

Estimation of radionuclides migration by the data from

$^{206}\text{Pb}/^{238}\text{U}$ - $^{207}\text{Pb}/^{235}\text{U}$ and ^{234}U - ^{238}U systems studies

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Taking into account the fact that pitchblende, the main uranium mineral of uranium deposits, has similar properties to synthetic uraninite, which constitutes 96% of spent nuclear fuel (SNF), uranium deposits of pitchblende ores can rightly be considered as natural analogues of underground SNF disposal and long-term storage facilities. Stability and disturbance of U-Pb and ^{234}U - ^{238}U isotopic systems at the sites of uranium mineralization development were investigated with the purpose of quantitative estimation of original uranium ores preservation degree. For this purpose a profile was selected through the ore zone in trachydacites of Streltsovskoe deposit (Western Transbaikalia, Russia) [1]. The profile is a typical cross section of a mineralized zone at a mining level at a depth of approximately 230 m from the surface. The profile was started near a fault, passed through a zone of stockwork-vein U mineralization ~2.5 m thick, and was ended in the host rocks at about 1.5 m from the interval with visible U mineralization (Fig. 1).

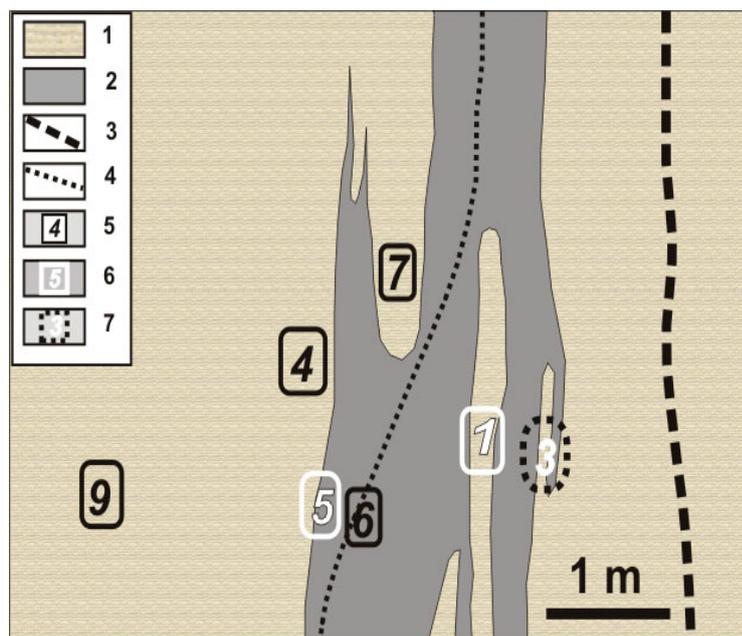


Fig. 1. Sampling scheme across the zone of uranium mineralization. The Streltsovskoye deposit.

1 – altered host rocks; 2 – zone of uranium mineralization; 3 – fault; 4 – linear zone of cataclasis; 5 – samples and their numbers in which $t(^{206}\text{Pb}/^{238}\text{U}) > t(^{207}\text{Pb}/^{235}\text{U})$ ratio was determined; 6 - samples and their numbers in which $T(^{206}\text{Pb}/^{238}\text{U}) < T(^{207}\text{Pb}/^{235}\text{U})$ ratio was determined; 7 - samples and their numbers in which both ratios of $T(^{206}\text{Pb}/^{238}\text{U}) > T(^{207}\text{Pb}/^{235}\text{U})$ and $T(^{206}\text{Pb}/^{238}\text{U}) < T(^{207}\text{Pb}/^{235}\text{U})$ were determined.

Our collection of samples (see Fig. 1) represents U mineralization in veinlets with quartz (samples 1, 3a and 5a), massive pitchblende aggregates (sample 3b), disseminated and impregnated U mineralization (samples 4a, 4b, 5b, and 7), hydrothermally altered trachydacites with U concentration close to the background (sample 9a), and a fragment of the mineralized zone with clearly pronounced post-mineral cataclasis (sample 6).

The predominant ore mineral in the profile is pitchblende. In intervals with disseminated mineralization, this mineral is accompanied by low-reflectance U oxides and U silicate.

Samples for our isotopic research were collected by sawing fragments 4-8 cm³ from large polished hand-specimens. Such a volume of the samples 30-50 g in mass contained various minerals, including proper U minerals. The samples were crushed to size less than 0.001 mm and then used (1) to determine their chemical composition by XRF, (2) to conduct their X-ray powder diffraction analysis, and (3) to examine their U-Pb and ²³⁴U-²³⁸U isotopic systematics.

Figure 2 shows the distribution of major components, U, Pb, and Zr in segments of the profile. The broad variations in the U concentration reflect the distribution of its predominant ore mineral (pitchblende) in the profile. Ca, Zr, and Pb are positively correlated with U, because the former two elements are accommodated as isomorphous components in the pitchblende structure, and Pb is produced in it by radioactive decay. Intervals with pitchblende are characterized by a decrease in the concentration of Na and, partly, also K. This fact is explained by the replacement of orthoclase and plagioclase by clay (montmorillonite) and mica (sericite) minerals in the trachydacites hosting the U ores. The same segments are marked by an increase in LOI values.

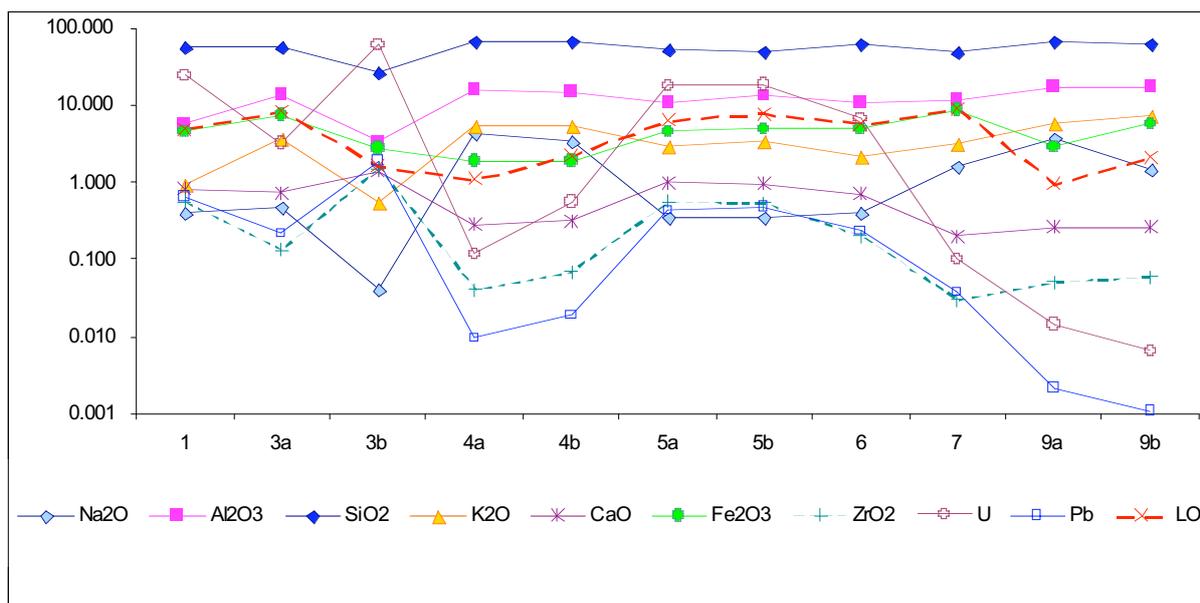


Fig. 2. Distributions of elements in the profile across the zone of U ore mineralization in trachydacites.

In studies of elements behavior in the process of post-ore transformations, the remarkable peculiarity of the U-Pb method was used, that is, the presence of two independent decay series (²³⁸U - ²⁰⁶Pb and ²³⁵U - ²⁰⁷Pb) with different half-life periods. ²³⁸U - ²⁰⁶Pb and ²³⁵U - ²⁰⁷Pb systems react differently on the processes that perturb their isolation what permits to obtain two different age values – T(²⁰⁶Pb/²³⁸U) and T(²⁰⁷Pb/²³⁵U).

All samples taken from the profile show discordant T(²⁰⁶Pb/²³⁸U) and T(²⁰⁷Pb/²³⁵U) age values, a fact testifying that their U-Pb systems were disturbed (Table). For comparison,

Table. U-Pb isotopic data obtained for our samples from the profile.

Sample	Concentration, wt. %		Isotopic composition of lead, %				Isotopic age, Ma		$^{234}\text{U}/^{238}\text{U}$ ($\times 10^{-5}$)
	U	Pb	^{208}Pb	^{207}Pb	^{206}Pb	^{204}Pb	$t(^{206}\text{Pb}/^{238}\text{U})$	$t(^{207}\text{Pb}/^{235}\text{U})$	
85	77.99	1.6341	4.833	6.198	88.84	0.126	135	135	5.508
1	24.06	0.6514	17.80	10.60	71.13	0.464	126	138	5.509
3b	61.16	1.8862	19.60	10.97	68.92	0.511	136	140	5.515
5a	18.12	0.4323	17.11	10.71	71.73	0.446	112	136	5.523
5b	19.00	0.4882	17.31	10.46	71.78	0.451	121	134	5.521
3a	3.285	0.2153	23.65	11.03	64.71	0.616	256	142	5.525
4b	0.565	0.0191	13.49	7.917	78.25	0.349	179	127	5.511
4a	0.119	0.0096	21.65	9.805	67.99	0.558	340	134	5.516
7	0.100	0.0374	37.89	15.65	45.47	0.987	717	142	5.506
9a	0.015	0.0022	44.29	18.35	36.23	1.135	165	142	5.523
6	6.731	0.2292	19.66	10.64	69.19	0.512	150	137	5.520

the table also lists data on pitchblende from previously examined sample 85 [2], whose U-Pb system has been closed since the time when the mineral was formed until nowadays.

The relations between the age values correspond to the inequality $T(^{206}\text{Pb}/^{238}\text{U}) < T(^{207}\text{Pb}/^{235}\text{U})$ for samples with high (>10%) U concentrations and to $T(^{206}\text{Pb}/^{238}\text{U}) \gg T(^{207}\text{Pb}/^{235}\text{U})$ for samples with lower U concentrations. The strong U-Pb isotopic discordance of the samples (>3%) provides evidence of a notable role of migration of intermediate decay products in the ^{238}U - ^{206}Pb system (RD^{238}U) migration.

U-Pb data for the samples collected from intervals with high U concentrations (samples 1, 3b, 5a, and 5b) and from those with low U concentrations (samples 3a, 4a, 4b, 6, 7, and 9a) are shown in a concordia plot (Fig. 3).

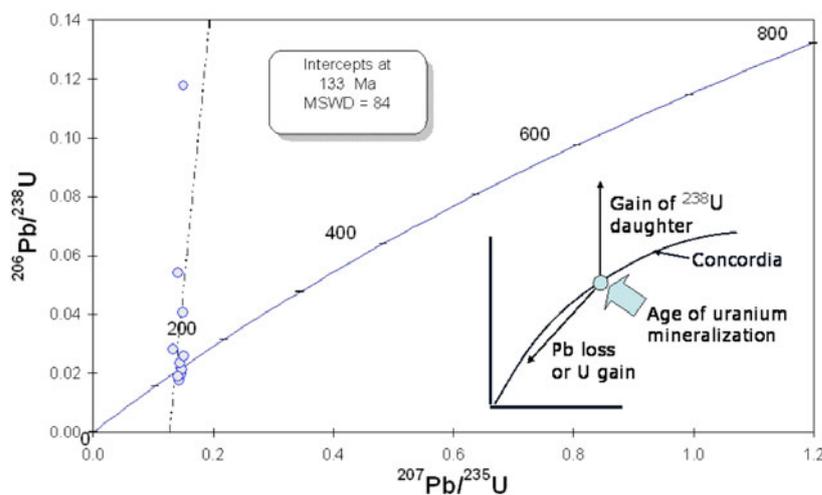


Fig. 3. Concordia plot for samples from the profile through the zone of U mineralization.

The discordia approximating our experimental data points is practically parallel to the $^{206}\text{Pb}/^{238}\text{U}$ axis, i.e., its orientation is notably different from that typical of usual situations with general Pb loss (or gain). These data provide direct evidence that these two groups of samples were affected by different processes. The discordance character of the first group, for which $T(^{206}\text{Pb}/^{238}\text{U}) < T(^{207}\text{Pb}/^{235}\text{U})$ and whose data points plot below the concordia, testifies that these samples lost their RD^{238}U during a long period of time. The discordance character of the second group, for which $T(^{206}\text{Pb}/^{238}\text{U}) \gg T(^{207}\text{Pb}/^{235}\text{U})$ and whose data points lie above the concordia, suggests the gain of RD^{238}U at these sites, which ensured an excess of the ^{206}Pb isotope. The aforementioned migration of intermediate decay products did not affect the ^{235}U - ^{207}Pb system, in which the lifetime of the intermediate decay products was four to seven orders of magnitude briefer than in the ^{238}U - ^{206}Pb system. Despite significant discrepancy of $T(^{206}\text{Pb}/^{238}\text{U}) - T(^{207}\text{Pb}/^{235}\text{U})$, the $T(^{207}\text{Pb}/^{235}\text{U})$ values fluctuate in a relatively narrow range and are close to the actual age of uranium mineralization. Maximal differences do not exceed 6% and apparently partly are caused by uncertainty of inherited lead correction. It allows to think, that there was no mass uranium migration. As the additional arguments in favor of absence of severe uranium migration are data of $^{234}\text{U}/^{238}\text{U}$ ratio measurements (see table). This ratio in all samples is close to equilibrium ratio which is $5.510 \pm 0.015 (\times 10^{-5})$ [3-5]. The analysis of the discordance character at different sites of the profile (Fig. 1) allows to say that isotopes deficit of U-Pb system at one site is compensated by its surplus at the neighboring one. Hence it follows that the migration of components of the U-Pb system (mainly ^{238}U -daughters) was limited by the scale of several tens of centimeters. It is obvious that, taking into account compensatory character of U-Pb system disturbance, the actual scales of uranium migration beyond the selected profile area should not exceed few percents.

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