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GENERATION OF CAVE AEROSOLS BY ALPHA PARTICLES: CRITICAL EVALUATION OF THE HYPOTHESIS

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The paper evaluates the feasibility of the hypothetical mechanism of cave aerosols generation under the action of natural radioactivity. Analysis has been performed from the standpoints of nuclear physics and aerosol mechanics. The hypothetical mechanism involves dislodgment of atoms and ions and knocking-out of larger fragments due to the bombardment of the bedrock by alpha-particles residing in the cave air. Calculations show that the largest amount of atoms and ions that could be generated by alpha-bombardment does not exceed 0.1 g from 1000 m^2 of the cave surface per 1 million years a quite negligible value. Presence of any water film thicker than 0.1 micron on the cave wall would completely prevent the dislodgment. The hypothetical mechanism, though physically plausible, cannot play any essential role in the generation of cave aerosols, and much less in the formation of speleothems.

The hypothesis about growth of certain types of speleothems from aerosols is becoming increasingly popular. One of the crucial problems with the aerosol mechanism is how does the speleothemic material (e.g., calcite or gypsum) get into the cave air and becomes an aerosol? The most intuitively comprehensible mechanism is the dispersion of water drops falling from a cave ceiling or generated by rapids and waterfalls in cave streams (Gadoros & Cher, 1986). This mechanism, however, is not applicable to many caves that do not contain any dripping or running waters. In 1994-1995 Klimchouk et al. published two papers (one in the *National Speleological Society Bulletin*) where, among other ideas, they discussed the possibility that cave aerosols are generated under the action of natural radioactivity. In the present article we will evaluate the feasibility of this hypothesis.

Four mechanisms of aerosol generation were suggested by Klimchouk et al. (Figure 1):

(1) Radioactivity (alpha, beta, and gamma) ionizes the cave air and the ions serve as condensation nuclei. Radon daughters can also behave as light ions and form molecular groups with water, oxygen, and other gas molecules, which can induce condensation.

(2) High-energy alpha particles formed in the cave air near the walls (zone 1 in Figure1) dislodge atoms and ions out of bedrock and these dislodged particles become aerosols.

(3) Alpha particles formed within the bedrock due to radioactive decay dislodge atoms and ions from a thin near-surface layer of the bedrock (zone 2 in Figure 1).

(4) Alpha particles knock out mineral fragments from the bedrock, generating small-size aerosol particles.

Now, let us quantitatively evaluate the possible contributions of each of these processes to the transfer of bedrock material into the cave air.

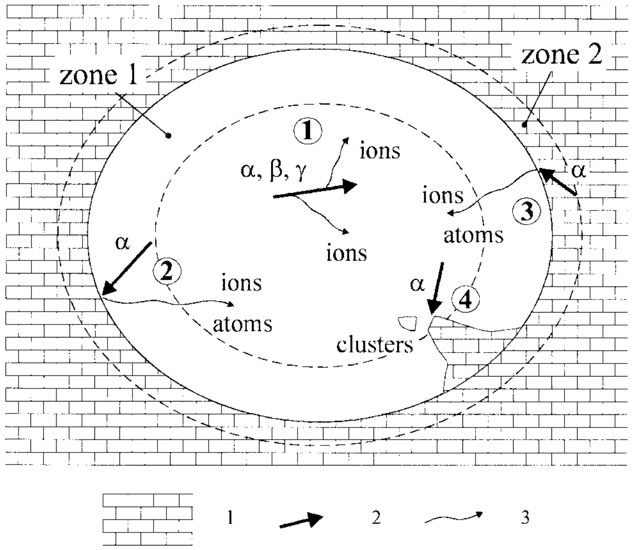

Figure 1. Processes responsible for generation of aerosols in caves by alpha particles as suggested by Klimchouk et al. (1995): 1 - bedrock; 2 - action of radiation; 3 - movement of atoms and ions. Numbers in circles correspond to the processes discused in the text. Zones 1 and 2 are explained in the text.

Process 1. Alpha particles, as they move through the air, create a dense chain of ions. The total number of ions created by a single alpha particle along its path (the mean free length for alpha particles in the air varies from 6 to 10 cm) reaches 10^5-$2 \cdot 10^5$ (Berthelot, 1948). Thus, in caves, where the radioactivity of the air typically varies from 10^4 to 10^5 Bq m^{-3} (Klimchouk et al., 1995), some 10^9-10^{10} pairs of ions are created in one cubic meter of the air every second. At high supersaturation levels these ions may serve as condensation nuclei for vapor present in the cave air, and thus, produce hydroaerosols. However, these supersaturation levels must be much higher than can be achieved in even the most humid cave in order for this condensation mechanism to become important (as in a Wilson cloud chamber). Also, as Klimchouk et al. admit, this process cannot play a significant role in the mass transfer of bedrock material because vapor in the cave air does not contain solutes.

Process 2. The number of atoms (n_{atom}) that can be dislodged by alpha particles out of bedrock may be defined as:

(1)

$$n_{atom} = B\frac{\pi}{8} N_{air} L_{air} St,$$

where B - is the number of atoms or ions dislodged by impact of a single alpha particle; N_{air} is the radioactivity of the cave air [Bq m^{-3}, or s^{-1} m^{-3}]; L_{air} is the mean free length for alpha particles in the air [m], S is the surface area of the bedrock exposed to radiation [m^2], and t is the time [s]. For further calculations we need to estimate B. Theoretically, its maximum value may be as high as 10^6. This number reflects a purely hypothetical situation in which the entire energy of the alpha particle (≈ 5 MeV) is spent for the dislodgment of atoms from a solid surface. In reality, however, such a situation is impossible because particles lose energy as heat and ionization while moving in the air and even more so in the near-surface layer of solids. As those losses are great (Radioactivity..., 1954; Novikov & Kapkov, 1965), the most conservative estimation would give B ≈ 10^3, which means that each alpha particle on impact with the cave wall can dislodge 1000 atoms of the bedrock material. Substituting of B = 103, N_{air} = 10^5 Bq m^{-3}, L_{air} = 0.1 m, S = 1000 m^2, and t = 1000 years in Eq. (1) gives for the number of dislodged atoms: $n_{atom} = 10^{20}$. In terms of the mass this corresponds to the removal of ≈ 0.01 g of the bedrock material from 1000 m^2 per 1000 years.

To enable aerosol mass-transfer it is necessary that atoms and ions are not only dislodged from the crystal lattice, but are also transferred into the cave air. Because of the quite-high diffusion mobility (≈ 0.05 cm^2 s^{-1}) of the atoms, the probability that such atoms will hit the wall and then get stuck back to it exceeds 99% (Foux, 1955). If this process of diffusion re-precipitation is taken into account, the proportion of dislodged material that can become aerosol is even less: ≈ 0.1 g from 1000 m^2 of the cave surface per 1 million years.

Process 3. Let us estimate how much material can be dislodged out of a cave wall surface by alpha particles formed due to radioactive decay inside the rock. The mean free length for alpha particles in solids varies from 20 to 50 microns (zone 2 in Figure 1), depending on density of solids. The radioactivity of the rock N_{rock} is defined as:

(2)

$$N_{rock} = \lambda_{Rn} C_{Rn}$$

where λ_{Rn} is the decay constant for radon [s^{-1}], and C_{Rn} is the concentration atoms of radon within zone 2 [m^{-3}]. Concentration of radon in the rock is described by equation (3) (Novikov & Kapkov, 1965):

(3)

$$C_{Rn}(x) = \frac{\lambda_{Ra} C_{Ra}}{\lambda_{Rn}} \left[1 - \exp\left(-\left(\frac{\lambda_{Rn}\eta}{D_{rock}} \right)^{1/2} x \right) \right]$$

where λ_{Ra} is the decay constant for radium [s^{-1}], C_{Ra} is the concentration of atoms of radium in the rock [m^{-3}], η is the porosity of the rock [parts of unity], D_{rock} is the coefficient of diffusion of radon in the rock [m^2 s^{-1}], and x is the distance from the cave wall into the rock [m]. Concentration of radon deep inside the rock ($x = \alpha$) is defined as:

(4)

$$C_{Rn}(\infty) = \frac{\lambda_{Ra} C_{Ra}}{\lambda_{Rn}}$$

Measurements in different geological settings yield D_{rock}/η ranging from $5 \cdot 10^{-2}$ to $2 \cdot 10^{-4}$ cm^2 s^{-1}, and the upper estimates of radioactivity of the rock, N_{rock} (a), do not, normally, exceed $4 \cdot 10^6$ Bq m^{-3} (Novikov & Kapkov, 1965).

The final equation defining the number of atoms (ions) dislodged out of solid surface by alpha particles acting from inside the rock may be written as:

(5)

$$n_{atom} = N_{rock}(\infty)\frac{\pi}{8} StB_{rock} \int_0^\infty \left[1 - \exp\left(-\left(\frac{\lambda_{Rn}\eta}{D_{rock}} \right)^{1/2} x \right) \right] dx$$

For further calculations we need to estimate B_{rock}, which is the analogue of B in Eq. (1). In solids, the take-off of atoms and ions is only possible from a very thin near-surface layer less than 0.01 micron thick (which is the maximum free length of recoil atoms; Radioactivity..., 1954). The majority of alpha particles in zone 2 lose their kinetic energy well before reaching this zone (the energy being spent for heat and ionization). Analysis of energy losses (Radioactivity..., 1954; Novikov & Kapkov, 1965) gives the most reliable estimate of B_{rock} ≈ 1. It can be computed from Eq. (5) that the mechanism under consideration may disperse some 0.003 g of solid material out of 1000 m^2 of the cave wall per 1000 years. Taking into account diffusion re-precipitation (see discussion of process 2), the fraction of this material which can become an aerosol will be ≈ 0.000003 g. In other words, the hypothetical process 3 may generate approximately 0.003 g of aerosol from 1000 m^2 of the cave wall per 1 million years.

Process 4. Klimchouk et al. (1995) hypothesize the "knocking-out" of mineral fragments from cave walls by alpha particle bombardment. The essence of this hypothesis is as follows. The adhesion energy of a particle 0.1 micron in size is ≈ 0.1 MeV, whereas the energy of alpha particle is ≈ 5 MeV. Hence, the alpha particle may easily dislodge such a particle from the surface, and, given that the circumstances are favorable, can do the same to 50 more adhesion particles. It is assumed here, however, that the particles are already removed from a crystal lattice (by some unspecified process) and are attached to the bedrock surface by adhesion only. Much greater energy would be required to "knock out" such large particles from the lattice. Besides, using only the energy conservation law, the authors of the hypothesis failed to consider the actual physics of the process. Having energies far exceeding that characteristic of crystal lattice bonds, alpha particles interact mainly with atomic nuclei, dislodging or shifting them by no more than 10-20 nanometers. Those effects are well-known in nuclear physics (Radioactivity..., 1954), and the theory of diffusion and recoil was developed in the late 1930s (Flugge & Zimens, 1939). In particular, it has been demonstrated in the later work that the recoil atom may penetrate through the solid no more than 10 nanometers, and most of its energy is spent for melting of an area of similar size. Taking all that into account, process 4 also does not look possible.

In all of the above, we have discussed only the interaction of alpha particles with a DRY cave wall. However, due to the fact that caves almost always possess high humidities, it is quite typical of cave walls to be wet. As mentioned above, the mean free length for recoil atoms, clusters, and fragments created by impact interaction of alpha particles with solid surface does not exceed 100 nanometers: hence, any water film thicker than 0.1 micron would completely absorb them.

CONCLUSION

Ideally speaking, the mechanism of aerosol generation by dislodging particles out of bedrock under the action of natural radioactivity, as suggested by Klimchouk et al. (1994; 1995), is physically plausible. However, quantitative estimations show that this mechanism cannot play any essential role in the generation of cave aerosols, much less in the formation of speleothems. We would like to emphasize that this conclusion does not imply that formation of speleothems by an aerosol mechanism is impossible in principle, and we will discuss this possibility in a later article in the *Journal of Cave and Karst Studies*.

REFERENCES

Berthelot, A. (1948). From Atom to Nuclear Energy. *Inostrannaja Literatura*. Moscow: 207 p. (In Russian. Available in French: *De l'atome a l'energie nucleaire*. Paris, 1947).

Flugge, S. & Zimens, K. (1939). *Z. Physik. Chem*., B42: 179.

Foux, N.A. (1955). Aerosol Mechanics. *Academy of Science of the USSR*. Moscow: 351 p. (In Russian)

Gadoros, M. & Cher, F. (1986). Aerosols in caves - theoretical considerations. *Proceedings of the 9th International Congress of Speleology 2*. Barcelona: 90-92.

Klimchouk, A., Nasedkin, V., & Cunningham, K. (1994). Cave Mineral Formations of Aerosol Origin: *Problems of Physical Speleology, Physico-Technological Institute of Moscow*. Moscow: 25-50. (In Russian)

Klimchouk, A., Nasedkin, V., & Cunningham, K. (1995). Speleothems of Aerosol Origin. *National Speleological Society Bulletin 57*: 31-42.

Novikov, G.F. & Kapkov, Y.N., 1965, Radioactive methods of exploration. *Nedra*. Leningrad: 759 p. (In Russian)

Radioactivity Applied to Chemistry (1954). Inostrannaja Literatura, Moscow, 562 p. (In Russian. Available in English: *Radioactivity Applied to Chemistry* / A.C. Wahl & N.A. Bonner, Eds. New York - London, 1951).