RESEARCH REACTOR SILICIDE FUEL REPROCESSING AT ORANO LA HAGUE

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ABSTRACT

On the basis of the extensive R&D program conducted by CEA and the detailed qualification program conducted by Orano that demonstrated the viability of silicide fuel reprocessing in the already-existing equipments of Orano La Hague plant, RR silicide spent fuel reprocessing at Orano La Hague Plant was granted in May 2017 after the Safety Report instruction by the French Safety Authority (ASN). This paper will present the latest evolutions on this major technological breakthrough, for the definition of spent fuel management strategy for Research Reactor operators including the feedback on first industrial operations. The range of silicide fuel that can be considered for reprocessing will be described, as well as the Orano La Hague reprocessing plant throughput and the perspectives for next decades of Research Reactor spent fuel sustainable management

1. Introduction

With the Research Reactor Spent Fuels (RRSF) management programs created under the Global Treat Reduction Initiative umbrella (currently named M^3 : Material Management and Minimisation), major part of research reactors operating with highly enriched fuel and new reactors operating with low enriched fuel use silicide-type fuel (U₃Si₂). Two strategies are available for management of RRSF (see Fig. 1): the once-through fuel cycle, with conditioning of the RRSF before disposal and the closed fuel cycle, with reprocessing of the RRSF meaning separating of reusable fissile material from the final waste, reuse of the fissile material under the form of fresh fuel and conditioning of the final waste for optimized disposal [1].

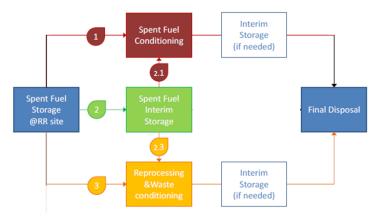


Fig 1. Two available strategies for management of RRSF

Since 1990's Orano has proposed sustainable and responsible solutions for RRSF management, including reprocessing at its La Hague¹ facility for aluminium-type fuels (UAI). In order to make U_3Si_2 users benefit from reprocessing solutions, Orano has been working over past years on the industrialisation of silicide fuel reprocessing at La Hague. After acknowledgement of U_3Si_2 reprocessing feasibility on all technical aspects, the administrative authorisation for reprocessing of one type of U_3Si_2 has just been granted by the French Safety Authority (ASN), further paving the way for reprocessing of all types of U_3Si_2 in the Orano La Hague plants.

This article gives update on current silicide fuel reprocessing solution in Orano La Hague plant, and on how to include reprocessing in research reactor's back-end strategy.

2. Reminder on the process

Reprocessing of RRSF at La Hague is based on the PUREX process for both UAI and U_3Si_2 spent fuels. In both cases the main reprocessing steps are:

- dissolution of the RRSF,
- mix with dissolution solution from Light Water Reactor (LWR) spent fuels (for aluminium management),
- liquid/liquid extraction and separation of U and Pu from Fission Products (FP) solutions and
- vitrification of the FP solutions after concentration.

However, for silicide spent fuels, a new step has to be added considering their high silicon content in the fuel meat. Indeed, this Si content leads to a high Si concentration in the dissolution solution which is not compliant with the PUREX liquid/liquid extraction process.

In order to meet the PUREX requirements the Si has to be separated from the dissolution and managed through a dedicated process flow.

This additional process step is performed thanks to the existing centrifugation equipment commonly used to separate the fines² during reprocessing operations for LWR spent fuels. Consequently, the separated silicon is managed through the fines line and is vitrified mixed with FP solutions at the end of the reprocessing operations: the vitrification step.

Fig. 2 reminds the whole reprocessing steps for U_3Si_2 RRSF including the added silicon separation step.

¹ The Orano La Hague plant is located in North-West of France, in the Normandy region, next to Cherbourg.

² The fines are small metallic parts not dissolved during the dissolution step.

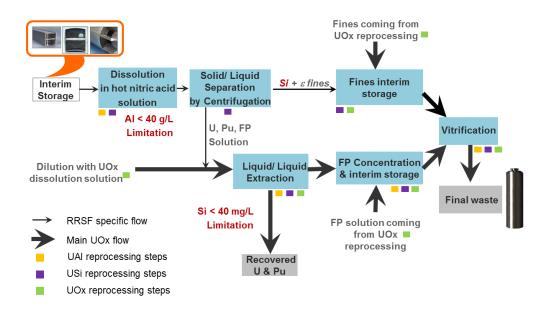


Fig 2. Process diagram for UAI and U₃Si₂ fuels reprocessing in Orano La Hague plant

3 Actions carried out and current industrial operations

3.1 From R&D phase to demonstration of the authorization application for U_3Si_2 reprocessing

In order to be able to reprocess silicide fuels at the La Hague reprocessing plants, an important R&D program has been carried out by the CEA and Orano [2]. The main goals of this R&D program were then:

- to characterise the behaviour of silicon from U₃Si₂ during dissolution,
- to characterise the behaviour of silicon from U₃Si₂ in extractions steps (PUREX process),
- to qualify the separation process of the silicon and the behaviour of the resulting silicon concentrated solution through the fines flow in the process.

This R&D program was completed end of 2013 and has finally demonstrated the feasibility of reprocessing operations in La Hague plants for spent silicide fuels. Following the R&D results, Orano focused in 2014 on the industrial qualification program in order to:

- take into account the process parameters coming from the R&D in the technical documents describing the industrial operating conditions for RRSF reprocessing,
- refine the reprocessing daily capacity and the annual capacity of reprocessing for silicide fuels,
- assess the impact of flows coming from silicide fuels dissolution on the whole La Hague processing activities.

The industrial feasibility and preliminary studies have been completed mid-2014. Operating ranges were successfully extended for the silicon separation step by centrifugation and the related management of silicon through the fines line, which today allow U_3Si_2 users to be offered attractive reprocessing solutions.

Following completion of the preliminary studies, the project has moved to the detailed studies phase during summer 2014. All the results and qualified operating ranges were taken into account in the process book dedicated to U_3Si_2 reprocessing operations. This batch of

documentation includes all the required documents for the main process steps (dissolution & silicon separation). For instance, unit description technical notes, process flow diagrams, chemical flow sheets, instrumentation process & automatism data sheets and process malfunction analyses are included. Studies to assess the impact of reference U_3Si_2 reprocessing operations on the whole La Hague plant activities (extraction, vitrification...) have been also performed and have concluded that this new qualified U_3Si_2 reprocessing fits with the whole plant operating & safety files.

These final studies were dedicated to finalizing of:

- detailed command and control systems studies considering that U₃Si₂ reprocessing operations are performed thanks to the same existing industrial equipments used to process UAI or LWR spent fuels; these studies lead to the final command and control softwares which are then used at industrial scale,
- the whole safety studies and the related U_3Si_2 reprocessing Preliminary Safety Report with the aim of considering operating ranges as wide as possible, and consistent with the reference U_3Si_2 spent fuel.

Thanks to the completion of this program, the U_3Si_2 process authorisation file has been submitted mid-2015 to the ASN.

3.2 Authorization for U₃Si₂ reprocessing and first industrial operations

After the technical exchanges between ASN and Orano and the completion of the public enquiry, the U_3Si_2 reprocessing at Orano La Hague plant has been granted by ASN on May 16th, 2017 for U_3Si_2 spent fuel coming from French SILOE and OSIRIS reactor³.

This authorization [3]:

- allows industrial U₃Si₂ reprocessing at La Hague facility for SILOE and OSIRIS spent fuel assemblies currently stored in La Hague storage pools,
- strongly facilitates and further paves the way for reception and reprocessing at La Hague plants of all types of U₃Si₂, starting for instance with U₃Si₂ coming from Australian OPAL reactor⁴ [4].

Based on this authorization, the first silicide RRSF reprocessing campaign has been successfully performed by Orano during summer 2017, thus industrially opening this new reprocessing solution. As expected, the dissolution of silicide spent fuels was complete, the silicon separation through centrifugation was very efficient and consistent with lab scale results and there were no additional impacts on the downstream operations in la Hague Plant.

³ <u>http://www.emtr.eu/osiris.html</u>

⁴ <u>http://www.ansto.gov.au/AboutANSTO/OPAL/</u>

4 Fuel characteristics consistent with the qualified process

4.1 General case

Considering the basic key steps of silicide fuel reprocessing operations (silicide dissolution, silicon separation, silicon management...), any type of silicide spent fuels can theoretically be reprocessed by Orano thanks to the reference qualified process described in the process book.

However, all the studies as described in 3.1 are based on a reference silicide fuel to be reprocessed in La Hague plant: the OSIRIS silicide fuel [5]. This allows Orano to define operating conditions ranges linked to fuel characteristics ranges. These ranges were described in the Preliminary Safety Report submitted to the ASN for obtaining of the U_3Si_2 reprocessing authorisation in the La Hague facility.

In any case, reprocessing of other silicide RRSF than the reference one will be subject to specific authorisation to be delivered after submission of a dedicated application file by Orano to the ASN. This situation leads to two cases:

- 1/ If the RRSF **is consistent** with all the acceptance and operating ranges described in the current authorization, the considered RRSF reprocessing application file will be a light dedicated one according to the authorization,
- 2/ If the RRSF is not consistent with all the acceptance and operating ranges described in the current authorization, it will be necessary to perform additional studies in order to assess the impact of its characteristics deviations on the reprocessing operations and its related cost in comparison with the reference fuel. If needed, an update of the reference process will have to be performed prior to the preparation of the dedicated application file for this spent fuel.

4.2 Relevant criteria for reprocessing scenarios assessment

To perform a reprocessing scenario assessment, Orano needs to obtain relevant information about the RRSF in order to:

- assess the reprocessing daily capacity, the annual reprocessing capacity and associated reprocessing costs for the RRSF,
- perform dedicated studies depending on RRSF specificities and/or if the characteristics deviation compared with the reference silicide fuel are significant, even if the core process operations are similar (centrifugation to separate the Si prior to U & Pu extraction). Once these preliminary studies are completed, it will be possible to assess the reprocessing daily capacity, the annual capacity and reprocessing costs for such silicide fuels.

A detailed description of the RRSF data needed for starting an assessment of possible reprocessing scenarios is provided in [2]. The following spent fuel characteristics are basically needed to be confirmed:

- geometry,
- chemical composition,
- standard chemical weight ratios (such as U_{initial}/AI or Si/AI)
- content of minor elements (for ex. magnesium, molybdenum, cadmium...)
- common information like burn-up, cooling time, initial and post irradiation composition, integrity (leakage)...

5 Perspectives for next decades of RRSF sustainable management

The U_3Si_2 reprocessing authorization is a major technological breakthrough, for the definition of spent fuel management strategy for RR operators. Based on this, the range of RRSF which can be reprocessed at La Hague is strongly increased, knowing that the capacity for reprocessing of silicide fuel is the same as the aluminium type fuel reprocessing capacity.

However, considering existing RRSF stockpile in La Hague storage pools, and existing reprocessing contracts, the La Hague RRSF reprocessing capacities are globally almost saturated. Orano already identified different ways to increase La Hague RRSF reprocessing capacities, especially:

- optimization/modifications of existing processes,
- implementation of a new RRSF reprocessing line.

These leverages have been assessed, and correspond today to well-identified projects at La Hague. Orano proposes today to RR operators interested in implementing RRSF sustainable management project to participate in this RRSF reprocessing capacity increase effort.

6 Performing reprocessing scenario assessment with Orano

Along with the reprocessing feasibility assessment and associated cost estimations, some other activities are to be looked at in order to set up a reprocessing project for silicide RRSF. These necessary activities are to be conducted in order to plan the transportation part, intergovernmental exchanges between the reactor's country and France, final waste management, and to set-up the overall project schedule.

6.1 Sites preparation and RRSF transportation

Transportation of silicide-type RRSF does not differ from transportation of aluminium-type fuels. In that regard, Orano has already acquired a worldwide experience in RRSF transportation (among others silicide-type), including the provision of several types of transport casks & baskets using multimodal transportation.

In order to assess transportation scenarios, RR operators have to select casks and transportation modes that meet their operational, regulatory and governmental constraints. The following needs to be reviewed for the RRSF shipment preparation [2]:

- compliance of package (cask loaded with RRSF content) with local (RR country), French and applicable international regulations,
- cask capacity in terms of number of RRSF elements which can be accommodated,
- cask compatibility with RR site and handling procedures,
- cask compatibility with La Hague site and handling procedures
- transportation mode selection knowing that both maritime and road transportation can be considered thanks to La Hague localisation by the sea, next to the Cherbourg harbour, designed for receipt and unloading of nuclear-material dimensioned-ships
- casks availability, long-lasting technology and safety of the cask design.

Following this review and preparation step, the transportation licensing phase is completed in order to allow for effective transportation to occur. For France, two agreements are to be granted by the ASN:

- transportation license, for transportation of the cask with the relevant RRSF content on French territory,
- license for receipt, unloading and reprocessing at La Hague, after required safety reviews.

For several RRSF transportation casks, these licenses are already available and consequently need to be slightly adapted for each RRSF specific content.

In the RR country, the same kind of transportation licences is necessary.

6.2 Intergovernmental agreement

According to European Directive⁵ and French law⁶, the introduction on French territory of spent nuclear fuels for a reprocessing purpose has to be framed by an intergovernmental agreement (IGA) between the SF country of origin and France. This agreement settles "a forecasted schedule for reception and processing of the material and, if any, the later planned use of the material separated during reprocessing". Article L542-2 of the French Environmental Code specifies also that disposal in France of radioactive waste from abroad is forbidden, including waste resulting from foreign RRSF reprocessing.

The above-mentioned IGA is to encompass the following items:

- Project description:
 - o Material owner,
 - Main stakes for the owner,
 - $\circ\;$ Location of the nuclear material,
 - $\circ\;$ Legal status and origin of the material,
 - o Material owner country presentation,
 - Planned contractual structure for material reprocessing, After RRSF reprocessing, the valuable material can be managed by Orano in order to be re-used in civilian purposes (fresh LWR fuels).
 - Planned scope of collaboration between the parties,
- Acceptability of reprocessing:
 - Type and characteristics of material to be reprocessed: design, total mass, mass of oxide (if any) and heavy metals, rate of combustion, cooling, initial enrichment,...
 - o Material transportation scheme (cask and transportation procedures),
- Schedule:
 - o Quantities to be reprocessed and timing,
 - Period of delivery of RRSF from the customer to Orano La Hague,
 - Period of reprocessing,
 - Period of waste return,
 - o Use /reuse of the recovered material,
 - Deadline for the last return of waste,
 - Destination of waste.

From Orano's experience on conducting this IGA process, between six months and two years are necessary to get the final agreement from all parties, starting from beginning of the official discussion between the countries. Consequently, this whole process has to be well included in RRSF reprocessing overall project.

A commercial transportation and reprocessing contract between Orano and a RR operator can be concluded before the end of the IGA process. Nevertheless, the IGA conclusion will be necessary in order to start transportation of nuclear material.

⁵ Council Directive 2011/70/EURATOM (07/19/2011) establishing a Community framework for the responsible and safe management of spent fuel and radioactive waste: <u>http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2011:199:0048:0056:EN:PDF</u>

⁶ French Environmental Code resulting from the law of June 28, 2006 on the sustainable management of radioactive materials and waste, and application decree no. 2008-209 of March 3, 2008 on procedures applicable to the reprocessing and recycling of foreign spent fuel and radioactive waste specifying certain conditions.

6.3 Final waste management

Another application of French law⁷ is about the final waste calculation method needed to define the waste type and quantity to be sent out of France after reprocessing of foreign spent fuel.

In order to comply with this regulation, Orano applies a material accountancy system including an activity unit for waste (UAR, *Unité d'Activité de Résidu*) and a mass unit for waste (UMR, *Unité de Masse de Résidu*). This system called EXPER (*EXPEdition des Résidus*) has been approved by decree, and has been implemented since October 2008 for all new RRSF reprocessing operations.

In the case of silicide-type RRSF reprocessing, if all the material is dissolved, the only remaining waste corresponds to the UAR system, based on the Neodymium quantities imported in France in the RRSF.

The UAR system implies two possible types of vitrified residues to be sent back: UC-V (Universal waste Canister, Vitrified type) and UC-U (Universal waste Canister, U vitrified type). The UC-V FP concentration is highly superior to the UC-U one, leading to a higher UC-V thermal power than UC-U's. According to each country regulation, UC-V and UC-U can be considered respectively as high level waste (HLW) and intermediate level waste (ILW).

Orano proposes to study the conditions under which the final waste can be managed with the RR operators and their regulatory bodies.

Two different examples can be underlined for final waste management:

- Belgium:

After reprocessing of BR2⁸ RRSF, UC-Vs have been jointly sent back to Belgium with residues from Belgian utilities SF reprocessing. As the LWR SF reprocessing results in much larger quantities of UC-Vs than RRSF reprocessing, the residues return was almost insignificant for BR2 operator.

- Australia:

Australia does not operate any nuclear power plant. Australia does not have any HLW to manage. The UC-U was consequently the best option for waste return to Australia after reprocessing of HIFAR⁹ SF. UC-U is indeed managed as ILW and does not need large investments for long term management (in comparison with final HLW disposal). The successful return of UC-U to Australia was performed end 2015 [4].

Orano proposes to adapt the final waste responsible and sustainable management to each country regulations and specificities [1].

6.4 Overall project schedule

The overall silicide RRSF reprocessing project can be separated into two major phases:

- the preparation phase (1 to 5 years), comprising of
 - o confirmation of reprocessing feasibility and cost estimation: 3 to 18 months,
 - transportation preparation: 3 to 24 months,
 - o Intergovernmental Agreement and related exchanges: 6 to 24 months,
- and the execution phase (10 to 30 years) with
 - RRSF evacuation from RR site,

⁷ French Environmental Code resulting from the law of June 28, 2006 on the sustainable management of radioactive materials and waste, and application decree no. 2008-209 of March 3, 2008 on procedures applicable to the reprocessing and recycling of foreign spent fuel and radioactive waste specifying certain conditions

⁸ https://www.sckcen.be/en/Research/Infrastructure/BR2

⁹ http://www.ansto.gov.au/AboutANSTO/HistoryatANSTO/Decommissioningearlierreactors/HIFAR/index.htm

- o transportation to the La Hague plant,
- unloading and interim storage of RRSF in La Hague SF pools
- performance of reprocessing operations directly or several years after receipt of RRSF, depending on the plant operational constraints and IGA-bound timelines
- o after reprocessing, storage of final waste for cooling
- shipment of the waste out of France (usually back to the RRSF country of origin) [1].

The IGA finalisation is a mandatory milestone between preparation and execution phases.

Depending on RR operators' needs and on the RR operator - Orano partnership, commercial contracts and commitments can be concluded for the overall project, or separately for each phase knowing that a contract for execution phase can be signed by the parties before IGA signature (the contract validity being conditioned by the IGA entry into force).

This overall timeline is described in the Fig. 4 below and is 10 to 40 years long depending on the reprocessing scenarios and the concluded IGA.



Fig 3. Timeline of the reprocessing project execution phase

7 Conclusions

In order to provide its customers with sustainable, cost-effective and responsible RRSF management solutions, Orano has commissioned silicide-fuel reprocessing at its La Hague plant. This new back-end solution is now industrially available for any U_3Si_2 RRSF types, after verification of the corresponding operating conditions, available capacities and associated costs, on a case-by case basis.

Based on this, Orano is ready to support RR operators in their back-end strategy definition for silicide fuels. Any silicide fuel reprocessing project can be implemented as soon as:

- safety authorisations for reprocessing and transportation are obtained, based on the current authorisation for reprocessing of French SILOE and OSIRIS reactor silicide fuel,
- IGA between the corresponding countries and France are finalised.

8 Acronyms

ASN CEA EXPER FP	French Safety Authority (<i>Autorité de Sûreté Nucléaire</i>) Atomic Energy Commission (<i>Commissariat à l'Energie Atomique</i>) Waste shipping system (<i>EXPEdition des Résidus</i>) Fission Product(s)
HLW	High Level Waste
IGA	InterGovernmental Agreement
ILW	Intermediate Level Waste
LWR	Light Water Reactor
M ³	Material Management and Minimisation
Pu	Plutonium
PUREX	Plutonium and Uranium Refining by EXtraction
R&D	Research and Development
RR(SF)	Research Reactor (Spent Fuel)
Si	Silicon
U	Uranium
U_3Si_2	Silicide type fuel
UAI	Aluminium type fuel
UAR	Activity unit for waste (Unité d'Activité de Résidu)
UC-U	Universal waste Canister, U vitrified type
UC-V	Universal waste Canister, Vitrified type
UMR	Mass unit for waste (<i>Unité de Masse de Résidu</i>)

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