INVESTIGATION OF THE URANIUM CONTENT OF COAL ASHES WITH NUCLEAR EMULSION

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Ashes of pulverized uraniferous brown coals were embedded in nuclear emulsion. We examined the active grains, and compiled statistics according to shape and number of the emitted tracks. The grains fused into spherical shapes are the carriers of the greater part (70,6%) of the total activity. The average uranium concentration of the spherical grains in our test substance was 0,65%.

Our investigations on the uranium content of Hungarian brown coals are closely connected with a research work of wider scope carried out first by the Institute of Experimental Physics of the University Debrecen and continued later by the Institute of Nuclear Research of the Hungarian Academy of Sciences Debrecen, established in 1954. These investigations [1—11] have for several years been systematically directed by one of the authors (A. SZALAY).

Departing from research methods so far applied, we are reporting in this paper on investigations with the photographic emulsion method. In some respects, this method has great advantages, of which we have quite profitably availed ourselves. The track of each α-particle emitted from grains of the substance embedded in the photoemulsion and containing radioactive materials becomes observable under the microscope. If we wait for a few weeks after the embedding and develop the plate only then, the sensitivity of the method strongly increases and individual observation of α-particles becomes practicable. Under the miscroscope not only the track, but also the grain becomes visible from which the track originates. Thus the method affords an opportunity even for morphological determinations.

This investigation had for its scope to decide whether the uranium content is uniformly distributed or whether it is present in a higher concentration in some morphologically describable grain types when uraniferous coal is burned in a powdered form and the ash grains obtained placed under the microscope, which enables discrimination of each individual grain.

For our tests we used ashes of pulverized coal from Ajka. When viewed by microscope, the grains of ashes may be roughly divided into two groups: irregular and spherical-shaped ones. Melting point of the coal particles corres-

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ponding to the latter is lower than the temperature of the interior of the furnace. This is the reason why they melt into spherical shapes.

We embedded the ashes in the nuclear emulsion by using the double-layer method of the autoradiographic technique. The only difference was that we applied liquid emulsion as the top layer. The emulsions were prepared by the Forte Industrial Research Laboratory of Photochemistry of Vác (Hungary) [12, 13].

The ashes to be tested were sprinkled over a horizontally placed emulsion of 100 μ thickness coated on a glass-plate support which was swelled beforehand. Then the emulsion for the top layer, previously kept at 5°C, was heated and poured on it in a layer of 40—50 μ thickness and finally dried by a cold air current. The sandwich was developed 100 days later, in ID 19 developer. The fading of silver grains during such a long exposure time is especially significant, and this is why we found tracks of various grain density.

The sandwiches were evaluated by a C. Zeiss LgOG microscope at 200× magnification.

The problems we wanted to solve by microscopic investigations, were the following :

1. which ash grains contain radioactive substances,

2. what percentage of the total activity is present in a certain type of ash grain,

3. what is the concentration i. e. uranium content of the individual ash grains?

For active ash grains we compiled statistics according to colour, shape and number of emitted tracks. The number of tracks which could be determined means a lower limit, because some of the tracks are obscured by the opaque grains. Besides, when grains have greater size, it may also occur that the whole track is included in the grain. We distinguished grains of spherical and irregular shapes. Within these categories, there were translucent and opaque ones. The classification according to shape was very strict and only grains of exactly circular cross section were classified as regular, since we found among the irregular active grains a considerable number of grains partly fused or spheres to which other smaller or larger ones of irregular shape were adhering. This examination yielded data for the distribution of the activity of grains (Table I).

The grains fused into spherical shapes are the carriers of the greater part of total activity (70,6 %) and the average number of tracks issuing from one grain is 9,6 % higher. 66,23% of the total activity is found in the opaque sphere, which amounts to 93,82 % of the total activity possessed by spheres. We further made investigations to ascertain, what percentage of the grains, fused into spheres, is radioactive. Using a magnification of 330 times, we observed 828 fused grains in 500 microscopic fields, under the square network placed into the ocular. Out of this number, we found 621 active grains after 100 days of irradi-

Table I
Distribution of activity in ash grains after an exposition of 100 days

Ash grain		Number of grains examined	Number of tracks emitted by ash grains	Average number of tracks [track/grain]	Percentage of activity [number of tracks / numb. of all tracks]
Sphere	opaque	723	3658	5,05	66,23
	translucent	37	241	6,51	4,36
Irregular	opaque	332	1552	4,67	28,10
	translucent	15	72	4,80	1,30

ation. The quantity of uranium in grains not showing any α-track is less than $1{,}15 \cdot 10^{-12}$ g, supposing that the uranium is in radioactive equilibrium with its daughter products. The uranium content of 75 % of the spherical grains emitting at least one α-track is higher than this.

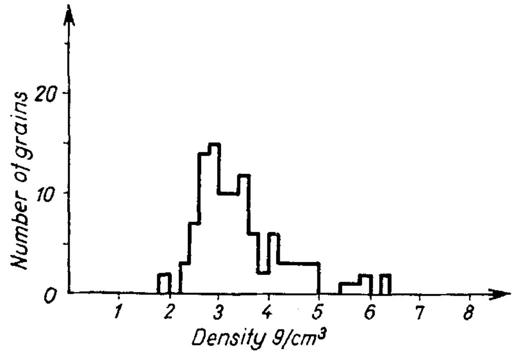

Fig. 1. Distribution of the density of grains fused into spheres

According to our measurements, the fused grains carry the greater part of the activity. The further aim of our investigation was to determine their uranium concentration. For this purpose we had to determine the density of the grains. For a preliminary inquiry, we made sedimentation tests in bromoform under the microscope. The majority of spherical grains sank in bromoform, their density is larger than 2,9 g/cm³. We repeatedly ascertained that grains remaining on the surface either contained gas bubbles, or had irregular grains of lower density adhering to them. The density of spherical grains with higher values than 2,9 g/cm³ was measured by Stokes law, determining their rate of sedimentation in pure glycerin. The viscosity of the glycerin was 12 Poise, at 18°C. The determination of the diameter and the rate of sedimentation was carried out by a horizontally placed microscope of 216 times magnification, in a special cuvette. The error was comparatively great (15%) owing to diffraction by the edges of the grains and the difficulty to determine the diameter during the motion of the grains. For the same reason, we could obtain reliable values only for grains of diameters greater than 25 μ. The result of the measurements made on 100 grains is shown in Fig. 1. The density of the majority of grains fused into spheres is ranging from 2,6 to 3,6 g/cm^3.

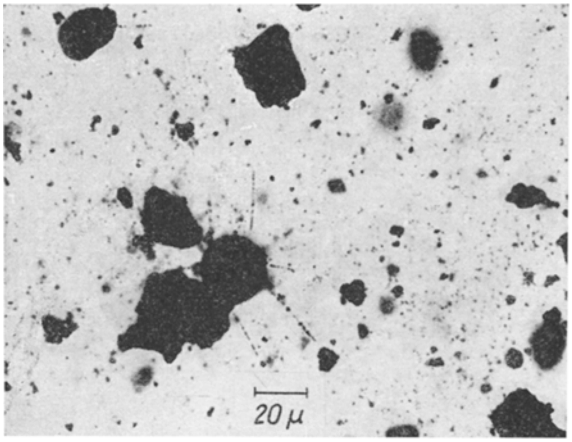

Fig. 2a. Microphotograph made from ash grains embedded in nuclear emulsions

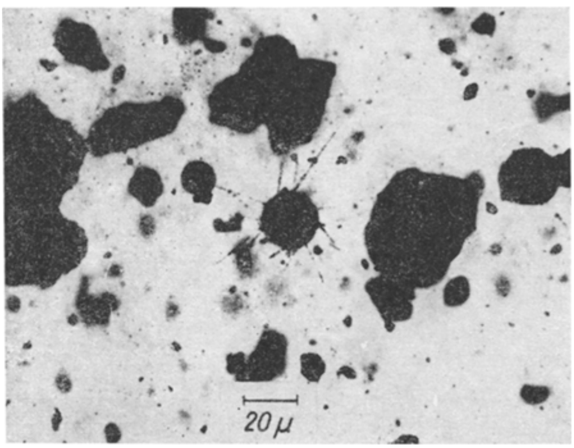

Fig. 2b. Microphotograph made from ash grains embedded in nuclear emulsions

Figures 2*a*—*c* are microphotographs made of active ash grains. It is clearly seen that the majority of irregular grains is inactive. By the number of α-tracks, we were able to estimate the uranium content of grains. (In the photograph, some of the tracks are not visible on account of the small depth of the focus). The number of tracks counted under the microscope is — for reasons already mentioned — the minimum value, thus the quantity of uranium determined in them should be regarded as lowest estimate. The diameter, mass (calculated from an average density of 3 g/cm^3) and uranium concentration of grains seen in the Figures are recorded in Table II.

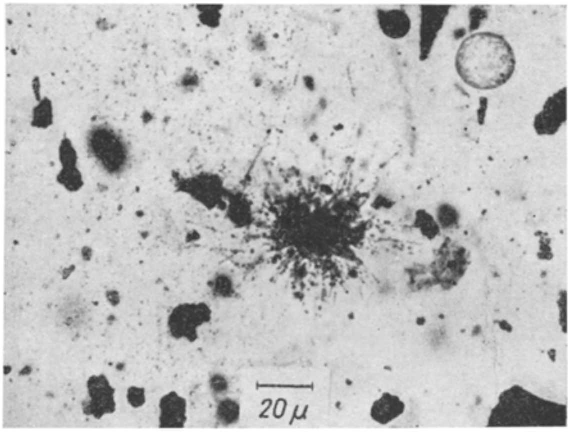

Fig. 2c. Microphotograph made from ash grains embedded in nuclear emulsions

Table II

Uranium concentration of active ash grains presented in Figures 2a—c

Fig.	Number of tracks	Diameter of grains in μ	Mass of grains 10^{-9} g	In case of pure U		In equilibrium of disintegration	
				quantity of U 10^{-9} g	percentage of U concentration	quantity of U 10^{-9} g	percentage of U concentration
2a	13	23	19,1	0,060	0,31	0,015	0,08
2b	24	19	10,7	0,108	1,01	0,027	0,25
2c	>50	12	2,7	>0,228	>8,44	>0,057	>2,11

Analytic investigations [10] proved that the activity of coal ashes is due to uranium, and they either contain no thorium at all, or if they do, it is at best 10^{-4}%. Even this latter value is rare. Consequently, we have not taken thorium into consideration when making our calculations.

For the uranium concentration we give two values. We obtained a minimum value by supposing the uranium in the grain to be in equilibrium of disintegration and supposing the recorded α-tracks to be emitted by the UI, UII, Io, Ra, Rn, RaA, RaC' and RaF. On the other hand, we obtained a maximum value by ignoring the equilibrium of disintegration and supposing only the presence of the UI and UII α-tracks.

Furthermore, we estimated the average U concentration of the spherical grains. Distribution of the diameter was determined by measuring 828 grains (Fig. 3). The diameter occurring most frequently was about 12 μ. The average number of tracks emitted by one grain was 5,13 (Table I). The calculations were based on an average density of 3,0 g/cm³ and the assumption that the uranium content of 25 % of the grains was negligible (less than $1{,}15 \cdot 10^{-12}$ g). We found the average uranium concentration of the spherical grains in our test substance to be 0,65 % for pure uranium, and 0,16 % when taking the equilibrium of disintegration into account.

Szalay and Almássy [10, 11] have ascertained that uranium is in radioactive equilibrium in coal and thus presumably, it is in equilibrium in the ashes

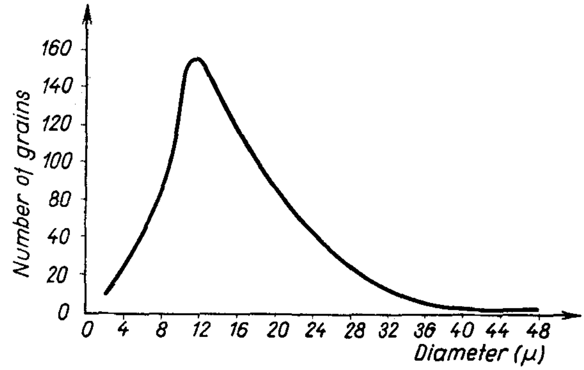

Fig. 3. Distribution of the diameter of grains fused into spheres

too. Therefore the reported values for the equilibrium of disintegration for uranium concentrations seem to be more reliable.

Summing up, we may state that in the ashes of pulverized uraniferous coals, uranium is mostly contained by the ash grains fused into spheres, which have a lower melting point. — Our next task is going to be the chemical and mineralogical determination of these ash grains.

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ИССЛЕДОВАНИЕ СОДЕРЖАНИЯ УРАНА В ЗОЛАХ УГЛЕЙ

Э. БУЙДОШО, Л. МЕДВЕЦКИ и А. САЛАИ

Резюме

Методом фотоэмульсии были исследованы активности зол разных углей. Мелкие частицы золы рассматривались под микроскопом; наблюдались две группы частиц: шарообразные частицы и частицы, имеющие неправильные формы. Шарообразные частицы имеют более низкую точку плавления по сравнению с температурой пространства горения котла, поэтому стали шарообразными.

Частицы золы вводились в ядерную эмульсию, изготовленную фотохимической исследовательской лабораторией завода Фортэ. После проявления фотоэмульсии считались трэки. Ряд исследований показал, что 70,6% от всей активности принадлежит шарообразным продуктам. Нами были проведены измерения по определению распределения диаметров и плотностей частиц.

Активность обнаружена в 25% от шарообразных частиц, отношение трэков к частицам в среднем 5,13 $\frac{\text{трек}}{\text{частица}}$. На основе наших измерений получилось среднее значение плотности частиц 3,02²см³, среднее значение диаметра 12 μ. Таким образом можно определить среднюю концентрацию урана в шарообразных частицах.

Содержанием тория в частицах можно пренебречь, поскольку аналитические исследования показали, что концентрация тория по крайней мере 10^{-4} %. Если предполагаем, что активность частиц вызвана лишь присутствием урана, концентрация урана 0,65%, при равновесном распаде 0,16%. Исследования Салаи и Алмашши показывают, что уран в угле находится в равновесии со своими продуктами распада, поэтому можно предполагать, что это имеет место и в золе, значит, 0,16% является наиболее вероятным значением.