Natural Neutron-Fission Wave

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The dynamics of Feoktistov's neutron-fission wave is considered. The possibility of this process under natural conditions, namely in the interior of planets, is assessed.
Key words: nuclear fission, neutrons, chain reaction, diffusion combustion, geophysics.

Feoktistov [1] showed that a self-propagating neutron-fission wave can be formed in a material incapable of a chain reaction under normal conditions (e.g., ^{238}U). For example, if half-space filled with uranium is irradiated with neutrons, plutonium is accumulated near the surface. Eventually, critical conditions can be reached and a reaction will start. Neutrons leaving the reaction zone are captured by subsequent uranium layers, in which plutonium concentration also increases. Under certain conditions, a steady wave can form that is independent on the initiation history. At the front of this wave, uranium is converted to plutonium by fission neutrons. The wave velocity is of the order of L/τ, where $L \approx 5$ cm is the diffusion length and $\tau = 2.3/\ln 2 = 3.3$ days is the time of plutonium formation from ^{239}U. Feoktistov [1, 2] used an uranium medium (or an uranium cylinder ≈ 1 m in diameter) as an illustrative geometrical model of a fast breed reactor that is internally safe owing to a large time τ of fuel activation and does not require an intermediate technology of plutonium isolation.

At the same time, Driscoll [3] and Herndon [4] put forward a hypothesis of actinoid concentration deep in the interior of planets during gravitational differentiation of their material. The same idea was developed independently and in more detail in [5, 6]. The Earth's crust is enriched with uranium and thorium as compared with the mantle. This concentration is considered to be a result of passing of these elements into a melt during the heating of the Earth's primary material and subsequent floating of lighter liquid fractions to the surface. However, there are some compounds for which the reverse drift direction is more natural. For example, uranium oxide (UO_2) is more refractory and denser than the material of the Earth's mantle and external core. Because of the relatively low solubility of UO_2 in the melts of the mantle and the core (as indicated by the data of [6]), one should expect sedimentation of uranium oxide particles onto the surface of the inner (hard) Earth's core, whose radius is ≈ 1200 km. At present, uranium oxide is inert, and all the more mixed with liquid iron, but in the past, when the fraction of ^{235}U was larger, a chain reaction was a natural consequence of the above-mentioned hypothesis. The aforesaid is also true for a number of other uranium compounds (UN, UC). Anisichkin [5] considered for the first time the feasibility of nuclear explosion of part of such a "deposit," for example, after a large asteroid impact, and possible planetary consequences of this event. Mitrofanov et al. [6, 7] gave experimental and theoretical grounds for this hypothesis and examined other models of nuclear energy release: relatively slow burnup maintained by settling-out of fresh fuel portions, a reaction coupled with the propagation of an internal gravitational wave of the level of the two-phase layer of uranium oxide–liquid metal, and nuclear detonation.

Undoubtedly, the presence of a concentrated uranium layer would Give a literal meaning to the concept of the neutron-fission wave. Since present-day uranium, even depleted, is in principle capable of reacting in such a wave (after initial ignition) with considerable energy release, it seems important to assess the real possibility of this wave regime. In the present paper, we consider characteristics of such waves and their possible manifestations.

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GOVERNING EQUATIONS

The problem of the excitation and propagation of a neutron-fission wave is considered within the framework of the approximations of [1]. In particular, the neutron capture cross sections for all substances, except for oxygen and iron, are considered to be equal to the cross section σ_{a} for ^{238}U. For iron, the absorption cross section is smaller by about an order of magnitude and the absorption by oxygen nuclei is ignored. For our goals, the equations of [1] are more conveniently written as

$$\varepsilon \frac{\partial n}{\partial t} = \frac{\partial^2 n}{\partial x^2} - n\left(1 - \frac{n_{\mathrm{Pu}}}{n_{\mathrm{cr}}}\right),$$

$$\frac{\partial n_8}{\partial t} = -n(n_8 - n_{\mathrm{Pu}} - n_9),$$

$$\frac{\partial n_9}{\partial t} = n(n_8 - n_9) - n_9, \quad \frac{\partial n_{\mathrm{Pu}}}{\partial t} = n_9 - n\frac{n_{\mathrm{Pu}}}{\gamma}. \quad (1)$$

Here the time t and coordinate x are measured in the units of τ and L, respectively, n_8, n_{Pu}, and n_9 are the dimensionless concentrations of ^{238}U, ^{239}Pu, and ^{239}Np, (in the initial state, $n_8 + n_{\mathrm{Pu}} = 1$), n is the neutron density in the units of $(\sigma_{\mathrm{a}} v \tau)^{-1}$, v is the characteristic neutron velocity, n_{cr} is the critical plutonium concentration, $\gamma = \sigma_{\mathrm{a}}/(\sigma_{\mathrm{a}} + \sigma_{\mathrm{f}})$, σ_{f} is the ^{239}Pu fission cross section so that the equilibrium plutonium concentration (at which Pu accumulation and fission by neutrons are in balance) is $\tilde{n} = \gamma n_8$. The necessary condition of the wave propagation is $\tilde{n} > n_{\mathrm{cr}}$; then, critical conditions can be attained during plutonium accumulation. In other words, transition through the critical concentration in the wave must proceed from below. For this, it is necessary that the critical concentration must be sufficiently low, at least, lower than the equilibrium concentration.

The value of the coefficient $\varepsilon = \lambda_{\mathrm{a}}/(v\tau)$, where λ_{a} is the absorption length, is quite small (for fast neutrons, it is of the order of 10^{-12}), so that the time derivative in the first equation of (1) can be omitted. This means that the diffusion is much faster than the kinetics. Feoktistov [1] took this approach and solved the steady-state problem, that is a system of ordinary differential equations. However, for our purposes, it is more convenient to retain the time-dependent form (1), which allows us to trace the wave formation and assess its stability. The diffusion equation was solved by a double-sweep method using an implicit scheme. In the calculations, the value of ε was so set small enough so that the term $\varepsilon \partial n/\partial t$ was negligible as compared with others but, at the same time, the numerical stability of the scheme was retained. Satisfactory results were obtained at $\varepsilon = 0.001$ but most calculations were performed at $\varepsilon = 0.0001$. In this case, diffusion remains

three to four orders of magnitude faster that the reaction.

In the present formulation of the problem, the of oxygen and iron affect only the model parameters, mainly the critical concentration n_{cr} (γ does not depend on dilution). In the calculations, the critical concentration was varied.

The roughest assumption of this model is a single-group treatment of neutrons. To a certain extent, it is possible to make the calculations more realistic by using, for example, a more detailed multigroup scheme to estimate n_{cr}. With infinite dimensions, the critical portion of ^{239}Pu in ^{238}U calculated using eight-group constants [8] is $\approx$3.7%. Dilution with oxygen to $\mathrm{UO}_2/\mathrm{PuO}_2$ increases the value of n_{cr} to 6.4%, primarily because of the neutron spectrum softening. The presence of iron in fuel pores with a typical "loose-packed" concentration of the fuel of $\approx$60% increases the critical portion of ^{239}Pu to 8.2% (densities of $\mathrm{UO}_2/\mathrm{PuO}_2$ and iron are assumed to be 19.5 and 12 g/cm^3, respectively, for conditions on the boundary of the hard core). The characteristic length L is approximately the same as in ^{238}U of normal density, i.e., roughly 6 cm (uranium dilution is compensated for by compression). Of interest for the present study is the case where the starting material is enriched with ^{239}Pu or ^{235}U. Within the framework of Feoktistov's approximations, we assume that these materials are equivalent. For comparison, we indicate the critical calculated concentrations of ^{235}U: 5.3% for metallic uranium, 7.85% for the oxide, and 9.8% for a mixture of the oxide with 40% (by volume) iron. Although plutonium is more reactive, this dependence is minor as compared with the rougher assumptions of the model and can be of significance only in boundary cases. The experimental value for metallic uranium is $n_{\mathrm{cr}} = 5.56\%$ [9], which indicates the quality of constants [8].

CALCULATION RESULTS

To test the computational scheme, we calculated the wave formation without initial enrichment [$n_8(x,0) = 1$ and $n(x,0) = n_{\mathrm{Pu}}(x,0) = n_9(x,0) = 0$], which corresponds to the formulation of [1]. A neutron flux was set on the left boundary of the calculation region ($x = 0$): at each time step, a certain constant quantity was added to $n(0,t)$ (0.1 per unit time). Here and below, we set $\gamma = 0.1$ (i.e., for this calculation in the initial state, $\tilde{n} = 0.1$). The critical concentration was varied. The calculated wave velocity was in good agreement with the data of [1] for $n_{\mathrm{cr}} = 0.04$–0.064.

Feoktistov [1] noted a similarity between the steady-state diffusion equation and the Schrödinger equation. This allowed the critical condition to be written as
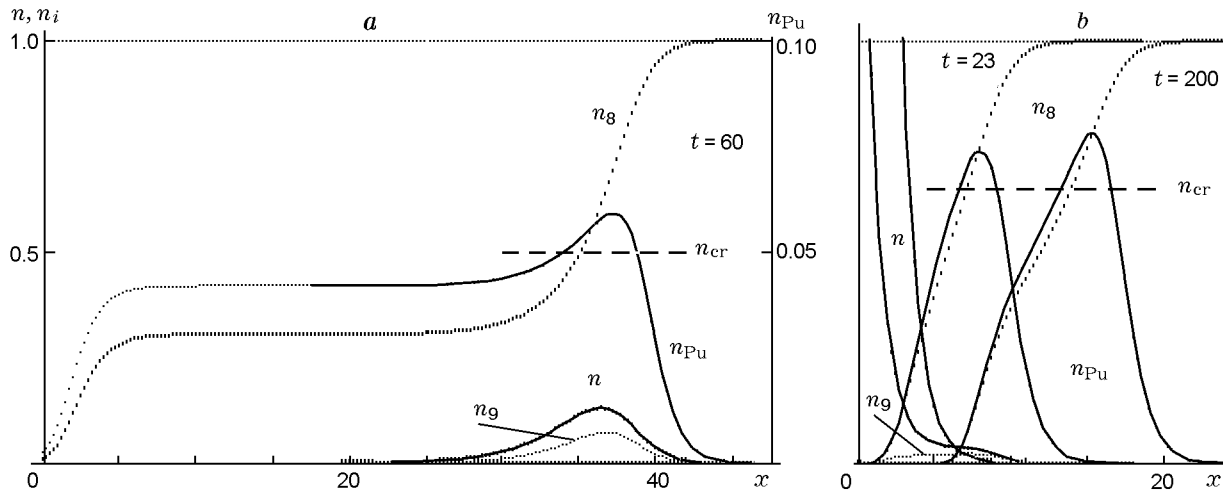
Fig. 1. Propagating wave (a) and "locked" wave (b): a segment of the curve of $n_{\mathrm{Pu}}(x)$ above the n_{cr} line is the active zone; the scales of n_{cr} and n_{Pu} are given with a $\times 10$ magnification.

$$I = \int \sqrt{\frac{n_{\mathrm{Pu}}}{n_{\mathrm{cr}}} - 1}\; dx = \frac{\pi}{2}, \qquad (2)$$

where the integral is taken over the supercritical region ($n_{\mathrm{Pu}} > n_{\mathrm{cr}}$). Equality (2) is similar to the Bohr–Sommerfeld condition in quantum mechanics. Here the potential well is the inverted plutonium concentration profile. Indeed, our calculations showed that in the steady-state regime, condition (2) is satisfied with accuracy up to several percent. There is no reason to expect higher accuracy because the quantization condition for the lower level is itself approximate.

Figure 1 shows the concentration distribution for $n_{\mathrm{cr}} = 0.05$ at $t = 60$. The wave velocity is $D = 0.625$. We note that in this regime, $\approx 70\%$ of $^{238}\mathrm{U}$ is burnt up.

If $n_{\mathrm{cr}} > 0.064$, the plutonium concentration can also exceed the critical concentration but the steady wave does not develop. Figure 1b shows calculation results for $n_{\mathrm{cr}} = 0.065$ at the times $t = 23$ and 200. At $t = 200$, even higher concentrations n_{Pu} were reached (and an even greater excess over n_{cr}) compared to the previous calculations but the length of this zone is small because the wave front moves extremely slowly. With time, the concentration n_9 drops to such a level that it becomes indistinguishable on the same scale. Such a "locking" can be quantitatively explained as follows. The rate of plutonium accumulation decreases as the limiting value of $\tilde{n}$ is approached; therefore, the concentration increases slowly. The condition to be imposed on the integral I for small values of the difference $n_{\mathrm{Pu}} - n_{\mathrm{cr}}$ requires that the supercritical region have a considerable width: $\Delta \sim \sqrt{n_{\mathrm{cr}}/(n_{\mathrm{Pu}} - n_{\mathrm{cr}})}$; however, the characteristic wave width does not grow with increase in n_{cr} but even decreases. As $^{238}\mathrm{U}$ is consumed, the equilibrium plutonium concentration also decreases:
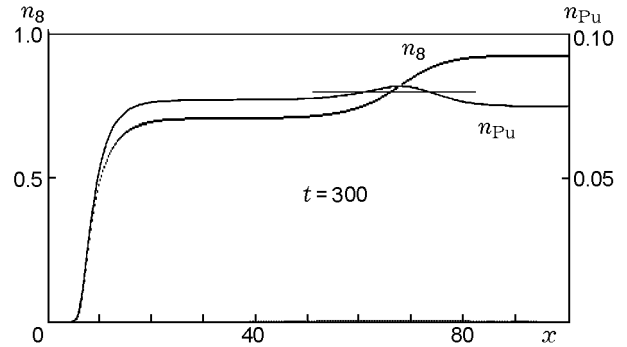

Fig. 2. Near-critical regime for the high-level initial enrichment: n_{cr} and n_{Pu} are given with a $\times 10$ magnification.

$\tilde{n} = \gamma n_8$. The scales of Figs. 1 and 2 are such that under our assumptions (for $\gamma = 0.1$), the equality $\tilde{n} = n_{\mathrm{Pu}}$ is reached upon intersection of the curves of n_8 and n_{Pu}. As can be seen from Fig. 1b, the concentration n_{Pu} starts to decrease from this moment (in Fig. 1a, a decrease in n_{Pu} occurs prior to the intersection because of a considerable concentration of the intermediate product n_9). As a result, the critical conditions are not reached in such a mobile "reactor," and the wave moves only due to the neutron flux from the left boundary. As the distance from the last diffusion increases, it becomes more difficult to maintain the wave motion by diffusion and the distribution "freezes." If the external neutron source is switched off, the wave is stopped earlier. Thus, for $n_{\mathrm{cr}} > 0.064$, it is not possible to form a self-sustained critical region.

Naturally, the presence of an active isotope in original uranium favors wave formation [2]. Figure 2 shows

the situation for $n_{cr} = 0.08$ and $n_{Pu}(x, 0) = 0.075$. A wave with velocity $D = 0.19$ was formed at the time $t = 300$. These conditions are nearly critical: at $n_{Pu}(x, 0) = 0.07$, the wave is not formed. It can be seen that for close values of n_{cr} and $n_{Pu}(x, 0)$, the wave is much longer. The concentrations n and n_9 are extremely low (amplitudes of the order of line thickness). The ^{238}U burning is less intense in this case than in the regime shown in Fig. 1 although it is still considerable (approximately 20%). As in Fig. 1, the plutonium concentration behind the wave is somewhat higher than the initial concentration.

At first sight, a wave regime can be attained even for $n_{cr} \to \tilde{n}$ by increasing the initial concentration $n_{Pu}(x, 0)$. Actually, however, values $n_{cr} < \gamma$ exist for which this cannot be done. For example, we were unable to find a proper value of $n_{Pu}(x, 0)$ for $n_{cr} = 0.09$. Neutrons from the external source first burn up plutonium on the boundary of the medium and convert uranium to ^{239}Np. With time, ^{239}Np begins to produce plutonium but it has no time to produce a sufficiently high concentration and the Pu production then decreases because of uranium burnup. An increasingly thicker layer containing no plutonium nor uranium is formed near the boundary (as can be seen in Fig. 2). Neutron diffusion through this layer cannot provide for the required plutonium concentration, and a wave does not form even for $n_{Pu}(x, 0) = n_{cr}$.

Thus, the initial enrichment with a reactive component extends the domain of existence of wave regimes. As the limit is approached, the wavelength becomes greater and the scales of all changes decrease. We note that the ratio of Pu/U increases considerably behind the wave. However, this does not mean an excess of criticality: uranium is replaced by fission fragments, which are treated as equivalent absorbents in the adopted approximation. Therefore, the material behind the wave is in a subcritical state. Considering the real properties of nuclides, the state of the material is even farther from criticality than the initial state. Moreover, the material behind the wave is incapable of maintaining the wave regime because of the worsening of the ratio of n_{cr} to $\tilde{n}$: the fission fragments cannot produce plutonium. However, if the fragments will be effectively removed (possible withdrawal mechanism is discussed below), the activity of the material behind the wave can exceed the initial level.

DISCUSSION

Initial enrichment of uranium provides an additional freedom in choosing uranium burning regimes in fast breeders reactors according to Feoktistov's concept. In particular, the "activation" effect in the approximation considered is sufficient for using the oxide fuel, whose critical concentration (6.4%), even without dilution with a coolant, almost coincides with the upper critical concentration for the wave regime. Presumably, this qualitative estimate remains true in the case of ^{235}U enrichment (taking into account the lower activity and other counter factors). More definite claims require detailed multigroup calculations using real nuclear constants.

Let us now consider the geophysical aspects. The UO$_2$/Fe layer can be $\approx$1 m thick (assuming the uranium abundance in the Earth of the order of $(2$–$3) \cdot 10^{-8}$ g/g [10], which provides for a satisfactory balance between the heat production from radioactive decay and the present value of the Earth's heat flow. Larger concentrations are also possible if we omit the requirement that the heat flow is steady-state, taking into account the immense heat inertia of the Earth. In the past, the uranium content and thickness of the layer could have been considerably larger (about twofold 4 billion years ago). Thus, because of the uncertainty of available data, a layer thickness can be of at least several meters whereas a thickness of even a few dozens centimeters makes the layer critical.

Kuroda [11] suggested the feasibility of chain nuclear reactions in rich uranium deposits in the remote past, indicating that the critical state was presumably reached approximately 2 billion years ago in the presence of water as a moderator. Later, a natural nuclear reactor, Oklo, was found that operated at exactly this time (see [12]). As regards the "deposit" inside the Earth's core, the conditions here seem even more favorable, the difference being that the reaction in this case involves fast neutrons.

The ^{235}U concentration dropped to the critical level (at infinite dimensions) of $\approx$10% approximately 3 billion years ago. After that, accumulation of a layer of considerable thickness became possible. In the further process, the concentration further decreased by $\simeq$2%, during a comparatively short period, which gave way to a neutron combustion wave. The characteristic wave velocity is of the order of L/τ, that is, a few centimeters per day. The combustion time of the layer on the surface of the inner Earth's core is of the order of million years. However, in the vicinity of the limit, the wave velocity is small and the combustion time will amount to about 10^7 years. This is a short period from a geological standpoint, and passage of the wave with a substantial burnup would have caused a geological catastrophe. At the same time, the regime considered is considerably slower than the explosive processes studied in [5, 6]. It can be compared to a slow reactor-type burning of ^{235}U [6] with the only difference that a considerable amount

of ^{238}U can also be in principle transformed in the wave.

Because the nuclear energy release is great, considerable heating of the medium occurs at an even very small reaction depth. In this case, as in conventional nuclear reactors, the heat removal limitations are significant. The problem is simplified to some extent by the low wave velocity. For example, in the situation shown in Fig. 2, the heat release density averaged over the supercritical region is 5 MW/liter. Approach to the propagation limit can decrease the power to the value of 1 MW/liter, typical of the active zone in the fast reactor technology [13]. One might expect that a natural "self-propelled" reactor is also capable of operating at this power level because the heat in it is also removed by liquid metal. Naturally, near the limit the wave will be sensitive to various fluctuations (in composition, layer thickness, etc.), which can lead to failure of the wave regime.

Far from the limit, convection flows inevitably arise, which disturb the geometry of the mixture layer and the fuel will be scattered around. However, this does not necessarily imply that the wave stopped propagating; a decrease in its average velocity is more likely. Apparently, the following scenario is possible. The reaction starts at a site where the critical conditions for ^{235}U have been reached. This natural reactor can operate at low power due to several stabilizing factors (thermal expansion, sedimentation of fresh fuel portions) or it can produce short-term flashes. In a few days, increased plutonium concentrations are produced in the vicinity of the site, and the reaction propagates laterally. Convection removes the reacting material, and the reaction stops in this place. After mixing with the ambient material and subsequent cooling, a mixture of uranium and plutonium oxides settles again on the layer boundary but mostly in the unreacted places. An increase in the layer thickness due to the addition of reactive material brings the conditions closer to criticality. This peculiar diffusion makes a flash possible at a new site. The characteristic period of such pulses is difficult to estimate; it is clear, however, that this process is definitely slower than combustion in a neutron-fission wave and this provides for sufficient average cooling. We note that the α-decay of plutonium (lifetime 34,600 years) will not be a limiting factor because the decay product is ^{235}U.

In the convection regime, one should expect self-separation of the active layer. Fission fragments forming lighter compounds float up while heavy actinoids go down [4]. To a lesser extent, this process can be take place in a "normal" wave. As noted before, in the wave, the plutonium concentration increases (see Figs. 1 and 2; in fact, this is the breeding effect). Taking into account this slag-disposal process, multiple propagation of combustion waves with rather small uranium burnup in each of them is more probable than a single disastrous event.

The role of thorium, which is $\simeq 4$ times more abundant than uranium, has not been mentioned so far. It can be expected that thorium compounds will also be involved in the formation of the dense layer, increasing significantly its thickness. If the materials are mixed, combustion of uranium alone is hampered by a reduction in effective enrichment (thorium does not have a natural fissile isotope). However, ideal mixing is hardly expected: macroscopic inhomogeneities, including regions with the predominance of uranium. Ignition of such active spots can involve thorium in neutron fission reaction, in which it is converted to ^{233}U (with a characteristic time of 39.5 days). The wave velocity limited by this time is an order of magnitude lower than that in uranium combustion (however, this ratio is hardly true for the pulsed regime). The ^{233}U lifetime is $2.3 \cdot 10^5$ years; i.e., the characteristic time range in which the process considered is potentially possible is also quite wide.

At present, the feasibility of the wave regime is doubtful (unless the isotopic composition of the active layer has been dramatically changed due to the breeding effect). As was mentioned above, even in the pure uranium oxide, the critical concentration coincides with the maximum permissible concentration in the approximation considered. The current fraction of ^{235}U is small and hardly facilitates the propagation conditions. Therefore, the fuel cannot be ignited even by a specially designed deep-buried reactor. The time interval in which fission waves could have been propagated can be estimated as 2–3 billion years ago. Selection of "candidates" among the geological events is beyond the scope of this paper. We note, however, that the periods of geological activity can be related to the cyclic nuclear energy release in the processes considered above.

The present calculations are based on the approximate model of [1] supplemented by equal treatment of ^{235}U and ^{239}Pu. Quantitative estimation of the effect of approximations is problematic because the wave regime is close to the propagation limit for the supposed composition of the active layer. More complete calculations taking into account all main isotopes and the neutron spectrum will be of great interest.

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