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NEW DATA ON THE POST-IMPACT MATERIAL IN RADIOLARIAN HORIZONS IN OUTER FLYSH CARPATHIANS AND SEARCH FOR A SOURCE CRATER

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Abstract: In the Outer Carpathians in Barnasiówka radiolarian shale formation, there is an intercalation underlied by bentonite. There were found very rare elements and minerals in this intercalation. It was recognized that this horizon has been filled with products of a big object collision with Earth. The age of the manganese-iron intercalation was determined by potassium-argon (K-Ar) dating of illites to be (89.3±1.2) Ma. Similar age, (85.2±0.6) Ma, was found for the post-impact glass from the Boltysh crater in Ukraine. It was concluded that the formation of this intercalation was synchronous with the Boltysh crater formation at the Cenomanian/Turonian boundary. The research for the K-Ar age of the crater creation in Logoisk (Belarus) established its formation to (124.2±1.2) Ma ago.

Keywords: Cenomanian/Turonian boundary, impact, crater, illite, K-Ar dating, Outer Carpathians

1. INTRODUCTION

Petrographic and geochemical examination of sampled rocks showed that the rocks which formed in the Outer Flysch Carpathians at the Cenomanian/Turonian boundary might be genetically associated with a collision of a large celestial body with the Earth's surface. The location of selected sites of the study area are shown in **Fig. 1**, whereas the schematic profile of these sedimentary rocks are shown in **Fig. 2**. In this finding, an increased content of some elements (like REE) and minerals (like bronzite and coesite grains) which rarely occur in the Earth crust, has been encountered. This thin layer reveals significant changes in the organic world (Kauffman, 1984; 1986; Raup and Sepkowski, 1986; Arthur *et al.*, 1987; Bralower, 1988; Sepkowski, 1990) – certain foraminifera species died out completely (e.g. *Rotalipora Gavelinella intermedia*) and were replaced by other organisms (radiolarians) which reached an abundant growth. Other premises, supporting an impact origin of the discussed layer, rely on the observed sedimentological phenomena which might be consequences of strong marine currents induced by propagation of a post-impact seismic wave. The anomalies occur within the "manganese layer" and in the surrounding rocks occurring in the radiolarian shales from Barnasiówka in the Silesian Unit (Bąk *et al.*, 2001).

A detailed examination of this formation as well as of its equivalents which are found in other units of the Outer Flysch Carpathians, showed that post-impact products and crumbs of a planetoid which struck the Earth about 90 million years ago, are likely to occur there.

Samples from the radiolarian shales from Barnasiówka were subjected to petrographic and geochemical examination that aimed to establish the age of the rocks as well as the chemical elements and mineral composition of the extraterrestrial matter out of which the planetoid was probably composed. This allowed the identification of the main crater that this planetoid had formed. The prospecting has been limited to two meteor craters known on the Earth. One of them was the Boltysh crater which, at the study onset, was believed to be almost the same age as that of the manganese layer of the Carpathians, while the second one was the Logoisk crater as the post-impact products studied in its interior were very similar to those

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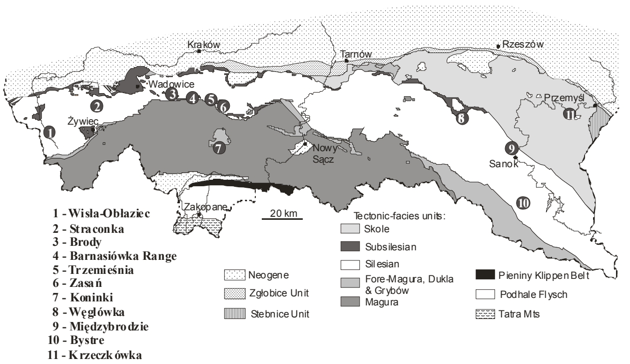

Fig. 1. *The study area with locations of selected sites.*

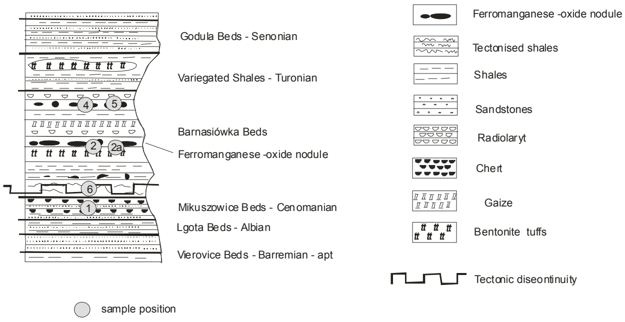

Fig. 2. *Position of radiolarian layers in the Silesian Series.*

present in the manganese layer from the Carpathians. The Boltysh crater, with its centre in Alexandrovka village, is in eastern Ukraine. The buried crater is dissected by the Tyasmin River which exposes an ejecta blanket of breccia at its banks.

Confirmation of a collision of a celestial body with the Earth's surface would be of a great significance for the explanation of many sedimentary events or even tectonic ones which took place in the Carpathian geosyncline at that time. It might also be an important contribution to the ongoing discussion on meteor causes of the event at the K/T boundary (Montanari and Koeberl, 2000; Sawłowicz, 1993). A fall of a planetoid or its shattered pieces likely took place in Eastern Europe that is evidenced by the Boltysh presumably formed at the same time as the sediments in the profile of the Beds from Barnasiówka.

Minor pieces of this planetoid fell into the Carpathian geosyncline. Large amounts of cosmic dust and rare elements accumulated in a thin "manganese layer" can provide evidence of a proximity to the collision site. Effects of this collision are not so clear in other parts of the world. However, the studies conducted on samples of rocks, which were created at the Cenomanian/Turonian boundary in North America and in Italy (Elder, 1987; Lopez *et al.*, 1988; Orth *et al.*, 1988; Poag, 1997a, b), have unambiguously demonstrated that changes ongoing at that time in the organic world were caused by a collision of a planetoid with Earth.

2. EARLY SEDIMENTOLOGICAL AND PETROLOGICAL STUDIES IN THE OUTER FLYSH CARPATHIANS

A complex of spongiolites, radiolarites, shales and sandstones with "manganese-iron" concretion described as "green and red shales", has been reported for the first time in the Outer Carpathians in the profile of the Skole series (Styrnałówna and Cizancourt, 1925). Later, these sediments were called "red radiolarites with red and green shales" or "radiolarites and siliceous marls" (Sujkowski and Różycki, 1930; Sujkowski, 1931). A horizon of the radiolaritic shale was also identified in the profile of the Silesian series (Burtanówna *et al.*, 1933). Subsequent sites, where this horizon occurred, were reported from the Silesian and Skole series, but the discussed complex was assigned various names: siliceous marls or jaspideous layers (Książkiewicz, 1951) as well as it was called a layer of radiolarites and spongiolites (Burtan and Ciszewska, 1956), greenish shales with radiolarians (Bieda *et al.*, 1963), radiolarian shales (Gucwa, 1966) or biochemical siliceous rocks (Burtan and Turnau-Morawska, 1978). The discussed layer of siliceous rocks was also studied by Koszarski *et al.* (1959), and Alexandrowicz (1973), Kotlarczyk (1978), Gucik *et al.* (1983), Geroch *et al.* (1985), Uličny *et al.* (1997), Paul and Wieser (2002). In the paper by Bąk *et al.* (2001) this layer is called a formation of radiolarian shales from Barnasiówka.

3. FORMATION OF RADIOLARIAN SHALES FROM BARNASIÓWKA – RESULTS OF THE PRESENT STUDY

The investigated layer of radiolarian shales from Barnasiówka occurs in the Outer flysch Carpathians. It lies in the Silesian series in the complex of layers which directly overly the Lgota Beds. Above, in the profile of this series, the Godula beds occur, see **Fig. 2**. Deposits of similar type are found in all the tectonic-facial units of the Outer Carpathians, i.e. in Skole, Sub-Silesian, Silesian and Magura series. In the case of the Sub-Silesian series this layer occurs above a gaize layers and it is covered with variegated shales and Węglówka marls. Lithology of the radiolarian shales is similar in all the series. These shales comprises a several meter thick packets of spongiolites, radiolarites and shales with manganese concretions. In the upper parts of this profile, singular, thin layers of sandstones appear. This radiolarian complex is covered with variegated shales, and Godula or Inoceramian beds.

Until now, this radiolarian layer has been identified in the following sites: Wisła, Straconce near Bielsko-Biała, Barnasiówka range, Brody near Lanckorona (Wieser, 1985a, b), Trzemeśnia, Zasań, Węglówka (Wieser, 1982a), Międzybrodzie near Sanok (Wieser, 1973 and 1982b), the Krzeczkówka stream near Przemyśl and in Bystre near Baligród (**Fig. 1**). The radiolarian layer is best recognized and most often described in the Silesian and Sub-Silesian series.

The most thorough petrographic and geochemical studies were conducted at outcrops of this layer in Brody, Trzemeśnia, Zasań and Międzybrodzie. Based on Energy Dispersive Spectrometry (EDS) tests, the following chemical elements have been identified: manganese, chromium and iron, scandium, rhenium, nickel, indium and dysprosium. These elements point to the presence of dust of cosmic origin in the sediments. In this case, a collided celestial body might be a planetoid.

We have attempted to estimate the time of impact by K-Ar dating of illite extracted from shales taken just above the iron-manganese layer. Illites were sorted by the Jackson method into 4 granular fractions as indicated in **Table 1**. The separation into granular fractions and potassium analysis (flame photometry) were performed in the Institute of Geological Sciences, Kraków, while the radiogenic argon was analysed in the Mass Spectrometry Laboratory at Maria Curie-Skłodowska University, Lublin. The results of analyses are shown in **Table 1**. The analytical uncertainty of K-Ar dating, in terms of relative standard error, is ca. 1%.

The interpretation of K-Ar dates of illites from the investigated layer is not unambiguous due to their contamination by older detritic material, which is reflected in large differences in age of particular fractions. However, on the basis of the finest illite fraction, we may estimate a possible age of diagenesis of this layer as (89.3±2.7) Ma. This date is the mean value for the finest grain fraction. In calculation of this mean value and standard deviation, the result for sample Trzemieśnia 6 was rejected, because this sample was taken from the part of the profile, where a thrust has been encountered, thereby the sample apparently was contaminated by a younger material, see **Table 1**. Note that the calculated standard deviation of dates of the finest fraction is only ca. 3 times larger than the analytical precision which is about 1 Ma. Thus, it is apparent that the dates of the finest fractions are similar and the calculated mean values are highly significant. However, a partial argon loss from the finest illite fraction cannot be excluded.

When considering the results of the illite analyses it is worth noticing that the established age of the finest illite fraction is similar to that Cenomanian/Turonian transisition and it agrees with the age obtained based on zircons examination by the fission-track method, (91.4±4.7) Ma, by Van Couvering *et al.* (1981). On the other hand, the age of Cenomanian/Turonian boundary is given as 90.4 Ma (Harland *et al.*, 1990) or 93.5 Ma (Gradstein and Ogg, 2004).

Smaller pieces of the considered planetoid have likely fallen onto the Carpathian geosyncline terrain. The microscopic examinations, which have been carried out recently, confirmed the presence of singular grains of bronzite in the manganese layer. The above can also be evidenced by modified quartz grains, of a specific light extinction, which were observed in Trzemeśnia, Łęki, Zasań and Koninki. Those grains could be suevites which preserved a relict, post-impact deformation, especially as some of them have black rims. These rims might be considered as traces of soot.

In the Outer Carpathians in the Sub-Silesian series, the site documenting the discussed layer is found in Zasań, in the upper course of the Zasanka stream. The rocks of the Sub-Silesian series belong to a belt of tectonic windows present in the Silesian unit just in front of the Magura overthrust. From the east, they are continued in the tectonic windows of Wiśniowa and Skrzydlna,

Table 1. *K-Ar dates of illite fractions.*

Sample	Grain size (μm)	K (%)	$^{40}Ar_{rad.}$ (pmol/g)	$^{40}Ar_{rad.}$ (%)	Age (Ma)
Trzemieśnia 1	2 – 0.2	2.536	928.6	91.2	199.7
	0.2 – 0.05	2.843	648.7	64.7	127.0
	0.05 – 0.02	2.300	449.0	58.7	109.2
	< 0.02	1.640	286.1	53.4	97.9
Trzemieśnia 2a	2 – 0.2	2.62	914.1	88.0	190.8
	0.2 – 0.05	3.05	696.8	63.9	127.2
	0.05 – 0.02	2.63	478.3	51.3	101.9
	< 0.02	1.86	295.5	48.4	89.4
Trzemieśnia 5	2 – 0.2	2.122	644.0	84.9	167.1
	0.2 – 0.05	2.78	626.1	64.8	125.4
	0.05 – 0.02	2.55	449.0	56.3	98.8
	< 0.02	1.86	275.6	48.1	83.5
Trzemieśnia 2	2 – 0.2	2.53	904.3	88.4	195.2
	0.2 – 0.05	2.76	636.2	75.3	128.3
	0.05 – 0.02	2.33	416.2	59.2	100.2
	< 0.02	1.67	256.6	56.7	89.5
Trzemieśnia 4	2 – 0.2	1.85	432.8	75.3	130.1
	0.2 – 0.05	2.64	644.0	63.0	135.5
	0.05 – 0.02	2.26	360.9	51.2	89.8
	<0.02	1.62	247.6	50.4	86.1
Trzemieśnia 6	2 – 0.2	2.43	158.3	54.3	37.2
	0.2 – 0.05	3.05	226.4	37.9	42.4
	0.05 – 0.02	2.574	139.7	29.6	31.1

while from the west in the tectonic windows of Myślenice and Jasienica. The Lower Cretaceous sediments of this series have, at their top face, a profile of siliceous rocks with radiolarites, spongiolites, bentonites and manganese layer.

The profile starts with black shales with rusty coating and fish pieces. Above, spongiolites occur which turn upward into greenish and almond radiolarites. The cement, apart from being chalcedony, can be rhodochrosite. Moreover, manganese oxides are also often present. Further up, there is a 3 cm thick, emerald-green bentonites layer present, where very fine pieces of micas and quartz are found.

Above the bentonites a layer of manganese concretions is located. The corns of these concretions are formed of hard marls with rhodochrosite and outward are covered with shells of black, non-transparent manganese oxides and iron hydroxides. Very tiny green and blue scales of amorphous copper carbonates are often found there. Above, there are thin layers of white carbonate gaizes interbedded with bentonites. The profile ends with radiolarites and spongiolites interbedded with bentonites and clayey inserts with china-clay, siderite and rhodochrosite.

EDS testing of the iron-manganese layer revealed the presence of the following oxides in 16-02 site (**Fig. 3**): SiO_2 – 89.56%; Al_2O_3 – 2.60%; K_2O – 0.19%; CaO – 0.00%; MgO – 0.12%; TiO_2 – 0.07%; SO_3 – 0.29%; MnO – 5.78%; Fe_2O_3 – 11.5%; InO – 0.20% in the upper part of the layer and SiO_2 – 6.57%; Al_2O_3 – 1.47%; K_2O – 0.40%; CaO – 0.19%; SO_3 – 0.45%; MnO – 51.41%; Fe_2O_3 – 39.14%; NiO – 0.15%; Sc_2O_3 – 0.08%; ZnO – 0.20% in another location.

In the Silesian series (Lanckorona facies), in the outcrop of the discussed layer in Trzemeśnia, above the

Fig. 3. *A piece of rock with the iron-manganese layer from Zasań with pointed location of sample 16-02 taken for analyses.*

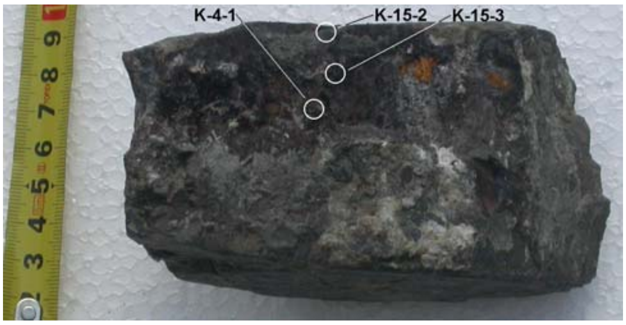

Fig. 4. *A piece of rock with the iron-manganese layer from Trzemieśnia with pointed locations of samples K-4-1 , K-15-2 and K-15-3 taken for analyses.*

Mikuszowice cherts (the uppermost part of the Lgota beds) there is a complex of thin-bedded radiolarites interlayered with black shales which contain organic remnants: shattered skeletons and scales of fish. Here, singular layers of glauconitic sandstones occur as well. Ca. 7 m above the top of the Mikuszowice cherts, a 0.5-3.0 cm thick layer of green bentonite is present. Ceosite has also been identified in bentonites.

Above, there is a several cm thick layer with manganese concretions present. These nodules are usually sphere-like. Particular nodules have concentric structures. The corn typically consists of gray clayey dolomites with tiny crystals of rhodochrosite which turn into a yellow aggregate of clayey minerals with pieces of volcanic glass. Manganese oxides and iron hydroxides make the concretion surface to be tar-black with a metallic glitter. Tiny scales and veins of green malachite coating are often noticed against this black background.

Above the manganese layer, there are spongiolites, radiolarites, gaizes and singular inserts of tufittes with feldspars, plagioclases, scales of micas and pieces of volcanic or post-impact glass.

EDS testing of the iron-manganese layer revealed the presence of the following oxides in K-4-1 site: SiO_2 – 4.61%; Al_2O_3 – 2.27%; K_2O – 0.75%; CaO – 2.64%; MgO – 0.77%; TiO_2 – 0.08%; SO_3 – 0.28%; MnO – 84.95%; Fe_2O_3 – 2.02%; ZnO – 1.63%, while at the coating of the concretion at point K-15-2 these were (**Fig. 4**): SiO_2 – 2.45%; Al_2O_3 – 0.94%; K_2O – 1.08%; CaO – 1.48%; MgO – 2.02%; TiO_2 – 0.05%; SO_3 – 0.38%; MnO – 84.82%; Fe_2O_3 – 5.98%; ZnO – 0.44%; Ni – 0.36%; ReO_2 and Sc_2O_3 have broad culminations, or in another part of this coating – in point K-15-3: SiO_2 – 6.70%; Al_2O_3 – 3.54%; K_2O – 0.97%; CaO – 1.46%; MgO – 1.41%; TiO_2 – 0.09%; SO_3 – 0.43%; MnO – 80.17%; Fe_2O_3 – 4.66%; ZnO – 0.36%; Ni – 0.12%; ReO_2 – 0.10%; Sc_2O_3 – 0.11%.

The inserts of tuff also occur in the overlying layer of the Godula variegated shales, e.g. outcropping in the nearby Bęczarka. A similar profile of this layer occurs in Brody.

The X-ray image (**Fig. 5**), obtained for „bentonite" directly overlying the iron-manganese layer, points to the presence of coesite. The latter is a polymorphic variety of quartz and forms only under influences of high temperature (500-800^{o}C) and pressure (35 kbar). Temperatures and pressures of such magnitude can be generated only during a collision of a cosmic body with Earth. Therefore, the layer which used to be called by former researchers "bentonite layer" is in fact an accumulation of impact products as it is the case of the iron-manganese layer.

4. METEORITE CRATERS IN BELORUSSIA AND UKRAINE – RESULTS OF THE STUDIES

Anomalous accumulation of elements pointing to a post-impact nature of the sediments used to be related to large craters in Logoisk in Belorussia and Boltysh in Ukraine (Masaitis *et al.*, 1980). These craters are 17 and 25 km in diameter, respectively. The performed examination of absolute age revealed that the Logoisk crater is much older than the sediments of the radiolarian layer and according to our study (**Table 2**) its age is (124.2±1.2) Ma, that is the transition from the Jurassic to Cretaceous. Until now, its age has been estimated as (40±5) Ma (Grieve *et al.*, 1995). This allows to conclude that this impact is reflected in the Carpathians in the Cieszyn beds. In the crater's interior, below the ejecta, glass, breccia and tagamite (glassy matrix impact melt rocks), gneiss has been drilled, which seemed to be formed after the planetoid struck granites of the East European platform. However, examination of micas has not confirmed the above assumption – gneiss tested by the K-Ar method revealed (1543.6±5.3) Ma, that is a possible age of formation of the East European platform granites. It is also likely that at this depth the temperature during the impact was not high enough to set the age of micas to zero.

On the other hand, the Boltysh crater, whose age according to Grieve *et al.* (1995) is (88±3) Ma, whereas in our study it is established as T = (85.2±0.6) Ma, was probably formed at the same time as the sediments in the

Table 2. *Dates of materials related with East European craters.*

Mineral	K (%)[*)]	$^{40}Ar_{rad.}$ (pmol/g)	$^{40}Ar_{rad.}$ (%)	Age (Ma)
Biotite from gneiss	7.818±0.007	33084	99.3	1543.6± 5.3
Ejecta glass from the Boltysh crater	2.964±0.003	448.3	69.8	85.2± 0.6
Ejecta glass from the Logoisk crater	2.950±0.003	657.5	54.2	124.2± 1.2

*) determined by isotope dilution method in Mass Spectrometry Lab., UMCS Lublin

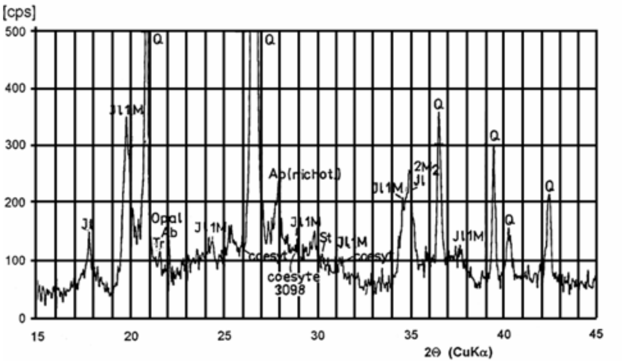

Fig. 5. *A part of X-ray spectrum of Trzemieśnia bentonite with the presence of the coesyte peak.*

profile of the radiolarian shales from Barnasiówka whose zircon age determined by Van Couvering *et al*., (1981) is (91.4±4.7) Ma.

5. CONCLUSIONS

The formation of the radiolarian shales from Barnasiówka, which occur in the Silesian series of the Outer Flysch Carpathians and which formed in the Carpathian basin at the same time, is isochronous with similar sediments from other Carpathian units. It comprises green and black shales, cherts and radiolarites, bentonites and iron-manganese layers. The test conducted on samples taken from the iron-manganese and bentonite layers revealed a higher content of the elements which rarely occur in rocks of the Earth's crust as shown by EDS testing.

The present K-Ar examinations of the age of the radiolarian shale formation are not unambiguous, but they suggest that this layer was formed ca. 90 million years ago. This result corroborates with the Cenomanian/Turonian boundary. The absolute age of the sediments sampled from the craters, which could have been responsible for the presence of traces of extraterrestrial matter in the radiolarian shales, eliminated a possible relation to the Logoisk crater. This crater turned out to be much older, (124.2±1.2) Ma, than the studied Carpathian sediments. On the other hand, the Boltysh crater is probably of the same age (85.2±0.6) as the studied Carpathian sediments.

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