THE CZECH NATIONAL R&D PROGRAM OF NUCLEAR INCINERATION OF PWR SPENT FUEL IN A TRANSMUTER WITH LIQUID FUEL

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Abstract

The principle drawbacks of any kind of solid nuclear fuel are listed and briefly analysed in the first part of the paper. On the basis of this analysis, the liquid fuel concept and its benefits are introduced and briefly described in the following parts of the paper allowing to develop new reactor systems for nuclear incineration of spent fuel from conventional reactors and a new clean source of energy. As one of the first realistic attempts to utilise 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 in [1], has been proposed for a deeper, both theoretical and experimental studies in [2]. There will be 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 [3].

Introduction

There are principle drawbacks of any kind of solid nuclear fuel listed and analysed in the first part of this 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 utilisation of atomic nucleus energy to a death end (not having been able to solve principle problems of the corresponding fuel cycle in an acceptable way). On the basis of this, the liquid fuel concept and its benefits are introduced and briefly described in the following part of the paper.

As one of the first realistic attempts to utilise 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 in [1], has been studied both theoretically and experimentally. There is a preliminary design concept of an experimental assembly LA-0 briefly introduced in the following para 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 characterised 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 partly even 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 analysed. 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.

Solid nuclear fuel concept drawbacks

In spite of the fact that all what is following is well known it seems to be worth to remind it in the new circumstances of nuclear power at the end of the 20th century while starting to search new nuclear energy systems and fuel cycle options for the 21st century. Since the discovery of the reaction of atomic nucleus fission, the main goal of all efforts was to utilise 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, immediately, 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, the reaching of 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 specifically defined size arranged in a heterogeneous lattice filled in by a solid (graphite) or liquid (heavy water) moderator with a certain pitch determined by optimal neutronic conditions. This arrangement has remained nearly exclusive one being used even in latter systems with fuels enriched by U235 content up to much higher levels than the content of natural composition of uranium. The reasons were 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 (neutron poison) 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 have been adopted in nuclear reactor technology . 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, reactors 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, another consequence in the greater amount of fuel being present in the core than really necessary for the demanded power and then the more products including actinides is generated in the system.
2. The original fuel is finally so heavily poisoned by the products that it cannot keep the self-sustained fission chain reaction any more and a further operation of the reactor under original conditions is impossible. A principle change in the operation and structure of the reactor becomes to be unavoidable what means an outage and exchange of at least a part of the original 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 basically 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.

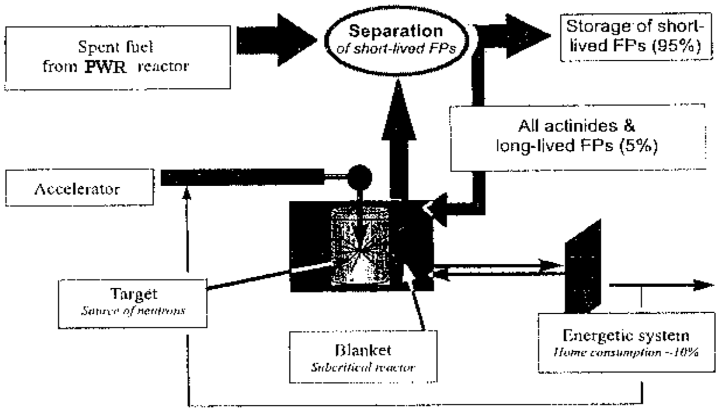
This fact contains one very controversial principle or better say a violence of a basic principle which can be described as follows: The energy generation in nuclear reactors utilises enforced nuclear process which are simultaneously producing products or nuclear waste (including secondary raw materials 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 one can utilise nuclear processes and which can transfer the high level and long-lived radionuclides towards short-lived or even stable nuclides-- the transmutation technology performed in a suitable nuclear reactor device and combined with an at least quasi-continuous separation of certain components of its core or reprocessing of the reactor fuel 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.

Liquid fuel concept for neutron source-driven transmutation technology

Molten fluoride salt fuel for neutron source-driven transmutation technology

The concept of a neutron source - driven subcritical blanket for a nuclear incineration of nuclear waste is well known for a several recent years [1] (see Figure 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 to approve its ability for an efficient realisation in the industrial scale.

Figure 1. **Principle scheme of a transmuter system**



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 either the outside of a close-packed set of cylindrical high-purity graphite blocks or inside cylindrical channels coaxially situated in e.g. hexagonal graphite blocks - Figures. 2 and 3.

Figure 2. **Single module of transmuter blanket**

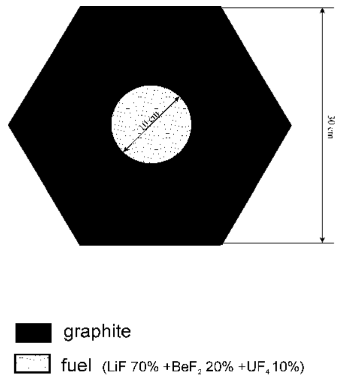
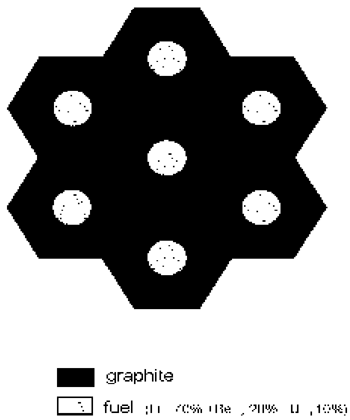


Figure 3. **The 7-module core of transmuter blanket**



There has been an experimental research system designed by the author preconceptually in [3] which should be developed and realised in the Nuclear Research Institute Rez 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 to reach the first results very cheaply and relatively soon. There can be an elementary, however, a sufficiently representative sample of the investigated reactor blanket lattice 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 LR-0 experimental reactor (a full-scale VVER type core modelling zero power reactor operated in Nuclear Research Institute Rez) or VR-1 (a training reactor operated at the Faculty of Nuclear Science and Physical Engineering of the Czech Technical University in Prague) which have been successfully operated for core analyses of thermal reactors since 1982 and 1990, respectively.

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, up to now usually used as 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 its all disadvantages (like e.g. the length ~ 1 km, the investments ~ U$ 1 billion, etc.) having been applied e.g. in the Los Alamos concept is not necessary more in the system and a low level parameter accelerator can be employed.

Neutron sources for transmuters based on low parameter accelerators

In various concepts of the accelerator driven transmutation technologies (ADTT) the distinct effort is devoted to an employment of external neutron source other than spallation reactions initiated on (mostly future proposed) high energy proton linacs (>1 GeV, 100 mA, 10^{19} n/s). The obvious reason is that the lengths (1 km) and expected cost (above 1 B U$) of these facilities, which are an inherent and insuperable weak point of high energy proton linacs [4], could make unreliable the wide application of the ADTT. In the “subcritical enhanced safety molten salt reactor concept” the effective way of reducing the external neutron source power (below 3.5×10^{16} n/s) is accomplished by the cascade neutron multiplication in the system of coupled reactors with suppressed feedback between them. For such a “burner” reactor scheme the possibility of replacing proton linac with 100 MeV electron linac (of substantially lower length and cost than proton linac) has been argued [4]. In the similar scheme an employment of the isochronous AVF cyclotron-based neutron source was also considered [5].

Although external beams in the mA range (10^{14} n/s) have been already demonstrated for conventional AVF cyclotrons, the cryogenic technology for compact cyclotrons and also a wide class of commercially available, low-cost (below 3 M U$) cyclotrons of a type CYCLONE seem to be well methodical and technical basis for further beam-intensity improvements. Nevertheless, for a significant increase of beam intensity and neutron source strength the drift tube of Alvarez linear accelerators with the RFQ (radio-frequency quadruple) injector are more promising. The most developed proposal for 3×10^{16} n/s source strength projected originally for fusion material irradiation tests (FMIT) is based on 35 MeV,100 mA deuteron linac [6]. A fast flowing lithium jet is considered to be the best target material [7] for megawatt powers of proton and deuteron beams with medium energy. The medium energy (E < 100 MeV) proton and deuteron induced reactions on thick Li target produce a forward-directed fast-neutron fields (the fluency averaged energy E_n of about 15 MeV is to be compared with E_n ~ 3 MeV which corresponds to the spectral yield from a spallation reactions). Therefore, it seems appropriate to perform any computational and experimental study of target - blanket systems employing primary neutron sources, which in general have energy spectra with suppressed contribution of low energy neutrons.

In the NPI Rez, the d(18 MeV)+Be neutron generator, originally developed for the military directed research is now being upgraded with the main task to take advantage of H^- and D^- negative ions recently implemented on the NPI isochronous cyclotron U-120M. Conversion of the cyclotron into H^-, D^- machine enabled to utilise a high efficient extraction by means of the stripping, which resulted in substantially increased extracted beam currents of positive p^+ and d^+ ions. Nowadays, up to 20 µA currents of the 15 - 30 MeV protons are routinely extracted to various types of targets for radionuclides production. The purpose of employing this beams for fast neutron production is to perform a broad range of experiments closely related to the activation analysis and ADTT program as well.

For ADTT empirical research in NPI, an experimental study of the spectral and yield characteristics of various neutron produced reactions between light nuclei is now under way, the main tasks of which are as follows: a) to verify yield calculations from cross-section data, b) to determine an empirical shape of spectral yield of neutrons from deuteron break-up processes so as it can not be predicted reliably from simple phase-space calculations, and c) to determine the contribution of the target- v station arrangement to the background part of produced neutron fields. Knowledge of these characteristics is needed to evaluate the target and beam options for the best simulation of ADTT external neutron source mentioned above. The results of first experiments were found to be in a good agreement with calculations based on updated cross-section database (EXFOR). The preliminary calculations show that for the present neutron facility the neutron source strength up to 6×10^{12} n/s.sr and fluency averaged neutron energy E_n =15 MeV could be achieved from thick deuterium target irradiated by 30 MeV protons.

Blanket concept of at Transmuter for PWR spent fuel incineration

There has been a convenient blanket concept for an efficient nuclear incineration of PWR spent fuel developed as a combination of those two ideas described in two paragraphs above. The concept is illustrated by the Figure 4, where two zones are indicated, one under-moderated and thus better equipped for actinides burning and the second well-moderated (fuel channel in a graphite block) and thus more convenient for fission products incineration.

Experience in fluorine technology and application of fluoride liquid fuel in ADTT

There has been a technological process worked-out for the separation of uranium and plutonium from the spent fuel by a dry (fluorine) method in the NRI, Rez, in a close collaboration with the Kurchatov Institute, Moscow, during the 80s. The whole process was upgraded to a pilot plant scale with a capacity of 1-3kg of processed fuel/hour. There was a part of the technological equipment built and verified at an inactive scale at the NRI Rez. The whole technological line called Fregat (Figure 5) was then realised in the Institute of Atomic Reactors at Dimitrovgrad, Russia, and all processes proved by reprocessing of spent fuel from the fast reactor BOR 60. All equipment including fittings, measuring instruments and accessories have been built in the former Czechoslovakia, the plutonium part of the pilot plant has been built in the former USSR. A certain experience has also been obtained on the uranium isotopes separation by ultra-centrifugation and electrochemical processes.

Figure 4. **The blanket concept of a transmuter**

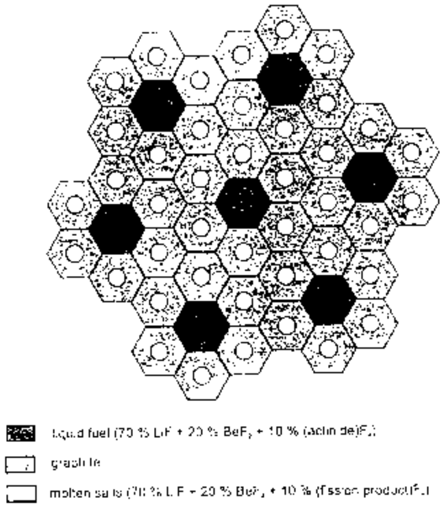
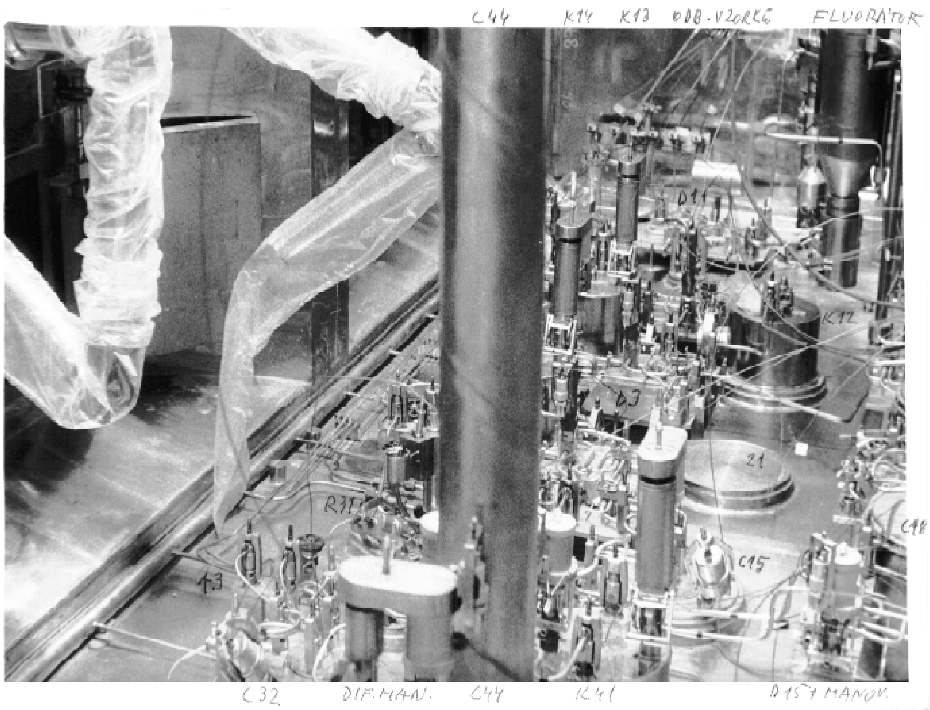


Figure 5. **The fluorine technology reprocessing line Fregat**



Technological development and testing

The experience gained in the course of the research is going to be applied in developing fluoride based liquid fuel as well as fluoride chemistry separation processes for the use in the Accelerator Driven Transmutation Technology (ADTT)

Beside this, the project "Experimental Molten Salt Loop for ADTT program" is carried out in the Škoda Works, Nuclear Machinery Ltd. in Pilsen, Czech Republic. In the frame of this project technological loop for studying of molten fluoride salts characteristics was designed and fabricated and then installed in the NRI Rez which will start up an experimental program till the end of 1998.

Conclusions

The analyses of spent fuel management from PWRs as well as all other nuclear reactors employing solid fuel concept have showed the principal drawbacks of that concept causing a series of consequences leading to a crucial issue of a nowadays nuclear power – spent solid fuel with accumulated actinides and long-lived fission products (without regard whether the open nuclear fuel cycle or a multiple reprocessing is applied). The necessity of an employment of nuclear processes and an adequate nuclear technology (nuclear incineration) in an efficient solution of that problem is definitely evident. The national R&D programme which was very briefly introduced in this paper is closely connected with the world - wide effort of research teams in leading countries of nuclear power like e.g. France, USA, Russia and Japan and forms a contribution to the common effort in the solution of that global issue. The overall co-operation in this field gives a real chance to make nuclear power acceptable as a clean source of energy for the 21st century.

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