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Status on Silicide Fuel Reprocessing at AREVA La Hague (2015)

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ABSTRACT

Silicide (U_3Si_2) fuels reprocessing qualification has been performed by AREVA considering the same process at La Hague as the one for UAl fuels, benefiting from decades of RRSF reprocessing experience. Some adaptations have been performed to deal with U_3Si_2 characteristics.

Based on CEA R&D fruitful program, AREVA has performed in 2014 and 2015 the industrial qualification phase to set up the silicide fuels reprocessing, including the safety analysis. The authorization file has been submitted to the French Authority in 2015, with an authorization expected by 2017. AREVA will be able to start industrial-scale U_3Si_2 fuel reprocessing after this ASN authorization.

AREVA is now ready to start reprocessing projects with its customers and partners for U_3Si_2 RRSF. To assess such opportunities, this paper encompasses:

- overview of U_3Si_2 RRSF reprocessing qualification,
- range of fuels for which reprocessing at La Hague will be available,
- identification of the necessary steps to be taken in a reprocessing project.

1. Introduction

With the Research Reactor Spent Fuels (RRSF) management programs created under the Global Treat Reduction Initiative (GTRI) 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 will use silicide-type fuel (U_3Si_2). The "take-back" option of M^3 programs will cease for RRSF irradiated after May 2016 [1], leaving no sustainable back-end option to research reactor operators who previously planned to benefit from GTRI/M^3 programs until shutting down of their facilities.
Since 1990's AREVA has proposed sustainable and responsible solutions for RRSF management, including reprocessing at its La Hague[1] plant for aluminium-type fuels (UAl).
In order to make U_3Si_2 fuel users benefit from reprocessing solutions, AREVA is currently finalizing the industrialisation of silicide fuel reprocessing at La Hague plant.
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 AREVA 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 will be 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 will be managed through the fines line and will be vitrified mixed with FP solutions at the end of the reprocessing operations: the vitrification step.
The following diagram reminds the whole reprocessing steps for U_3Si_2 RRSF including the new step of silicon separation.

[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.

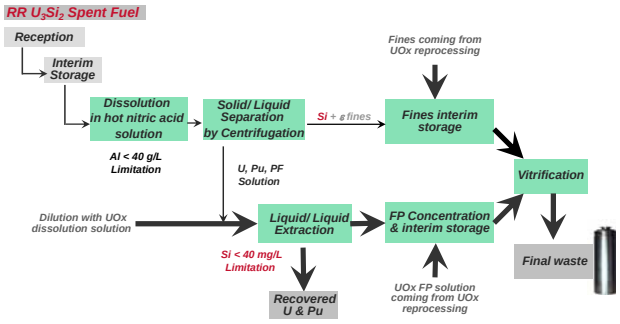

Fig.1: Process diagram for Silicide fuels reprocessing in AREVA NC LA Hague plant

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 fuel reprocessing

In order to be able to reprocess silicide fuels at the AREVA La Hague reprocessing plant, an important R&D program has been carried out by AREVA and the CEA [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 plant for spent silicide fuels.

3.1.2. Industrial feasibility and preliminary studies performed by AREVA:

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 AREVA 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 will allow AREVA to offer attractive reprocessing solutions to its customers. Based on their results, AREVA has moved from preliminary studies 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.

3.2. Current status of detailed studies carried out by AREVA

Since the detailed studies phase started (mid-2014), AREVA has finalized all the technical documents in order to perform U_3Si_2 reprocessing in its La Hague plant.
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 have been successfully completed. Studies to assess the impact of reference U_3Si_2 reprocessing operations on the whole AREVA 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 finalize:

- detailed command and control systems studies considering that U_3Si_2 reprocessing operations will be performed thanks to the same existing industrial equipments used to process UAl or LWR spent fuels. These studies will lead to the final command and control softwares which will be used at industrial scale,
- the whole safety studies and the related U_3Si_2 reprocessing Preliminary Safety Report (RPS) with the aim of considering operating ranges as wide as possible, and consistent with the reference U_3Si_2 spent fuel, has also been achieved.

Thanks to the completion of this program, the U_3Si_2 process authorisation file is submitted for approval in 2015 to the French Safety Authority by AREVA.

3.3. Schedule overview

The following timeline summarizes the current tentative schedule that will conduct to industrial reprocessing of silicide fuel at AREVA La Hague plant.

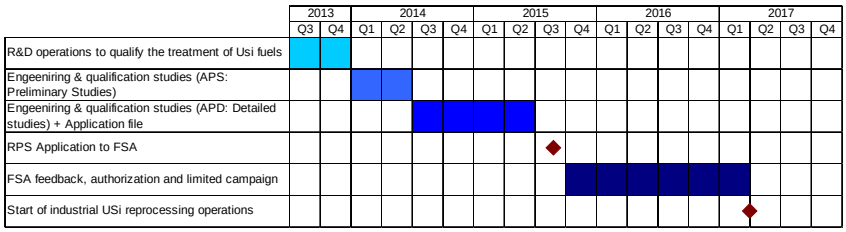

Fig.2: Tentative timeline of the industrial qualification program prior to process authorization

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 performed by AREVA are based on a reference silicide fuel to be reprocessed in La Hague plant. This allows AREVA to define operating conditions ranges linked to fuel characteristics ranges. These ranges are described in the Preliminary Safety Report (RPS) to be submitted to the French Safety Authority (ASN) in order to get the authorization for reprocessing silicide fuels in the AREVA La Hague plant as mentioned above.
In any case, reprocessing of other silicide RRSF than the reference one will be subject to specific authorization to be delivered after a dedicated application file by AREVA to FSA.
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). One 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.

The first criteria are the geometrical characteristics of the spent fuel (diameter, length). Indeed, the current process consists in dissolving the SF in a dissolution pit that imposes some constraints on the SF to be reprocessed (maximum acceptable diameter and length). If the fuels size is not consistent with this “physical entrance” range, additional operations will have to be considered such as prior cutting operations for instance. Detailed characteristics (such as, but not limited to plates thickness,…) are also useful to perform the capacity assessment.
The other important characteristics for the reprocessing assessment are linked to the chemical composition of the spent fuel. Basically, the type of Al alloy (cladding and fuel meat matrix) is a key parameter for the capacity assessment as it is linked to the dissolution kinetics. It can be a key point for additional study especially if the type of alloy is not included in the considered alloy range of the process book or if it includes a chemical element which can have a strong impact on downstream reprocessing operations (extraction, concentration or vitrification).
Standard chemical weight ratios for RR spent fuels (such as $U_{initial}$/Al or Si/Al) are very important to assess the reprocessing daily capacity and especially the silicon ratio in case of U_3Si_2 SF. The amount of silicon to be separated from the dissolution solution has an impact on

the dissolution/centrifugation stage (number of operations and reprocessing rate) and leads to dilution of “usual” reprocessing flows with consequences on the fission products concentration capacity and on the vitrification rate. Regarding Al total mass, possibilities of upstream cropping operations in order to reduce the total mass to be reprocessed can be taken into account in a scenario assessment as an optimisation.
The content of minor elements (ex. magnesium, molybdenum, cadmium…) is also helpful for reprocessing scenarios assessment considering the fact that such minor elements can have an impact on the reprocessing daily capacity. Such elements can have a link with corrosion concerns, deposit formation, management of releases in the environment, vitrified residue specification… ; that is why it is necessary for AREVA to get the RRSF chemical composition as detailed as possible.
Obviously, other common information such as burn-up, cooling time, initial and post irradiation composition, integrity (leakage)… are useful for the scenario assessment, for the comparison with the reference ranges given in the current U_3Si_2 process book, and more generally with all the reprocessing steps at La Hague, from receipt of the RRSF to final waste production.

4.3. Requested information

The following table presents the basic data information requested to start an assessment of possible reprocessing scenarios. This list has to be considered as a starting point for exchanges with AREVA on RRSF reprocessing, and can be updated according to the SF characteristics, or after first exchanges between AREVA and the RR operator.

Table 1: Basic data information needed for U_3Si_2 reprocessing scenarios assessment by AREVA.

Basic information Datasheet								
Reactor Name - type of fuel (standard, control, USi, UAl, …)								Comments
1	Burn-up rate (average and max)	average			max			
2	Cooling time (min and max)	min			max			
3	De-activation date							
4	Amount of cycle							
5	Spent fuel integrity (any leaking, disassembled fuel?)							
6	Do sipping tests have been conducted? If yes, can you please give procedure and results?							
7	α and $\beta\gamma$ spectra							
8	Total α and $\beta\gamma$ activity per FA (Bq/FA)							
9	Thermal power							
10	Contents of U and Pu post-irradiation	U (g)			Pu (g)			
11	Initial/Post irradiation enrichment of U and Pu	Enrich.	Initial	U_{tot} (mass)				
				U^{235}/U				
			Post	U_{tot} (mass)				
				U^{235}/U				
12	Si and Al content	Before cutting	Al (g)					
			Si (g)					
		After cutting	Al (g)					
			Si (g)					
13	Structure, cladding, fuel meat matrix & spacer material	Structure						
		Cladding						
		Fuel meat matrix						
		Spacer						
14	Other items in assembly (including materials, quantities, dimension (mm) and weights (g))							
15	Presence and detailed weight percentage of any other minor elements (Mg, Mo, B, Se, Cd…)							
16	Do you have any possibility to cut pieces (head or foot) on site? If yes, could you please describe parts that can be cut?							
17	Exhaustive fuel specifications and drawings	Circular section, length, thickness… (FA, plates) Before and after cropping if any						
18	Total mass for each fuel element (g) (before and after cutting)							
19	Any useful information regarding the fabrication process specificities							
20	Amount of element to be transported/treated / mid-long term inventory planning							
21	Absorbed dose at 1m from surface of the spent fuels							

5. 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.

5.1. Transportation

Transportation of silicide-type RRSF does not differs 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 transportation of the RRSF needs also to be plan early enough within reprocessing scenarios assessment.

5.1.1. Site preparation and interfaces with selected transportation casks

Transportation cask selection is key in the overall reprocessing project timeframe. The type of cask will have direct impacts on the transportation frequency, possible modifications of RRSF storage and handling procedures at reactor site, possible modifications of RRSF receipt facilities and handling procedures at the AREVA La Hague plant.
The following criteria are to be reviewed in order to prepare a reprocessing project and to assess the possible options for transportation:

- Compliance with local (RR country), France, and applicable international regulations and technical requirements. The competent Safety Authorities will have to review the cask safety files and provide agreements for transportation of each specific RRSF content.
- Capacity, i.e. amount of RRSF per cask, is also a key factor in order to reduce the amount of transports to be performed and the transportation frequency during RR lifetime. Along with public acceptance, the cost for transportation will directly be impacted by this parameter.
- Compatibility with RR site and handling procedures. Several criteria such as the cask weight, cranes and floors capacities, handling tools and procedures are to be looked at in order to identify the range of RRSF transportation casks that suits with the constraints of each reactor site. In some cases, adaptations of RR site may be needed in order to allow using the selected transportation cask. AREVA can provide, when applicable, complementary and flexible transfer systems in order to simplify loading of RRSF in transportation casks.
- Compatibility with the AREVA La Hague site and its handling procedures. RRSF unloading operations are performed under-water in the NPH pool at La Hague. This facility has been designed for flexible operations and can consequently adapt to several RRSF transportation cask designs. Nevertheless, safety studies and adaptation of equipments can be required to unload new cask designs.

5.2.2. Transportation of RRSF

Before effective transportation, the transportation licensing phases are to be completed.
For France, two agreements are to be granted by 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.
Transportation modes also have a strong influence on a reprocessing project.
As La Hague is located by the sea, next to the Cherbourg harbour, designed for receipt and unloading of nuclear-material dimensioned-ships, both maritime and road transportation can be considered.
For European RR, the road transportation mode is often selected. It may raise some additional licensing costs (for the countries the truck drives through).
For overseas RR, the transportation casks capacities and availability in the project time-frame become major issues in order to reduce project uncertainties and associated costs.
The casks availability, the long-lasting technology and safety of the cask design are to be considered during scenario assessment.

5.2. Intergovernmental agreement

According to European Directive[3] and French law[4], the introduction on French territory of spent nuclear fuels for a reprocessing purposes has to be framed by an intergovernmental agreement (IGA) between France and the SF country of origin. 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 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 (new LWR or MOX 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 and heavy metals, rate of combustion, cooling, initial enrichment,...
  - Material transportation scheme (cask and transportation procedures),
- Schedule:
  - Quantities to be reprocessed and timing,

[3] 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

[4] 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 specifies certain conditions

- 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 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 owner 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.

5.3. Final waste management

Another application of French law[5] concerns the final waste calculation method.
In order to comply with this regulation, AREVA applies a material accountancy system including a unique activity unit for waste (UAR, *Unité d'Activité de Résidu*) and a unique mass unit for waste (UMR, *Unité de Masse de Résidu*).
This system allows AREVA to calculate the amount and type of waste to be sent back to its customers. 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 Nd quantities imported in France in the RRSF.
The UAR system implies two possible types of vitrified residues: CSD-V (*Conteneur Standard de Déchets Vitrifiés*) and CSD-U (*Conteneur Standard de Déchets Vitrifiés de type U)*.
The CSD-V concentration in FP is highly superior to the CSD-U one. The thermal power is consequently higher in CSD-V than is CSD-U.
According to each country regulation, CSD-V and CSD-U can be considered respectively as HLW and 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 RRSF, CSD-Vs have been jointly sent back to Belgium with residues from Belgian utilities SF reprocessing. As the LWR SF reprocessing results in much higher volumes of CSD-V than RRSF reprocessing, the residues return was almost insignificant for the BR2 operator (SCK).
- Australia:
  Australia does not operate any Nuclear Power Plant. Australia does not have any HLW to take manage. The CSD-U was consequently the best option for Australia as it is managed as ILW and does not need large investments for long term management (in comparison with final HLW disposal).

AREVA proposes to adapt the final waste responsible and sustainable management to each country regulations and specificities.

5.4. Overall project schedule

The overall silicide RRSF reprocessing project can be separated in two major phases: the preparation phase, and the execution phase.
Depending on RR operators' needs and on the AREVA-RR operator partnership, commercial contracts and commitments can be concluded for the overall project, or separately for each phase.

5.4.1. Preparation phase

The preparation phase is mainly composed of the above-mentioned steps (see paragraphs 4 and 5).
The following timeline can be considered in a silicide RRSF reprocessing project preparation:

- 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.

These timelines are to be adapted for every single case with regards to all aspects of the reprocessing project. Of course, all these activities can be run in parallel in order to shorten the preparation period.
Even if the IGA finalization is a mandatory milestone between preparation and execution phases, it is reminded that a commercial contract for execution phase can be signed by the parties before IGA signature.

5.4.2. Execution phase

The execution phase starts with RRSF evacuation from RR site, transportation to the AREVA La Hague plant, unloading and interim storage of RRSF in AREVA La Hague SF pools.
Depending on the AREVA operational constraints and IGA-bound timelines, reprocessing operations can be performed directly, or several years after receipt at La Hague.
After reprocessing, the final waste is stored for cooling.
As the final waste quantities are very low after RRSF reprocessing, the residues return can be optimized for the entire RR-reprocessed inventory in order to perform as less transportations as possible for a dedicated country, for example by mutualizing it with other transportations.
The residues are sent back conditioned and packaged in compliance with the country of destination's waste management policy: in dual-purpose casks for transportation and long-term storage or in transportation casks for transfer to a storage/disposal facility.
This overall timeline is described in the Fig.3 below and is 10 to 40 years long depending on the reprocessing scenarios and the concluded IGA.

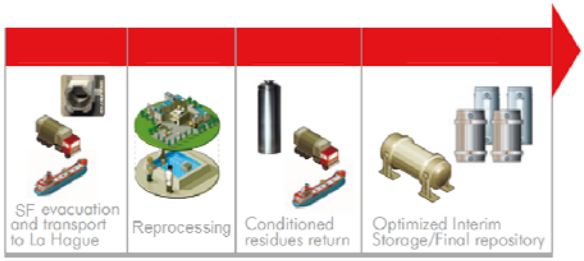

Fig.3: Timeline of the reprocessing project execution phase

6. 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 will be 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.
AREVA is ready to support RR operators in their back-end strategy definition for silicide fuels as of today. The first transportations will be possible starting 2017, after Safety authorities authorizations for reprocessing and transportation, and after IGA finalization between France and the corresponding countries.

7. References

[1] C.E. Messick, J.J. Galan, *"Global Threat Reduction Initiative"*, Transactions of RRFM2014, p. 274-279, Ljubljana, Slovenia, March, 30 – April, 3, 2014.

[2] C. Eysseric, A. Juvenelle, N. Reynier-Tronche, JP. Feraud, T. Randriamanantena, D. Ode, B. Lorrain, J.F. Valery, X.Domingo, *"Status of silicide fuels reprocessing at La Hague plant", RRFM2014, Ljubljana, March 30th - April 3rd 2014, Transactions, p. 298-307*