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Research Article

Conceptual potential of a pyroelectrochemical technology for the thorium engagement in the fast neutron fuel cycle*

Valery S. Naumov[2]

1 *National Research Nuclear University «MEPhI» (Moscow Engineering Physics Institute), 31 Kashirskoe sh., Moscow, 115409 Russia*

Corresponding author: *Valery S. Naumov* (valnau387@yandex.ru)

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Abstract

The use of thorium in combination with plutonium in nuclear power generation offers a solution to the problem of reducing the accumulation of long-lived transplutonium nuclides. Along with this, the existing uranium fuel cycle (UFC) has such disadvantage as the vulnerability to unauthorized use of nuclear materials. The thorium fuel cycle (TFC) is devoid of these drawbacks.

The engagement of thorium in nuclear power is possible provided the availability of an appropriate technology for reprocessing irradiated thorium. A fuel cycle based on thorium oxide may not differ in principle from the already developed pyrochemical fuel cycle involving uranium and plutonium oxides. Thorium oxide is most commonly obtained in compact state by electrolysis of molten salts from thorium-containing electrolytes. The most thorough studies of physical and chemical and electrochemical behavior of thorium in molten haloids of alkali and alkaline-earth metals were conducted in the 1960ies and the 1970ies.

Since extensive experimental material has been accumulated by now for justification of the use of pyroelectrochemical and chemical processes for regeneration of fuel in molten salts, then it has also been proposed that technologies for fuel reprocessing in molten chlorides of alkali metals should be applied resulting in a crystalline product that can be used for the fuel element fabrication.

Unlike uranium and plutonium, thorium behavior in molten salt environments is less complex. In molten salts, thorium exists predominantly in the form of Th^{4+}, and the mixture of uranium and thorium dioxides with ThO_2 content reaching up to 50 % can be obtained by electrolysis of molten salts.

Therefore, the existing amount of knowledge about the chemistry of thorium allows regarding the use of pyrochemical processes in production of thorium oxide as highly promising, and the available data on the physical and chemical properties of thorium and its compounds in high-temperature molten salts makes it possible to state that the pyroelectrochemical technology can be potentially used in production and reprocessing of thorium fuel.

Keywords

Thorium; oxide fuel; thorium fuel cycle; options; pyroelectrochemical processes; electrochemical behavior of thorium; fuel reprocessing

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Introduction

Implementation of thorium-based nuclear fuel cycle for nuclear power generation (NPG) is important both from the viewpoint of the general concept of development of nuclear power generation and the development of specific types of fuel cycles as applied to this or that type of already existing and/or currently developed nuclear reactor types.

The issue of development of dedicated reactors for incineration of transplutonium elements (TPE) including plutonium emerged in the conditions of ever expanding involvement of available inventories of plutonium (weapons plutonium during the last four – five years). As the result, the issue of involvement of thorium in nuclear fuel cycle is not particularly urgent from the viewpoint of provision of nuclear raw materials. At the same time, examination of potential use of thorium seems to be quite logical within the framework of the emerging trend in nuclear reactor development. Application of thorium in combination with plutonium resolves such important problems as the reduction of accumulation of long-lived transplutonium nuclides in nuclear fuel, enhancement of environmental safety and reliability of the nuclear fuel cycle. The concept of thorium fuel cycle deployment is configured depending on the degree of elaboration and real possibility of implementation of thorium fuel cycle in this or that reactor type (Murogov et al. 1983).

Russian studies of thorium fuel cycle are mainly conducted at the level of conceptual studies and laboratory experiments. At the same time, significant scope of research of nuclear data for thorium fuel cycle has been implemented at the level of laboratory studies including the following: neutronics of nuclear reactor with thorium-containing fuel loads, irradiation and analysis of obtained samples using physical and radiochemical methods. Here, investigation of irradiated samples of thorium also allowed obtaining the technological experience of thorium reprocessing and uranium-232 handling (Kang and Von Hippel 2001).

Special attention paid to thorium fuel cycle is further motivated by the factors which are maybe important to a different degree for different countries, but the general interest (possibly, not all-encompassing) includes the following:

- Expansion of the base of fuel resources for nuclear power generation; advancement of different types of nuclear reactors with respect to the efficiency of fuel use, safety, and stability against diversion of nuclear materials;
- Possibility of creation of flexible fuel cycles with involvement of all fissile and fertile materials.

Favorable situation has evolved at present encouraging the search for additional approaches to deployment and use of thorium in NPG. Contemporary status of NPG development temporarily removed the urgency of the problem of exhaustion of uranium fuel resources. Or, to put it more accurately, more time is available now for resolving this problem than it was originally expected. Research can be implemented without any rush during the available time period and with acceptable intensity of investment of funds allocated for the solution of fundamental problems of NPG for the purpose of creation of the structure of optimal NPG fuel cycle with involvement of uranium, thorium, accelerator-driven and fusion neutron sources and resolving the problem of development of NPG fuel cycle producing no actinide radioactive wastes (Alekseev et al. 1999).

Since thorium fuel was never fabricated and reprocessed in industrial conditions, there exists some uncertainty with respect to its practical implementation, especially during reprocessing SNF using pyroelectrochemical technologies. Nuclear power generation is based in many countries on the uranium fuel cycle (UFC), i.e. on the use of uranium fuel. The most developed countries achieved significant results in covering the energy needs of significant fraction of consumers due to the incineration of uranium-235 from natural resources in thermal nuclear reactors.

However, the existing UFC has the following significant shortcomings:

- Low inherent protection of the fuel cycle from unauthorized use of nuclear materials;
- Accumulation in the process of power generation of long-lived fission products (FP) and minor actinides (MA).

The above considerations cause public concern because they slow down the progress of nuclear power generation.

With transition to thorium fuel cycle (ThFC) nuclear power generation will not have these shortcomings. High degree of self-protection of ThFC from unauthorized use of fissile materials is explained by the presence in the fuel, along with U-233, of U-232 isotope. This makes the fuel not applicable for military purposes mostly due to its high radiotoxicity.

Thanks to the absence in the fresh fuel of U-238 MA accumulation in ThFC is at a lower level as compared with fuel cycle with uranium fuel. This property of ThFC must be used for eliminating long-lived FP and MA accumulated within the UFC and, potentially, for eliminating negative consequences of the use of UFC and of nuclear power generation as a whole.

Thus, the situation has evolved at present when all the necessary prerequisites are in place for starting ThFC for the purposes of both involvement of considerable thorium resources in the fuel cycle and for the reduction of environmental hazard of nuclear power generation. Deployment of thorium in nuclear fuel cycle (NFC) must be aimed at the harmonious enhancement of efficiency of nuclear power generation.

Because of the absence in the nature of uranium-233 use of thorium in power generation requires closed fuel cycle. During initial phases of development of ThFC it is advisable to use the following two compositions of nuclear materials as the nuclear fuel:

- Th-^{235}U (possibly with addition of MA);

- Th-^{239}Pu (possibly with addition of MA which may be loaded in the reactor in the form of oxide, nitride or carbide mixtures, metal alloys, as well as melts of halogenides of alkali and/or alkaline-earth metals.

Therefore, the purpose of the present study was to implement analysis based on the available data on physical and chemical properties of thorium and to provide substantiation of the possibility of involvement of thorium in fuel cycle of fast reactor with obtaining oxide fuel using pyroelectrochemical technology of fuel reprocessing.

Prerequisites for pyroelectrochemics production of thorium oxide

U-233 extracted from spent nuclear fuel will be used for gradual transition to the Th - U-233 fuel composition inherent to ThFC to which plutonium, long-lived FP and MA will be added.

Experience of development and economic appraisal by ANL (USA) (Chang 1989), SSC RF-RIAR (Russia) (Skiba et al. 1993), PNC and CRIEPI (Japan) (Inoue and Tanaka 1997) of technologies based on non-aqueous pyrochemical processes demonstrated that these processes are more cost-effective because they involve less number of operations, they are devoid of the problem of radiolysis of working solutions, and the wastes in these processes are predominantly generated in solid compact form. At the same time "dry" technologies guarantee non-proliferation of nuclear materials, because they ensure purification of nuclear materials from fission radionuclides by not more than two – three orders of magnitude.

In our opinion, it is advisable in the development of ThFC to make use of the latest developments in non-aqueous technologies with full implementation of the advantages offered by high-temperature processes.

Molten-salt high-temperature processes of nuclear fuel fabrication and reprocessing must be attributed to the most thoroughly studied and tested in practice (USA (Chang 1989), Russia (Skiba et al. 1993), Japan (Inoue and Tanaka 1997)).

Salt melts as the media for pyrochemical and pyrometallurgical methods of spent nuclear fuel reprocessing possess the following advantages:

- High radiation resistance allowing reprocessing fuel with burnup of 100 MW×day/kg and more;
- High thermal stability allowing reprocessing fuel with specific heat release capacity in excess of more than 1 MW/kg and decay time of less than one year;
- It is possible to perform operations with salt melts with high concentrations of fissile materials, because salts applied in fuel reprocessing are not neutron moderators;
- Chemical processes in molten salt environment proceed at high rates and practically do not limit the kinetics of technological operations.

Due to the above listed advantageous properties of salt melts pyrometallurgical and pyrochemical processes attain the following significant positive features:

- Simplicity of technological process layouts and compactness of equipment;
- Minimization and compactness of radioactive wastes;
- Fire and explosion safety.

The widest practical experience of use of molten-salt systems was gained in Russia and, in particular, at the SSC RF-RIAR with participation of institutes of the Russian Academy of Sciences during the development and implementation of pyroelectrochemical technology of fabrication and reprocessing of uranium and mixed uranium-plutonium oxide fuel (MOX-fuel) for fast reactors. Extensive experimental studies of behavior of uranium, plutonium, fission radionuclides and structural materials in molten halogenides of alkali and alkaline-earth metals (first of all molten chlorides) were performed in the process of development and physical and chemical substantiation of the suggested technology.

Studies of thorium behavior in non-aqueous media and, in particular, in molten salt electrolytes were performed in Russia and in some foreign countries. However, as a whole, the level of investigation of thorium chemistry in such electrolytes is less thorough than uranium or plutonium chemistry. Information about physical and chemical and electrochemical behavior of thorium in molten halogenides of alkali and alkaline-earth metals is to a considerable extent based on the studies implemented during the 1950-ies – 1970-ies (Smirnov and Yudina 1959, Smirnov et al. 1970, Smirnov et al. 1976, Kudyakov et al. 1968, Kudyakov et al. 1972, Emelyanov and Evstyukhin 1956a, Emelyanov and Evstyukhin 1956b, Vokhmyakov et al. 1973, Smirnov et al. 1978, Chiotti et al. 1975).

The following options of fuel cycles are addressed in the estimations of the potential of thorium application in nuclear power generation:

- uranium-uranium ($^{235}UO_2$ - $^{233}UO_2$ fuel);
- uranium-thorium ($^{235}UO_2$ - ThO_2 fuel);
- uranium-thorium (U - Th alloy).

Since all the above fuel compositions have high melting points it is logical enough to examine the possibilities of their obtaining by pyrochemical methods, for instance, in molten salt systems.

Pyroelectrochemical reprocessing using molten salts is applied both to oxide fuel (SSC RF-RIAR, Russia) and to metal fuel (Argonne National Laboratory, USA). Such efforts are implemented during already long enough period with conducting experimental studies for substantiating the fuel cycle based on the use of the following elements:

- "Dry" pyroelectrochemical reprocessing of irradiated fuel with its conversion to the form applicable for recycling;
- Use of fuel elements with vibropacked fuel;
- Organization of recycling processes using the simplest method and remotely controlled production lines.

The following properties of components of irradiated fuel must be taken into consideration in the development of pyrochemical process for obtaining oxide thorium fuel or mixed oxide thorium fuel with addition of uranium and plutonium:

- Uranium ions U(III), U(IV), U(V), U(VI) are equally stable in molted chlorides of alkali and alkaline-earth metals; plutonium ions in highest oxidation states Pu(V) and Pu(VI), as well as Th(IV) and Th(II) are stable within certain area of ratios of oxidation and reduction potentials;
- Specific electric conductivity of ThO_2, as well as of UO_2 - ThO_2 at working temperatures suitable for their possible extraction from molten salts (600 – 750°C) is significantly lower than that for uranium and plutonium oxides.

Therefore, it is important to know the conditions of crystallization and electrolytic deposition of oxygen containing forms with extraction of thorium and plutonium or of thorium and uranium dioxides in the development of technological processes of fabrication of oxide fuel. Evaluation of thermodynamic parameters of the process can be performed according to the following reactions:

$MeCl_n + n/4\ O_2(gas) \rightarrow MeO_{n/2}(solid) + n/2\ Cl_2(gas)$,

$MeCl_n + l/2\ O_2(gas) \rightarrow MeO_l Cl_m(solid) + 0.5(n - m)\ Cl_2(gas)$.

Possibility of obtaining oxide fuel on the basis of ThO_2 in molten salts

There exist a number of publications on the investigation of physical and chemical and electrochemical behavior of thorium in molten halogenides of alkali metals (Smirnov et al. 1970, Smirnov et al. 1976). Wide range of quantitative regularities in the variations of valence states of thorium and its electrochemical behavior were established on the basis of experimental data (Kudyakov et al. 1968, Kudyakov et al. 1972).

However, in contrast to uranium and plutonium, thorium behavior in molten salt media is more straightforward.

The following must be attributed to the most important aspects of thorium chemistry:

- Thorium exists in salt electrolytes predominantly in the form of Th^{4+}, Th^{2+}form and is produced in equilibrium with metal; oxygen-containing forms of thorium soluble in molten halogenides are not present in the technology in question in significant quantities;
- Electric conductivity of thorium dioxide is lower by $10^3 – 10^4$ times within the temperature range from 600 to 1000°C than that for uranium dioxide;
- Reduction of $ThCl_4$, ThF_4 salts and ThO_2 dioxide to thorium metal is possible only with the help of active metal, for the most part of calcium;
- Thorium metal in compact form with removal of impurities can be obtained by electrolysis of molten salts;
- Thorium dioxide interacts with oxides of different metals; with uranium dioxides it forms a continuous series of solid solutions; state transition diagrams can be found in reference literature for UO_2 - ThO_2 and UO_2 - ThO_2 - ZrO_2 systems (Voronov et al. 1971);
- Thorium dioxide can be produced by deposition from molten haloids by carbonates of alkali metals, oxides of alkali and alkaline-earth metals or by oxygen-containing gas mixture;
- Mixture of uranium and thorium dioxides with thorium concentration up to 50 % can be obtained by electrolysis of molten salts; purification from the main mass of rare-earth elements (neutron poisons) can be achieved in the process.

It is known that in electrolytic process of sedimentation of UO_2 - ThO_2 cathode deposits from molten LiCl - KCl electrolyte and improvement of physical and chemical properties of fuel, in particular, increasing fuel density, it is preferable to arrange in the molten medium of exchange reaction of the following type:

$$UO_{2\,т} + YMCl_{4(molt.)} = YMO_2(1 - Y)UO_{2(s.s)} + YMCl_{4(molt.)}.$$

Information about thorium potentials in salt solutions available in the references (Smirnov and Yudina 1959, Smirnov et al. 1970, Smirnov et al. 1976) is important for extraction of thorium on cathode in the fuel regeneration processes. Thorium equilibrium electrolytic potentials in molten electrolyte containing 11.2 mol. % of thorium cited in Ref. (Smirnov and Yudina 1959) are described within the temperature range of 653 – 800 K relative to chlorine electrode by the empirical equation $E = –3{,}092 + 6{,}5\times10^{-4}\ T \pm 0{,}01$ V. These results are in good agreement with observations and regularities of electrolytic deposition of cathode products (metals and uranium dioxide) formulated in (Baraboshkin 1976, Baraboshkin et al. 1971).

Systems containing three and more components can be used as the fuel along with two-component solid solutions (Kotelnikov et al. 1978). However, the data on their electrolytic extraction from salt solutions are not available.

Investigation and experimental studies on the substantiation of electrochemical technology for obtaining uranium and mixed uranium-plutonium fuel for fast reactors in chloride solutions are practically completed in Russia. The fuel cycle technology for production of thorium oxide fuel can also be based on the pyroelectrochemical production of thorium dioxide in molten chlorides according to the following two mutually complementary processes:

- Pyroelectrochemical fabrication and reprocessing of MOX-fuel;
- Fabrication of fuel elements by the vibropacking method.

While the electrochemical process of production of individual uranium and plutonium dioxides, as well as of their homogenized mixture with plutonium concentrati-

on up to 45% allows ensuring production of polydisperse crystalline fuel powders with density of granules close to the theoretical value, the question whether electrolytic depositions of UO_2 - ThO_2 with preset composition and preset density can be obtained from molten chlorides remains not clear. Also, information about the structure of UO_2 - ThO_2 electrolytic depositions and specific features of their production on the cathode is missing.

As a whole, investigated processes of electrochemical deposition of UO_2 or ThO_2 fuel allow already now discussing the possibility in principle of reprocessing thorium fuel using molten salts and incorporating such operation in the technological layout of thorium fuel regeneration.

Similarly to the fabrication and reprocessing of uranium or mixed uranium plutonium oxide fuel, the technology can include the following stages:

- Separation of fuel from cladding can be achieved both using the existing aqueous radiochemistry methods based of the cutting-and-leaching method, and using already well-enough elaborated pyrochemical methods with application of non-aqueous reagents (liquid metals or molten salts or combinations thereof);
- Dissolution in molten salts and performing in the same equipment of subsequent operations – extraction of noble FP, electrolysis of UO_2 or ThO_2 and concentration of FPs;
- Crushing oxides after their extraction, vacuum processing and preparation for vibropacking;
- Vibropacking of oxide fuel in fuel elements.

Knowledge of physical and chemical properties of fissile materials and FP, as well as experience of mastering MOX-fuel fabrication processes allow already at present presenting in general terms the possible technological reprocessing or the technology of pyroelectrochemical regeneration of thorium fuel, as well as defining those scientific and technological studies which are required for their development:

- Investigation of physical and chemical properties of thorium oxide deposition from molten salts;
- Investigation of co-deposition of thorium and plutonium oxides from molten salts;
- Investigation of behavior of fission products in molten salts containing thorium.

Electrochemical regeneration process provides the possibility to restore physical mechanical and nuclear physics properties of fuel with the final product of reprocessing – regenerated nuclear fuel – obtained in the granulated form applicable for charging fuel elements.

Taking into account the potential capabilities of thorium fuel cycle and the duration of stages of implementation of new fuel technologies it is recommended to continue R&D efforts for selecting optimal use of thorium both in already operated and in future advanced nuclear reactors. Already during the current stage of scientific research of the multi-component structure of nuclear power generation for the development of future technologies it is advisable to use the “cross-coupled” closed fuel cycle with fuel exchange between uranium thermal reactors and plutonium fast reactors with thorium blankets.

In such fuel cycle fast reactors with breeding ratio in the reactor core » 1 will, while maintaining plutonium fuel in the core, breed U-233 in the thorium blankets for starting up and re-loading uranium thermal reactors operated on uranium fuel made of the mixture of U-233 and U-238 isotopes accumulating plutonium for starting up new fast reactors. Besides the above, the problem of protactinium poisoning of thermal reactors taking place in thorium-uranium fuel cycle is eliminated (Alekseev et al. 1999).

Conclusion

It can be noted that all the required prerequisites for commencing the development of pyroelectrochemical technologies (or certain reprocessing operations) for fabrication and regeneration of thorium-based fuel compositions are in place, namely:

- Sufficient knowledge of physical and chemical properties of thorium and its compounds in a number of the most important solvents within wide temperature range is available;
- Thorough knowledge of physical and chemical properties of practically all components of fuel within ThFC including actinides and fission products is available;
- There exists vast experience of development of high-temperature processes of fabrication and regeneration of MOX-fuel for fast reactors;
- There exists vast experience of development of automated and remotely operated and maintained equipment for production and regeneration of fast reactor MOX-fuel, as well as for fabrication of fuel elements and fuel assemblies by the method of vibropacking of powders.

This potential must be proactively used in the implementation of thorium fuel cycle in the structure of nuclear power generation.

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