

CURRENT STATUS OF CZECH R&D PROGRAM IN PARTITIONING AND TRANSMUTATION

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Abstract

Czech research and development program in the area of P&T is grounded on the Molten Salt Transmutation Reactor System concept with fluoride salt based liquid fuel, the fuel cycle of which is based on integrated pyrochemical fluoride partitioning processes. The attractiveness of Molten Salt Reactors is not only in the transmutation of transuranium elements and long-lived fission products, but also in their possibility to work under the $^{232}\text{Th} - ^{233}\text{U}$ fuel cycle with minimized production of transuranium elements in their waste stream. The experimental and theoretical works in the area of R&D of Molten Salt Reactor technologies and pyrochemical partitioning processes are oriented mainly towards the technological research of Fluoride Volatility Method and electroseparation method, measurement of molten-salt neutronic characteristics accompanied by the development of computer codes and the development of selected equipment and structural materials for molten-salt technology.

Introduction

Sustainable development of nuclear power is one of the key points of the National Energy Policy for next 30 years adopted by government of the Czech Republic in the beginning of 2004. The crucial present-day issue, on which the whole further development of nuclear power is conditioned, is finding of a satisfactory solution of the back-end fuel cycle problem. The problem is of high strategic importance as for state institutions as for electricity generating companies operating nuclear power stations. One of the possible solutions offers the closing of nuclear fuel cycle with the application of a partitioning and transmutation (P&T) technology. The realization of such a technology, in the first instance in conjunction with the development and deployment of advanced nuclear power reactors, is considered to be a promising way for the future use of nuclear power. Development of P&T technology is also in a good agreement with the National concept of radioactive waste and spent fuel management adopted by Czech government in 2002. The support of national P&T program is here considered as a development of alternative concept to the concept based primary on the non-reprocessed spent fuel disposal into the underground repository.

The only company generating electricity in nuclear power plants in the area of Czech Republic is the Czech Power Company CEZ, which operates two NPPs with six PWR (VVER) units. The nuclear share of total electricity generation in the Czech Republic reaches now about 40 %, however the further accrual is anticipated due to intention to construct other new nuclear reactors during forthcoming decades. At present, the spent fuel from Czech NPPs, after discharging from reactors, spending from five to six years cooling period at reactor pools, is stored in the Spent Fuel Storage Facility. According to the Czech Atomic Act, the State guarantees the safe disposal of all radioactive wastes including spent nuclear fuel. The disposal of the non-reprocessed spent fuel in the underground repository is described as the solution, feasible by current sum of scientific and technical knowledge and expresses the certainty of safe solution of the fuel cycle back-end. For this case, the realization of an underground repository is preliminary planed with the possibility to accept the first spent fuel after 2065. Because of the final decision how to dispose the spent fuel will have to be made in tens of years, the chance to develop technically, economically and socially more attractive solution than underground disposal of spent fuel is in the foreground of professional interest for present and next decades. Mainly now, after adopting the National energy policy, the need of the closing of the fuel cycle rises up significantly.

Czech R&D Program in Partitioning and Transmutation

The Czech research and development program in the field of partitioning and transmutation is grounded on the Molten Salt Transmutation Reactor system concept with fluoride salts based liquid fuel, the fuel cycle of which is based on pyrochemical and/or pyrometallurgical fluoride partitioning of spent fuel. The choice of Molten-Salt Reactor (MSR) came out from the attractiveness of this type of reactor system to be used as for transmutation of actinides from spent nuclear fuel as for the possibility of MSR to work under $^{232}\text{Th} - ^{233}\text{U}$ fuel cycle with minimized production of nuclear waste. Both features can make MSRs attractive also for electricity generating companies, which must now pay a fee corresponding to the production of radioactive wastes. The last but not least reason for the MSR choice has been a historical experience of several Czech institutes and companies in the development of nuclear fluoride technologies.

The experimental and theoretical works in the area of research and development of Molten Salt Reactor technologies and pyrochemical partitioning processes in the Czech republic are oriented mainly towards the following fields:

- technological research and development of “Fluoride Volatility Method” for spent fuel reprocessing
- laboratory research on electrochemical separation processes using fluoride molten salt media
- development of computer codes for calculations of neutronic characteristic of Molten Salt Reactors
- experimental prediction of neutronic characteristics for the validation of computer codes
- development and verification of structural materials for molten fluoride salt medium and development of selected equipment for MSR technology and for fluoride partitioning technology

R&D in the field of Fluoride Volatility Method is concentrated to the development and verification of experimental semi-pilot technology for PWR spent fuel reprocessing, which may result in a product the form and composition of which might be applicable as a starting material for the production of liquid fluoride fuel for Molten-Salt Transmutation Reactor. The Fluoride Volatility Method is based on the direct fluorination of powderized spent fuel with fluorine gas in a flame fluorination reactor, where the volatile fluorides (represented mainly by UF_6 , partially NpF_6) are separated from the non-volatile ones (e.g. PuF_4 , AmF_3 , CmF_3 , fluorides of majority of fission products), and on the subsequent purification of the volatile components by using technological operations of condensation, rectification and sorption. Consequently, the objective is a separation of a maximum fraction of uranium component from plutonium, minor actinides and fission products. The method should be placed in the “Front-end” of the MSTR fuel cycle. The other important mission of the process is also the conversion of the oxide form of spent fuel into the fluorides, which represents the chemical form of the liquid fuel for Molten-Salt Reactors. The main research work in the area of Fluoride Volatility Method is focused to the experimental program carried out at the technological line FERDA (Fig. 1) in the Nuclear Research Institute Rez plc. The three-year test program is focused mainly to the study of flame fluorination process, which is considered to be the crucial unit operation of the technology. Then the line should be appropriately modified for replacement into hot cells and the verification program should continue with irradiated fuel.[1]

Fig. 1: Upper part of UF_6 , NpF_6 condensers with piping and measurement system



The R&D on electrochemical separation technologies from fluoride melt media is aimed to the final partitioning of selected actinides from lanthanides, passed from the Fluoride Volatility Process or to the “on-line” reprocessing (partitioning) of the circulating fuel in Molten-Salt Reactors. This technology is considered to be applicable as for Molten-Salt Transmutation Reactors (MSTRs) (plutonium and minor actinides burners) as for Molten-Salt Reactors working under the $^{232}\text{Th} - ^{233}\text{U}$ fuel cycle. It is considered that the “on-line” reprocessing based on the electrochemical separation processes should be, in addition to the volatilization techniques, accompanied by molten salt / liquid metal reductive extraction process. This technology is not under the development in the Czech Republic so far, however, the international co-operation between the NRI Rez plc and CEA Marcoule in France, where this process is under the study, allows us to use the results achieved also in this area. The main effort in the R&D of electrochemical separation technology has been focused to the development of two electrochemical separation techniques – **cathodic deposition method** and **anodic dissolution method**, both methods are under the study for various types of molten fluoride salt media. Several mixtures of fluorides (LiF-NaF-KF , LiF-NaF , LiF-BeF_2 , LiF-NaF-BeF_2 , LiF-CaF_2) have been selected and tested as individual carrier molten fluoride salts. Some detailed results of the program are presented in the R. Zvejskova paper.[2]

Besides the two main experimental partitioning activities, the flow-sheeting research is also in the focus of interest in the NRI Rez plc. Flow-sheets proposed as for MSTR as for MSR represent the important clue for experimental scientific and engineering staff not to leave the assigned R&D line. The simplified schematic flow-sheet of the MSTR fuel cycle, based on the Czech P&T concept is shown in Fig. 2., the conceptual flow-sheet of MSTR on-line reprocessing is shown in Fig. 3.

Fig. 2: Simplified scheme of the MSTR fuel cycle

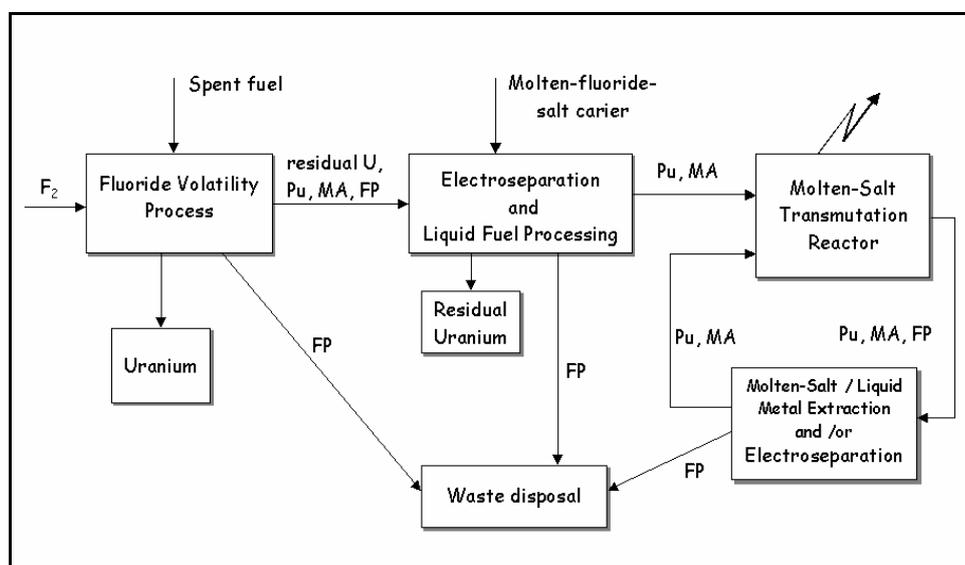
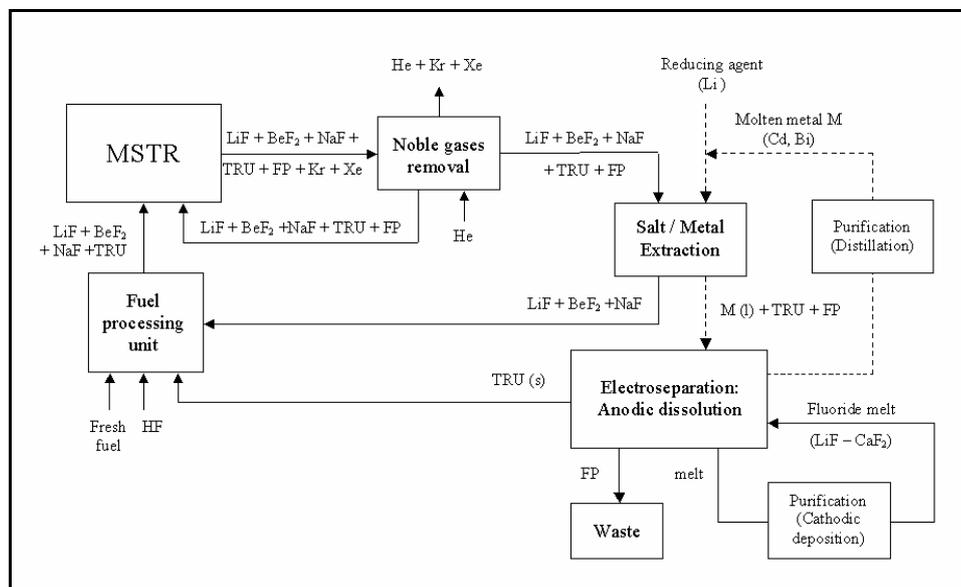


Fig. 3: Conceptual flow-sheet of MSTR on-line reprocessing (TRU burner without fertile material, multiple recycling)



The activities in the area of transmutation are oriented to the development of modified computer codes for calculations of neutronic characteristics (including time behavior) of the Molten-Salt Reactor systems taking into account the specific features of circulating liquid fuel based on molten fluorides. The developed code OGAR calculates the evolution of fuel composition during the burnout and permits to implement different fuel feed and reprocessing strategies. Burnout calculations are done in two steps. First, the neutronic cross sections are evaluated using a popular Monte Carlo MCNP code. These cross sections are then inserted in a rate equation and the fuel evolution is calculated. The change of cross sections with the fuel composition evolution is controlled and the cross sections are recalculated if necessary. The code is mainly used for optimization of reactor core geometry, to study neutronic properties of different carrier salts and to study the influence of different reprocessing strategies.

The theoretical program is supplemented by experimental prediction of neutronic characteristics and other corresponding characteristics (including time behavior) both on experimental zero power experimental reactor VR-1 at the Technical University in Prague (by the methods of inserting zones and oscillation of samples as well as neutron sources) and on the LVR-15 research reactor at the Nuclear Research Institute Rez plc (by irradiation of samples of the transmuter core in high neutron fluxes) performed for validation of the modified computer codes employed in core analyses of the reactor systems in question. The main effort represents the experimental test program of instrumented BLANKA probes (Fig. 4 and Fig 5). [3]

The main aims of the BLANKA program are:

- Experimental verification of long time behavior of transmuter blanket which contains molten fluoride salts as a fuel and graphite as a moderator or reflector
- Material research – behavior of materials in neutron and gamma fields, and materials interactions on high temperature conditions
- Validation of computational code

All the irradiated samples of individual compositions have been precisely analyzed afterwards and both the methods and techniques are currently being adjusted to the specific demands of the proposed experimental program and its efficient fulfillment and reaching its main aims.

Figure 4: The scheme of the BLANKA probe

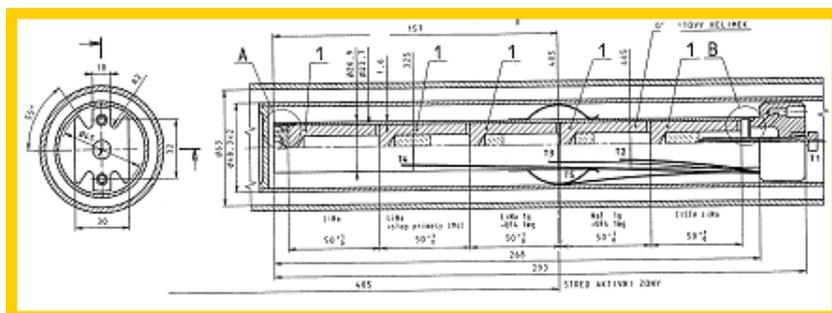
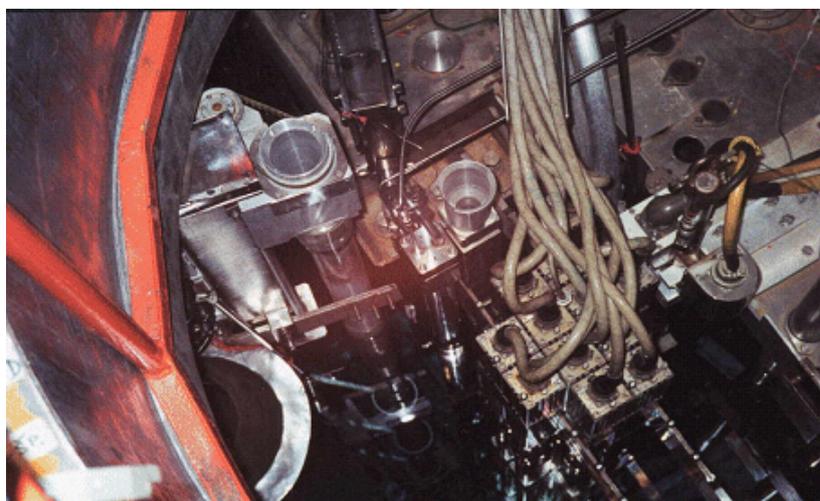


Figure 5: View of BLANKA probe in the LVR-15 reactor



Development and verification of structural materials for molten fluoride salt media is focused mainly to the further development of special nickel superalloy MONICR experimentally produced by SKODA Nuclear Machinery company. The verification program covers the production of several MONICR sheets, tubes and other samples, corrosion and irradiation tests and a machinability of the material. The development of special equipment for MSR technology and fluoride partitioning technology has been focused mainly to the design and construction of pumps (impellers) for molten fluoride salt medium (Fig. 6) and to the design and construction of apparatuses for fluoride volatility technology for fluorine gas medium and for design and manufacture of measuring components for electroseparation processes.

Fig 6: View on EVM-4-MS impeller of the molten-salt test loop



Conclusion

The major part of the P&T program in the Czech Republic is performed as a component of the SPHINX project supported by the Ministry of Industry and Trade of the Czech Republic. The project SPHINX (**SPent Hot fuel Incineration by Neutron fluX**) covers the R&D of selected parts of MSR technology including MSR fuel cycle technology. The solvers of the project, grouped within the “Transmutation Consortium”, are the Nuclear Research Institute Rez plc held by the Czech Power Company CEZ, SKODA Nuclear Machinery, Energovyzkum Brno Ltd and the Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University Prague. The complementary support of Czech P&T program comes from the Czech Nuclear Account controlled by the Radioactive Waste Repository Authority and from other domestic sources. Significant importance plays also the participation of several Czech institutions in projects of 5th and 6th Framework Programme of EC/EURATOM, mainly in the projects PYROREP, EUROPART and MOST. The R&D work performed in the frame of these projects by Czech institutions is in a good agreement with the Czech national program on P&T and therefore the international co-operation contributes reciprocally to the faster problem solving of the fuel cycle back-end.

References

1. M. Marecek, F. Lisy and J. Uhlir, “*Technological development of fluoride volatility method for front-end of molten salt transmutation reactor fuel cycle*”, Proc. of “InWor for P&T and ADS 2003”, Mol, Belgium, October 6 – 8, 2003
2. R. Zvejskova, K. Chuchvalcova Bimova, F. Lisy, P. Soucek “*Study of Electrochemical Processes for Separation of the Actinides and Lanthanides in Molten Fluoride Media*”, OECD/NEA “8th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation”, Las Vegas, Nevada, U.S.A., November 9-11, 2004
3. M. Hron, M. Mikisek, “*Czech Experimental Program for Molten Salt Transmuter SPHINX*”, IAEA Meeting “Perform an Assessment of Accelerator Driven Systems (ADS) Dynamics and Safety Physics”, Rez, Czech Republic, September 29 to October 1, 2003.