RESEARCH REACTOR SILICIDE FUEL REPROCESSING AT AREVA LA HAGUE

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ABSTRACT

After an extensive R&D program conducted by French CEA that demonstrated the viability of silicide fuel reprocessing in the already-existing equipment, and after a detailed qualification program that aimed at integrating the lab-scale results into the industrialized processes of AREVA La Hague plants, AREVA submitted the Safety Report seeking for authorisation of the French Safety Authority (ASN) to reprocess Research Reactor silicide fuel at La Hague site in October 2015.

This paper will present the latest evolutions on this major technological breakthrough, for the definition of spent fuel management strategy for Research Reactor operators. The next steps include the ASN reprocessing authorisation, feedback on the first reprocessing campaign, and start of industrialized operations.
The range of silicide fuel that can be considered for reprocessing will be described, as well as the AREVA La Hague reprocessing plants 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 (U_3Si_2). 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].

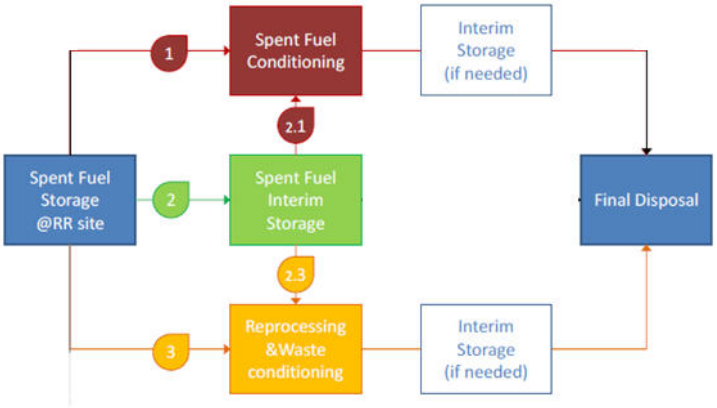

Fig 1. Two available strategies for management of RRSF

Since 1990's AREVA has proposed sustainable and responsible solutions for RRSF management, including reprocessing at its La Hague[1] facility for aluminium-type fuels (UAl).
In order to make U_3Si_2 users benefit from reprocessing solutions, AREVA 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 AREVA La Hague plants.
This article will update the reader on the schedule for silicide fuel reprocessing solution availability, 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 UAl 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 (see 3.1.1.).
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 fines[2] 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 below reminds the whole reprocessing steps for U_3Si_2 RRSF including the new silicon separation step.

[1] The AREVA 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.

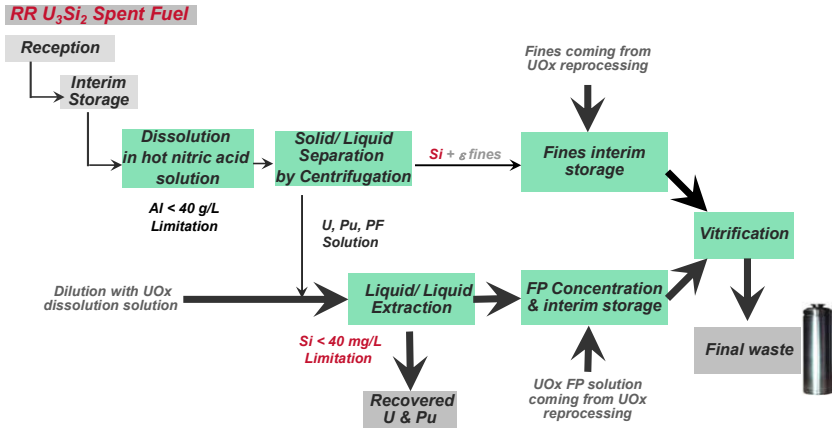

Fig 2. Process diagram for Silicide fuels reprocessing in AREVA La Hague plant

3. Update on ongoing actions and schedule for industrial commissioning

3.1. Past R&D and studies

3.1.1. R&D program for the scientific and technical feasibility demonstration of the 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 AREVA [2].
The main goals of this R&D program were then:

- to characterise the behaviour of silicon from U_3Si_2 during dissolution,
- to characterise the behaviour of silicon from U_3Si_2 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.

3.1.2. Industrial feasibility and preliminary studies

Following the R&D results, AREVA 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.

3.1.3. Detailed studies

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 U_3Si_2 reprocessing operations are performed thanks to the same existing industrial equipments used to process UAl 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. Administrative authorisation for U_3Si_2 reprocessing

Technical exchanges between the ASN and AREVA were completed by end of 2016. Public enquiry was achieved in February 2017. A few weeks later, the ASN granted the administrative authorisation for reprocessing at La Hague of U_3Si_2 spent fuel coming from French OSIRIS reactor[3].

This authorisation:

- allows for first industrial U_3Si_2 reprocessing campaign to be performed at La Hague facility in 2017, with management of 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 U_3Si_2, starting for instance with U_3Si_2 coming from Australian OPAL reactor[4] [3].

Fig. 3 below summarizes the completed steps which conduct to industrial reprocessing of silicide fuel at AREVA La Hague facility.

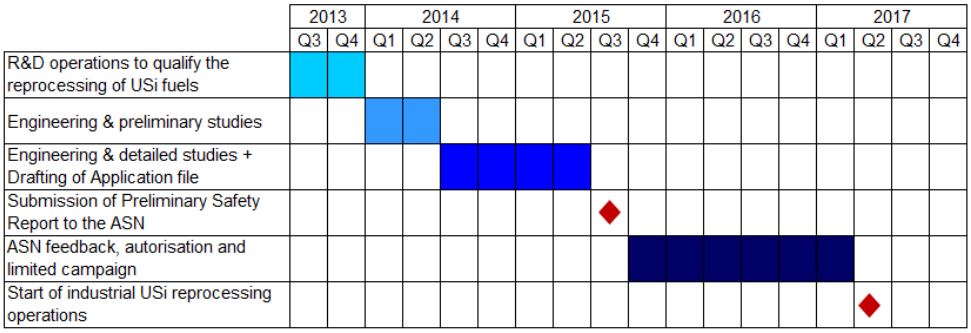

Fig. 3 Qualification program schedule leading to industrial reprocessing of U_3Si_2 at La Hague

[3] http://www.emtr.eu/osiris.html
[4] http://www.ansto.gov.au/AboutANSTO/OPAL/

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 AREVA 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 [4]. This allows AREVA to define operating conditions ranges linked to fuel characteristics ranges. These ranges are described in the Preliminary Safety Report submitted to the ASN for obtaining of the U_3Si_2 reprocessing authorisation in the La Hague facility (see 3.2).
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 AREVA 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 application file (for reference silicide fuel), the considered RRSF reprocessing application file will be a light dedicated one according to the current reference fuel application file,
- 2/ If the RRSF **is not consistent** with all the acceptance and operating ranges described in the current application file, 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, AREVA 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 $U_{initial}$/Al or Si/Al)
- 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 authorisation 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.
AREVA 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. AREVA 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 AREVA

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, AREVA 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 Directive[5] and French law[6], 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:
  - Material owner,
  - Main stakes for the owner,
  - Location of the nuclear material,
  - Legal status and origin of the material,
  - Material owner country presentation,
  - Planned contractual structure for material reprocessing,
    After RRSF reprocessing, the valuable material can be managed by AREVA 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,...
  - Material transportation scheme (cask and transportation procedures),
- Schedule:
  - Quantities to be reprocessed and timing,
  - Period of delivery of RRSF from the customer to AREVA La Hague,
  - Period of reprocessing,
  - Period of waste return,
  - Use /reuse of the recovered material,
  - Deadline for the last return of waste,
  - Destination of waste.

From AREVA'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 AREVA 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.

6.3. Final waste management

Another application of French law[6] 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, AREVA 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*

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

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).
AREVA 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 BR2[7] 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 HIFAR[8] 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 [3]**.**

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

[7] https://www.sckcen.be/en/Research/Infrastructure/BR2
[8] http://www.ansto.gov.au/AboutANSTO/HistoryatANSTO/Decommissioningearlierreactors/HIFAR/index.htm

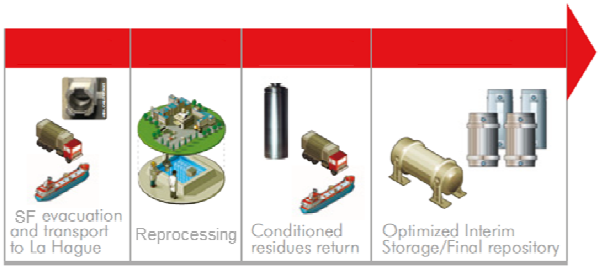

Fig. 4 Timeline of the reprocessing project execution phase

7. Conclusions

In order to provide its customers with sustainable, cost-effective and responsible RRSF management solutions, AREVA has been developing silicide-fuel reprocessing at its La Hague plant. This new back-end solution is available as from 2017 for 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, AREVA 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 authorities authorisations for reprocessing and transportation are obtained, based on obtaining of administrative authorisation early 2017 for reprocessing of French OSIRIS reactor silicide fuel,
- IGA between the corresponding countries and France are finalised.

8. Acronyms

ASN	French Safety Authority (*Autorité de Sûreté Nucléaire*)
CEA	Atomic Energy Commission (*Commissariat à l'Energie Atomique*)
EXPER	Waste shipping system (*EXPEdition des Résidus*)
FP	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
UAl	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 (*Unité de Masse de Résidu*)

9. References

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[4] S. Loubiere, G. Bignan, ND. Iracane, *“Sustaining material testing capacity in France: from OSIRIS to JHR”, IGORR 2009, October 2009*