# R&D OF PYROCHEMICAL PARTITIONING IN THE CZECH REPUBLIC

**Jan Uhlir**
Nuclear Research Institute Řež plc
250 68 - Řež, Czech Republic

## Abstract

The Czech national research and development programme in the area of "Pyrochemical Partitioning" is directed primarily on the development of the "front-end" part of the fuel cycle technology for the molten salt reactor systems with a liquid fuel based on fluoride melts. The present research is directed particularly on the development of suitable fluoride separation technology based on "fluoride volatility method" the target of which is the removal of the uranium component from spent nuclear fuel and on the research of the electroseparation procedures and further on the development of appropriate construction materials and equipment for the fluoride molten salt technologies.

## 1. Introduction

Nuclear waste, especially spent fuel from nuclear reactors containing long-lived radionuclides represented mainly by actinides and long-lived fission products, presents a common problem for all countries operating nuclear power plants. Satisfactory solution of this problem is a factor limiting to a considerable extent the further nuclear power industry development, namely in developed countries. The present technical and technological sum of knowledge indicates that this problem could be largely resolved by using the transmutation technology so that the nuclear power industry may become widely acceptable for the public.

The use of transmutation reactors with fluoride salts based liquid fuel might be one of the possible answers. In the near future, these systems might be conceived as critical ones and successively as sub-critical accelerator driven reactor systems. In addition to the nuclear burning of plutonium and minor actinides produced in the U-Pu cycle these reactor systems might consecutively operate within the U-Th cycle as well.

The advantage of the molten salt transmutation reactors (MSTR) demonstrates itself above all in connection with a continuous or at least quasi-continuous chemical separation process.

For such a compact coupling of MSTR with chemical reprocessing it will be very appropriate to keep fuel in one chemical form, as far as possible, in the course of the entire fuel cycle. Accordingly, if the MSTR fuel will be based on fluoride melt then the separation processes should also be based on separation techniques from fluoride melt media. Pyrochemical and pyrometallurgical technologies comply generally with this requirement [1].

## 2. Czech research and development programme

The Czech research and development programme in the field of pyrochemical and pyrometallurgical separation is based first of all on the experience acquired in the past in the development and realisation of a pilot-plant fluoride technology for the reprocessing of spent fuel from the Russian BOR-60 fast reactor [2]. At present, this experience is utilised for the development of suitable separation processes and technologies for the fluoride based MSTR fuel cycle.

Experimental and theoretical studies in the field of pyrochemical technology development for the MSTR fuel cycle are oriented in particular to the following areas:

- Technological research in the field of the “Fluoride volatility” method directed at the suitability verification of a technology for thermal or fast reactor 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 MSTR. Consequently, the objective is a separation of a maximum fraction of uranium component from Pu, minor actinides and fission products. Integral part of this research also is the flowsheeting research – working out a proposal for a suitable technological flowsheet for treating spent fuel into a form fitted for MSTR including the separation procedures before transmutation (front-end) and separation processes after passage of fuel through the transmutor (back-end).
- Research on material and equipment for fluoride salts media connected with the experimental programme on ADETTE technological loops. Objective of the programme is verification of the new developed construction material for fluoride melts, development of selected devices

(first of all pumps) for fluoride melts and acquirement of practice in fluoride melts handling and manipulation in greater amounts.

- Laboratory research on electro-separation methods in fluoride melts media in relation to the study of their properties. The effort in this field is aimed first of all to the determination of optimum conditions for uranium and fission product separations and to the selection of suitable electrolyte composition based on fluoride salt mixture.

At present, the greatest attention is aimed to the realisation of the pilot-plant technology for the separation of the uranium component from other spent fuels components by the modified "Fluoride volatility" method. The other two areas mentioned are also intensively studied.

## 3. Technological research in the area of "fluoride volatility" method

Technological research of the "fluoride volatility" method can be ranked into the "front-end" area within the MSTR fuel cycle. It is a separation process for reprocessing spent fuel from thermal or fast reactors into a form suitable for the MSTR.

The main operations of this process are:

- Removal of the cladding material from spent fuel.
- Conversion of the fuel into a powder form (oxides) of a granulometric composition suitable for the fluorination reaction.
- Fluorination of the fuel (the purpose of this operation is the separation of the uranium component from plutonium, minor actinides and most of fission products).
- Uranium component purification.

The essential research activities are centred upon the technological verification of items 3 and 4 leading to the removal of the main portion of the uranium component from spent fuel. The proposed technology is based on the spent fuel fluorination with gaseous fluorine in a flame fluorination reactor, where the volatile fluorides are separated from the non-volatile ones, and on the subsequent purification of the component by using technological operations of condensation and distillation and also of sorption. As the fluorination reaction is suggested as a flame reaction, the necessary size of the flame fluorination reactor (Figure 1) is a critical parameter influencing dimensions of the entire experimental technological line.

According to the process design, up to 95-99% of uranium in the form of volatile $UF_6$ will be removed from the non-volatile fluorides of plutonium ($PuF_4$), minor actinides and majority of fission products. In this way, the component representing the greatest share of the spent fuel will be removed.

The technological research is closely associated with the "flowsheeting research", the aim of which is also to act as an unifying framework of the individual research activities, in addition to the working out of the technological flowsheet. In Figure 2 a simplified flowsheet is presented of the designed MSTR fuel cycle, the "front-end" of which is based on the "Fluoride volatility" method.

Figure 1. **Experimental flame fluorination reactor**

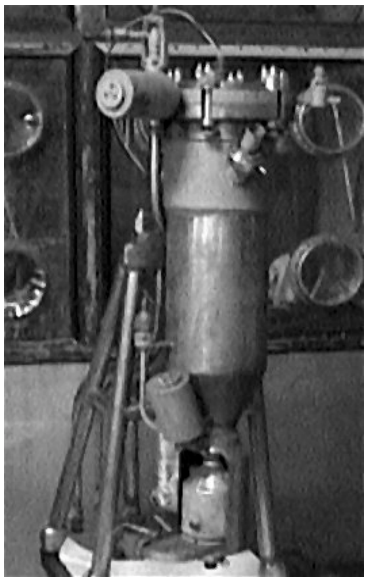
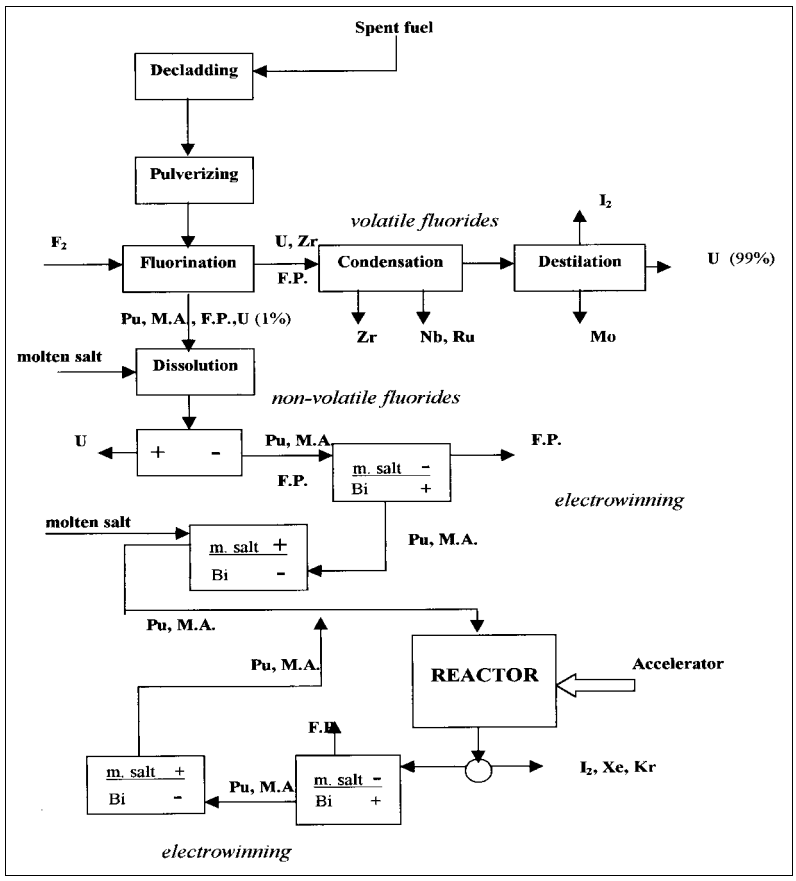


Figure 2. **Simplified scheme of fuel cycle of MSTR**



## 4. Research on material and equipment for fluoride technologies

Development of material and equipment for molten fluoride salts based technologies is connected first of all with the ADETTE technological loop programme [3] (Figure 3). The main objectives of the ADETTE loops experiments are:

- Testing of construction materials – corrosion research including the stress corrosion.
- Testing of welds.
- Research of fluoride melts thermohydraulics.
- Testing of pumps and valves for fluoride melt medium.
- Testing of measuring sensors and of the methods of measurement and control.
- Collection of data for the development and design of apparatuses for ADS technology with fluoride melts.

Special attention has been paid to the testing of the new corrosion resistant alloy MONICR SKODA. This high nickel content alloy is designated as a structural material for fluoride experimental loops for the operation temperatures of approx. 700$^{o}$C.

The MONICR material is also intended for the construction of specific components of the loops like molten salt pump (impeller vertical pump with a flange-mounted electric motor), control and closing valves, molten salt storage tanks, heat exchangers etc.

Figure 3. **Experimental molten salt loop ADETTE-0**

## 5. Laboratory research on electroseparation methods

Laboratory research in the area of electro-separation methods is directed first of all at the determination of optimum conditions for residual uranium and fission product separation from fluoride melt and further on the selection of a suitable composition of the electrolyte based on fluoride salts mixture. As the fluoride melt should be able to dissolve sufficient amounts of plutonium and minor actinide elements, mixtures of LiF–NaF and LiF–NaF–KF type are in the foreground of interest. The assumed ability of sodium and potassium fluorides to form co-ordination compounds with transuranium element fluorides and so to significantly increase their solubility in the melt was the reason for choosing the mixtures mentioned.

The research programme in this area is further directed to the determination and study of selected physicochemical properties of fluoride melts, particularly to the:

- Solubility of lanthanides in molten fluorides.
- Standard redox potentials of individual elements.
- Melting points of the molten salt mixtures.
- Density and viscosity of the supporting fuel matrices.
- Data on corrosion resistance of structural materials.

## 6. Conclusion

Research and development in the area of “Pyrochemical Partitioning” is carried out in the Czech Republic as a component of the national P&T programme within the framework of the “Transmutation” consortium. The programme is funded first of all by the Ministry of Industry and Trade and by the Radioactive Waste Repository Agency. The participants of the programme are, in addition to the NRI Řež, SKODA Nuclear Machinery, Energovyzkum Brno, Nuclear Physics Institute of the Czech Academy of Sciences and Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University Prague.

Experimental research work related to the development of pyrochemical technologies is concentrated mainly in the Fluorine Chemistry Department of the NRI Řež.

The Czech national conception in the area of P&T research issues from the national power industry programme and from the Czech Power Company intentions of the extensive utilisation of nuclear power in our country. The Czech Republic, as a relatively small country, has an understandable interest in a wide integration into the solution of problems associated with spent nuclear fuel in the international context, first of all in co-operation with the EU member countries. Involvement into the 5th Framework Programme EC is an evidence of it, viz. in the area of “Pyrometallurgical Partitioning” where, for example, the PYROREP project is complementing very well with the research activities within the national programme framework.

## REFERENCES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