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Ya B Zeldovich and nuclear power

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Abstract. The idea on a homogeneous nuclear reactor, first suggested by Ya B Zeldovich and Yu B Khariton in 1939, has since had its ups and downs and is now re-emerging, enriched with the knowledge and experience accumulated over the years having past. One of the current versions of the idea, the fast molten-salt reactor with a U–Pu fuel cycle, is presented in this paper.

The news of the discovery of uranium fission reached Leningrad in March 1939, and shortly after the studies were reported by Frisch, Peierls, Fermi and Szillard, Joliot-Curie and collaborators, and some others, from which it followed, in particular, that 2–3 neutrons were released when a nucleus of uranium underwent fission, i.e., in principle, a sustained chain reaction in uranium could take place, involving the release of an enormous amount of energy. The question of chain *chemical* reactions was well known to Ya B Zeldovich and, especially, to Yu B Khariton as N N Semenov's research fellow, and they immediately applied their knowledge to study the possibility of realizing a chain *nuclear* reaction in uranium. By the end of their investigations, a detailed review was published by N Bohr and J Wheeler on the fission of uranium nuclei, but instead of the actual mechanism of fission, Zeldovich and Khariton were interested in the kinetics of this process. In their first short article, "On the chain decay of the main isotope of uranium" [1] (received by the editorial board of *JETP* on 7 October 1939), they dealt with the possibility of realizing a chain reaction in an infinite medium of uranium, taking into account the moderation and absorption of neutrons. At that time, Bohr's hypothesis that practically all the observed fission fragments belonged to the rare isotope ^{235}U, instead of the 'main isotope' ^{238}U, had not been yet experimentally confirmed, so Zeldovich and Khariton considered the fission of ^{235}U only to introduce "quite a small additional number of neutrons." No definite conclusions were made in article [1], since at that time no reliable data either on the number of secondary fission neutrons (ν) or on their initial energy existed yet, and, besides, the cross sections of fission (σ_f) and neutron absorption (σ_c) in uranium (especially in the resonance region) were not known well, either (Fig. 1). However, the research strategy has already been clearly outlined in the article, and even the criteria were formulated in order to achieve experimental proof of the chain reaction being possible in uranium. (The conclusion that a chain reaction was not possible in natural uranium was made by the authors in the comment added during proof reading of the manuscript.)

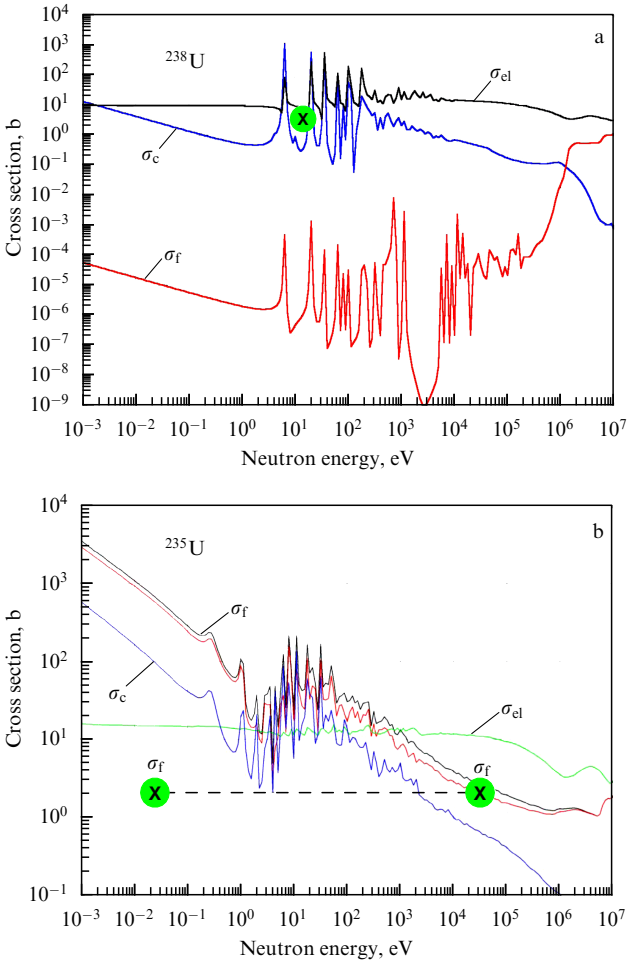

Figure 1. Cross sections of nuclear fission (σ_f), neutron absorption (σ_c), and neutron elastic scattering (σ_{el}) in isotopes (a) ^{238}U, and (b) ^{235}U. Circles indicate the cross section values used in Refs [1–3].

Already two weeks later (on 22 October 1939), a second article entitled "On the chain decay of uranium under the action of slow neutrons" [2] was submitted for publication. In this article, the difference between fission under the action of fast and slow neutrons was underlined for the first time, and resonance neutron absorption in uranium in the presence of a moderator (hydrogen), as well as the conditions in which a chain reaction is possible in ^{235}U, were dealt with methodically. The notations ν, υ, φ, well known to every student

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specializing in nuclear physics, were also first introduced in the same article, as was the condition necessary for a chain reaction to originate: $\nu\nu\varphi = 1$. In spite of the scarcity of experimental data, the value of $\varphi = 0.844$ (the probability of a neutron avoiding resonance capture in ^{238}U) calculated in the article is in quite good agreement with the results of significantly later calculations. In article [2], a confident conclusion was made concerning the impossibility of triggering a chain reaction in a homogeneous mixture of water and uranyl nitrate; however, a comment was made that if ordinary water (H_2O) were replaced by heavy water (D_2O), or if natural uranium were replaced by uranium enriched with the isotope ^{235}U (up to approximately 1–2%), a chain reaction would become possible. As we now know, both of these possibilities were subsequently realized.

Finally, in April 1940, a detailed review, "The fission and chain decay of uranium", written by Ya B Zeldovich and Yu B Khariton, was published in *Uspekhi Fizicheskikh Nauk* [3]. By then, quite a significant number of experimental and theoretical studies devoted to the physics of the fission of uranium had been carried out (the bibliography of the review contained 85 titles), so with account of the new results, the conclusions reached in their articles published in *JETP* [1, 2] were corrected, although the main assertions remained intact:

- no chain reaction is possible in a homogeneous mixture of natural uranium and water;
- to trigger a chain reaction, enrichment of natural uranium with the ^{235}U isotope is necessary, or ordinary water must be replaced by heavy water.

In May 1941, Ya B Zeldovich and Yu B Khariton were joined by I I Gurevich, and together they carried out a study, "The critical dimensions and mass, required for chain fission of nuclei induced by neutrons", which was immediately classified (it was to be published only half a century later [4]). In these calculations, the values selected for ^{235}U were $\sigma_c = 0$, $\sigma_f = 3 \times 10^{-24}$ cm^2, the value of ν was made to vary between 2 and 4, while the elastic scattering cross section was varied within the limits of $\sigma_{el} = (3-6) \times 10^{-24}$ cm^{-2}. The values obtained under these assumptions by Zeldovich, Khariton, and Gurevich for the critical mass of ^{235}U in a 'fast neutron reactor' (in other words, an atomic bomb) were reasonable: between 0.9 kg and 16.6 kg. The same article dealt with the conditions for reaching criticality in a thermal neutron reactor with water as a moderator, and it was shown that, in the case of uranium enriched by the ^{235}U isotope up to approximately 4–5%, criticality could be reached in such a homogeneous reactor with a ratio between uranium and hydrogen concentrations of about 0.1–0.2. (Notice that in those times the idea of a heterogeneous reactor had not yet become conventional, and only the homogeneous liquid mixture of a moderator with uranium salt dissolved in it was considered.)

As is known, a chain fission reaction in natural uranium was first realized by Fermi in a heterogeneous reactor, which represented a lattice made up of uranium blocks immersed in a thick of graphite. Several such uranium–graphite reactors (RBMK in *Russ. abbr.*) are still in operation at nuclear power plants (the uranium utilized is enriched with the ^{235}U isotope up to $\approx 2\%$). But nuclear power is mostly ($\approx 80\%$) based on solid-fuel water–water power reactors (VVER in *Russ. abbr.*), pressurized water reactors (PWRs), and boiling water reactors (BWRs), in which water serves simultaneously both as a moderator and as a coolant, while uranium tablets enriched with the ^{235}U isotope up to approximately 3–5% are used as the fuel. At present, about 440 nuclear reactors with a total power of ≈ 370 GW produce $\approx 14\%$ of the electric energy in the world, and, moreover, about 500 naval reactors are in operation on board ships and submarines.

Given the success achieved in nuclear engineering, the idea of a homogeneous reactor was left behind, even though the first such 5 kW reactor was constructed in Los Alamos back in 1944, and Fermi very actively took part in creating it. (The reactor had the shape of a sphere 30 cm in diameter, filled with a ^{235}U salt solution in water.) Emilio Segre testified that even during his hard work on the atomic bomb in Los Alamos, Fermi often discussed the idea of a homogeneous reactor (he called it 'boiling') [5, 6]. Fermi's pupil and a participant in the commissioning of the first reactor, Alvin Weinberg, achieved this dream in 1965: he launched the molten-salt reactor experiment (MSRE) in Oak Ridge (USA), in which the fuel consisted of a melt of the $2LiF-BeF_2$ salt and of uranium and thorium fluorides (UF_4 and ThF_4), respectively, dissolved in it, while graphite served as a moderator [7]. The thermal neutron spectrum in this reactor was oriented toward the Th – U-cycle and most of the time it was operated precisely in this cycle. The MSRE reactor was in operation for about five years without any trouble, but was shut down owing to a project of a fast neutron reactor. At present, a sole 20 kW molten-salt reactor, ARGUS, is in operation in the world; its fuel is a water solution of uranyl sulfate (UO_2SO_4) with a 90% ^{235}U enrichment (the NRC 'Kurchatov Institute').

The release of the energy of nuclear fission is the most important achievement of humankind since the taming of fire. But already Fermi understood that nuclear power (NP) based on thermal reactors consuming the rare ^{235}U isotope had no stable future without the resolution of four key problems, which did not usually even come to anyone's mind in the military race frenzy:

- providing supplies;
- safety;
- nonproliferation of fissile materials;
- closing the nuclear fuel cycle.

The first constitutes a key problem: the world supplies of commercial ^{235}U (of which there is only 0.72% in natural uranium) are estimated to amount to approximately 50 thousand tons. The present-day consumption amounts to about 600 tons per year and by the middle of this century it will increase to approximately 1000 tons per year [8], i.e., the resource of modern NP based on thermal neutrons does not exceed the time interval of 50–100 years. It is well known, however, that the problem of supplies for NP can be resolved by moving from thermal reactors, consuming ^{235}U, to fast reactors that consume ^{238}U. There are also serious arguments indicating that at the same time the problem of dealing with long-lived radioactive NP waste can also be resolved if a fast reactor with inherent safety is created [9].

Such a reactor must fulfil, in addition to other requirements, the following [10]:

- it must provide a minimum excess of reactivity;
- it must exhibit negative temperature and void reactivity coefficients;
- no fire-hazardous or chemically active coolant must be present;
- there must be no pressure in the first circuit.

In a fast reactor, fission takes place not only of ^{235}U nuclei, but of ^{238}U as well, and also of ^{239}Pu, which is produced in the neutron capture reaction $n + {}^{238}U \rightarrow {}^{239}Pu + e + \tilde{\nu}$. When ^{239}Pu undergoes fission, $\nu = 2.84$ secondary neutrons

are produced instead of $\nu = 2.42$, as in the ^{235}U nucleus fission. Moreover, in the case of ^{239}Pu nuclear-fuel burning in a fast reactor, the ratio $\alpha = \sigma_c/\sigma_f \approx 0.1-0.2$ (instead of $\alpha \approx 0.4$, as in a thermal reactor), resulting in an additional generation of neutrons which can be used, in particular, for extended reproduction of ^{239}Pu. However, the idea of utilizing the produced ^{239}Pu in thermal reactors turned out to be flawed, since in this case nearly 30% of the ^{239}Pu turns into the ^{240}Pu and ^{241}Pu isotopes, while the last of these turns into the transuranium elements Am and Cm, i.e., the most dangerous radioactive waste (in fast reactors, this part is reduced to $\approx 10\%$).

It also turned out that an equilibrium mode of nuclear-fuel burning is possible in a fast reactor with a concentration of $\approx 10\%$ of ^{239}Pu (or $\approx 13\%$ of ^{235}U) in a mixture of U and Pu: it just suffices to feed the reactor with ^{238}U, since this isotope effectively reproduces the burned up ^{239}Pu when a neutron is captured. Such an industrial 350 MW reactor was first constructed in the USSR in 1972, and was successfully in operation for 25 years. At present, in Russia, a single industrial 600 MW fast reactor in the world, BN-600, is in operation, and its coolant is fire-hazardous sodium; a fast reactor, with a lead coolant is under development [11]. Two of the key problems can be resolved in these reactors at the same time: that of supplies (both ^{239}Pu and ^{238}U are burnt instead of ^{235}U), and that of getting rid of the transuranium elements. However, for achieving closure of the nuclear fuel cycle (i.e., the reuse of spent fuel), it is necessary to resolve the quite difficult engineering and technological problem of producing fresh solid-fuel elements from the highly active spent nuclear fuel (SNF) discharged from the reactor.

This is only one of the reasons to revive the idea on a homogeneous reactor, in which a melt of fluoride salt with uranium and plutonium salts dissolved in it serves as the nuclear fuel. Such a reactor does not require solid-fuel elements to be manufactured, but for closure of the nuclear fuel cycle it is necessary that the neutron spectrum of the molten salt reactor (MSR) be fast. This, in turn, is only possible if the concentration of uranium and plutonium atoms in the salt melt is not inferior to 10 at.% (about 50% in mass). No such salt, however, had been known until recently (the solubility of PuF_3 in a melt of the $2LiF-BeF_2$ salt does not exceed ≈ 1 mol.%), and only during the past two years was it established that at a temperature of 700 °C (the MSR operating temperature) approximately 45 mol.% of UF_4 and ≈ 30 mol.% of PuF_3 dissolve in the LiF-NaF-KF (FLiNaK) eutectic (Fig. 2) [12–15]. The neutron spectrum of such a molten salt reactor is quite close to the spectrum of a fast solid-fuel reactor (Fig. 3) [16, 17].

MSR satisfies all the aforementioned requirements for reactors exhibiting 'inherent safety' and, first of all, the key requirement: the occurrence of negative temperature and void effects, which excludes severe accidents, such as the Chernobyl one. The fuel cycle of a molten-salt fast reactor (MSFR) is essentially simplified, since it requires no manufacturing of fuel elements from highly active SNF, and the actual processing of the nuclear fuel becomes possible at the very nuclear power plant, which, in turn, reduces the risk of unauthorized proliferation of fissile materials.

Moreover, the utilization of an MSR is at present considered one of the most promising ways of getting rid of long-lived radioactive waste (mainly Am) accumulated during the past 60 years of NP existence. Already now ≈ 240 thousand tons of SNF has been accumulated in the

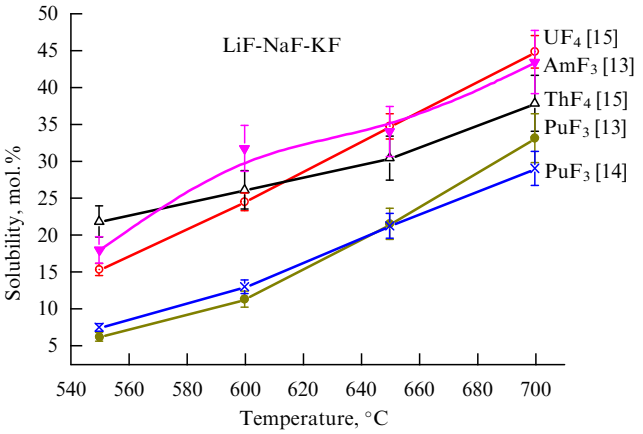

Figure 2. Solubility of actinide fluorides in FLiNaK.

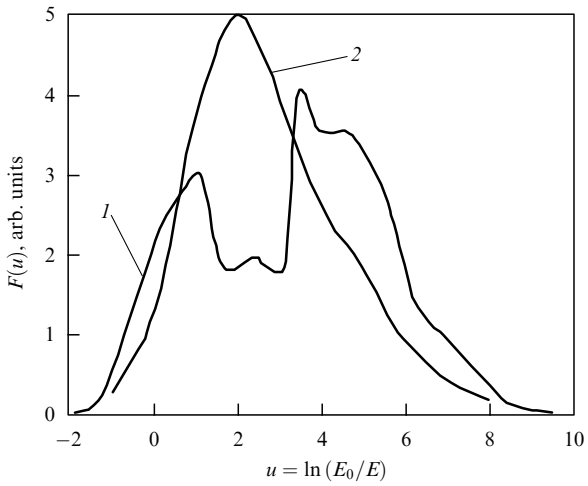

Figure 3. Neutron spectra $F(u)$ in MSFR (curve *1*) and in a fast reactor (curve *2*) as functions of lethargy u ($E_0 = 2$ MeV) [16, 17].

world, and it contains about 300 t of ^{241}Am, and every year this mass increases by approximately 10 t owing to the ^{241}Pu $\rightarrow$ ^{241}Am decay [17]. To eliminate this waste, MSRs are most appropriate: during one year of operation, one MSR of thermal power 1 GW controlled by an accelerator is capable of 'burning up' about 300 kg of Am, i.e., the Am annual production from about 90 thermal reactors of the same power [18].

At present, FLiNaK is the only known salt exhibiting such a high solubility of actinide fluorides. It cannot be ruled out that with time other salts with the same property will be found, and, possibly, they will be more suitable from an economic point of view and more convenient for processing, exhibiting lower corrosivity and so on; but the main idea of the MSFR here remains intact: *a fast reactor with liquid fuel and a* U–Pu *fuel cycle*. Numerous scientific, technological, and engineering problems will have to be resolved along the path toward realization of this idea, but the ultimate goal is quite worthwhile.

Over 70 years have passed since publication of the first articles by Ya B Zeldovich and Yu B Khariton; during this period, numerous processes taking place in a nuclear reactor have been thoroughly studied, and now any student knows more about them than all the authors of articles published in those times (see Fig. 1). Nevertheless, the fundamental problems of nuclear power engineering have still not been

resolved: no safe and economically acceptable fast nuclear reactor has been constructed, its closed fuel cycle has not been developed, and the problem of dealing with radioactive waste has not been resolved. One of the reasons for this is the lack of scientists of the superior intellects similar to Ya B Zeldovich and E Fermi in the enormous army of engineers and technologists engaged in the nuclear industry. Having created nuclear weapons, they forever left the field of knowledge associated with nuclear power, within which technological and engineering problems gradually started to prevail over scientific ones. Here, the understanding was lost that the creation of sustained nuclear power represents first of all a *scientific* problem, over and over again exceeding in complexity (and significance) the problem of developing nuclear weapon. And, if in the near future the tendency that has taken shape does not change, we risk irretrievably losing the possibility of taking advantage of nuclear power — the sole long-term (for several hundred thousand years) source of energy, without which our civilization will turn out to be nonviable [19].

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