EXPERIMENTAL VERIFICATION OF SELECTED TRANSMUTATION TECHNOLOGY AND MATERIALS FOR BASIC COMPONENTS OF A DEMONSTRATION TRANSMUTER WITH LIQUID FUEL BASED ON MOLTEN FLUORIDES (DEVELOPMENT OF NEW TECHNOLOGIES FOR NUCLEAR INCINERATION OF PWR SPENT FUEL IN THE CZECH REPUBLIC)

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Abstract

Development of the project SPHINX (spent hot fuel incinerator by neutron flux) is based on the utilisation of a subcritical nuclear reactor system with liquid fuel based on molten fluorides. Existing experimental facilities have been used for experimental testing of the system, individual technological components and newly developed materials that should be resistant in the environment of molten fluorides at operational temperatures. Examples of facilities include the experimental reactors LR-0 and VR-1 with inserted zones for the investigation of neutronic characteristics of transmuter blanket cores, research reactor LVR-15 with instrumented probes for transmuter blanket sample testing in high neutron flux conditions, and experimental loops with molten fluoride salts for an investigation of transmuter system operational conditions. This paper describes some of the experiments performed and results obtained thus far.

Introduction

During the last decade, there have been new tendencies of significant decreasing of amount and potential danger of nuclear waste getting up in the world of nuclear power. The development of a convenient technology to resolve this issue has involved the rehabilitation of nuclear transmutation technologies, which have been well known since the beginning of the first nuclear era. However, at that time such technology was evaluated as unacceptably demanding from technological and economical perspectives. An attempt at the end of the first nuclear era to develop modifications of this technology under different conditions was supposed to be promising and acceptable for the efficient closing of the nuclear fuel cycle. The newly modified transmutation technologies should make it possible to return a fuel component, which is contained in nuclear fuel and burned up in conventional nuclear reactors, back to the nuclear fuel cycle and to turn a significant part of long-lived nuclear waste (heavy metals from the transuranium region) into useful energy without rests and in an irreversible way. Besides other positive features, this could be a way to exclude or at least minimise the undesirable misuse of such nuclear materials.

A series of national projects were started at the end of the 1980s and at the beginning of the 1990s. Of these projects, at least two should be mentioned – the Japanese OMEGA project and LANL's (USA) ADTT project. In the middle of the 1990s [1] in the Czech Republic, a similar project was begun following the tendencies established by Charles Bowman of LANL. The project, which was later on called SPHINX (spent hot fuel incinerator by neutron flux), has focused on the development of a burner for spent fuel from PWRs that have been operated in this and certain other central European countries. The principle feature of the adopted transmuter concept has been, since the very beginning of the project, the concept of liquid fuel based on molten fluorides. The proposal and organisation of the project was based on the activity of the national consortium, Transmutation, established in November 1996 by four leading institutions in nuclear research: Nuclear Research Institute Rez plc, the Nuclear Physics Institute of Academy of Sciences, SKODA Nuclear Machinery plc and the Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague [with whom the Technical University in Brno (specialised in secondary circuit problems) became associated in 2000].

The first stage of the project (mainly R&D) was proposed in 1997 and once approved, started under the acronym LA-0 in 1998. The focus was concentrated on broad computing analysis of the proposed system and elaboration of complex input for designing an experimental transmutation system on a zero power level. After successfully completing the first stage in 2000, the second stage was approved for the period of 2000 to 2003, which focused on experimental verification of selected transmutation technology for design development of basic components of the LA-10 demonstration transmuter at a 10 MW power level. This activity is based on the utilisation of existing experimental facilities as well as long-term experience and know-how, which include the following:

- Neutron source facility NG1 on external beam line of the isochronous cyclotron U-120M of the Nuclear Physics Institute (NPI) of the Czech Academy of Sciences in Rez (variable energy up to 35 and 17 MeV of protons and deuterons, respectively), which is based on the d (17 MeV) + Be (thick target) reaction.

- Experimental reactors LR-0 and VR-1 with inserted zones for the investigation of neutronic characteristics of transmuter blanket cores.

- Research reactor LVR-15 with instrumented probes for transmuter blanket sample testing in high neutron flux conditions.

- Experimental loops with molten fluoride salts for an investigation of transmuter system operational conditions.

- Know-how in the field of fluorine technology gained in the frame of long-term R&D activity on spent fuel reprocessing by dry method.

The governmental authorities of the Czech Republic as well as the future utilities of the developed technology have been providing the necessary funding. The utilities include: Radwaste Repository Authority, CEZ a.s. (Czech Electricity Generating Co.), SKODA Works Company Nuclear Machinery Branch plc and some other entities. A substantial part of the project has been incorporated in suitable forms of international collaboration such as the European Union's Framework Programmes, which are considered to be the most convenient. Individual parts of the project have been incorporated into corresponding tasks of the MOST (molten salt technology) project of the EU's Fifth Framework Programme. This collaboration began in November 2001 and lasted for three years. The convenient incorporation of the whole complex into the preliminarily proposed projects of the Sixth Framework Programme for a period of four years (2004-2008) as well as into other forms of multinational co-operation is desirable.

Principle features of the SPHINX project

The principle scheme of the SPHINX project is introduced in Figure 1. One of the main features of the project is the front end of its fuel cycle – the dry technology of reprocessing spent fuel from commercial reactors based on a fluoride volatility process is employed. This simultaneously represents the process of preparation of the transmuter fuel (in the form of molten fluorides) for the transmuter fuel cycle, supposing a continuous circulation of liquid fuel and at least quasi-continuous cleaning up during the transmuter operation. We should underline that the majority (~95%) of the inventory of spent fuel from the uranium-plutonium fuel cycle of conventional power reactors (nearly all uranium inventory as well as the majority of short-lived fission products) is supposed to be taken out for the remainder of the transmuter fuel cycle whereas all transuranium as well as long-lived fission products should remain in the mentioned forms of fluoride.

The second main feature of the SPHINX project is the developed concept of the transmuter and its blanket (Figure 2).

Figure 1. The principle scheme of the SPHINX project

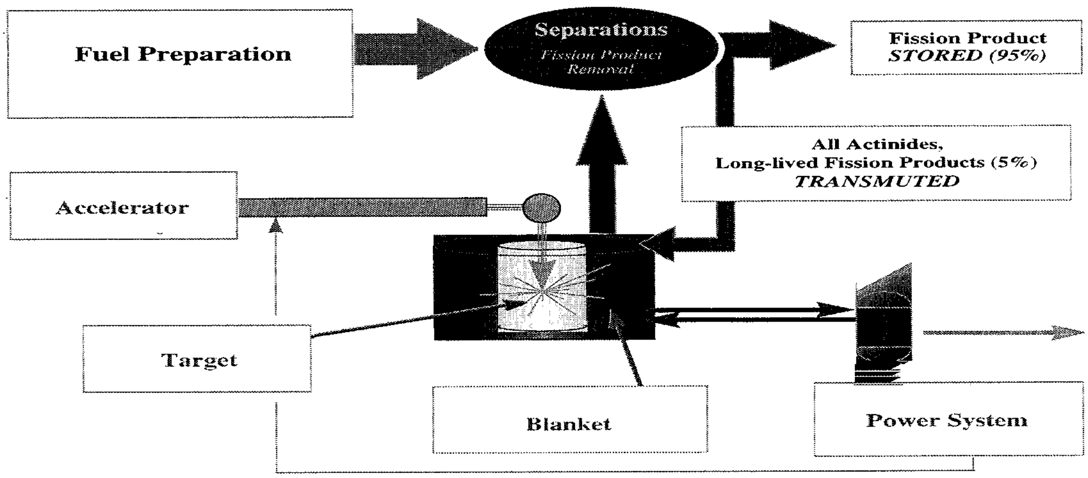
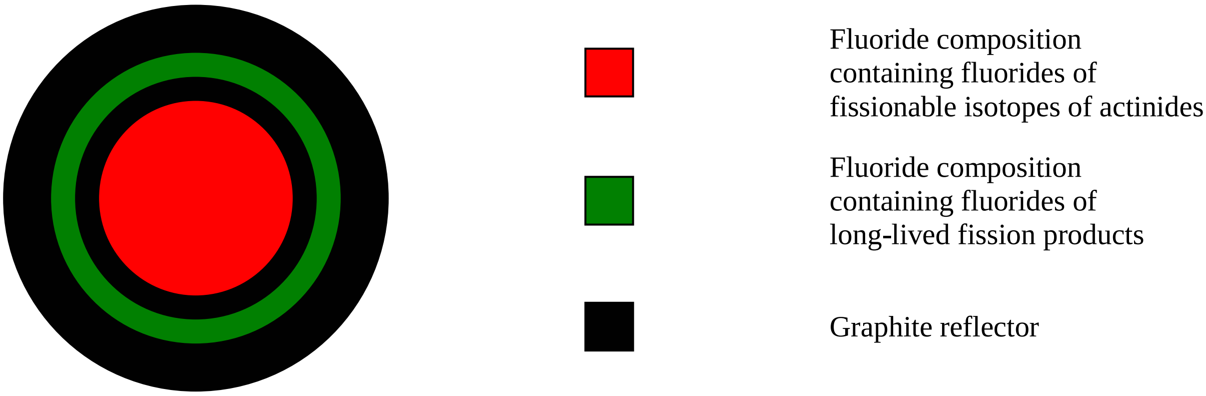


Figure 2. The schema of a transmuter elementary module with two primary circuits of molten fluorides

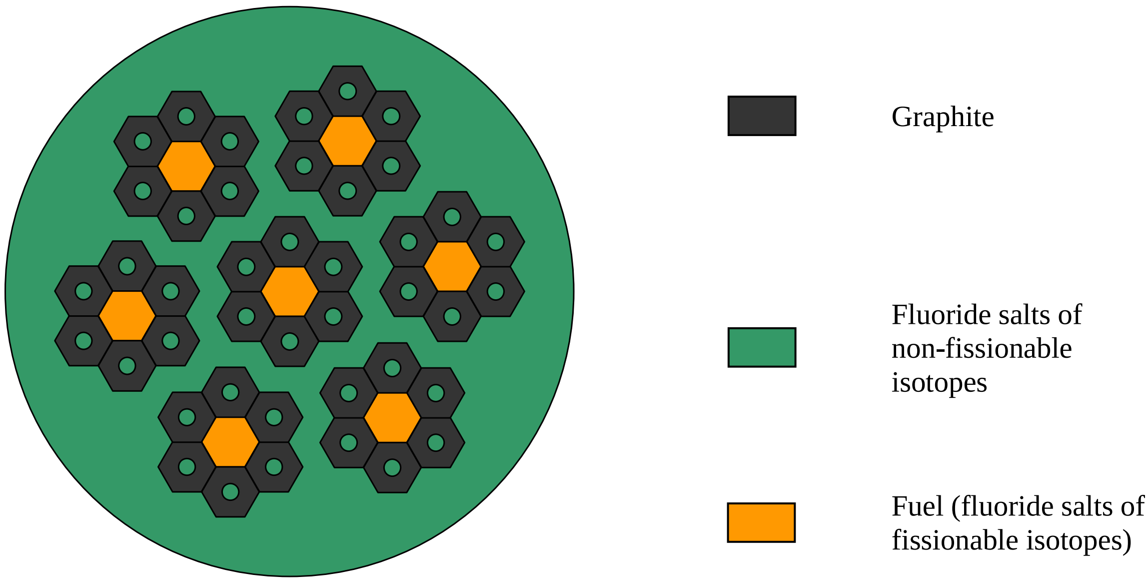


This concept is based on the idea of utilisation of efficient burning of transuranium nuclei in a large size fuel channel in the epithermal energy range where the majority of isotopes have resonances in their neutron cross-sections. It might be a critical system with reactivity and power controlled by specific flowing liquid fuel control systems (based on control of liquid fuel composition or its flow characteristics) or a subcritical system (with the subcriticality margin on the order of several betas) whose reactivity (keeping the prescribed level of subcriticality) is controlled in the same manner as in the above-described case of a critical system. Furthermore, the system is kept in a steady state and its power is controlled (driven) by an external neutron source or by a driving zone (part of a conventional reactor core).

The elementary module can be designed as a subcritical hexagonal fuel channel surrounded by six hexagonal graphite blocks, equipped with coaxial tubes (where molten fluorides of long-lived radionuclides may flow) that allow for efficient transmutation of long-lived radionuclides by an intensive flux of well-thermalised neutrons. These elementary modules represent an intensive source of energy and epithermal neutron flux that might be slowed down in graphite blocks and allow for very efficient incineration of at least some of the long-lived fission products in well-thermalised neutrons escaping the transmuter core. It should be noted that these elementary modules can be closely packed (joint) into a critical system or arranged in a complex of autonomous subcritical elementary modules that are driven by a set of external neutron sources or by a driving zone (a reactor core of another type surrounding the subcritical assembly of the molten fluoride elementary modules).

The two purposes of the proposed blanket system utilisation make usage possible as an efficient nuclear incinerator, actinide burner and, simultaneously, efficient energy source. Let us note that once such a system is developed and proven, it might serve as a new clean source of energy just by switching over from a uranium-plutonium fuel cycle to a thorium-uranium fuel cycle and thus excluding the generation of actinides. The main features of the SPHINX concept and the proof of individual processes as well as the corresponding technological and operational units of the nuclear "jeep" should be verified by the operation of a demonstration complex with power output on the order of 10 MWt. The concept of an experimental assembly of that type is introduced in Figure 3. Before we are able to design such a device, we must perform a broad experimental verification of design inputs for credible designing of a demonstration transmuter. The process of experimental verification recently (2002) began in the scope of the LA-10 project on the basis of preliminary preparation of an ideology of the experimental programme that was formulated earlier (in the scope of the LA-0 project in 1999 and 2000); some of its principle features were proven during this period. The experimental programmes and projects that have so far been developed/started will be described in the text that follows.

Figure 3. Subcritical dual-purpose experimental transmuter concept



Main experimental programmes started in the scope of the SPHINX project

The two specific features of the SPHINX project mentioned above, namely the chemical technology of the front end of the fuel cycle process and the neutronics (both static and kinetic) of the multi-purpose blanket with flowing liquid fuel, represent sufficiently new concepts to be calculated by modified computer codes that should be experimentally verified. Regarding the fluoride volatility process, we use as a base the knowledge gained from experience accumulated during the 1980s when we worked in close collaboration with the Russian Kurchatov Institute (RKI) in Moscow. During this time, such technology was developed and a pilot line was built and prepared for testing by processing hot spent fuel from the BOR60 fast reactor in the Atomic Reactor Research Institute (ARRI) in Dimitrovgrad. During the 1990s, this line was innovated and installed at NRI Rez and the first stage of testing for the SPHINX project was begun.

The new blanket concept was developed and proven by broad computer analyses in the second half of the 1990s. In the same time period, experimental programmes for verification of main neutronic characteristics including time behaviour were proposed. For this purpose, there are several large experimental devices operated in the scope of the Transmutation consortium. First of all, the experimental reactor LR-0 at NRI Rez and VR-1 at FNSPE are at our disposal as well as the NPI cyclotron accelerator that was equipped with a new target serving as an intensive neutron source. The VR-1 experimental reactor was equipped with the oscillator for the purpose of SPHINX-type blanket testing (namely, time behaviour and important safety characteristics). The first stage of experimental verification of some basic neutronic characteristics, reactor equipment and materials technology, and measurement techniques has already begun and involves experiments with inserted zones containing materials typical of our blanket concept and simulating neutronic features in a simplified model.

Technology of the new non-traditional materials in the SPHINX blanket is the subject of a broad R&D programme including experimental testing. For this purpose, a series of technological loops was erected by SKODA Nuclear Machinery plc, starting with laboratory equipment and continuing in a semi-pilot scale. The same is valid for the development of a technology for continuous cleaning up of circulating liquid fuel in the transmuter internal fuel cycle based on electro separation methods.

In 2000, the Technical University in Brno (TUB) was associated into the Transmutation consortium for the studying and testing of secondary cooling circuits of the LA-10 transmuter. The first step in an experimental programme in that field, the measuring bench, was erected and will serve for both measurement of basic technological characteristics of fluoride compositions and verification of design input for an auxiliary circuit of a molten fluoride media pantry in large loop testing complexes, which are being developed jointly by SKODA NM and Energovyzkum (EVM) Ltd. (a daughter company of TUB), as well as for the LA-10 demonstration transmuter.

The most important experimental program was started in 2001 and was based on experimental irradiation of blanket samples in the high neutron flux of research reactors. This programme is being carried out in close contracted collaboration with the RKI project AMPOULE and involves the development and validation of a modified reactor computer code based on a stochastic Monte Carlo model of neutron field long-term time behaviour in a system with circulating liquid fuel based on molten fluorides. The experimental irradiations are simultaneously performed at NRI and RKI in the first stage. The second stage representing irradiation of samples containing fluorides of actinides (Np, Pu, Am and Cm) was agreed to be performed and finalised jointly at RKI in 2002/2003. Some of the results of the preparatory (first) stage of this programme, which have already been obtained in NRI Rez in the scope of the BLANKA project, will be briefly introduced in the following section. We note that the proposal to start broad experimental verification of design inputs for a demonstration transmuter of the mentioned type was already put forth at the Fourth International ADTT&A'01 Conference in Reno, Nevada at the end of 2001 [3]. All of these experimental programmes were also (just recently) proposed to the Sixth Framework Programme of the European Commission.

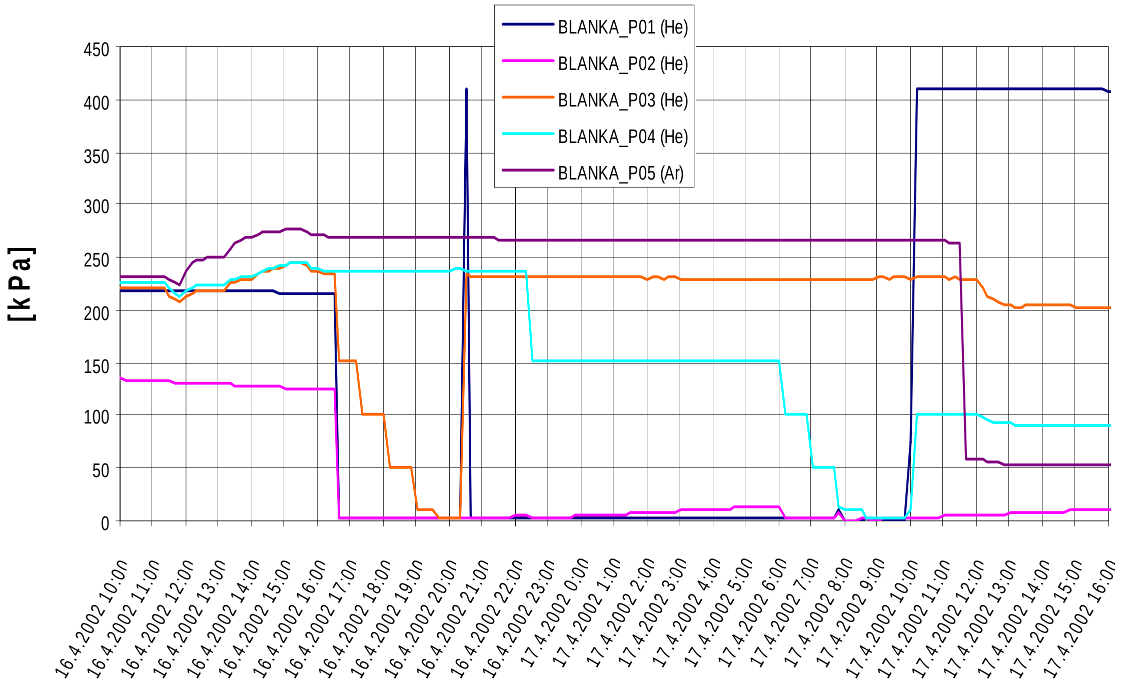
The programme of irradiated probes and BLANKA instrumented rigs

The very first idea for this programme was initiated by RKI as early as the late 1980s when some very preparatory irradiations and measurements were performed in the Tashkent research reactor. On the basis of the obtained experience, the project AMPOULE was developed at RKI in the 1990s. After a period of pre-contract negotiations in the second half of the 1990s, the bilateral contract was finally agreed upon and signed in July 2001. Immediately thereafter the first stage of the joint development of the modified reactor computer code ISTAR (based on a stochastic Monte Carlo model of neutron field long-term time behaviour in a system with circulating liquid fuel based on molten fluorides) started in autumn 2001. At the same time, preliminary irradiations, measurements and evaluations were performed on the LVR-15 research reactor at NRI with the aim to be able to design and perform the regular programme of irradiated BLANKA probes, serving as validation of the developed computer code.

During the same time frame, an elementary metallic probe and a complex of irradiated probes were performed, which lead to the realisation of BLANKA instrumented rigs. The first series, BLANKA 100 (identical in size to one fuel assembly of both the VR-1 experimental and LVR-15 research reactor), was designed and tested at NRI during 2001. Preliminary tests of the rig neutronic characteristics started in December 2001 on the experimental reactor VR-1 at the Czech Technical University in Prague. Then, a series of first irradiations were performed on the NRI research reactor LVR-15 in January and February 2002. The series of irradiation tests called BLANKA 101 were curried out with empty probes (no fluoride content), serving just for the tuning of all measuring channels and verification of design-predicted characteristics (temperature and pressure fields in particular).

After an evaluation of the obtained results, the first experiments were performed in April 2002 using probes containing pure NaF and/or NaF with an admixture of metallic Mo powder, modelling the presence of fission product. The probe, BLANKA 102, was inserted in the core of the research reactor at its very edge with a low value of neutron flux where heating of the probe is generated nearly by gamma capture only. The temperature field was measured by a set of five independent thermocouples located in different positions in the elementary probe room. Figure 4 shows the results of a continuous measurement of gas pressures in individual layers of the BLANKA 102 probe during irradiation. The decrease of the pressure of the noble gas (Ar) filling the elementary probe at 11:30 on April 17 is caused by the testing of a controlled discharge of expected gaseous fission products in the case of the fluoride composition containing components of fluorides of fissionable metals.

Figure 4. Results of measurements of gas pressures in individual layers of BLANKA 102 probe



After evaluation of BLANKA 101 and 102 test results, some convenient small rearrangements were applied in the prepared new version of the BLANKA 103 probe, which were used in the experimental programme performed during the first half of 2003. The majority of the irradiated samples were based on a composition of ^{7}LiF + NaF. The admixture of modelling fission products and finally even an admixture of a small amount of UF_4 were incorporated into the individual sample compositions. An exact description of the BLANKA 103 sample content can be seen in Figure 5.

Figure 5. Contents of the BLANKA 103 sample pots

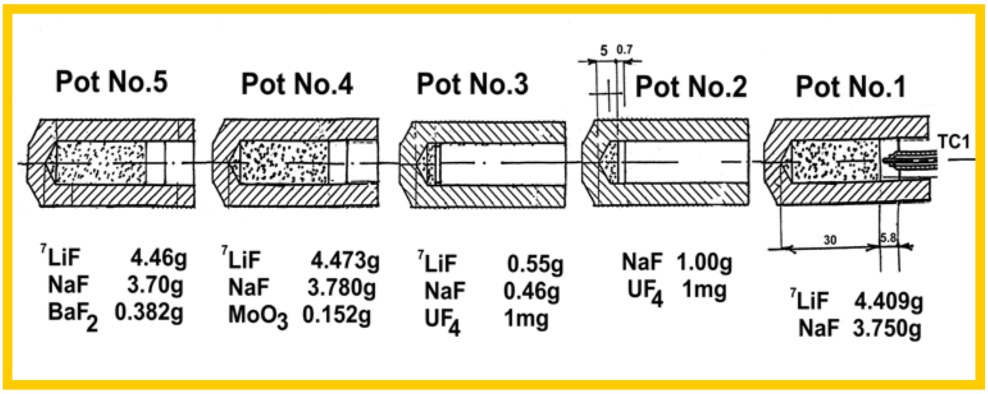
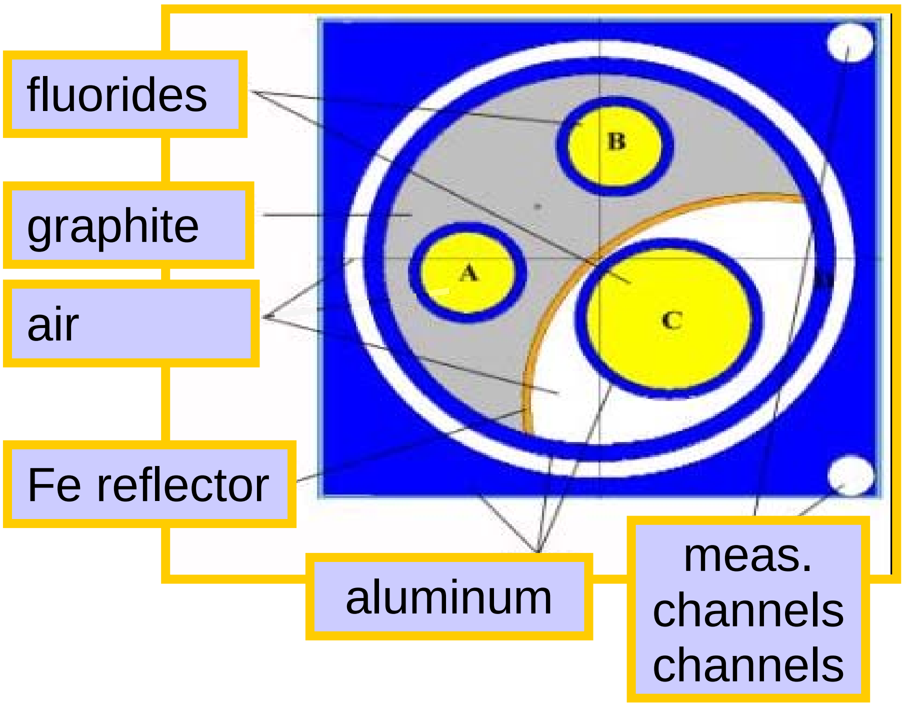


Figure 6. The simplified model of the BLANKA 400 rig for neutronic tests at room temperature



Following the experience accumulated during the BLANKA 100 instrumented rigs, the larger size series (identical to the area of four fuel assemblies) called BLANKA 400 was designed and preliminary tests started on the VR-1 experimental reactor at the end of 2003 under room temperature and low level of neutron flux conditions. For these neutronic tests, the simplified version manufactured from nuclear aluminium structural material (see Figure 6) was used. Once again, the first series of experiments was performed with pure NaF in tubes A, B and C, starting at the beginning of 2004 and due to finish by 31 May 2004. The same model was to serve for an adjustment of the testing facility with the external neutron source NG1 on external beam line of the isochronous cyclotron U-120M at NPI during the second half of 2004. During this time, neutronic experiments with BLANKA 400 filled in by LiF + NaF compositions in various ratios were forecast to take place at the VR-1 experimental reactor.

Conclusions

Principle features of the SPHINX project involving the development of a transmuter with liquid fuel based on molten fluorides and a connected R&D programme were described in necessary detail. The experimental part of the R&D programme of the SPHINX project was briefly introduced, which serves as verification of design inputs for designing a demonstration unit of a transmuter with liquid fuel based on molten fluorides. Due to the current status of the experimental programme, performance was focused on the irradiation of samples of molten salt systems as well as structural materials proposed for the blanket of the SPHINX transmuter in the field of high neutron flux of research reactors (attention focused on the latter part). We should underline that the main aims of this programme (called BLANKA irradiated probes) are the following:

- Experimental verification of long-time behaviour of transmuter blanket, which contains molten fluoride salts as fuel and graphite as moderator or reflector.

- Material research on behaviour of materials in neutron and gamma fields, and material interactions in high temperature conditions.Validation of computational codes (system of codes NJOY – preparation of data library for MCNP; MCNP – computation of cross-sections for the code ISTAR being developed for the computation of actinide concentration in long-term operation of the transmuter).

Preliminary experiments have been performed by irradiation of simplified samples of fluorides of non-fissionable metals since mid-2001. The first experiment with the instrumented rig BLANKA 101, which served for the tuning of measuring methods and channels, started in December 2001. In this experiment, the probe without fluorides was inserted into the core of the LVR-15 research reactor in NRI Rez for a few days of irradiation on different levels of the reactor power (neutron flux). The second BLANKA 102 experiment was conducted in April 2002 when two samples were irradiated – one containing pure NaF and another containing NaF with an admixture of Mo metallic powder (modelling a fission product). Measurements and experience collected in this stage fully confirmed the principle correctness of design and operational properties of the programme of BLANKA instrumented rigs, serving as irradiation tests of transmuter blanket material samples under conditions close to operational.

After the evaluation of BLANKA 101 and 102 test results, some convenient small rearrangements were applied in the prepared new version of the probe BLANKA 103 and they were used in the experimental programme performed during the first half of 2003. Nearly all of the irradiated samples were based on a composition of ^{7}Li + NaF. The admixture of modelling fission products and finally even an admixture of a small amount of UF_4 were incorporated into individual sample compositions. There was also the insertion of structural material samples (also a subject of broad non-active and pre-irradiation technological testing), which were supposed to be irradiated and preliminarily tested in these irradiations. At the same time, the preliminary neutronic tests of the larger sized version (equal to the area of four quadratic fuel assemblies of VR-1 or LVR-15 reactor) of the BLANKA 400 rig started using a simplified aluminium model at room temperature inserted into the VR-1 experimental reactor and into the testing facility with the external neutron source NG1 on the external beam line of the isochronous cyclotron U-120M in NPI. We will be able to report later this year on results obtained in these experimental programmes.

Acknowledgements

The SPHINX project was proposed at the end of 1996 by the Transmutation consortium established by four leading nuclear research bodies in the Czech Republic (Nuclear Research Institute Rez plc, SKODA Nuclear Machinery plc in Pilsen, the Nuclear Physics Institute of Academy of Sciences in Rez and the Technical University in Prague) to whom Technical University in Brno (specialised in secondary circuit problems) associated in 2000. The project has been supported by the Ministry of Industry and Trade of the Czech Republic, CEZ a.s. (Czech Power Company) and RAWRA (Radwaste Repository Authority). At present, two agreements on multi-national co-operation in this field have been signed – one with the European Commission (project MOST) and one with the Russian Kurchatov Institute. The experimental programme of project SPHINX was proposed to the Sixth Framework Programme of the European Commission for the period 2004-2008.

The authors would like to acknowledge all the support the project has received so far and to express their hope that understanding will continue as progress is made towards the efficient resolution of crucial issues on nuclear power and as successful development takes place in this new era.

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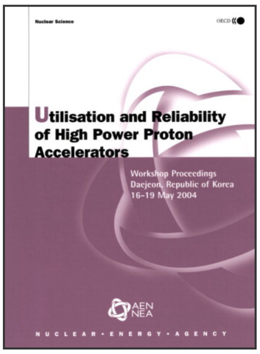

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