



ASGARD

Advanced Fuels for Gen IV Reactors: Reprocessing and Dissolution

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SHORTNAME OF LEAD PARTNER (CHALMERS)

Author(s): Barbara Oberlander, Kastriot Spahiu

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SAC Member	SAC Leader	Coordinator
Prof. Kastriot Spahiu	Dr. Barbara Oberländer	Ch. Ekberg
Kastriot Spahiu	Barbara Oberländer	

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Dissemination level			
PU	Public		
RE	Restricted for specific group		
CO	Confidential (only for ASGARD partners)	X	Scientific Advisory Committee to Coordinator, Governing Board and PCC.

Version control table

Version number	Date of issue	Author(s)	Brief description of changes made
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2.0	28/06/2016	P. Koran	AMO check and formal correction, finalization

Relevant domain(s) and workpackage(s)

Tick **ALL** or select in the following table:

DM	WP		
DM 1 <input type="checkbox"/>	WP 1.1 <input type="checkbox"/>	WP 1.2 <input type="checkbox"/>	WP 1.3 <input type="checkbox"/>
DM 2 <input type="checkbox"/>	WP 2.1 <input type="checkbox"/>	WP 2.2 <input type="checkbox"/>	WP 2.3 <input type="checkbox"/>
DM 3 <input type="checkbox"/>	WP 3.1 <input type="checkbox"/>	WP 3.2 <input type="checkbox"/>	WP 3.3 <input type="checkbox"/>
DM 4 <input type="checkbox"/>	WP 4.1 <input type="checkbox"/>	WP 4.2 <input type="checkbox"/>	WP 4.3 <input type="checkbox"/>

Project information

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Coordinator:	Christian Ekberg
EC Project Officer:	Michel Hugon / Roger Garbil
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Coordinator contact:	+46-31-7722801, info@asgardproject.eu
Administrative contact:	+420 245 008 599, amo@asgardproject.eu
Online contacts:	www.asgardproject.eu

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CONTENT

- EXECUTIVE SUMMARY..... 4
- 1 INTRODUCTION 5
- 2 SCIENTIFIC ADVISORY COMMITTEE FINAL REPORT, JUNE 2016..... 6
 - 2.1 SCIENTIFIC PROGRESS DM2: INERT MATRIX FUELS. 6
 - 2.2 SCIENTIFIC PROGRESS DM3: NITRIDE FUELS..... 7
 - 2.3 SCIENTIFIC PROGRESS in DM4: CARBIDE FUELS 8
 - 2.3 SCIENTIFIC PROGRESS DM1: EDUCATION, TRAINING AND DISSEMINATION 9
- 3 CONCLUSIONS..... 9
- 4 SCIENTIFIC RECOMMENDATIONS AND GUIDANCE..... 10

EXECUTIVE SUMMARY

This is a report compiled by two members of the Scientific Advisory Committee ; Barbara Oberländer (IFE, Norway) and Kastriot Spahiu (SKB, Sweden), for the Coordinator, the Governing Board and the Project Coordination Committee.

SAC members are impressed by the efficiency of the project concerning the management, execution and reporting. The overall impression of the scientific performances within the four domains of the Asgard project is a good one in terms of milestones achieved, scientific results obtained and deliverables produced. These achievements are even more impressive in view of the unexpected shutdown for several months during the second year at one of the important hot labs (NRG). Even though some work could not be performed during the several months shut down of NRG, most of the deliverables have still been realized and only a few have been affected. This has been possible through an effective management by the project coordination committee as well as through the willingness of several partners to be flexible about when and how the work is performed.

1 INTRODUCTION

Bellow follows a short description of the EU-Project Asgard.

COORDINATOR

Chalmers (SE) with prof. Christian Ekberg is coordinating the Asgard project.

The members of the EU-project Asgard (consortium) are presented shortly below.

1 Chalmers, SE	5 Paul Scherrer Institut(PSI), Ch	9 Ceske Vysoke Uceni Technicke v Praze, CZ	13 Institut National de Cercetare-Desvoltare Pentru Tehnologii, RO
2 Forschungszentrum Jülich(FZJ), DE	6 Nuclear Research and Consultancy Group (NRG), NL	10 Kungliga Tekniska Hoegskolan(KTH), SE	14 University of Leeds, UK
3 Instytut Chemii I Techniki Jadrowej, PL	7 Karlsruher Institut für Technologie (KIT), DE	11 Evalion, CZ	15 University of Manchester, UK
4 National Nuclear Laboratory (NNL), UK	8 Commisariat a l'energie atomique et aux energies alternatives(CEA), FR	12 Westinghouse Electric, SE	16 University of Cambridge, UK

MEMBERS OF SAC

Members of the Scientific Advisory Committee (SAC) contributing to the compilation of this report were Dr. Barbara Oberländer (IFE, Norway)-SAC-chairman and Prof. Kastriot Spahiu (SKB, Sweden). SAC members which did not participate in the report compilation are Grigore Horhoianu (Nuclear fuel engineering laboratory, Institute for nuclear research) and prof. Hamit Ait-Abderrahim (SCK.CEN, Belgium).

MANDATE OF SAC

The mandate of SAC is to give scientific recommendations and advice to the project Governing Board (GB) and Project Coordination Committee (PCC).

2 SCIENTIFIC ADVISORY COMMITTEE FINAL REPORT, JUNE 2016.

WORK:

The members of SAC have followed with pleasure and great interest the last ASGARD meetings in UK, RO, DE and SE, the schools and reports/deliverables published on the project's electronic database "PingPong".

SCIENTIFIC CONTENT

The four domains of the ASGARD project include the following scientific topics:

2.1 SCIENTIFIC PROGRESS DM2: INERT MATRIX FUELS.

The work in this work-package includes both the production of inert matrix fuels (CERCER and CERMET) and their dissolution for recycling. Given the high content of inert matrix in these fuels (MgO or Mo), special head end treatments after dissolution are necessary in order to remove the inert material in order to simplify the separation of actinides and to improve the final conditioning of the remaining fission products before vitrification. Electrospray ionisation mass-spectrometry (ESI-MS) has been used successfully to study potential formation of mixed actinide (IV)-Mo species. Such species could be detected for Th(IV) in Mo containing solutions. The investigation of solutions containing Mo and Th at 0.5, 1 and 3 M HNO₃ was performed successfully. The relative abundance of mixed species decreases with decreasing Th concentration and increasing acid concentration.

The separation of Tc from Mo-containing solutions has been studied through static and dynamic tests with a suitable solid phase extractant, A336-PAN. The kinetics was relatively fast, an uptake of 98.1% was reached in 30 min.

The modified procedure proposed for preparation of materials having high content of actinides (up to 50%) via composite materials with polyacrylonitrile (PAN) binding matrix was used to prepare pellets containing minor actinides in the MgO matrix. Dried beads of 80 %MgO and 20% of binding polymer or of a mixture of MgO and CeO₂ were characterised and from the porosity a maximum content of Eu or Ce was calculated when the porosity was filled with an infiltrated concentrated solution of Eu or Ce. The following products were prepared and characterised: MgO pellet prepared from MgO powder, pellet prepared from (MgO+CeO₂)-PAN beads (CeO₂ content 10, 20 or 30%), pellet after infiltration of Eu-nitrate into MgO-PAN, pellet after infiltration of Ce-nitrate into MgO-PAN, pellet after infiltration of (Ce+Eu)-nitrate into MgO-PAN and pellet after infiltration of Eu-nitrate into (MgO+CeO₂)-PAN. The pellets have been characterised by XRD, SEM, HRTEM for homogeneity and the density has been determined.

The internal gelation method has been tested for U-Nd particles with extended Nd-content (up to 81%), which have been characterised by X-ray adsorption spectroscopy

XANES/EXAFS at the European Synchrotron Radiation Facility (ESFR). Data treatment in order to determine U-O and Nd-O bond length as well as oxygen to metal ratio is under way.

At ICHTJ, the reduction of gels obtained by the sol-gel method at higher temperatures (1100 C-1200 C) has been tested and the obtained material was characterised by SEM/EDX and XRD.

The photochemical precipitation of actinides from formate containing solutions has resulted in the precipitation from uranyl and thorium from concentrated solutions, which were converted to oxides. CeO_2 , Eu_2O_3 and mixed $(\text{Ce,U})\text{O}_2$ and $\text{Eu}_2\text{O}_3\text{-UO}_2$ oxides were also prepared and characterised.

The experimental work in this work package is almost finished and the last period has been dedicated to reporting: several deliverables have been delivered and several reports have been prepared and are under internal review. Only WP 2.2 ((Pu,Am) O_2 pellet preparation and dissolution) has a certain delay in this work package due to the unavailability of the actinide laboratory and resulting in two delayed deliverables; most of the due deliverables are submitted or under review. Two Ph.D. theses have been completed in this work package and one of them has been defended in December 2015.

Partners in this domain are: INE-KIT (DE), FZ Jülich (DE), KTH (SE), NRG (NL) CTU (CZ), CTH (SE), ICHTJ (PL)

2.2 SCIENTIFIC PROGRESS DM3: NITRIDE FUELS

The scientific work concerns nitride fuels production both on the dry and the wet route and their dissolution for recycling. The production of fuels by the sol-gel route and the fabrication of nitride fuels, including enrichment of nitrogen in N-15 are important challenges. Challenges are also the dissolution of irradiated nitride fuels, the recovery of N-15 and the fabrication of nitride fuels using N-15 gas.

Dissolution tests of fresh and irradiated nitride fuels have been completed.). High density uranium nitride pellets with variable impurity levels of O and C have been produced at KTH. Pellets of the following nitride fuels have been fabricated: UN, (U,Zr)N, PuN and (Pu,Zr)N.

An N-15 production facility using sulfuric acid was installed and tested at INCDTIM both at atmospheric and up to 2 atm. pressure. Actually, the N-15 recovery tests have been completed and a N-15 conserving manufacturing process has been successfully tested. The N-15 requirements for Gen-IV reactors have also been determined. A stable catalyst for sulphur dioxide production has been identified.

Partners in this domain are: KTH (SE), CTH (SE), NRG (NL), PSI (CH), INCDTIM (RO).

2.3 SCIENTIFIC PROGRESS IN DM4: CARBIDE FUELS

The work includes both productions of carbide fuels by different routes and their dissolution for recycling. As with the nitride fuels there are some issues with the production and recyclability of the carbide fuels. Challenges are carbide fuel design and production, reprocessing of carbide fuels, including molten salt reprocessing, oxidative pre-treatment, direct dissolution as well as studies on carbide fuel pyrophoricity and pellet-cladding interactions.

For carbide fuel production, powder mixing and conventional press and sinter methods have successfully been used at NNL to obtain both dual density pellets and pellets with added Ni as sintering aid. At PSI, microwave internal gelation has been used to obtain excellent microspheres with very narrow size distribution in tests with ceria as a Pu-analogue. The Pu-containing spheres are awaiting allowance from the Swiss authorities and this has caused a certain delay for deliverables D4.1.2. and D4.1.4. Novel carbide production methods have been tested at ICHTJ by using ascorbic acid as a carbon substrate for carbide materials. The conditions for carbothermic reduction have been optimized to give a good control of the stoichiometry of the final carbide and also to reduce the temperature to 1400 C (cf. 1650 C for conventional reduction). The work with carbothermal reduction of various U precursors to obtain uranium carbide and of CTU with carbonization of PAN beads containing U or Eu by the carbothermal reduction of oxides obtained UC₂ and UC carbides is now completed.

CEA has tested fuel cladding interactions for TiC cladding, finding good performance of TiC cladding and tested carbide fuel pyrophoricity in low oxygen atmospheres and air. PSI has tested pre-oxidation of UC fuel in CO₂ at 1000 C, removing significantly carbon content, important for further dissolution. CEA has performed tests on direct dissolution of uranium carbide beads. The organics generated during carbide dissolution could be destroyed by using a boron doped diamond electrode in conjunction with a Ag(II) catalyst down to 0.6% of the original carbon content in the UC pellet. Solvent extraction experiments have been performed to demonstrate that the residual organics maintain Pu in the aqueous phase suggesting that organic complexation interferes with solvent extraction. When the organics are destroyed with the electrode, the extraction of Pu is roughly the same as when no organics are present. NNL has tested the reactivity of SiC and TiC in molten CaCl₂ based salts and shown that conversion of SiC to Ca-silicates is feasible in CaO+CaCl₂ melts. A test of the chemical pre-treatment of carbide fuels in molten salts has been carried out with three types of TRISO fuel with alumina kernel, thermally treated for three hours at 1000 C in molten chlorides. Characterization by SEM/EDX shows stripped coated layers of TRISO particles, confirming the oxidation degradation of the fuel under the applied thermal conditions. The β-SiC based cladding dissolved in molten CaCl₂ containing 12.5% CaO at 1000 C for 12 h, demonstrating that the molten salt method works for SiC cladding. A pellet of uranium carbide has been treated thermally for 30 min at 800 C in molten CaCl₂-NaCl with 10 % CaO under argon

and gamma peaks of U-235 detected by high resolution HPGe demonstrate transfer of uranium from the pellet to the melt.

Electrolytic dissolution of UC giving U(III) in molten LiCl-KCl eutectic has been tested successfully, producing black particulates, most probably insoluble carbon materials, which float on the melt surface. Electrodeposition of U will be tested with larger quantities of U material and also attempts to characterize the black material. Some delays in the deliverables concerning carbide pre-treatment in molten salts are due to difficulties in obtaining access to an active SEM facility and will be completed in 2 months.

Partners in this domain are: PSI (CH), CEA (FR), NNL (UK), ICTHJ (PL), UMAN (UK), UCAM (UK)

2.3 SCIENTIFIC PROGRESS DM1: EDUCATION, TRAINING AND DISSEMINATION

A working mobility scheme has been established with 2 persons founded to work in other laboratories and several publications and other dissemination actions have been achieved during the whole project. Summer schools have been organized by KTH, CTU, CEA and EVALION. E-learning modules and courses have been organised by CTU and INCDTIM. The Second ASGARD International Workshop was organised in connection to the Topfuel 2015 Conference in Switzerland. The Second AGARD Summer School was run as a joint SACSESS, ASGARD, and CINCH II school with a topic “Working with Pu” at Chalmers in May 2015.

Partners in this domain are: CTH (SE), CTU (CZ), NNL (UK), KTH (SE), Westinghouse (SE), INDCTIM (RO), KIT-INE (DE), PSI (CH), NRG (NL), CEA (FR) EVALION (CZ).

3 CONCLUSIONS

Within the ASGARD project the overall impression of the scientific performances within the four domains is a good one in terms of milestones achieved, scientific results, dissemination reported, networking and training of young scientists and deliverables produced.

4 SCIENTIFIC RECOMMENDATIONS AND GUIDANCE

Domain 1 (Project management, education training and mobility, dissemination, exploitation and networking. **Coordinator** is Teodora Retegan, Chalmers.

SAC Observations and Recommendations: Activities in domain are following the progress plan. SAC members can contribute in reviewing scientific papers resulting of the ASGARD project.

Domain 2 to Domain 4 (Oxide fuels Coordinator is Eva de Visser-Tynova, NRG alt. Andreas Geist, KIT, **Nitride fuels Coordinator** is Janne Wallenius alt. M. Jolkkonen, KTH and **Carbide fuels Coordinator** Marc Sarsfield NNL)

SAC Observations: Commissioning of suitable lab facilities is in good progress or completed. Some delays are reported, mainly related to difficulties in work with highly active samples. Activities, milestones and reports are following the general progress plan. The NRG partner had a temporary institutional situation with a temporary shut-down of the nuclear laboratories, which was appropriately handled by PCC minimizing the consequences.

Interesting scientific results according to the ASGARD project plan are on fuel fabrication and dissolution of carbide fuels, oxide inert matrix and nitride fuels.

Fuels as obtained by synthesis of carbide fuel by co-milling of UO_2 and graphite & gradient pellet pressing seem presently promising candidates for high temperature, Gen IV fast reactor concepts.