LIQUID FUEL CONCEPT BENEFITS

M. HRON
Nuclear Research Institute,
Řež, Czech Republic

Abstract

There are principle drawbacks of any kind of solid nuclear fuel listed and analyzed in the first part of the paper. One of the primary results of the analyses performed shows that the solid fuel concept, which was to certain degree advantageous in the first periods of a nuclear reactor development and operation, has guided this branch of a utilization of atomic nucleus energy to a death end (not having been able to solve principle problems of the corresponding fuel cycle in an acceptable, for public namely, way). On the background of this, the liquid fuel concept and its benefits are introduced and briefly described in the first part of the paper, too.

As one of the first realistic attempts to utilize the advantages of liquid fuel, the reactor/blanket system with molten fluoride salts in the role of fuel and coolant simultaneously, as incorporated in the accelerator-driven transmutation technology (ADTT) being proposed and currently having been under development in the Los Alamos National Laboratory [1], will be studied both theoretically and experimentally. There is a preliminary design concept of an experimental assembly LA-0 briefly introduced in the paper which is under preparation in the Czech Republic for such a project.

Finally, there will be another very promising concept [4,5] of a small low power ADTT system introduced which is characterized by a high level of safety and economical efficiency. This subcritical system with liquid fuel driven by a linear electron accelerator represents an additional element -nuclear incinerator- to the nuclear power complex (based upon thermal and fast critical power reactors) making the whole complex acceptable and simultaneously giving an alternative also very highly acceptable nuclear source of energy and even other products (e.g. radionuclides, etc.). In the conclusion, the overall survey of principal benefits which may be expected by introducing liquid nuclear fuel in nuclear power and research reactor systems is given and critically analyzed. The other comparably important principles (e.g. the general subcriticality of reactor systems principle) are mentioned which being applied in the nearest future may form a basis for an absolutely new nuclear reactor concept and a new nuclear power era at all.

1. INTRODUCTION

Since the discovery of the reaction of atomic nucleus fission, the main goal of all efforts was to utilize it for an energy generation. As one of the most important conditions for an efficient achievement of this goal self-sustaining of fission chain reaction was demanded in an assembly containing fissionable nuclei of nuclear fuel without an external source of neutrons. If this was reached, the assembly was defined as being critical. Let us note that it was by definition (theoretically) critical on prompt neutrons released from fission reactions only. Very early, it was observed experimentally that the assembly reaching criticality is in fact very slightly subcritical on prompt neutrons and that there is a not very strong natural source of delayed neutrons originated from radioactive decay of some of the fission products always added (which, fortunately, allowed easier control of the system).

At the early stages, reaching criticality was one of the most difficult tasks and all the effort and ideas had been devoted to this aim. The reason was that there were only small amounts of fissionable materials available in those times in the form of the low (0.7%) content of U235 in natural uranium. Therefore, solid phase metallic uranium with highest as possible density was used and in the form of blocks with a specific definite size arranged in a heterogeneous lattice in a solid (graphite) or liquid (heavy water) moderator with a certain pitch determined by optimal neutronic conditions. This arrangement has remained nearly exclusive even in latter systems with fuels enriched by U235 content up to much higher levels. The reasons had been of different nature, however, the designs have mostly started from what became already an approved conventional principle - solid fuel blocks in a heterogeneous lattice - which has been kept even in the case of pure or high enriched fuel in a fast neutron system without moderator.

One of the next consequences of the adoption of the solid fuel concept has been a type of control system which has been mostly applied for a short term control of nuclear reactors - the concept of solid absorbers - and what is more the concept of a negative neutron source at all. This and a number of other consequences can be traced to start all from the initial tension in neutron economy when the principle of a self-sustaining fission chain reaction and consequently the concept of a critical reactor are adopted. They all begin to form a magic circle of convention in which the short term and finally even long term operational behaviour of nuclear, namely power, reactor is being imprisoned and limited in its ability to give a positive and broadly acceptable development. Let us explain this thesis in some following more see-through examples.

The adoption of the solid fuel concept leads to the principal necessity to keep the fuel blocks at a certain position in the reactor core for a shorter or longer period of time. This in-core residential time is especially long in power systems where at least a quasi-continuous exchange of fuel would be very complicated and expensive. Therefore, the following very inconvenient consequence arises: the whole time, the block of solid fuel remains at a certain position in the reactor core, there are fission fragments and by neutron capture induced radionuclides (let us call them altogether products) being accumulated in the volume of the fuel block. There are several secondary consequences caused by this fact which contribute to the above mentioned magic circle forming:

1) reactivity margin for a short term as well as long term negative influence of the increasingly accumulated products has to be applied which has to be compensated by another artificial negative source of neutrons. It has in principle a consequence in greater amount of fuel being present in the core than really necessary for a demanded power and then the more products including actinides is generated.

2) the original fuel is finally so heavily poisoned by the products that it cannot keep the self-sustained fission chain reaction more and a further operation of the reactor under original conditions is impossible. There is an unavoidable principle change in the operation and structure of the reactor which means an outage and exchange of at least a part of the fuel charge.

3) the most controversial problem what to do with spent solid fuel arises and a vicious circle has been closed or a solid fuel concept "trap" snapped.

The above briefly described solid fuel concept shows its most important and sensitive drawbacks:

1) continuous accumulation of products during the whole residential time of fuel blocks in the core,

2) following necessity to stop the operation, discharge spent fuel and store it for a necessary period of time (in order of magnitude of years until it reaches a desirably low level of radioactivity) in a specific storage,

3) the last and the most difficult drawback is the need of an optimal decision of the following destiny of spent fuel.

Up to now, the only two possible solutions were developed either to reprocess (chemically) it and to prepare next generation of solid fuel (it means with principally the same class of drawbacks) or to dispose it in a depository of a corresponding quality (which sometimes is called repository because a possible reuse of the disposed product is supposed). In the former case mostly chemical methods and processes are applied. In the latter, a lot of branches is involved, however, nearly all of them are of a classical (non-nuclear) nature. The only nuclear process which is employed is the natural radioactive decay.

This fact contains one very controversial principle or better say a violation of a basic principle which can be described as follows: The energy generation in nuclear reactors utilizes enforced nuclear process which are simultaneously producing products or nuclear waste (including secondary raw materials e.g. actinides). The treatment of the products needs to apply an adequate technology in an adequate scale. This principle has not been applied and fulfilled in those so far developed and designed systems for spent solid fuel management. There is an adequate technology which only utilizes nuclear processes and which can transfer the high level and long-lived radionuclides towards short-lived or even stable nuclides-transmutation technology performed in a suitable nuclear reactor device and combined with a continuous separation of certain components of its core or reprocessing of the reactor fuel so as to avoid the consequent induction of radioactivity by neutron irradiation of stable and short-lived nuclides. One of the principle concepts allowing to reach such a technology in an industrial scale is the concept of liquid nuclear fuel.

2. PRINCIPLES OF DIFFERENT TRANSMUTATION SYSTEMS

The first candidate for this purpose is a nowadays called classical nuclear reactor itself. Its ability, to certain degree, to burn fission products is natural and well known. The first limitation and even barrier we certainly approach in this attempt is the neutron economy of conventional reactor systems. The next attempt could be an actinide fuelled (critical) reactor. Then another problems arise. First of all, the problem of control of such a system. It is well known that the fissile isotopes of Neptunium, Americium and Curium have a considerably smaller fraction of delayed neutron emitters in comparison to the common fuels based on Uranium isotopes and the essential role which the fraction of delayed neutrons is playing in the control of nuclear reactor in critical state is not necessary to recall here. Beside this, there are other problems with a small Doppler effect and a positive sodium void coefficient arising in this case as well.

To overcome these problems, there have been various concepts of subcritical systems driven by a suitable neutron source proposed in the recent past, aiming at the transmutation of actinides and long lived fission products. The suitable neutron source, it means the source with a sufficient yield of neutrons allowing to avoid the principle barrier of neutron economy, appeared in the concept of spallation reaction induced by highly accelerated charged particles, e.g. protons:

1) The idea of a direct exploitation of the spallation process to transmute actinides and fission products had to be given up because of a barrier of necessary particle currents which are much larger than the most optimistic theoretical accelerator designs show around 300 Ma. It had been reported [1,2] that the destruction rate of the largest possible proton accelerator would correspond only to a fraction of the amount of fission products

generated by one thermal reactor of 1000 MW_e in the same period of time.

2) The idea of a direct use of spallation neutrons was tested. The fission products were supposed to be placed around a proton target to use only the spallation neutrons as they are generated in the target. In dependence on the characteristics of the material which is to be transmuted, either the fast neutrons would be used as they are emitted from the target or they would be slowed down by a moderator to energy levels with higher transmutation cross sections like the resonance or thermal energy ranges. The following simple estimate shows the substantial disadvantage of such a system. Assuming that it is possible to make all the spallation neutrons available for the transmutation process:

$$E_{fp}=q_{fp}\frac{P_b}{\eta_{sp}}\frac{1}{\eta_b\eta_T}\,[MW] \tag{1}$$

where E_{fp} is the amount of energy which is necessary to transmute the fraction
q_{fp} of radionuclides per fission process in a nuclear energy system,
P_b is proton energy,
η_{sp} is the number of neutrons generated by one proton,
η_b is the efficiency of converting electricity into proton beam energy (~0,5) and
η_T is the efficiency of converting thermal energy into electricity (~0.33).

For example, in the case of a 1,5 GeV proton beam emitting 50 neutrons per spallation in a lead target, the transmutation of ^{99}Tc, ^{129}I, ^{135}Cs, ^{90}Sr, ^{85}Kr and ^{93}Zr (constituting 28% of all fission products) would require 51,3 MeV to transmute the fission product fraction of one fission process. This is ~26% of the total power production of the energy system under consideration and the real percentage of energy required will be even higher due to the very optimistic assumptions made in this estimate. Together with the cost for reprocessing it would make this type of accelerator transmutation prohibitively expensive, at least in a commercial nuclear energy system.

3) The idea of a neutron source driven subcritical reactor-type assembly to improve neutron economy remained. Technically, this can be realized by surrounding a proton target region by fissionable material in a cooling system. For removing of the high specific heat released in the target, there is a liquid metal alloy mostly used. However, we should mention that the specific heat production per neutron is considerably lower than in a fission process (30 MeV against 80 MeV).

Let us quantify the power production P_{fi} of a subcritical assembly fed by spallation neutrons:

$$P_{fi}=\eta_{sp}\frac{a-k}{\mu(1-k)}\frac{i}{C}E_f \tag{2}$$

where k is the multiplication factor, a is the importance of the target position (usually $a>1$ for a central target position), μ is the mean number of neutrons in a fission process, E_f is the power release per fission (=3,2.10^{-11}Ws), η_{sp} is a neutron yield from one proton, i is proton current, C is proton charge (=1,6.10^{-19}As). The dependence of the power production of an accelerator driven facility upon its subcriticality $(1-k)$ assuming a proton beam of 1 GeV and 1 mA bombarding a Pb-Bi target releasing neutrons per spallation with an importance $a=1$ illustrates that near criticality an 1 mA current already generates a relatively high fission power and for $k=0,97$ more than 100 MW can be achieved.

In this case, the additional neutrons from the subcritical system as well as its fission power, which can be also transformed into electricity, can be exploited to run the transmutation process. The energy required to transmute a fraction q_{fp} of fission products in such a system can be expressed as follows:

$$E_{fp}=\frac{\eta_{sp}\frac{k}{\mu(1-k)}E_f-\frac{P_b}{\eta_b\eta_T}}{\eta_{sp}[(1-k/\mu)\eta_{fp}+\frac{k}{1-k}((1-k/\mu)\eta_{fp}-\frac{q_{fp}}{\mu})]} \tag{3}$$

where $\eta_{fp}=\frac{\Sigma_a(FP)}{\Sigma(FP+fuel+constr.mats)}$

The positive sign of E_{fp} means that there is even a surplus of energy, while the negative sign indicates a need of energy which should be added to the system from outside.

In spite of these very promising features, the fact that the amounts of long lived fission products generated by currently operated thermal reactors are relatively very high and corresponding huge amounts of neutrons are required to their transmutation results to a certain stress in the overall neutron economy of those designed systems and requires a relatively high power of the accelerator for spallation neutron source. One of the possible improvements in this situation is a higher degree of multiplication of spallation neutrons from the source by the subcritical reactor. In the deeply subcritical transmuter with $k_{eff}=0.7$, the proton beam power of 300 MW is required to transmute ~17% of LLFP, such as ^{99}Tc, ^{129}I, ^{85}Kr and ^{93}Zr, created by a 1 GW$_e$LWR. When the spallation source neutrons are multiplied by a transmuter which is closer to criticality (with e.g. $k_{eff}=0.99$) and when the ratio n_f of neutron capture by LLFP to the total is 20%, the proton power becomes 4 MW.

There has been a series of deep analyses and evaluations of various transmutation systems feasibility performed in the recent of few last years. In spite of the great variety of different concepts, most of them reached a consensus on the principal structure of a system being divided into two basic technological parts:

a) The source unit. (Mostly based upon neutrons yielded from either direct or spallation reactions induced by accelerated charged particles in nuclei of some heavy elements. There is , in general, an annulus of some high - Z material, such as lead, for further multiplication of the source through (n,xn) and (p,xn) reactions employed here or they may be multiplied by a fissile target core - so called active target.)

b) The subcritical reactor/blanket unit which contains a moderated heterogeneous lattice, using a flowing liquid which contains actinides. There may be separate regions in the blanket where long-lived fission products (LLFP) are transmuted. The structures of the heterogeneous core, which were competitive in the previous stages, were either a heavy water moderated aqueous slurry system or a graphite moderated molten salt system. The former has been connected with a demanding continuous chemical reprocessing which became its drawback. The latter can utilized a mechanical (e.g. centrifuge) separation of individual components of the fluid fuel and has also a higher thermal efficiency. Therefore, we will concentrate in our further analysis and a development of a design concept on such a type of systems.

3. LIQUID FUEL CONCEPT FOR NEUTRON SOURCE-DRIVEN TRANSMUTATION TECHNOLOGY (NSDTT)

3.1. The Los Alamos Molten Fluoride Salt Fuel for Accelerator-Driven Transmutation Technology (ADTT)

The LANL concept of an accelerator-driven subcritical blanket for a nuclear incineration of nuclear waste has been known for several recent years [1]. Let us recall at least very briefly the main features of the last developed version of this concept and let us show a part of a proposed research program of an approvement of its ability for an efficient realization in the industrial scale.

The fuel material is in the form of the fluoride salt AcF_4 dissolved in a molten salt carrier whose composition is a mixture of ^7LiF and $^9BeF_2$. The carrier's melting point and operating temperature are about 500°C and 650°C, respectively. The molten salt flows over the outside of a close-packed set of high-purity graphite blocks. One of the possible arrangements is graphite

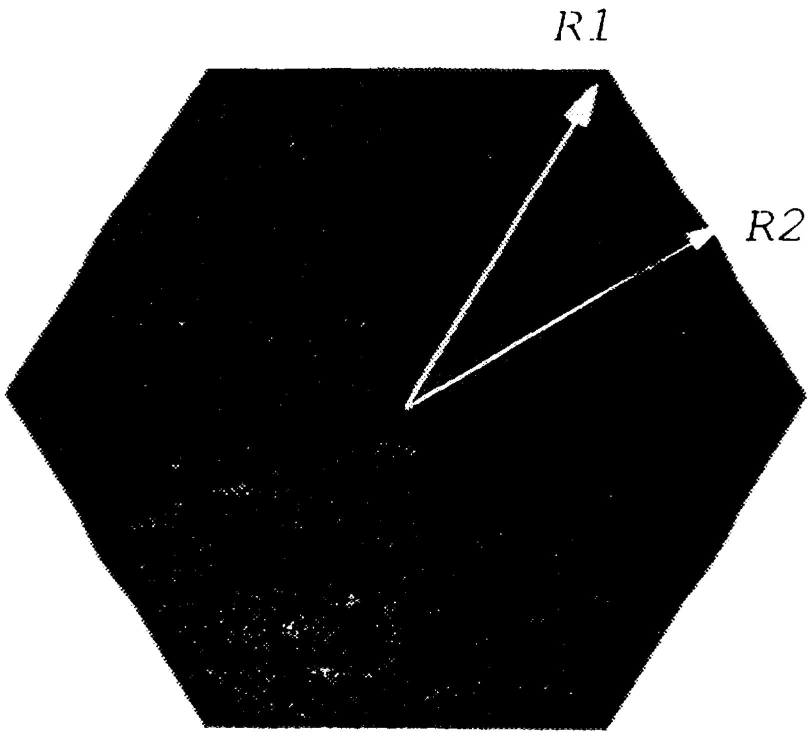

FIG. 1. Experimental Assembly LA-O Elementary Cell.

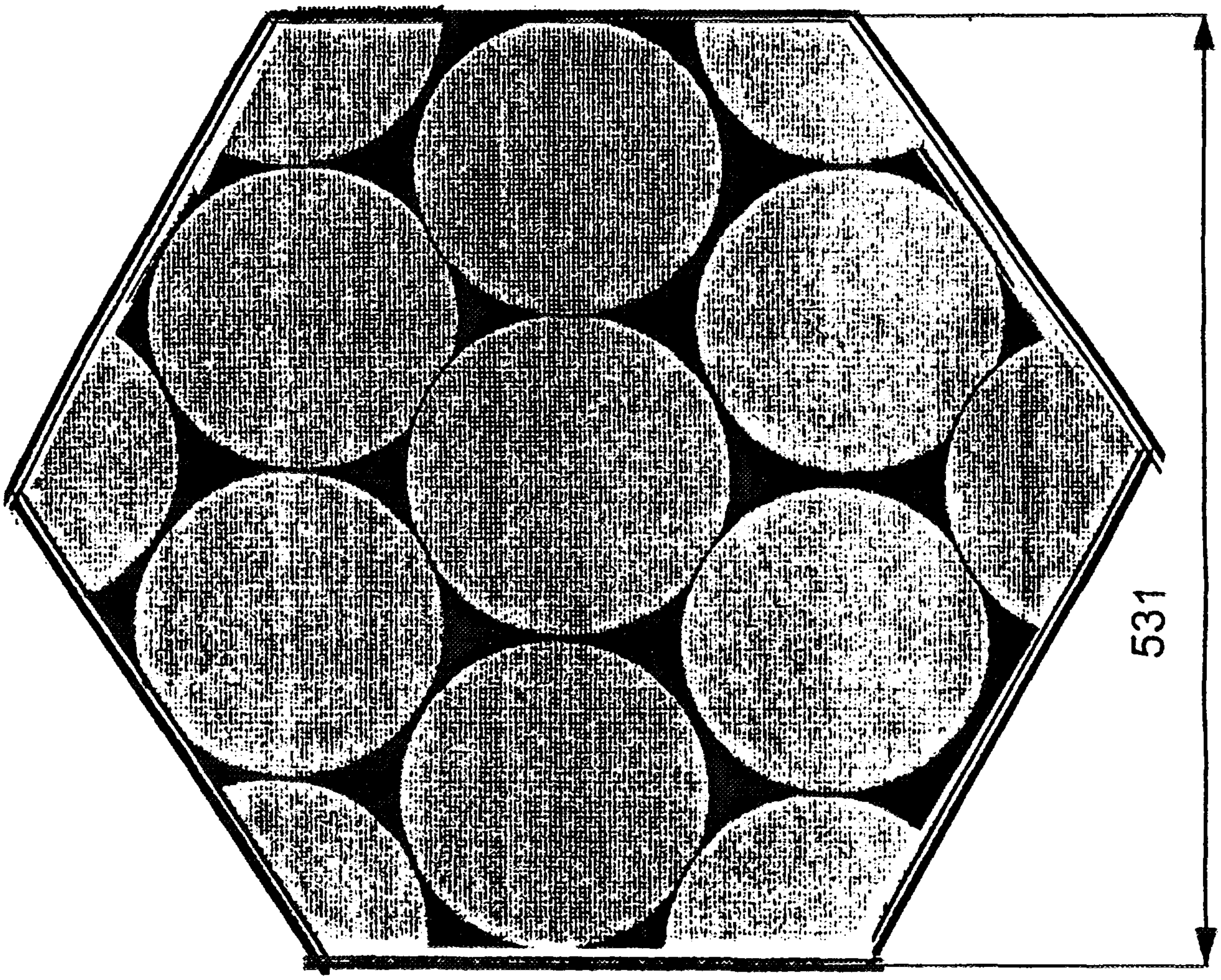

FIG. 2. Experimental Assembly LA-O Elementary Module.

blocks in the form of cylindrical rods arranged in a regular triangular lattice. There were simple elementary analyses performed which has lead to a preliminary accepted set of optimal values of the graphite rod radius R1=8.66 cm and the elementary cell hexagon outer circle radius R2=10.0 cm (Fig. 1). There has been an experimental research system called LA-0 preconceptually designed by the author in LANL [3] which should be developed and realized in the Nuclear Research Institute Řež plc in the Czech Republic. The final purpose of the system would be an experimental testing of a given type of transmuter reactor/blanket core neutronics and possibly also other physical and technological characteristics and properties including time behaviour. For the very first stage, the following scheme can be applied which will allow the first results to be reached very cheaply and relatively soon. There can be an elementary, however, a sufficiently representative sample of the investigated reactor blanket lattice (an elementary module - Fig.2) inserted into an existing experimental reactor core serving like a driver and the basic set of its characteristics can be experimentally measured and verified. The suitable experimental reactor can be e.g. the experimental reactors LR-0 (full-scale core modelling in Nuclear Research Institute Řež) or VR-1 (training reactor at Czech Technical University Praha) which have been successfully operated for core analyses of thermal reactors since 1982 and 1990, respectively.

3.2. Low power ADTT system

The molten salt reactors (MSRs) with the continuous control of nuclide composition almost do not require an initial reactivity margin. In such reactors, subcriticality may be reduced up to the minimum value ß where ß is the effective delayed neutron fraction. However, with such a small subcriticality and in view of available uncertainties in nuclear data and nuclide concentrations, the difference between subcritical and critical MSR in a great extent disappears: in both cases the nuclear safety is ensured by the large negative temperature reactivity effect. The deeper subcriticality is of course substantiated by the fact that under such conditions we exclude the necessity to control a reactor - burner in a dynamic mode, that is a bit difficult and poorly known.

In this case, the e.g. accelerator - driven positive source performs only one of the usual functions - the function of a reactor control system without inertia, an alternative to the usual reactor control organs, negative sources like e.g. absorbers or decreasing of the dimensions of the system, etc.

The high level parameter proton accelerator with all its disadvantages (like e.g. the length ~ 1 km, the investments ~ U$ 1 billion, etc.) having been applied in the Los Alamos

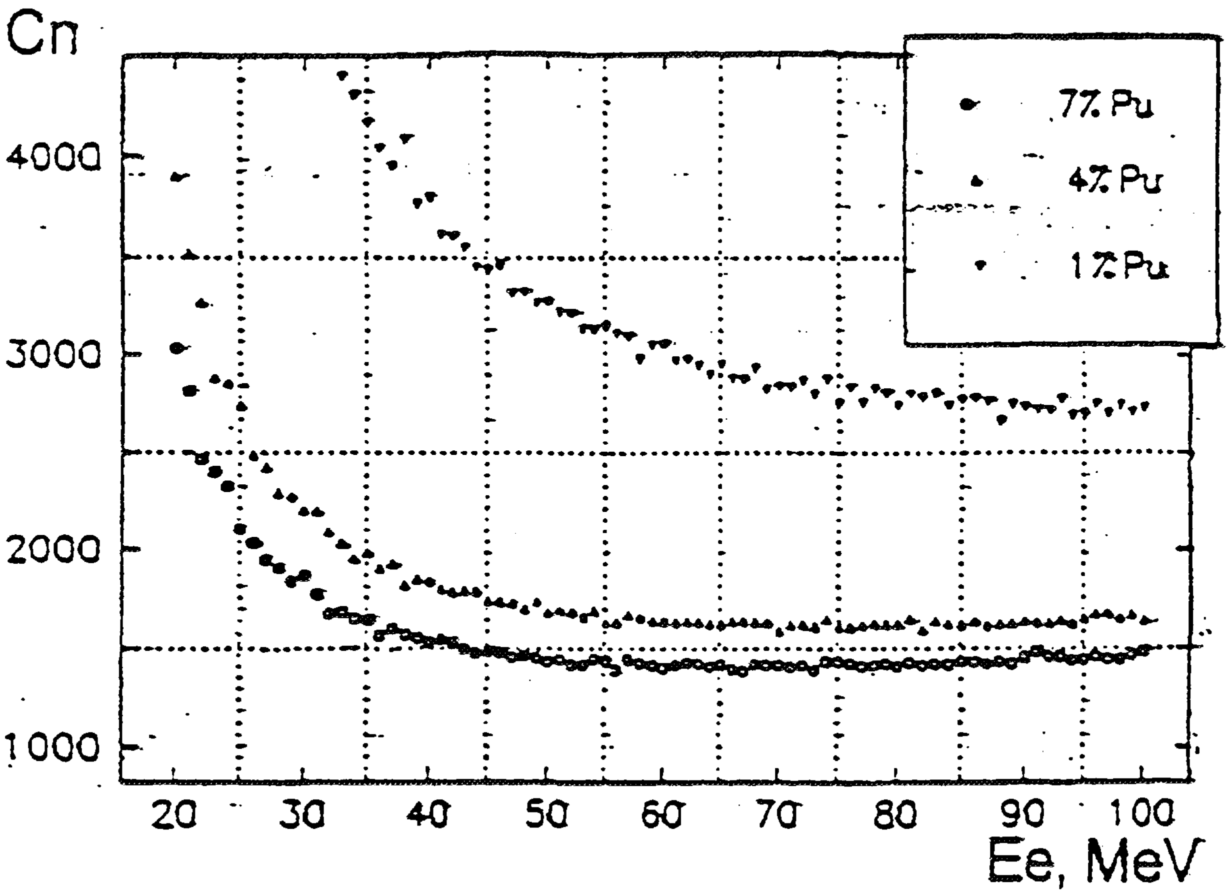

FIG. 3. Electron beam energy necessary for one photonuclear neutron generation.

concept is not necessary in the system and a low level parameter accelerator can be employed. There has been a very efficient concept proposed by Russian scientists [4,5] recently utilizing an electron accelerator. The electron beam is supposed to enter directly the molten salt environment in a homogeneous core where it generates photonuclear neutrons by e - n - τ scheme. The Fig. 3 shows a calculated photonuclear neutron energetic cost in molten salt with equal numbers of Li, Be and F nuclei for compositions with 1, 4 and 7 mol.% of actinides. The energetic cost of a photonuclear neutron makes Cn ≈ 1300 ÷ 1400 MeV that is 25 times higher than that of a electronuclear neutron generated by a proton. However, the decision on the acceptability of a neutron source may not be taken only on the basis of the Cn value.

There may be a scheme for a significant and simultaneously safe neutron multiplication device that would allow to apply electron accelerators effectively. One very convenient scheme of such a type has again been introduced in [4,5]. The main idea of this concept is a use of the principle of cascade neutron multiplication on subcritical cores known for a long time. The subcritical MSR is analogous to fast-thermal critical coupled

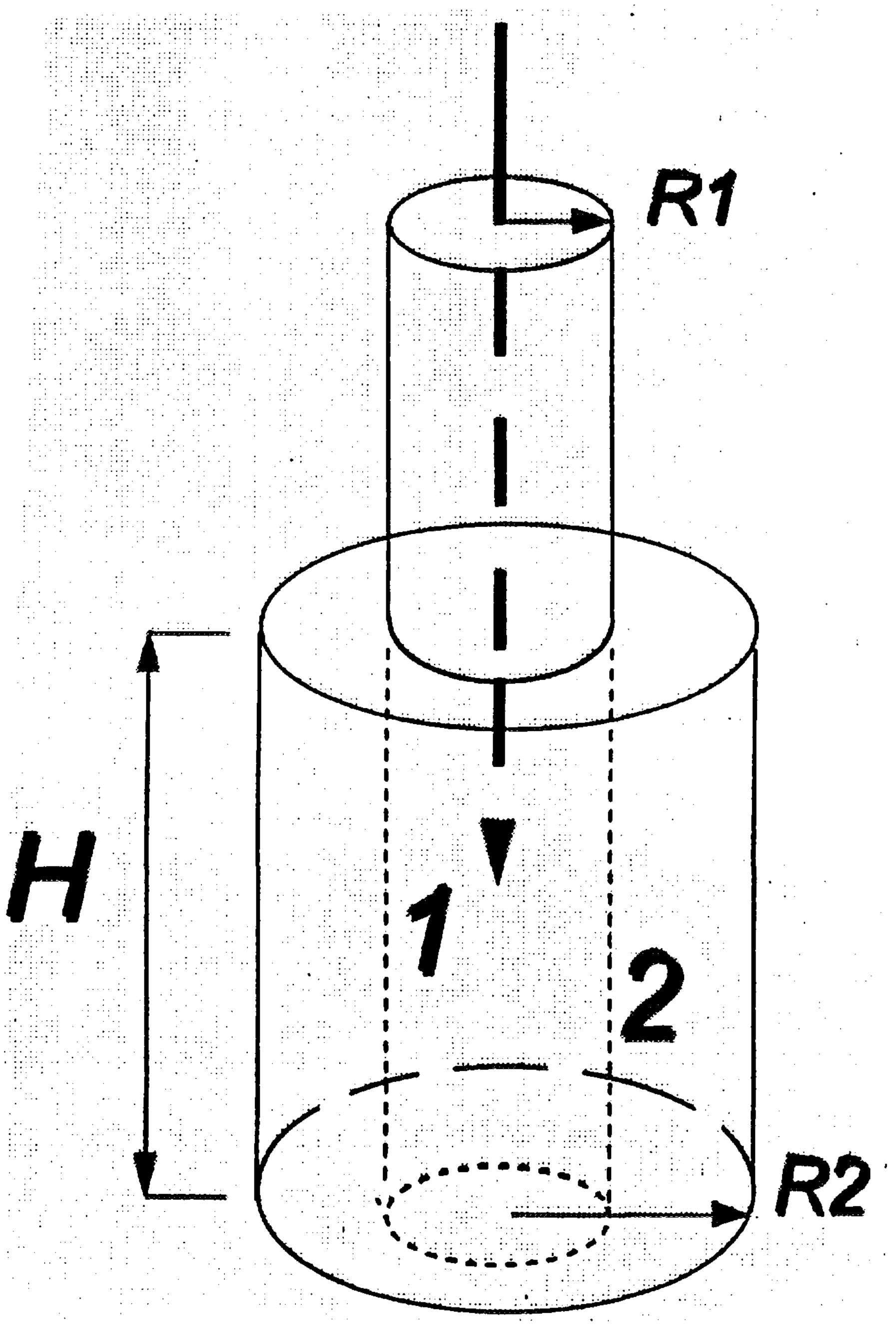

FIG. 4. Principal scheme of a cascade MSR.

reactors [6,7]. The theory of coupled reactors was developed and experimental results for such systems were described. Each reactor in a fast-thermal critical system is subcritical. The system cannot be critical only with fast neutrons. There was a good agreement between theory and experiment obtained. It shows that no unexpected and unknown phenomena occur in similar systems. There is a sufficient number of experimental results available which were performed on fast-thermal critical assemblies in USA (ZPR-III, ZPR-VI), Russia (SPECTR), France (ERMINE-2), Switzerland (PROTEUS) and Belarus (BTS).

The design introduced in [4,5] is a subcritical MSR of 1250 MW_t which consists of two cylindrical cores (Fig. 4). The first core is a fast system with a fuel salt composition of 53NaF + $41ZrF_4$+ + $6XF_4$ which is separated by a cylindrical wall from the second pool-type thermal core with fuel salt composition of 69LiF + $28BeF_2$ + $3XF_3$ and with an external graphite reflector compatible with fuel salt at high temperatures. The principle scheme of the system's function is as follows:

Fuel salt 1 enters the core 1 bottom through a pipe and due to heating elevates in a 12-m raiser with the 30-m diameter, in which the external source channel is located, and then enters the separate part of the intermediate heat exchanger 1HX.

Fuel salt 2 enters the core 2 top, comes down in a peripheral annulus, elevates in a 12-m raiser with a 1.2-m outer diameter and then enters 1HX.

There is a secondary coolant of the composition of $92NaBF_4$+ + 8NaF flowing through 1HX pipes. To avoid a fuel salt leak the reactor is placed into a 10-cm-thick double wall vessel. The gap between the inner and outer walls is filled with nitrogen. There is a gas volume above the fuel salt free surface. The pipelines are made of hastelloy-N - heat/proof material containing 15 - 18% Mo, 6 - 8% Cr and additionally alloyed with 1% of nitrogen to prevent interaction tellurium.

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