [4]. The site of the event was visited only later. No fallen material was found, the trees of the forest were hurled down and they are lying radially.

Computer simulation [5] for a comet with relative velocity not more than 72 km/sec and with a  $5 \times 10^{13}$  g mass could describe the observations – with fusion energy release in the gas cap only  $5 \times 10^3$  Joule. The arising inclined cylindrical shock front caused the trees to fall away from the centrum (Fig. 4). The pressure reached 25,000 atm. Between the body and the shock wave the temperature could reach 400,000 K. But another computer simulation [6] exclude the possibility of a comet, because it should have been exploded 2-3 times higher. The body could be rather a stony asteroid, because of its 6-9 km explosion height (an iron asteroid, on the contrary, would explode even lower).

## The traces of a grazing impact in Argentina

The crater field at Rio Cuarto city, Argentina [7, 8] (Fig. 5, 6), counts at least 10 long elliptical features the largest of which being  $4.5 \times 1.1$  km. It is a remnant of a grazing impact of an asteroid that also exploded coming through the atmosphere. The identification happened recently, in spite of the fact that geophysicists recognized it already earlier, but they thought it was created by water or winds. Such elliptical craters are known on other planetary bodies as well, for example many on Mars, some on Venus and on the Moon, but on Earth, besides the above mentioned, only one has been discovered in Campo del Ciclo, Argentina.

## Traces of the atmospheric effects on impacts on Venus

Among the impact craters found on Venus [9] one can easily observe the effect of the atmosphere because of its massive nature. (The surface pressure is about 100 atm.)

The craters with a diameter of more than 15 km are similar to those found on other bodies in the Solar System but their ejecta blanket differ very much. The inner part of the ejecta blanket – between 0.5 and 0.8 crater diameter – is ballistic and similar to those found on other bodies. But there is an outer part of the ejecta until 2.5 (instead of 1.4) crater diameter, that is superballistic. Here only smoother material can be found. The outermost part (until 3-4 crater diameter and even beyond) is very smooth i.e. radar dark (Fig. 7). This smooth ejecta blanket is obviously overlain by the strong winds that blew radially from the impact site at the time of the impact. The flower shape outer feature of the superballistic ejecta blanket can also be caused by the turbulence of the atmosphere after the impact, that was throwing down the picked up matter cauliflower-like at the edges of the ejecta blanket (Fig. 8). The turbulent movement of the atmosphere can be the cause of the lack of the ejecta blanket in some directions as well. The direction of the lack indicates the direction of the approach of the incoming body, if it was coming in obliquely.

If the crater diameter is smaller than about 15 km, than 50%, if smaller than 12 km than almost all of the craters are irregular (Fig. 9) indicating the synchronous impact of several chunks – or the craters are in clusters (Fig. 10) referring to the explosion of the impactor close to the surface. Smaller than 3 km diameter craters do not exist indicating that bodies smaller

than about 100 m can not survive the transit through the atmosphere. But there are a lot of radar-dark (that is smooth) areas without craters (Fig. 11). These are very probably the traces of so called "tunguska events" when the incoming body exploded in the atmosphere, and the arising shock wave was strong enough to smooth the boulders and the soil out.

## Conclusion

The events reported in this paper – the SL-9 impact in particular – indicate that the breaking apart and the explosion of a relatively large interplanetary body during the transit of a dense atmosphere seems to be inevitable. If this happens at least partially, then evolving temperatures are high enough to modify principally both the physics and chemical composition of the entering body and its surroundings. Therefore one should not expect to find the original molecular composition in the remnants of such an impact: neither H<sub>2</sub>O in the traces of comet SL-9 after its impact on Jupiter, nor chemically intact traces of interplanetary matter in spherules previously modified by an explosion in the terrestrial atmosphere.

**Acknowledgement:** The author is indebted to Mr. P. Decsy for his able help in the preparation of the paper.

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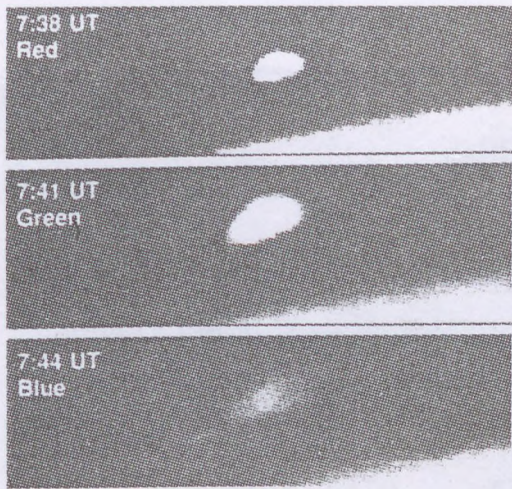


Fig. 1. Above the disk of Jupiter the fireball glares after the impact of the first fragment (designated by A) of comet Shoemaker-Levy 9 as observed in different wavelengths by the Hubble Space Telescope.

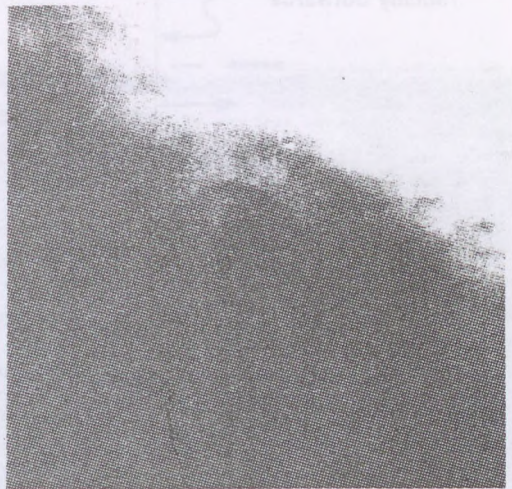


Fig. 2. The dark spot and the egg-shaped ring after the impact of fragment G of comet Shoemaker-Levy 9 as observed by the Hubble Space Telescope. The smaller dark spot to the left is the aftermath of fragment D.

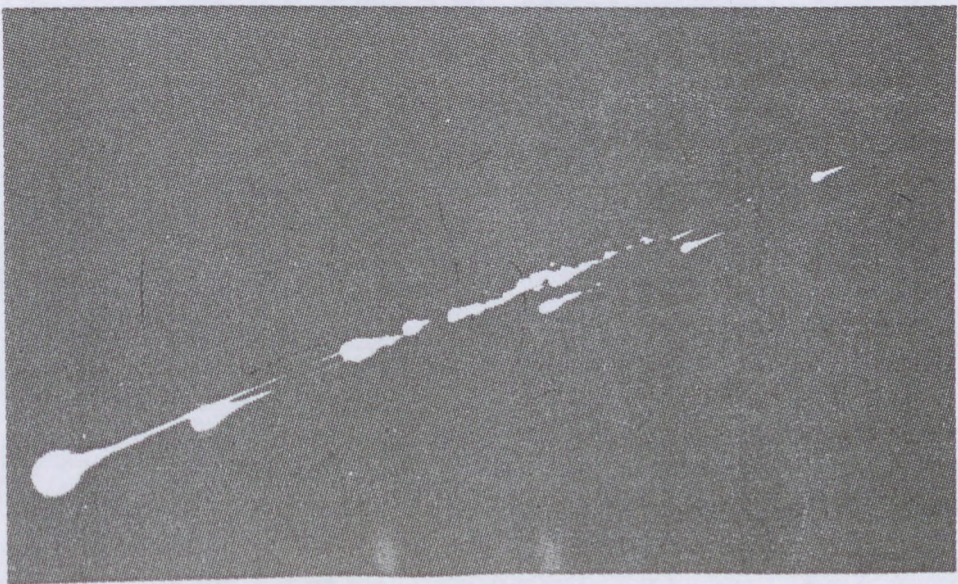


Fig. 3. The breakup of the Peekskill meteorite's parent body. Photo S.E. Ruck.

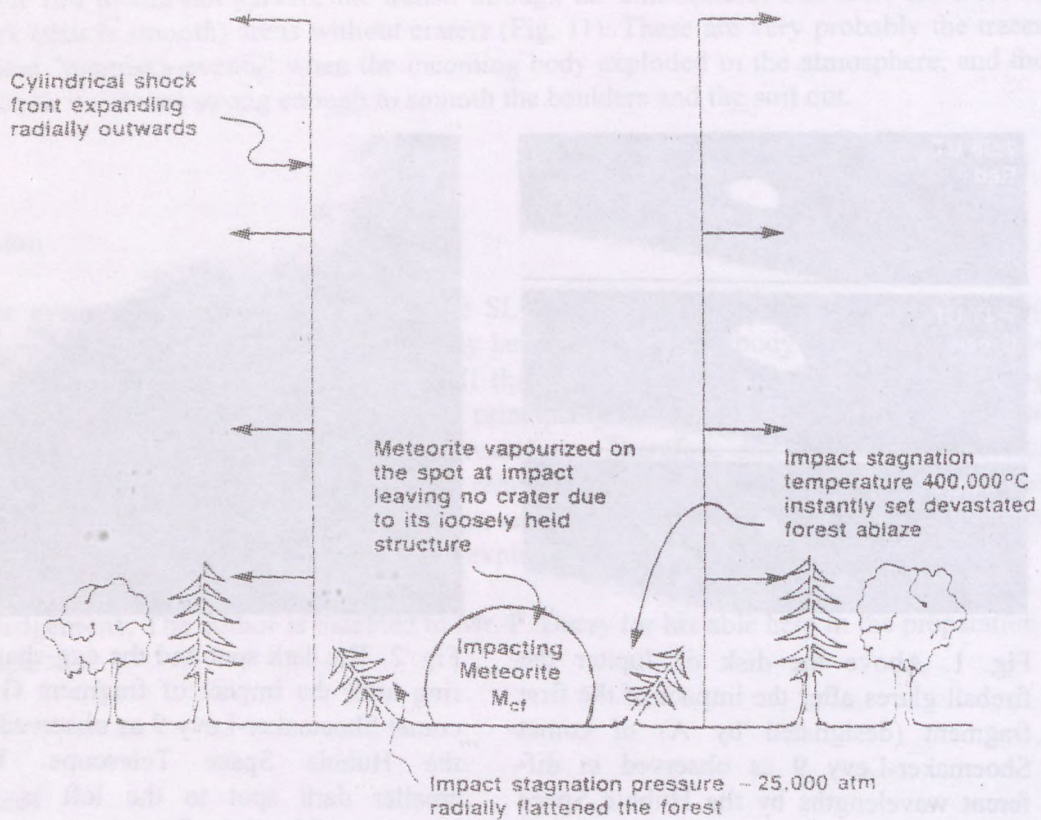


Fig. 4. A Conception how the Tunguska Event has flattened the forest radially [3].

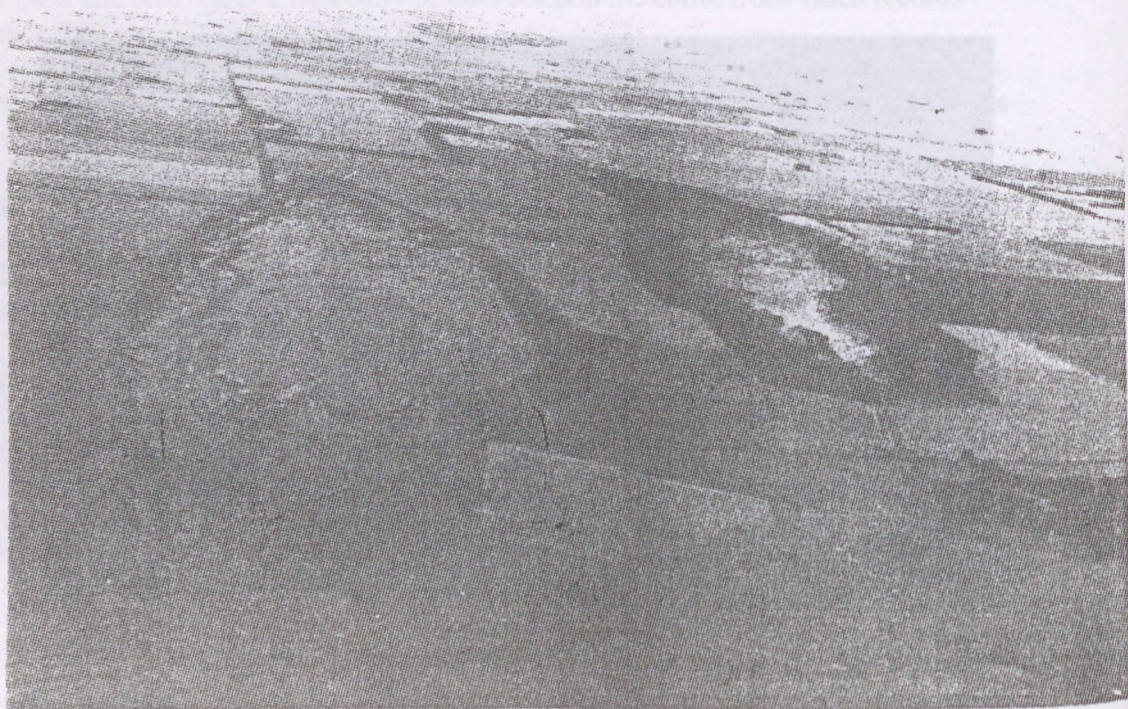


Fig. 5. Aerial view of two elliptical features on the crater field at Rio Cuarto city, Argentina [6].



Fig. 6. Inside panorama of one of the craters within the craterfield at Rio Cuarto city, Argentina [6].



Fig. 7. A typical larger impact crater on Venus. Its inner part is ballistic, its outer part is superballistic and the outermost ejecta blanket is radar-dark. Magellan radar image.



Fig. 8. A typical larger impact crater on Venus with couliflower-like edges and a missing sector of the ejecta blanket in one direction. Aglaonice crater, Magellan radar image.



Fig. 9. A typical irregular impact crater on Venus suggesting the synchronous impact of several fragments of a body that exploded just before the impact. Magellan radar image.



Fig. 10. A typical crater cluster on Venus suggesting the explosion of the impactor close to the surface. Magellan radar image.



Fig. 11. Typical traces of "tunguska events" on Venus with radar-dark, i.e. smooth areas - some of them without any crater (upper part), some with a small impact crater inside (lower part). Magellan radar image.







## The StaTOR program

The StaTOR program is a reticular, hierarchic, dynamic text data base software, which is often identified as a hypertext system. I put it – because of its original aim – rather to the CATH – Computer Aided THinking – category. We could simply call it a mind-manager, the software of thinking people.

### The disadvantages of the software used to solve the same problems as StaTOR:

- they are expensive (0.5-1.5 million HUF),
- they are complicated (they come out with several hundred/thousand pages long reference guides);
- they are slow (the user often has to wait),
- they have many limits (e.g. for the length of data),
- there are lots of things which cannot be written down in them (only a tree shaped data base can be created, while the required data system is usually more reticular- or hierarchic-like),
- the find is done by text samples (whose outcome is that we don't find lots of things (conjugated forms, synonyms, words written in foreign languages etc.). but we get many undesired, unnecessary data, 10-100 times more than the required information, and this find is time and money consuming, the system does not learn, does not get smarter after a find).

### The results until now:

The StaTOR program is an original, totally new program which does not have the problems of other softwares, and is giving opportunities for future applications, also right now. The StaTORs features are totally different from other software used to solve the same problems.

- the data recorded with the StaTOR can be recollectd by its CONTENTs,
- THERE IS NO ACTUAL FINDING PROCEDURE, so it is very fast,
- the hierarchic, reticular, dynamic information storage GIVES BACK OUR THINKING, THE LOGICAL STRUCTURE OF THE WORLD SENSED BY US, so everything that can be written down in it can be efficiently represented,
- the information can be found without unnecessary information.

# StaTOR

- only the important parts can be seen from the data, the data structure is well organized, can be changed easily, can be extended and can also be reorganized,
- it has only a minimal limit (at present(!), the number of texts that can be stored is 32.000, the length of each text is 32.000 characters),
- it is simple: knowing only 8 commands – with the help of a 2 pages long guide – it can be used for months.

### People use the program (within a very short time, in more than 15 places) for the following purposes:

- a researcher for processing his whole scientific lecture notes (including also processing of literary references, terminology, dictionary, encyclopedia, automatic characteristic selection, etc. applications), for replacing card-indexing and for writing articles, etc.,
- for creating historical handicraft data base,
- a university professorate for creating an encyclopedia,
- a biochemist for his scientific work,
- a big museum for the purposes earlier mentioned by the researcher, and e.g. for analysing religious texts,
- a philosopher for representing other philosophers' ideologies,
- for representing a wider family tree than earlier.

---

**StaTOR — the software of thinking people**

- for analysing the stylistics of old religious texts,
- for creating terminologies (with their connections),

- a veterinarian co-operative for preparing a publication of 1000 products for the automatic organization of the contents of the publication from many views, (the publication will also be made available on floppy disks),

- a philosopher and others use the program for creative thinking that has more significance beyond the traditional methods,

- a big research institute for organizing researches,

- for organizing conferences,

- for stock taking,

- a big computer firm for office organizing and for manager functions,

- and not in the least there has been a successful experiment on perfecting an allocation project with an automatic power source.

The philosophy of the StaTOR makes it possible for everybody to build up his own world (whatever can be written down can be efficiently represented, processed and created).

#### The StaTOR basically can be used as follows:

- the hierarchic representation of the texts,
- automatic processing of structural data (for the registration of books, objects, doctors, pharmacies etc.),

- binding information to a conceptual net as a frame, and after that a look-up by CONTENTS, writing of articles, etc.,

- for modern thinking, for creative intellectual work, for joining our knowledge recorded by the program and our thoughts more effectively, for building up our own world.

#### Reference places:

- Central Research Institute for Physics
- Veterinary Pharmacological and Toxicological Society,

- Ethnographical Museum,

- Regional History Collection of Obuda,

- Alternative Secondary School for Economics

- Hungarian TV

- Archives of Budapest

- and more private persons (researchers, authors).

#### The expected development of the software:

The development is partly theoretical, partly technical.

- the program should not only be able to run on stand alone IBM PCs, but also on networks. and the Windows version and also the English and German version should be completed. Also the present text object limit of 32.000 should be extended to 1 million objects. The memory and background storage management should be fixed by unified power-resource management,

- theoretical development – which is almost totally completed also right now (we haven't yet found a counter example), that ANYTHING should be able to be presented in the data base created by the StaTOR, that can be said or written down, in such a way that it should give back the real connection of the things, the human point of view, maybe exceeding this point of view,

- the extension of the earlier hierarchies built up of text objects, and nets with the possibility of giving the form of connection between the objects,

- extension with graphical elements (which is intensively demanded – by the users),

- the further development of the special data structure in such a way that it would make it possible to solve problems which belong to the field of artificial intelligence,

- creation of conceptual hierarchies (thesauruses) for users of different fields,

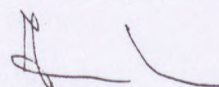
- the research for different data types, completion of solutions of data representation,

- the research for the StaTORs usage fields and methods.

A creative and high level data base can be formed with the StaTOR. The data base formed earlier with another program can be loaded/is to be loaded into the StaTOR, or a logical structure, a thesaurus can be created for the old one.

Szentendre, 25. August, 1993

DEVELOPER:



TOR Thought ORiented Software

#### DEALER AND INFORMATION:

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Kiadja a Központi Fizikai Kutató Intézet  
Felelős kiadó: Lukács Béla  
Szakmai lektor: Bíró Tamás  
Nyelvi lektor: Horváth István  
Példányszám: 191  
Készítette: Prosperitás Nyomdaipari Kft  
Budapest, 1996. július hó