

## On the Nuclear Physical Stability of the Uranium Minerals

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**A**N attempt is made in this paper to apply the nuclear reactor theory in geochronology and to explain certain interrelations between the age and the nuclear physical stability of the uranium minerals, as well as the geological environments of the mineral formation.

The infinite multiplication constant,  $k_{\infty}$ , may be considered as an indicator of the stability of the uranium minerals, which are the natural assemblages of uranium, moderator, and impurities. We may consider a system to be quite "stable," when the infinite multiplication constant of the assemblage is far less than unity. The system will be nuclear physically "unstable," when the infinite multiplication constant is greater than unity.

According to the nuclear reactor theory,

$$k_{\infty} = \epsilon p f \eta, \quad (1)$$

where  $\epsilon$  is the fast fission factor,  $p$  is the resonance escape probability,  $f$  is the thermal utilization factor, and  $\eta$  is the number of fast neutrons available per neutron absorbed by uranium.

When dealing with geological events, the change of the uranium enrichment as a function of geological time should also be taken into consideration. The major neutron sources in minerals are the spontaneous fission and the  $(\alpha, n)$  reactions.

The values of  $p$  and  $f$  can be calculated if the chemical composition of the mineral is given,  $\epsilon$  is always close to unity, and  $\eta$  as a function of the uranium enrichment is known.<sup>1</sup> Hence the value of  $k_{\infty}$  of a mineral at any geological time can be calculated.

Table I shows the calculated values of  $p$ ,  $f$ ,  $\eta$ , and  $k_{\infty}$  of a sample of Johanngeorgenstadt pitchblende.<sup>2</sup> Similar calculations show that most of the uranium minerals were nuclear physically "stable" during the past 2800 million years, provided the water content of the minerals had remained unchanged during the geological past. It is worthy of note, however, that a slight increase of the water to

TABLE I.  $p$ ,  $f$ ,  $\eta$ , and  $k_{\infty}$  of a Johanngeorgenstadt pitchblende. Chemical composition of the mineral: 1 mole  $\text{UO}_2$ ;  $\frac{1}{2}$  mole  $\text{H}_2\text{O}$ ; 0.0  $n$  moles  $\text{PbO}$ ,  $\text{CaO}$ ,  $\text{MgO}$ ,  $\text{As}_2\text{O}_5$ ,  $\text{SiO}_2$ ; 0.00  $n$  moles  $\text{Al}_2\text{O}_3$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{CuO}$ ,  $\text{MnO}$ ,  $\text{Bi}_2\text{O}_3$ ,  $\text{V}_2\text{O}_5$ ,  $\text{MoO}_3$ ,  $\text{WO}_3$ , alkalis,  $\text{SO}_3$  and  $\text{P}_2\text{O}_5$ .

Geological time (million years)	0 (present)	700	1000	1400	2100	2800
$\text{U}^{235}$ enrichment (percent)	0.7	1.3	1.6	2.3	4.0	7.0
$p$	0.47	0.45	0.43	0.42	0.38	0.34
$f$	0.93	0.95	0.96	0.97	0.98	0.99
$\eta$	1.32	1.57	1.66	1.77	1.91	1.98
$k_{\infty}$	0.58	0.67	0.69	0.72	0.71	0.67

TABLE II. The water-uranium ratio and the values of  $p$ ,  $f$ ,  $\eta$ , and  $k_{\infty}$  (Johanngeorgenstadt pitchblende, 2100 million years ago).

$n$	1/4	1/2	1	2	3	4	5	10
$p$	0.29	0.47	0.62	0.74	0.79	0.82	0.84	0.86
$f$	0.99	0.98	0.97	0.95	0.93	0.91	0.89	0.81
$\eta$	1.91	1.91	1.91	1.91	1.91	1.91	1.91	1.91
$k_{\infty}$	0.55	0.88	1.15	1.34	1.40	1.42	1.43	1.33

uranium ratio could have easily caused a sharp upward change of  $p$ , without affecting  $f$  considerably, and the result of which could have been enough to make the system nuclear physically "unstable."

It is generally accepted that the deposition of the uranium minerals took place at the pegmatitic-pneumatolytic and early hydrothermal stages. Hence, one may consider that the crystallization of the uranium minerals represents the following sequence of events. An aqueous solution of uranium ( $\text{U}^{235}$  enriched) is gradually converted to an assemblage of uranium plus  $n$  moles of water ( $n=1, 2, 3, \dots n$ ) and finally to an almost water-free uranium mineral.

Let us imagine that the crystallization of the Johanngeorgenstadt pitchblende took place 2100 million years ago. The calculated values of  $p$ ,  $f$ ,  $\eta$ , and  $k_{\infty}$  are shown in Table II. Table II shows that the assemblages of the Johanngeorgenstadt pitchblende plus water were nuclear physically "unstable" 2100 million years ago, and the critical uranium chain reactions could have taken place, if the size of the assemblage was greater than, say, a thickness of a few feet. The effect of such an event could have been a sudden elevation of the temperature, followed by a complete destruction of the critical assemblage.

The effect of the ground water or the water vapor from the molten magma could have resulted in the formation of a nuclear physically "unstable" assemblage of uranium plus  $n$  moles of water. Such mechanism might explain the fact that the ages of the large uranium deposits never exceed 2000 million years, or the marked discrepancies exist between the  $\text{Pb}^{206}/\text{U}^{238}$  age and the  $\text{Pb}^{207}/\text{Pb}^{206}$  age of the uranium minerals.

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<sup>1</sup> The method of calculations is described in the following reference books; Samuel Glasstone, *Principles of Nuclear Reactor Engineering* (Princeton University Press, Princeton, 1955); Raymond L. Murray, *Introduction to Nuclear Engineering* (New York, 1954); Richard Stephenson, *Introduction to Nuclear Engineering* (New York, 1954).

<sup>2</sup> F. W. Clarke, *The Data of Geochemistry* (Washington, 1924).