Management of Spent Fuel from Nuclear Power Reactors

Learning from the Past, Enabling the Future

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MANAGEMENT OF SPENT FUEL FROM NUCLEAR POWER REACTORS

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Marketing and Sales Unit, Publishing Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
1400 Vienna, Austria
fax: +43 1 26007 22529
tel.: +43 1 2600 22417
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Session 4.1: Recycling as a spent fuel management option

Session Chairs: C. Roussel (France) and A. Kirkin (Russian Federation)

Session 4.1 comprised of five presentations, two from France, two from Russian Federation and one from Japan.

- **Paper ID#127 by B. Morel (France)** presented an overview of the solutions brought by R&D programs to maintain a high level of reliability in La Hague reprocessing plant. Removing clogging from dissolution equipment, columns of extraction have been done thanks to basic rinsing operations. To avoid corrosion issues, metallic materials are carefully selected. Corrosion issues bring the development of tele manipulator very resistant to radiation to repair equipment in very hostile environment and of the cold crucible melter to vitrify the fission product solution rich in molybdenum.
- **Paper ID#53 *(Invited)* by A. Sheremetyev (Russian Federation)** presented the experience of Mayak Production Association. Various spent nuclear fuel can be treated thanks to the adaptation of facilities for VVER-1000, fast reactors spent nuclear fuel, or spent nuclear fuel from research reactor facilities or propulsion facilities and defective spent nuclear fuel. According to the author the line will be equipped with a heavy-duty cutting machine and a voloxidation unit that allows containing tritium.
- **Paper ID#33 by H. Asano (Japan)** presented the Japanese research programme which aims to optimize the fuel cycle conditions regarding the environmental load reduction of the geological disposal for high level radioactive waste. Fuel burnup, spent fuel cooling period, waste loading of vitrified waste and separation of minor actinides have been studied and compared to the present conditions and their impact on geological repository.
- **Paper ID#126 by P. Breitenstein (France)** presented a solution developed to manage damaged fuel assemblies: packaging, transport logistics, wet or dry storage, and reprocessing. These damaged fuel assemblies are from different types of reactors and from various countries. Various cask designs exist, and specific operations are developed to manage defective fuels to decrease global risk.
- **Paper ID#19 by A. Rodin (Russian Federation)** presented an approach based on an adaptation of existing methodology of explosion safety assessment considering the specificity of nuclear industry. The method could allow researchers to evaluate the safe conditions of chemical processes of nuclear fuel cycle facilities and it can be used as a basis for further safety requirements development.

Session 4.2: Recycling as a spent fuel management option

Session Chairs: A. Khaperskaya (Russian Federation) and Y. Guoan (China)

Session 4.2. comprised of five presentations, three from France and two from Russian Federation.

- **Paper ID#72 *(Invited)* by C. Delafoy (France)** presented the French experience in using MOX fuel in LWR and new approaches to Pu multi-recycling strategies in LWRs with the CORAIL and MIX fuel assembly. The paper also contents the adaptations to be implemented at the MELOX production plant to face the degradation of the Pu isotopic vector of MOX fuel and its higher Pu content. The CHROMOX product which involves Cr_2O_3 doping is characterized by an enhanced homogeneity of the Pu distribution in the fuel and an increased matrix grain size. With these evolutions, larger

TRACK 4: RECYCLING AS A SPENT FUEL MANAGEMENT OPTION

ID#127 La Hague reprocessing plant: a mature technology continuously enhancing its performances through industrialization of R&D programs
B. Morel, A. Salvatores

ID#53 Experience and prospects of spent nuclear fuel reprocessing at Mayak
A. Sheremetyev, D. Koulupaev

ID#33 Cross-sectoral study on nuclear energy system for less-impacted radioactive waste management. Effect of various spent nuclear fuel properties and reprocessing conditions on geological disposal
H. Asano, T. Okamura, E. Minari, M. Nakase, K. Takeshita

ID#126 Solution for management and disposal of multiple design defective fuel elements
P. Breitenstein, V. Cholot, V. Vo Van

ID#19 Reactive hazards during SNF reprocessing as possible causes of radiation accident
A. Rodin, A. Ponizov, E. Belova

ID#72 Plutonium recycling through LWR MOX fuel: Today and tomorrow
C. Delafoy, J. Jonnet, Y. Rugama, V. Garat

ID#55 Evaluation of technical and economic indicators of REMIX fuel application in the LWR fuel cycle
A.V. Grol, E.A. Bobrov, A.V. Gurin, V.A. Nevinitsa, A.Yu. Smirnov, P.S. Teplov, P.A. Fomichenko, N.D. Goletskiy, N.V. Kovalev, A.B. Sinyukhin, B.Ya. Zilberman

ID#92 An oxidizing digestion process applied to SFR MOX fuel recycling to recover plutonium and reduce solid residue volumes
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ID#94 Prospective inventory of radioactive materials and waste produced by the 327odell nuclear fleet according to different plutonium multiple recycling options in the frame of the 327odell law for waste management
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ID#29 Core 327odelling and source term calculations using MCNPX code for Fukushima Daiichi unit-1 nuclear power plant accident
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ID#16 A new design of a PWR fuel assembly for direct recycling of spent fuel
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ID#59 A new approach to nuclear fuel recycling for LWR REMIX fuel concept: Status and trends
N.V. Kovalev, N.D. Goletskiy, B.Ya. Zilberman, A.B. Sinyukhin

ID#155 Increasing plutonium disposition rate in the thermal reactors (VVER and RBMK)
S. Dawahrah

ID#13 Complete co-processing of spent fuel as a back end fuel cycle strategy
B.B. Acar, H.O. Zabunoglu

REACTIVE HAZARDS DURING SNF REPROCESSING AS POSSIBLE CAUSES OF RADIATION ACCIDENT

A. RODIN
Scientific and Engineering Centre for Nuclear and Radiation Safety
Moscow, Russian Federation
Email: rodin@secnrs.ru

A. PONIZOV
Scientific and Engineering Centre for Nuclear and Radiation Safety
Moscow, Russian Federation

E. Belova
A.N. Frumkin Institute of Physical chemistry and Electrochemistry
Moscow, Russian Federation

Abstract

The experience of nuclear fuel cycle facilities operation shows that explosions during reprocessing of radioactive material could lead to release of radioactive elements with consequences for environmental. The root cause of many of them is chemical interactions with heat and gas generation. Hazard identification methods have been developing for many years, but the specifics of the nuclear industry requires to adapt the approaches. The chemical processes are going to be used for solving the radioactive waste problems that means that the agreed by the scientific community approaches of the safety assessment should be developed.

1. INTRODUCTION

The development of nuclear energy is associated with solving spent nuclear fuel and waste problems. One of the option is the reprocessing of spent nuclear fuel including actinides recycling and partitioning of radioactive waste to minimize the amount or completely eliminate it with the implementation of the transmutation concept. Nowadays the most developed technology of radioactive elements partitioning is based on hydrometallurgical processes [1, 2].

Also non-water methods are researched, but an operating experience of prototypes is not enough to state that the required indicators will be achieved. However, the water methods of reprocessing include disadvantages related to safety. One of them is using of nitric acid and formed nitrates that are known as strong oxidants.

Modern radiochemical reprocessing of SNF and RW consists in the selective separation of the elements by using of extraction methods, sorption, and precipitation. Implementation of advanced fuel cycles is related to increasing the degree of separation required increasing of reprocessing steps and using of additional equipment and reactants. Used during reprocessing of waste and SNF chemicals could interact with nitric acid and nitrates and produce heat and gases. In some abnormal regimes of operation of nuclear fuel cycle facilities (NFCF) this interaction may lead to destroying of equipment and release of radionuclides, radiation accident.

The analysis of incidents [3] in NFCF shows that some of them were caused by interaction of organic components with solutions contained of nitrate ion.

For example:

— 12/01/1953 Savannah River Plant, USA – Chemical explosion at a TNX evaporator;
— 29/09/1957 Southern Urals, USSR – A tank filled with a highly active solution exploded;
— 20/11/1959 Thorex Pilot Plant, USA – A chemical explosion in an evaporator;
— 6/11/1963 Plutonium processing facility, Hanford, USA – Exothermic reactions in a plutonium-loaded anion exchange resin;
— 12/02/1975 Savannah River, USA Tributyl-phosphate and uranyl nitrate was thermally decomposed in a denitrator;
— 30/08/1976 Hanford, USA - Chemical reactions of nitric acid with cation ion-exchange resin;

- 6/04/1993 Tomsk-7, Russia – An exothermic chemical reaction between an organic compounds and concentrated nitric acid in the tank;
- 17/07/1993 Mayak, Russia - Thermal-chemical explosion at an ion-exchange column.

In addition, the consequences of the incidents had radiological impacts that means the chemical interactions should be taken into account as possible reasons of Design Basis Accidents or Beyond Design Basis Accidents.

Therefore the safety assessment of NFCF at which are possible such accidents should include solution of two main aim. One of them is to determine of technological parameters that could lead to explosion and other one is to predict of the radiological consequences that based on damage prognosis. Taking into account the tendency to increasing the depth of burnout, reducing the storage time of SNF and reprocessing of a fast reactor SNF, it is increasing of radiation effect on media lead to producing a large number of degradation products. The presence of this products complicates to safety assessment of NFCF.

2. SAFETY ASSESSMENT OF REACTIVE SUBSTANCES

According to the document [4] the type of chemical interaction between components that is not absorbed by the environment (equipment) and leads to explosions could be defined as result of reactive hazards of substances. The approaches of reactive hazards assessment of unstable chemicals have been developing for many years. Simplified theories include the analytical solutions and criteria such as were made by Semenov [5], Frank-Kamenetsky [6] or Todes [7]. But nowadays calculations based on finite element methods are more often used that is related to developing of computers techniques. All of this safety assessment approaches based on determination of conditions at which the rate of heat generation in an object begins to exceed the heat losses. The last one allows a safety specialist to create any reactor geometry and use more complicated chemical kinetic equations in comparison with simplified solutions at which all of that are strictly defined. Nevertheless most of the developed methods could be used after analysis of a specific case and choose of a safety factor on limiting parameter especially when simple kinetic models of chemical reactions describe well an observed heat generation. At the same time there are no any analytical solutions that could be used for prediction of temperature changes in technological media when it is necessary also to take into account radioactive heat sources. In some cases such features as intensive radioactive decay should not be ignored and it requires to use mathematical models with linear heat sources independed on temperature and also calculation by computers codes.

3. POSSIBLE APPROACH FOR NFCF

To ensure the safety assessment of typical hydrometallurgical processes for radioactive materials treatment specific guides are being developed. They include methods and criteria for making decision. One of them is «Fire- and explosion safety assessment of sorption systems for reprocessing SNF» [8]. The scheme of safety assessment is shown in Fig 1.

The implemented in the document approach is based on gradually decreasing of conservatism in assessment. On the one hand it assists to save resources when parameters of processes are far away from the critical points, but on the other it allows the safety specialist to set operational limits on acceptable level and justificate them. The scheme could be divided in five general parts.

The first essential part (1) is executing of analysis of the processes such as sorption, desorption, flushing for determination of potential hazardous chemicals. Firstly it is recommended to find the most dangers substance. It may be mixtures with highest concentration of reductants and oxidants, or heat generating elements, or the most degradeted one, or with lowest heat transfer coefficient or else. After the parameters such as adiabatic temperature (Tad) and volume of gaseous products (V) should be calculated by using some of conservative approach. It is needed to make a decision about acceptance of decomposition this substance by comparison of the parameters (P) with safety limits (SL) such as pressure of sorption column destruction or others. If it is acceptable there is possible to go to the last part of assessment (5) - analysis of deviations in the process. Such events as operator mistakes, equipment failure and other one including a complex of them should be considered on this step. Under failure condition of the sorption system safety of the processes with new initial parameters should be assessed from the first step. If on the first part the parameters of decomposition exceed safety limits the second one should be carried out.

The second part (2) is focused on determination of time to maximum rate of chemical decomposition under adiabatic condition (τad). In this part also an information about kinetic of chemical reactions occurred in

technological media is needed. There is essential step of assessment to provide the developing of mathematical kinetic model of chemical reactions and also that is one of the most complicated one. In the article [9] one way of model creation problem solving for ion-exchange resin was demonstrated.

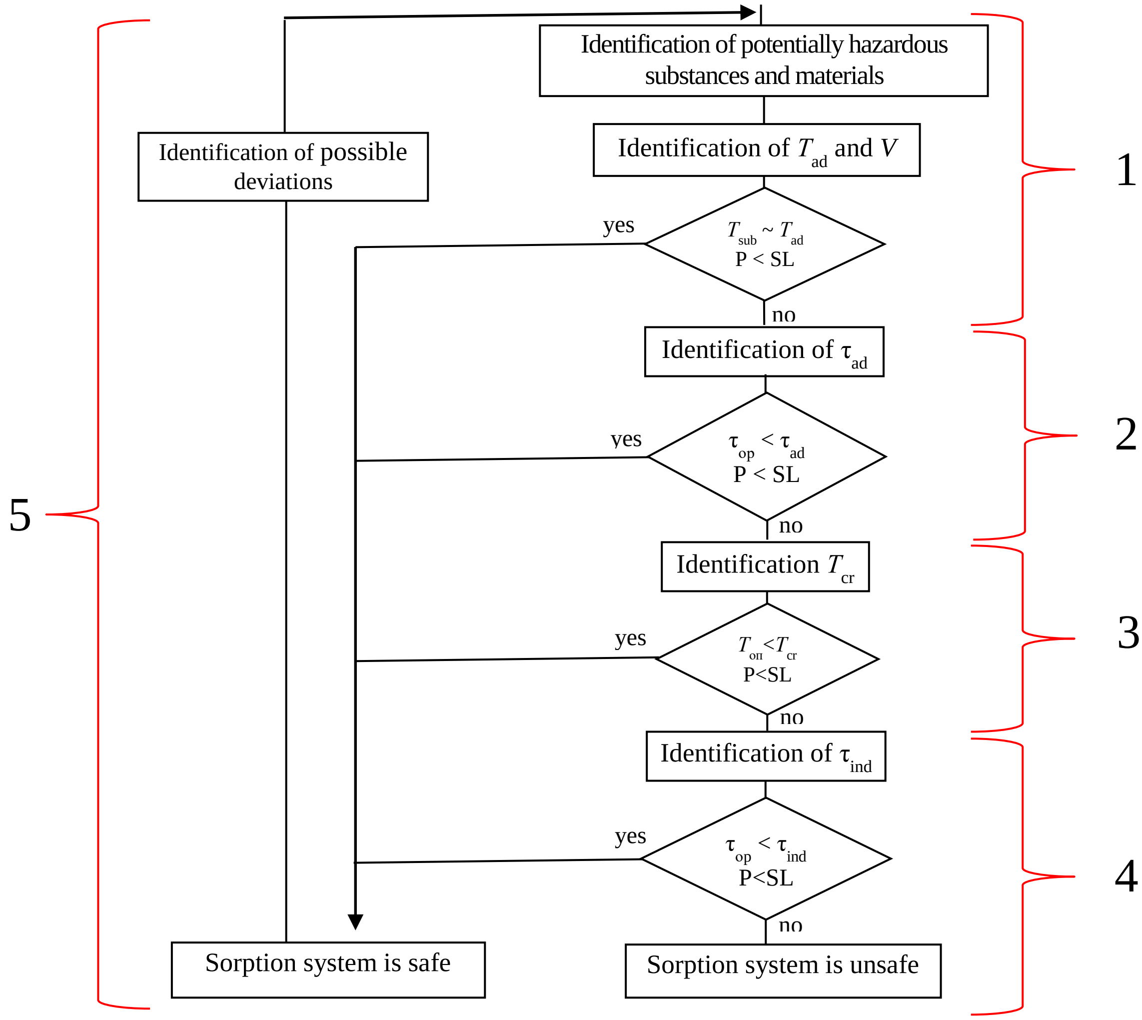

FIG. 1. The safety assessment scheme for sorption systems is implemented in the safety guide.

Using of formal kinetic approach that involves steps of investigation by methods of thermal analysis could be implemented. The prediction of runaway reaction in grams scale tests by using kinetic model based on differential scanning calorimetry data is shown in Fig 2.

Besides this example shows that critical conditions may be found with an acceptable for technical purpose precision. It was demonstrated that obtained by math modeling critical temperature is about 240 °C and experiments confirmed that at 230 °C there is no explosion, but at 243 °C it is observed. It shows the possibility of developing kinetic models that could be used for safety assessment.

In this part of the assessment (2) the calculation of temperature and gas release evolution under adiabatic conditions of substance are recommended. Using adiabatic conditions is a conservative simplification that permits not consider the heat losses influence. The scheme of making decision in second part of assessment is slightly changed. The comparison of estimated τad with time for normal operation (τop) and achieved parameters (P) by this time (τop) with the safety limits (SL) are supposed. The decision about acceptance of the parameters has to be made. If it is acceptable the last part of assessment (5) or if not the next (3) should be carried out.

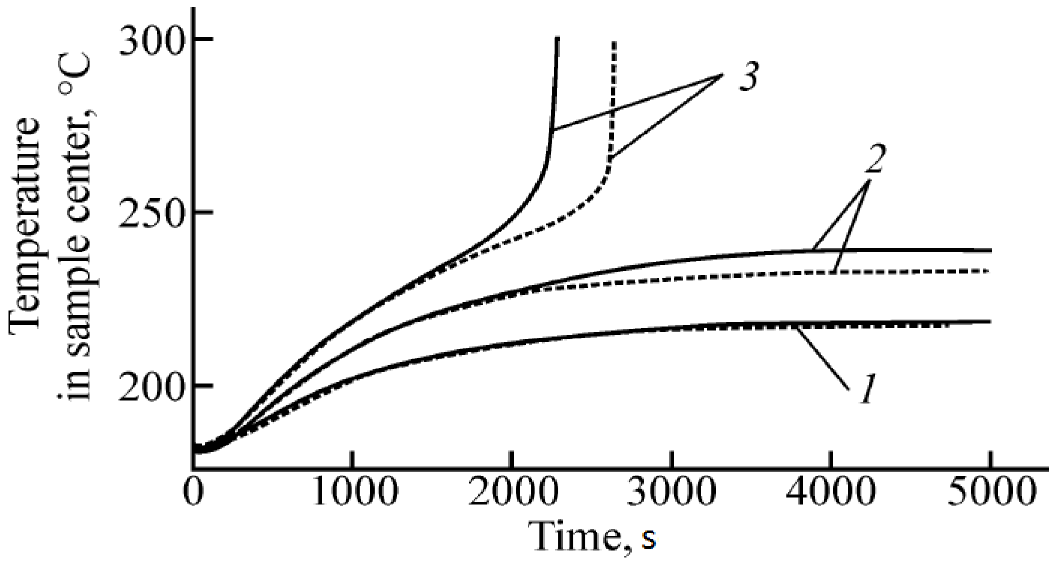

FIG. 2. Experimental and calculated variation of temperature in the center of a sample of VP-1AP resin in the nitrate form at different thermostat temperatures. Thermostat temperature, °C: (1) 220, (2) 230, and (3) 243. Solid lines: estimation by the model; dashed lines: experimental data.

The third and the fourth part is related to each other and both of them need to create a full model of the process. This model should include all of the heat sources and losses. The criteria are similar to previous part but in the third part should be additional estimated the general possibility of explosion under initial condition. That means the critical temperature (Tcr) has to be found and compared with operational one (Top).

An example of temperature curves for super- and sub- critical conditions are shown in Fig 3.

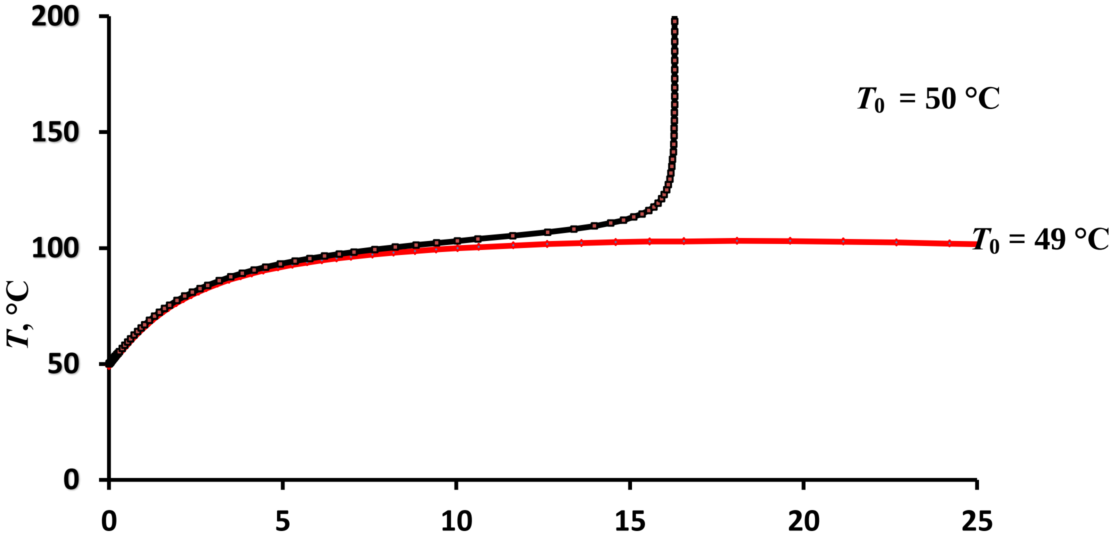

FIG. 3. The evolutions of temperature for super- and sub- critical conditions.

τ, h

For supercritical conditions the temperature increases exponentially when for subcritical one the reactions slows down without an explosion.

Both of the third and the fourth parts supposed to calculate achieved parameters for normal time of operation (τop) and compare with the safety limits (SL). If after fourth part the safety limits are violated the initial conditions are recommended to be considered as dangerous.

The developed approach allows operator to find the unacceptable conditions that could be used for setting of additional safety limits. Also such estimated parameters as the pressure and the temperature of gaseous products are initial data for providing the damage prediction.

4. SUMMURY

It is shown that reactive substances pose a danger to nuclear fuel cycle facilities that requires to improve the methods of identification and analysis. The developed approaches particularly allows to achieve the general aims of safety assessment. Developing of reactive hazard identification approaches can be associated with improvement of calculations methods that could solve the problems of the initial separation of a potentially hazardous substance from a safe one. Also it is related to improving of methods of kinetic parameters determination that will allow to get information about mechanisms and at the same time work with small sample quantity that is important for radioactive materials.

Improving the methods of reactive hazard analysis should focus on ways of consequences determining such as calculating of shock waves parameters and releasing of radioactive elements. The probabilistic approach should also be used to determine ways to improve the safety of nuclear fuel cycle facilities.

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