

INFORME **DFN/RA-02/SP-23**

REF. EXTERNA

**Innovación en tecnología de fabricación nuclear.
Innovation Of Nuclear Manufacturing Technology (IONMAT).**

DIVISIÓN DE FISIÓN NUCLEAR


Ciemat Centro de Investigaciones
Energéticas, Medioambientales
y Tecnológicas

CSV : GEN-b5ea-ae89-1a43-4f71-c9fb-bee1-a341-084f

DIRECCIÓN DE VALIDACIÓN : <https://sara.ciemat.es:8443/csv/CsvRecoverService?csv=b5eaae891a434f71c9fbbee1a341084f>

Código seguro de Verificación : GEN-b5ea-ae89-1a43-4f71-c9fb-bee1-a341-084f | Puede verificar la integridad de este documento en la siguiente dirección : <https://sara.ciemat.es:8443/csv/CsvRecoverService?csv=b5eaae891a434f71c9fbbee1a341084f>



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 1 DE 29
	REF. EXTERNA	REVISIÓN 0

TITULO:

Innovación en tecnología de fabricación nuclear. Innovation Of Nuclear Manufacturing Technology (IONMAT).

AUTORES:

N. Rodríguez-Villagra, S. Fernández, A. Núñez, J.M. Elorrieta, A. Milena-Pérez, L. J. Bonales, L. Gutiérrez, L. Serrano, S. Durán, L. Anta, H. Galán

ABSTRACT:


Uno de los mayores retos de la industria nuclear actual es el diseño de nuevos tipos de pastillas de combustible que garanticen al mismo tiempo la calidad y los requisitos regulatorios según las especificaciones del combustible, en el marco de la gestión del combustible nuclear. La propuesta del proyecto IONMAT pretende llevar a cabo actividades de investigación que ayuden a mejorar el reto de obtener una energía nuclear segura, sostenible y limpia para la sociedad. Este proyecto, financiado por la Agencia Estatal de Investigación (AEI) con referencia: PID2021-124913OA-I00, se basa en la utilización de sistemas tradicionales y avanzados de fabricación de combustibles nucleares cerámicos. La propuesta se divide en tres tareas fundamentales. En primer lugar, se plantea la fabricación de análogos de combustible nuclear irradiado y el desarrollo de métodos de conversión por vía húmeda de actínidos en matrices inertes. En segundo lugar, se plantea el desarrollo de combustibles tolerantes a accidentes (ATF) capaces de mejorar el rendimiento (permiten aumentar el grado de quemado) y seguridad, complementado con estudios de degradación. En último término, se realizará una caracterización microestructural y morfológica exhaustiva de los materiales obtenidos, evaluando asimismo la efectividad de los procesos de fabricación aplicados.

	NOMBRE	FIRMA	FECHA
Preparado por:	N. Rodríguez		
Revisado por:	H. Galán		
Conf por A. C.:	S. Fernández		
Aprobado por:	E. González Romero		

FECHA DE EMISIÓN

--	--	--	--	--	--



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 2 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

Index

1. INTRODUCTION4

2. BACKGROUND, CURRENT STATUS AND JUSTIFICATION OF THE PROJECT5

 2.1. GENERAL BACKGROUND AND CURRENT STATUS5

 2.2. BACKGROUND OF THE RESEARCH GROUP6

 2.3. JUSTIFICATION OF THE PROJECT8

3. OBJECTIVES, METHODOLOGY AND WORK PLAN11

 3.1. STARTING HYPOTHESIS AND GENERAL GOALS11

 3.1.1. Specific Goals12

 3.2. SCIENTIFIC METHODOLOGY AND DESCRIPTION OF WORK13

 3.3. RESOURCES20

 3.4. CHRONOGRAM22


4. SCIENTIFIC-TECHNICAL IMPACT, SOCIAL AND/OR ECONOMIC IMPACT FROM THE RESULTS.
INTERNATIONALIZATION.23

5. ACKNOWLEDGEMENTS25

6. REFERENCES26

Código seguro de Verificación : GEN-b5ea-ae89-1a43-4f71-c9fb-bee1-a341-084f | Puede verificar la integridad de este documento en la siguiente dirección : <https://sara.ciemat.es:8443/csv/CsvRecoverService?csv=b5eaae891a434f71c9fbbee1a341084f>



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 3 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

Index of tables

Table I CIEMAT facilities available within the HLWU.....	21
Table II Methods and techniques available within the HLWU.	21
Table III Chronogram of IONMAT project.....	22

Código seguro de Verificación : GEN-b5ea-ae89-1a43-4f71-c9fb-bee1-a341-084f | Puede verificar la integridad de este documento en la siguiente dirección : <https://sara.ciemat.es:8443/csv/CsvRecoverService?csv=b5eaae891a434f71c9fbbee1a341084f>




Ciemat DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 4 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

1. INTRODUCTION

This document presents and details the research proposals of the High Level Waste Unit (HLWU) of CIEMAT within the framework of the PID2021-124913OA-I00 project (IONMAT project) funded by the Spanish State Research Agency (*Agencia Española de Investigación, AEI*).



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 5 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

2.BACKGROUND, CURRENT STATUS AND JUSTIFICATION OF THE PROJECT


2.1.GENERAL BACKGROUND AND CURRENT STATUS

About 20% of the total electricity in Spain has been produced by nuclear energy in the last ten years, contributing with a solid industrial system to the safety of energy supply and the reduction of GreenHouse Gas (GHG) emissions. In general, nuclear fuel safety has steadily improved in recent years. In this context, it is worth mentioning that due to possible economic stresses caused by the COVID-19 pandemic, there could be an impulsion to adopt solutions that hinder sustainable development and intensify the impacts on climate change. Fundamental knowledge on technologies that can lead towards a green recovery is required (1). Several options exist to decrease GHG emissions caused by energy production, one of them is nuclear energy. Over the past 50 years, nuclear energy production has reduced CO₂ emissions, being the world's second-largest source of low-carbon electricity behind hydropower. This mature and proven technology has many sustainable options for a waste management strategy. In addition, a range of small modular reactor (SMR) and advanced reactor designs are presently under development. Attending to UNECE (The United Nations Economic Commission for Europe) (2), "*nuclear energy is complementary to renewable energy sources*" to achieve "*decarbonized electricity systems at low cost to consumers – as has been proven by France and Sweden*". Even more, UNECE Expert Group also referred that nuclear power technology is progressing with a range of new reactor technologies. High Level radioactive Waste (HLW) from nuclear energy production needs to be responsibly managed before disposal. In this sense, innovations such as new fuel designs can expand the efficiency of nuclear power plants (NPP). There is an immense potential to support the development of sustainable nuclear energy.

Closely, the mitigation and control of fuel defects are driven by economic and safety issues. With the objective of "*zero fuel failures*", a number of attempts have been made by all sectors of the nuclear industry (suppliers of products, vendors, engineering companies, electricity companies, agencies and regulators) regarding the identification and reduction of the safety risks directly associated with its design; as well as to technological innovation and optimization of new processes (including existing ones) that can be part of the Advanced Nuclear Fuel Cycles (ANFC). The latter would include the "*recycling*" strategies of the radiotoxic inventory that remains in the nuclear fuel after its irradiation in the NPP.

For the aforementioned reasons, one of the biggest challenges of the nuclear industry is the design of new types of fuel rods and pellets, through different R&D&i programs and in collaboration with research centres, while simultaneously guaranteeing quality to assure the regulatory requirements according to the fuel specifications under the management frame for nuclear fuel (3). In order to reach this goal, the national context regarding the management of HLW and Spent Nuclear Fuel (SNF) has to be taken into account. Spanish regulation (4) defines the concept of radioactive waste as "*any waste material or product for which no further use is foreseen and that contains or is contaminated by radionuclides in concentrations or levels of activity higher than those established by the Ministry of Industry, Tourism and Commerce (MITYC), following a report by the Nuclear Safety Council (CSN)*".



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 6 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023


The management of the HLW in Spain has also been decided by means of centralized temporal interim storage (ATC or CTSF, Centralized Temporary Storage Facility), or in a number of locations, for about 60 years, after which the best solution for the final disposal will be implemented. The international consensus is that Deep Geological Repository (DGR, English acronym of *Almacenamiento Geológico Profundo*, AGP) is the best technical option for the final disposal of HLW in those countries which have decided not to further use irradiated fuel. An alternative to the direct DGR of SNF is the ANFCs involving the Partitioning and Transmutation strategies (P&T) by using fast reactors (Gen-IV like the sodium or lead cooled fast reactors) and Accelerator Driven Systems (ADS) or SMR. These ANFC strategies would allow a significant reduction of the radiotoxicity and the heat load of the ultimate waste and therefore an enhanced safety of their management; therefore it cannot be ruled out based on new technological solutions developed in the uncertain prospect. For this reason, the European Commission has recently launched in 2019 a new *Strategic Energy Technology (SET) Plan* which identifies a list of competitive and sustainable low carbon energy technologies to be developed and deployed in Europe (5). In the SET Plan, a list of current priority areas are identified, and includes “*safe management of radioactive waste and decommissioning*”, and “*efficiency and competitiveness of current and innovative technologies* “. Nuclear waste reduction could be implemented in each country or centralized in a few countries with long-term nuclear deployment. To properly choose the best technology and option for HLW, it is essential to maintain R&D in these advanced systems during, at least, 60 years of temporary storage. All this, without losing the perspective of a definitive location.

2.2.BACKGROUND OF THE RESEARCH GROUP

Our research group in CIEMAT’s Nuclear Fission Department has a long trajectory in the field of nuclear fuel related to Spent Fuel Stability under interim and final disposal conditions, and P&T in the last 20 years. Particularly, the High Level Waste Unit (HLWU) has reached recognition at the international level over the past years, which evidences its skilful status for the achievement of this R&D program. We are involved in European Union Programs as well as in Spanish R&D programs financed by ENRESA (*Empresa Nacional de Residuos Radiactivos S.A.*) and National Research Programs (SYTRAD2), which finished in 2021. The HLWU of CIEMAT participates in:

- Development of ANFC for assisting the radioactive waste management and improving the sustainability of nuclear fission as an energy source;
- Support and provision with the scientific and technical expertise required for adequate radioactive waste management and long-lived radionuclide partitioning processes;
- Achievement of the best characterization feasible of the initial state of SNF and HLW in those variables that might become critical during conditions of dry storage or final disposal;
- Investigation into SNF alteration, behaviour and evolution in simulated interim storage or final disposal conditions. The experimental activities have been so far focused on providing a thorough characterization of surrogate fuel samples submitted to different boundary conditions and improving the knowledge and its performance assessment relative to its short, medium, and long-term stability.



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 7 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023


The role of CIEMAT is to offer scientific-technical support for current and future decision-making in Spain, by means of participation in both national and international R&D projects. In this respect, the HLWU has been working for suitable management of SNF, seeking sustainability and social acceptance of the nuclear fuel cycle, in collaboration with ENRESA, and has provided helpful information on waste management. In recent years, the greatest efforts of the HLWU have been focused on the physico-chemical characterization of radioactive waste and the development of ANFC that go hand in hand with innovation in the field of new types of fuels. Moreover, the HLWU counts with a unique facility in Spain for nuclear material handling that valorizes the singular experimental installations of CIEMAT. The research group has facilities to manipulate/analyze long live radionuclides (a radioactive laboratory IR-30 and IR-35, being the last one under licensing process). On the other hand, conventional laboratories (named “cold” laboratories) are also available for surface characterization techniques with no radioactive isotopes or “*exempt*” radioactive material, dissolution and fabrication tests, as well as the synthesis, purification, and cold analysis of organic ligands included in all radionuclide processes of separation.

Currently, the main scientific-technical areas of HLWU include the next research lines (including relevant publications of the research group during the last ten years):

- **Fabrication** and characterization of advanced fuels, high density fuels, irradiated fuels, and analogues of irradiated fuels. Development of innovative processes and preparation methods of ATF materials and ceramic transmutation blankets (waste minimization targets) (6-12).
- **Characterization** of irradiated nuclear fuel and high level wastes (both packaged and conditioned solid wastes) and their stability under temporary or repository storage conditions, focussing on its alteration and its potential subsequent radionuclides release (13-26). Behaviour and safety of SNF at present reactors or of enhanced safety (ATF) during transport, dry storage, or final disposal (11, 12, 17-19, 21-23, 27, 28).
- Research and optimization of new liquid-liquid **extraction processes** for the recycling of long-lived minor actinides (MA) in two scenarios: heterogeneous recycling (Am/Cm) to obtain dedicated fuel or targets with a high proportion of An; the recovery of all An together (Pu, Am, Cm, Np) for homogeneous recycling as part of mixed oxide fuel (UO₂-An) keeping a low percentage of An. The long-term behaviour of new extraction systems and safety requirements at the industrial scale are comprised, and also optimization of conversion systems (29-39).

On the other hand, CIEMAT, as energy reference centre in Spain, has a proven experience in R&D on safety of the SNF, particularly during transportation and storage in the interim dry storage (the CTSF) foreseen in the present and new “*ENRESA's Research and Development Plan*”, PGRR, sanctioned by the government. ENRESA's 6, 7 and 8th Research and Development Plans (40) explicitly include activities related to the understanding of the physico-chemical properties of the components of radioactive waste, in addition to the evolution over time and the impact of irradiation history of HLW and SNF on the CTSF. Activities related to the characterization of SNF and its behaviour under temporary or repository storage conditions, comprise fresh nuclear fuel and SNF surrogate materials. It should be mentioned that, although focused on the fuel types used in Spanish NPPs, some attention has to be paid to ATFs. In particular, in the field of potential improvements on



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 8 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

fuel design and which factors affect the UO_2 matrix alteration under a range of conditions. These innovative fuels (ATF) are being investigated, by modifying their physico-chemical properties to improve their thermo-mechanical resistance. Besides, surrogate samples are needed to develop measurement procedures that can be applied to investigate chemo-physical, microstructural, and mechanical properties of SNF.

Our participation in international projects: FIRST-NUCLIDES (ID: 295722), REDUPP (ID: 295722), DISCO (ID: 755443), EJP-EURAD (ID: 847593), PATRICIA (ID: 945077), and GENIORS (ID: 75517); and in national programs: OCATS-ENRESA (ID: 079-CO-IA-2018-0007), SYTRAD-2 (ID: ENE2017-89280-R), SOPSEP-ENRESA (ID: 0079000269), ACESCO-ENRESA (ID: 079000189), CeluCem (ID: CTQ2011-28338), SYTRAD (ID: ENE2014-55140-R), among others, provided this team with the know-how and experience to identify the relevance and potential impact of the progress in nuclear data evaluations for the fuel cycle and the final disposal.

IONMAT project would be the first step to apply in the type of "*Oriented Research*", specifically in the priority topic "*Climate, Energy and Mobility*" described in the "*State Research Plan*".

2.3.JUSTIFICATION OF THE PROJECT

Most of the above mentioned research lines require producing representative surrogate fuel samples to be used, mastering a comprehensive set of experimental techniques, and setting up suitable devices to simulate the postulated experimental conditions. Originally, the target framework for doing so is reproducing and improving industrial manufacturing methods of nuclear fuel, paying special attention to:


- Uranium- or Thorium -based (fertile) fuels (MOX , $(U,Pu)O_2$ with MA);
- Candidates for the so-called Inert-Matrix Fuels (IMF) to host Pu and MA from reprocessing; ATF;
- SNF surrogate pellets, with physico-chemical features similar to that reported for irradiated nuclear fuel at medium and high burn-ups, including microstructural and mechanical characterization.

Fuel fabrication processes by traditional ceramic processing of powder with UO_2 , as conventionally applied on an industrial scale, involve obtaining high-density pellets of fresh fuel and irradiated fuel analogues, necessary to mimic micro and macroscopic fuel properties. The production of fuel pellets using dry powder technology has been performed at the HLWU facilities by two manufacturing approaches:

- 1) *Dry method*, traditionally used in the industry for the preparation of UO_2 fuel pellets (11);
- 2) A two-step process in which the inner ring of the pellet was obtained by axial pressing and the outer ring by *slip casting* using well-dispersed, concentrated aqueous suspensions, as an analogue preparation of ceramic nuclear fuel pins (6).

In this framework, the challenge is to optimize these processes in order to reach four **fundamental objectives**:



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 9 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

- 1) To implement processes free of radiotoxic dust, avoiding the risks inherent to the handling of powdered material; in this sense, different concepts are today feasible, based on co-precipitation operations, matrix infiltration, thermal co-denitration, or other related.
- 2) To obtain ATF fuels with properties that improve their behaviour during irradiation in thermal reactors, such as decreasing the release of fission gases and fuel pellet-cladding interaction (12).
- 3) To condition waste streams coming from selective recovering of MA and other long-lived fission products (FP) for a successful conversion method to suitable solid matrices, in an appropriated form of advanced/innovative fuels for their reincorporation to the nuclear fuel cycle in future NPP (candidates for transmutation targets). One of the tentative options of IMF could be in a ceramic form to burn these actinides (An) in existing Light Water Reactors (LWR) or GenIV reactors.
- 4) To study the effect of the composition during the conversion process, from streams to solid forms, to explore the suitability with the previous separation process. The conversion needs to be applicable to industrial processes and economically viable, which in the end results in remote-controlled automated equipment for fuel reprocessing and fuel pin manufacturing. It is known that the use of powder metallurgy to produce oxide fuel is the most mastered process and can be readily implemented. However, the great complexity in managing dust at all stages, where powder form is used (grinding, sieving, loading the presses), must be taken into account (41). This project would be the first step, on a laboratory scale, for the safe development of nuclear materials fulfilling the industry requirements.


Looking toward the sustainability of nuclear energy, the recycling of radionuclides aims to reduce the volume, the inventory and the long-term radiotoxicity of irradiated fuels before final disposal. It will also contribute to a better and complete exploitation of the inherent power in the waste. Advanced fuel design concepts are being investigated in order to minimize the MA stockpiles to be managed (42, 43) and to maximize the transmutation rate and energy yield. The public and stakeholders are more likely to accept this management option if they anticipate economic benefits. A number of IMFs compositions and geometries are considered to transmute the MA (3). "Inert" concept means low thermal neutron absorption cross-section of the elements constituting the matrix. IMF requirements are related to the term "durability" owing to irradiation or physico-chemical processes. The following general criteria are used to select appropriate IMFs:

- 1) Allow a high density of fissile elements;
- 2) Good compatibility with coolant (up to 500°C) and structural materials;
- 3) Good radiation stability (α -, β -, γ -radiation and heavy ion irradiation avoiding phase transformations);
- 4) Low neutron absorption and swelling; and
- 5) Keep suitable physical, mechanical and thermal properties.

In addition, other requirements for IMF are also needed:

- a) Fabrication processes to be implemented in hot cells with heavy shielding and remote handling, and, if possible, avoiding powder handlings should be economically viable and competitive;




 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 10 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

Irradiated IMF should be compatible with the fuel reprocessing, if MA are going to be recycled back, and;

- b) The selected matrix should provide a first fuel containment barrier against possible leakage of FP and other radionuclides.

Finally, in case the IMF final option will be new reprocessing, the matrix should be soluble in HNO_3 to allow a multi-recycling scenario; however, in case of direct disposal, IMF should have enough stability to ensure the very slow rate of long-term corrosion under repository conditions and retention of trace elements because of low diffusivity. The need for DGR remains in spite of the "closed nuclear fuel cycle" and the role of P&T. There are several candidates of fuel types being examined as IMF: *Homogeneous* distribution of MA throughout the fertile fuel phase (Solid solution) or *Heterogeneous* fuels (composite fuels, i.e. CER-CER: composite ceramics of MA embedded in another ceramic, both insoluble in each other; CER-MET: composite ceramics embedded in a metal phase like Mo). Examples of fuel target matrices as ceramic oxides in solid solutions are zirconia, ceria, yttria and magnesia (44, 45).



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 11 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

3.OBJECTIVES, METHODOLOGY AND WORK PLAN

The project is part of the Program "*Proyectos de Generación de Conocimiento*" (PEICTI: 2021-2023), area of "*Energía y transporte*" (EYT), subtask "*Energía*" (ENE). In the "*Plan Estatal de Investigación Científica, Técnica y de Innovación 2021-2023*" one of the strategic actions is "AE5: Clima, Energía Y Movilidad". Specifically, it is part of the action line (1) climate change and decarbonisation, in the area of "*eficiencia energética y descarbonización, nuevas fuentes de energía sostenible*".


3.1.STARTING HYPOTHESIS AND GENERAL GOALS

This proposal is based on the hypothesis that the use of safe and profitable production of nuclear energy entails materials adapted to existing consumption needs and social demand; and the upcoming future of advanced NPP. The accident at the Fukushima Daiichi NPP has revealed the need for improved safety margins of current nuclear energy, which can be ensured by the development of ATF. This fact, together with the need for improved fuel performance in the reactor, results in an increased fuel burnup as well as improving its alteration performance during and after irradiation. In addition, it has triggered us to rethink the large amount of highly radioactive waste that would need to be stored or even better, to be "*recycled*" within P&T strategies to reduce nuclear wastes. "*Recycling*" long-life radiotoxic elements (transuranic ,TRU) in suitable matrices for use in advanced reactors, would result in a benefit for present generations (with the consequent reduction in the volume and inventory of MA in waste to be stored; reduction of the time interval required to control the disposal), minimizing the risks for future generations. Recycling TRUs by means of P&T requires an integrated effort at different scales.

Therefore, **the main goal of this project** is to contribute to the design of a global management strategy at the Spanish and European levels, finding a solution to the minimization of the risk on HLW management. The general objective is to evaluate potential ways of obtaining nuclear fuels, simulated irradiated fuels (not irradiated) and doped matrices. To follow through with the development of this large-scale strategy, in this project, we will focus on two relevant aspects for closing the circle on the applicability of advanced reprocessing strategies and future reactor designs.

On one hand, the strategic research line "Product Conversion" essentially implies the link between separation and fuel fabrication of new concepts proposed in the ESNII initiative (46). On this task, it will be addressed the conversion study of the actinide, lanthanide, and FP streams (coming from the hydrometallurgical chemical separation processes of SNF) to oxide solid forms, whatever its ultimate goal it was (both for waste disposition and recycling of some long-lived radionuclide fractions back into transmutation fuels or targets). On the other hand, it will be approached the design of the ATF concepts proposed in the strategic agenda of CEIDEN platform and SNETP initiative (47). Fabrication of doped UO₂ with inactive elements (FP and lanthanides, Ln) would be included within this line to simulate physico-chemical properties of irradiated fuel.



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 12 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023


3.1.1. Specific Goals

IONMAT project proposal intends to carry out research activities that could help to improve the challenge of obtaining secure, sustainable, and clean nuclear energy for society. This project is based on the use of advanced systems to fabricate ceramic nuclear fuels with a dual purpose: 1) obtaining new types of considered pellets, which are more efficient for energy generation. Evaluation of the influences and variables involved in wet manufacturing and conversion from the waste stream of aqueous reprocessing, considering the weak acid resin process and gellification process. Viability in the manufacture of solid fuel matrices of interest and; 2) Production and characterization of irradiated nuclear fuel surrogates. Mimic the physico-chemistry of conventional irradiated fuel based on UO_2 , in order to be used in corrosion and degradation experiments. Therefore, the specific objectives of the IONMAT project are described next:

1. **Fabrication and characterization of SNF surrogates**, improvement of industrial manufacturing methods of nuclear fuel, paying special attention to ATF and to SNF surrogate's pellets, with physico-chemical features similar to that reported for irradiated nuclear fuel at medium and high burn-ups, including microstructural and mechanical characterization (participation in EURAD H2020 EURATOM project). Implementation of **advanced and innovative conversion techniques** to feed and support the integration studies related to reprocessing and fabrication routes (supporting the participation in PATRICIA H2020 EURATOM project). Application of the know-how about extraction process (composition, stability, etc) to the obtaining of UO_2 -An matrix and IMF. For reaching this goal, different co-precipitation methods will be explored.
2. The development of **manufacturing processes to produce of ATFs** is of utmost importance. To a good extent, the relevance of experimental observations in CIEMAT's laboratories depends on a suitable representation of those fuels, as presently CIEMAT cannot work with irradiated fuels of any kind. Samples produced with simulated features of SNF will be used for more realistic studies on SNF corrosion behaviour. For reaching this goal, different methods of pins manufacturing will be developed via powder metallurgical routes, fabrication route of co-precipitation of uranyl cations with other cations present in the irradiated fuel matrix; followed by calcination and sintering, optimizing atmospheres and temperatures. The materials proposed for this work are pure UO_2 and doped UO_2 . Dopants to be used are: Gd (as burnable neutron absorber), Rare Earth oxides (Eu, La, Nd) as non-radioactive surrogate metals that are representative of uranium, and FP, Ce (plutonium and americium surrogate), Cr (used as dopant in ATF), Zr (assuming Zircaloy-Fuel gap closure) or other potential inert matrices (MgO, CeO_2 , ZrO_2).
3. **Solid characterization**: Samples prepared will be described, evaluated and discussed in terms of microstructure and homogeneity.

To go further in the development of these global approaches, in IONMAT we will focus on two relevant aspects for the applicability of advanced fabrication technologies in the field of reprocessing strategies and therefore, future reactor designs; and because of the inconveniences for handling radiotoxic SNF, wet routes will be also applied for ATF and simulated irradiated fuel (microstructure, burnup and inventories). On one hand, it will be addressed a state of the knowledge on novel technologies for nuclear fuel production as well as its viability for remote fabrication of MA-bearing fuel to be applied as an industrial process. Then, the design of fabrication



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 13 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

of MA bearing fuel system will be approached, following the one proposed in the ESNII-SNETP initiative (46). The aforementioned particular objectives represent improvements that these developments would entail directly from setting-up of new methodologies or the modification of existing ones, including new research lines related to the improvement of nuclear fuel properties. In the stability studies of highly active radioactive waste, the samples produced would serve as support for the experiments that simulate the conditions of a CSF and DGR. As follows, materials will be tested under a variety of conditions and parameters (dry, wet, partial oxygen pressure, temperature, groundwater, etc.) for a better understanding of the integrity and behaviour of radioactive waste. The systematization of the recycling of nuclear material, essentially U, will allow better sustainability of inner resources and self-sufficiency for manufacturing SNF analogues through a highly qualified technical support staff.

3.2.SCIENTIFIC METHODOLOGY AND DESCRIPTION OF WORK

In concordance with specific goals, the current project focuses on three sublines of research that are defined to succeed in fulfilling the above-mentioned objectives:


1. **Task 1:** Fabrication of irradiated nuclear fuel analogues as pellets. Study and development of actinide conversion by the wet route. Influence of fabrication method on the stability of the final fuel pin.
2. **Task 2:** Fabrication of Advanced Tolerant Fuel pellets and degradation studies.
3. **Task 3:** Characterization and examination of the properties of irradiated nuclear fuel analogues.

The current project focuses on recovered MA and FP from both heterogeneous and homogeneous recycling, advanced fuels and surrogates of irradiated fuel. The three purposes have in common the utilization of “nuclear manufacturing” processes. To understand and improve the methodology, innovative systems are ongoing and lay in the scope of CIEMAT capabilities. Based on previous results, first, the work will be focused on extended studies involving the “dry” powder-pellet route, which are needed to understand and compare the physico-chemical properties by means of a simple way. Changes in powder production due to co-precipitation will likely include the production of an homogeneous distribution of chemicals. Therefore, the possibility of conversion and recycling of MA and FP will be explored. To further the demonstration of these fabrication processes, the relevance of liquid-liquid extraction processes in the recovery of MA must be understood and controlled. Consequently, in the last part, the effort will be focused on a detailed and comparative study of the influence of alternative routes on important physico-chemical properties.

Task 1: Fabrication of irradiated nuclear fuel analogues as pellets. Study and development of actinide conversion by wet route. Influence of fabrication method on the stability of the final pin.

To improve the knowledge of SNF performance in storage systems, the impact of oxidizing/reductant environments and the chemical systems involved must be assessed to clearly establish their safety, not only during a normal storage operation, but also in case of mal-operation, to identify any unexpected behaviour. During interim storage and during transport to the final disposal facility, the SNF will have a completely different structure to non-irradiated fuel. Nuclear



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 14 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

fuels are subjected to high levels of radiation, which results in substantial modifications of the initial fuel microstructure such as damage in the material, local defects like interstitials, loops and vacancies. During fuel operation, in the central sections of the fuel pellet (at a higher temperature) grain growth, porosity build-up and larger gas release will occur. On the contrary the original microstructure transforms into a nano-porous matrix highburn-up structure (known as the rim effect) on the periphery. Therefore, it is necessary to produce suitable non-radioactive analogues that closely resemble nuclear fuel in terms of crystallography and microstructure so that we can gain a better understanding of how changes in the sample surface affect.

The materials proposed for this work will mimic: a) chemical composition; b) pellets with radial porosity to reproduce the “*High Burnup Structure*” observed in irradiated fuels; c) chemical fuel-cladding interaction by studying UO_2-ZrO_2 systems.

Regarding the conversion process (conversion ratio) which involves demonstration of co-precipitation on a significant scale, pellet fabrication prior to industrialisation and guaranteeing a high homogeneity of the pellet will be done. Therefore, manufacturing of CER-CER (ceramic-ceramic) will be performed by using inert matrix material suitable for GenIV reactors (transmutation blankets).

The work plan destined to get on task 1 objectives will be divided in three main subtasks:

1. **Subtask 1.1.** Production and characterization of irradiated nuclear fuel surrogates by the dry powder method.

The methodology to be applied will consist of a classic powder metallurgy system. It will be focused on mimic doped- UO_2 pellets by means of grinding / sieving, pressing green pellets and after calcining and sintering at high temperature and under controlled atmosphere conditions, pellets will be obtained;

The materials proposed for this subtask are:

- Pure UO_2 .
- Doped UO_2 pellets by using the corresponding non-radioactive surrogates of FP and Ln / MA.
- Ethylene-bis-stearamide (EBS, $C_{38}H_{76}N_2O_2$) as both binder and lubricant.


Parameters and conditions to be tested:

- Effect of additive content and dopant: variable up to the solubility limit.
- Sintering atmosphere: reducing ($5\%H_2/N_2$).
- Sintering temperature/time: $1675^\circ C$ for 1-10 h.
- Pressing Pressure (uniaxial) : between 700 and 1000 MPa.

Techniques:

- Pellet fabrication techniques: furnace, polisher, mixer mill and uniaxial press.
- Characterization techniques of starting materials and obtained pellets: particle size (laser diffraction), geometric and immersion density measurements (Archimedean test), heat



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 15 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

treatments with atmosphere control (thermogravimetry TGA, differential scanning calorimetry DSC), X-ray diffraction (XRD), specific surface area BET by physisorption (nitrogen), optical and scanning electron microscopy (SEM) and Raman spectroscopy.

Milestone 1.1: to obtain doped- UO_2 sintered pellets by dry route.

2. Subtask 1.2. Radial Porosity simulation.

Another key issue is the role of the porosity on the physical properties of irradiated fuel, with changes in its properties as a consequence of the fission gas behaviour and the disappearance of the grain structure on the fuel periphery, in relation to a strong increase of the porosity in the rim. The analogue must also have a microstructure similar to a typical UO_2 SNF porosity: 95%TD (theoretical density) in the central part of the fuel pellet and 75%TD in the rim, in the outer part of the pellet. Tailored porosity fabrication devoted to the optimisation of different microstructures will make use of a pore former (i.e. U_3O_8 or fructose) to favour the creation of interconnected porosity.

The methodology to be applied will be a combination between classic powder metallurgy and wet system as slip casting.

The materials proposed for this subtask are:

- Pure UO_2 .
- Pore former U_3O_8 .
- Ethylene-bis-stearamide (EBS, $\text{C}_{38}\text{H}_{76}\text{N}_2\text{O}_2$) as both binder and lubricant.
- Ammonium salt of a polyacrylic acid PAA (Duramax D3005, Rohm & Hass, PA, USA).

Parameters to be tested:


- Porosity (%) in a single specimen: 75-95%TD.
- Radial porosity in a single specimen: 95%TD (inner) and 75%TD (outer).
- Sintering atmosphere: reducing ($5\%\text{H}_2/\text{N}_2$).
- Sintering temperature/time: 1675°C for 4 h.
- Pressing Pressure (uniaxial) : between 700 and 1000 MPa.

Techniques:

- The inner green cylinders are obtained by pressing. For the outer ring, the slip casting technique.
- Pellet fabrication techniques: rheometer, ultrasonic homogenizer sonicator, propeller stirrer, mixer-homogenizer furnace, polisher, mixer mill, uniaxial press.
- Characterization techniques of starting materials and obtained pellets: particle size (Laser Diffraction), geometric and immersion density measurements (Archimedean test), heat treatments with atmosphere control (TGA), XRD, BET, SEM and Raman.

Milestone 1.2: To provide adequate UO_2 sintered pellets simulating radial porosity found in real irradiated fuels.



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 16 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

3. **Subtask 1.3.** Preparation of mixed oxide fuels containing U.

Preparing an oxide fuel comprising uranium and surrogates of actinide and/or lanthanide element by two methods: 1) Preparing a feedstock solution consisting of a nitric solution containing actinide and/or lanthanide nitrates and uranium in the form of a hydroxylated uranyl nitrate compound/ uranyl cations with other cations contained in the irradiated fuel matrix; 2) Passing solution on a cation-exchange resin including carboxylic groups, whereby the actinide and/or lanthanide in the cationic form and the uranium in the uranyl form remains fixed to the resin; and thermally processing resin in order to obtain fuel.

The materials proposed for this subtask are :

- Pure UO₂.
- Doped UO₂ pellets: Gd, Eu, La, Zr.
- Resine.

Parameters and conditions to be tested:

- Effect of Additive Content and Dopant: variable up to the solubility limit.
- The impact of the separation streams on the conversion process.
- Sintering atmosphere: reducing (5%H₂/N₂).
- Sintering temperature/time: 1675°C for 4 h.
- Pressing Pressure (uniaxial): between 700 and 1000 MPa.

Techniques:


- Cation-exchange resin methodologies.
- Pellet fabrication techniques: Rheometer, Ultrasonic Homogenizer Sonicator, Propeller stirrer, mixer-homogenizer Furnace, Polisher, Mixer Mill, Uniaxial Press.
- Characterization techniques: particle size (Laser Diffraction), geometric and immersion density measurements (Archimedean test), heat treatments with atmosphere control (TGA), XRD, BET, SEM and Raman.

Milestone 1.3: To establish a systematic procedure to obtain doped UO₂ pellets by wet route.

4. **Subtask 1.4** Inert matrices preparation by wet route.

It is known that the previous steps and conditions in the separation process affect the subsequent conversion into solid forms suitable for fuel fabrication. This step plays an important role by closing the actinide cycle. The relevance of liquid-liquid extraction processes in the recovery of MA requires the use of both: characterization techniques and its purification. Considering these, the following processes will be developed: the conversion process (conversion ratio) which involves demonstration of co-precipitation process on a significant scale, pellet fabrication prior to industrialisation guaranteeing a high homogeneity of the pellet. The materials proposed for this work will mimic inert matrix material suitable for GenIV reactors (transmutation blankets) or for a



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 17 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

HLW conditioning matrix, such as urania, ceria or zirconia. Particularly we will be focused on zirconia materials.

The methodology will be based on *co-precipitation concept* for mixed oxide fuels containing Zr. Preparing an oxide fuel comprising Zirconia-based inert matrix fuel and surrogate of actinide and/or lanthanide elements: preparing a feedstock solution consisting of a nitric solution containing actinide and/or lanthanide nitrates and uranium in the form of a hydroxylated uranyl nitrate compound. The impact of the separation streams on the conversion process will be also explored. As a contingency measure, in case there were no available resins, other alternatives like pure acid streams will be used.

The materials proposed for this subtask are :

- Pure ZrO₂.
- Doped ZrO₂ with different element contents.

Parameters to be tested:

- Effect of feedstock solution .
- Effect of resin.
- Calcining and sintering atmosphere: reducing and partially oxidizing.
- Calcining and sintering temperature/time: 1000-1700°C for 1-10 h.
- Pressing Pressures (uniaxial): between 700 and 1000 MPa.

Techniques:

- Characterization techniques: ICP-MS, HPLC-MS, α and γ -spectrometry, UV-vis, pH meter and titrator, SEM, Raman and DRX, Laser diffraction, BET.
- Pellet fabrication techniques: Furnace, Polisher, Mixer Mill, Uniaxial Press.


Milestone 1.4: To obtain doped ZrO₂ pellets by wet route.

Deliverable of task 1: Report on the influence of dopants, porosity and fabrication methodology on the physico-chemical changes of UO₂ sintered pellets.

Task 2: Fabrication of Advanced Tolerant Fuel pellets and degradation studies.

This task will be focused on Cr doped-UO₂ pellets to study the influence of the fabrication system and parameters (powder, mixing technology, sintering atmosphere, pressure, additive content) on the grain growth of UO₂. The aim of additives in UO₂ fuel is to increase the fuel resistance by means of an enlargement of the average grain size on the matrix (densification) and the plasticity of the fuel in present LWRs. It is known that this change in grain size alters the diffusion phenomena of FP inside the fuel pellets, causing an increased retention of the gaseous products inside the ceramic. Among additives considered in the literature, chromia (Cr₂O₃) doped fuel has turned out to be an especially effective alternative to traditional UO₂ fuel (today used in LWRs) in the grain growth process during sintering, also showing a great improvement on pellet-cladding



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 18 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

interaction and fission gas retention. Based on terms of safety, economy, and compatibility, in this task, the effects of different powder materials origin/synthesis/pretreatment, Cr content, fabrication method (mixing methods, processes, pressure, and sintering atmosphere) on the grain growth of UO_2 will be systematically studied. The materials proposed for this work will mimic the manufacture of ATF (Cr- UO_2 doped to reinforce the integrity of the UO_2 in operation during its irradiation). Moreover, the back-end cycle of these novel ATFs needs to be properly assessed (either at dry storage or at long-term waste disposal). One of the main problems to deal with at dry storage conditions is the potential oxidation of the fuel, in case of cladding failure. When UO_2 (cubic) incorporates certain trivalent cation dopants, the oxidation behaviour is slower than pure UO_2 , due to the stabilization of cubic U_4O_9 or UO_{2+x} phase. In order to study the effect of delayed oxidations, Cr-doped UO_2 pellets will be exposed to the oxidizing environment (air). These results have direct application in safety terms, and as far as our knowledge, no previous investigations of Cr-doped UO_2 fuel oxidation have been carried out.

The work plan destined to get on task 2 will be divided into two subtasks:

1. **Subtask 2.1.** Production and characterization of ATF by dry powder method.

The goal is to study the influence of preparation parameters (sintering atmosphere, temperature, time and Pressing Pressures) and Chromium content on the grain size of UO_2 . The grain growth of Cr_2O_3 -doped UO_2 will be studied under different oxygen potential sintering atmospheres.

The methodology to be applied will consist on *classic powder metallurgy* system. It will be done by means of grinding / sieving, pressing green pellets and after calcining and sintering at high temperature and under controlled atmosphere conditions, pellets will be obtained;

The materials proposed for this subtask are: Cr-doped UO_2 pellets with different Cr_2O_3 contents: 0 – 6000 ppm.

Parameters to be tested:

- Effect of Cr_2O_3 content: 0 – 6000 ppm.
- Sintering atmosphere: reducing ($5\%H_2/N_2$) and partially oxidizing ($x\%O_2/N_2$ or $x\%CO/CO_2$).
- Sintering temperature/time: 1000-1700°C for 1-10 h.
- Pressing pressures (uniaxial): between 700 and 1000 MPa

Techniques:


- Pellet fabrication techniques: rheometer, ultrasonic homogenizer sonicator, propeller stirrer, mixer-homogenizer furnace, polisher, mixer mill, uniaxial press.
- Characterization techniques: particle size (laser diffraction), geometric and immersion density (Archimedean test), heat treatments (TGA), XRD, BET, SEM and Raman.

Milestone 2.1: To obtain Cr-doped UO_2 pellets (ATF) by dry route.

2. **Subtask 2.2.** Co-precipitation concept for ATF fuels.

The goal is to study the influence of preparation parameters (mixing process, powder pretreatment and wet systems) and Chromium content on the grain size of UO_2 . The grain growth of



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 19 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

Cr₂O₃ doped UO₂ by this powder synthesis will be studied under different oxygen potential sintering atmospheres.

Methodology: prepare a feedstock solution consisting of a nitric solution containing Cr nitrates and U in the form of a hydroxylated uranyl nitrate compound (uranyl nitrate).

The materials proposed for this subtask are :

- Pure UO₂
- Cr-doped UO₂ pellets with different Chromium contents.
- Sintering atmosphere: reducing (H₂/N₂) and partially oxidizing (x%O₂/N₂ or x%CO/CO₂).

Parameters to be tested:

- Effect of Cr Content: 0 – 6000 ppm.
- Sintering atmosphere: reducing (5%H₂/N₂) and partially oxidizing (x%O₂/N₂ or x%CO/CO₂).
- Sintering temperature/time: 1000-1700°C for 1-10 h.
- Different Pressing Pressures (uniaxial) : between 700 and 1000 MPa.

Techniques:

- Synthesis techniques.
- Pellet fabrication techniques: Rheometer, Furnace, Polisher, Mixer Mill, Uniaxial Press.
- Characterization techniques: particle size (Laser Diffraction), geometric and immersion density (Archimedean test), heat treatments (TGA), XRD, BET, SEM and Raman.

Milestone 2.2: To obtain Cr-doped UO₂ pellets (ATF) by wet route.

3. **Subtask 2.3.** Oxidation behaviour of ATF fuels under dry storage conditions.

One of the problems to deal with at dry storage is the potential oxidation of the fuel, in case of cladding failure. Thus, in this subtask we use the samples prepared and characterized in previous subtasks, consisting of a series of UO₂ and (U,Cr)O₂ pellets covering all possible scenarios concerning the solubility of Cr in the UO₂ matrix, to study the influence of Cr addition in the oxidation behaviour of these new fuels. The goal is to study the influence of Chromium on matrix oxidation.

Methodology: oxidation of Cr-doped UO₂ in order to obtain a complete picture of ATF oxidation.


The materials proposed for this subtask are :

- Pure UO₂.
- Cr-doped UO₂ pellets with different Chromium contents.

Parameters to be tested:

- Effect of Cr Content: 0 – 6000 ppm on oxidation degree.
- Temperatures: 200-700°C.
- Oxidizing atmosphere: air (21%O₂/N₂) and partially oxidizing (x%O₂/N₂).



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 20 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

Techniques:

- Oxidation experiments: thermobalance under a constant airflow ($60 \text{ mL}\cdot\text{min}^{-1}$).
- Analytical techniques: XRD, SEM and Raman, as a sensitive technique for distinguishing between UO_2 -based fuels .

Milestone 2.3: To evaluate the influence of Chromium on matrix oxidation.

Deliverable of task 2: Report on the impact of fabrication methodology on the microstructure of Cr- UO_2 sintered pellets and the effect of dopant concentration.

Task 3: Characterization and examination of the properties of irradiated nuclear fuel analogues.

For comparison, pellets prepared by the same technological process (within tasks 1 and 2) will be described, evaluated and discussed, i.e. dry powder method or co-precipitation, in terms of microstructure and influence on: lattice parameter by XRD, and Raman spectroscopy; particle size (laser diffraction); geometric and immersion density (Archimedean test); heat treatments with atmosphere control (TGA-DSC); BET by physisorption (nitrogen), surface morphology by SEM. The grain size of the samples will be estimated by means of the linear intercept method.

Milestone 3: To evaluate the effect on the microstructure of the fabrication procedure and assess the changes in the composition and morphology of pellets. To write up results, complete papers, write up a final report and organize sessions for dissemination of results. Everybody in the research team, the working team will participate in data processing, statistical analyses and writing of papers.

Deliverable of task 3: Final Report on relevant-process conditions for the industrial applicability of different fabrication methods of nuclear fuels.


3.3.RESOURCES

For the accomplishment of the objectives, the team of the HLWU will make use of his expertise in a number of methodologies and state-of-the-art to provide the required results. The laboratories and the techniques/equipment available for the project aim to cover the current requirements and future needs as unique laboratories in Spain provide this kind of services and scientific support to public institutions, NPP and industries.

HLWU facilities include five laboratories: two of them are hot-laboratories (IR-30 facilities and the new facility IR-35, currently under licensing process, where it is possible to perform the hot-experiments) and three "cold-lab". Relevance of this new IR-35 facility is focused on higher activity limits for handling radioactive materials than those in IR-30, and enabling equipment's that currently can only work with "exempt" samples for currently running investigations getting CIEMAT's activities closer to what is expected in real scenarios.

The lab (IR-30) is divided in two laboratories: a) with 3 glove-boxes, 5 fume cupboards, an UV-Vis spectrometer, a potentiometric titrator, oscillating shakers, centrifuges, furnaces, press; b)



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 21 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

devoted to analysis and equipped with 2 gamma spectrometers (Ge detectors), an alpha spectrometer, ICP-MS and HPLC-MS. Moreover, HLWU has laboratories equipped to perform the organic synthesis and purification of needed molecules, a Raman spectrometer, SEM and XRD.

Additionally, CIEMAT facilities include a γ -irradiation site. Likewise, all of the laboratories, expertise, and unique high-security-facilities available are proper scenarios for the aforementioned experimental methodologies to be carried out (summarized in Table I).

Table I CIEMAT facilities available within the HLWU.


Facilities	Laboratories	Characteristics of emitters/equipments
IR-30	EXPERIMENTAL LAB.	Storage: 5 GBq β/γ ; 5 MBq α and 20 MBq natural U and Th. Process: 10 MBq β/γ ; 5 MBq α and 2 MBq natural U and Th. 5 fume hoods; 3 anoxic glove boxes; Equipments involved in this CT: uniaxial press (25Tn), furnace (1700°C); mixer mill, polisher + metallographic surface preparation, autoclaves and thermal treatments, etc.
	ANALYSIS LAB	Process: 0.5 GBq β/γ ; 0.5 MBq α and 2 MBq natural U and Th. Equipments involved in this CT: acid purifier system, ICP-MS, etc.
IR-35	Laboratory of tests and characterization	In construction. 5 fume hoods; 3 anoxic glove boxes; Storage: 14.5 GBq β/γ ; 40 MBq α and 20 MBq natural U and Th.
Non-radioactive laboratories		Equipments involved in these labs: Rheometer, XRD (IR-09), BET, Raman spectroscopy, SEM-EDX, laser diffraction, OM-Microdurometer, microbalance, TGA-DSC, density by Archimedean immersion, UV-VIS, rotary evaporator, flash columns, etc.)
	<ul style="list-style-type: none"> • Laboratory S1.25 • Laboratory S1.20 • Laboratory S1.18 	

All the listed methods/techniques (see Table II) are adapted for handling both radioactive and exempt materials:

Table II Methods and techniques available within the HLWU.

Technique	Parameter	Equipment
BET	N ₂ -BET Surface Area	ASAP 2020 (Micromeritics) ISO 12800E
XRD	Crystalline structure	D8 ADVANCE Eco (Bruker) – ICCD
Raman spectroscopy	Structural / speciation	LabRam HR Evolution (HORIBA) Linkam stage
SEM	Surface morphology/grain size Lineal intercept method (Heyn)	TM4000 Plus 15 kV (Hitachi). ASTM E 112-96 ;UNE-EN ISO 643:2012
Laser diffraction	Particle size	Malvern Mastersizer 3000 Hydro EV and Aero S
Archimedean test	Density	Sartorius kit
TGA-DSC	Thermal analysis	Q50 and Q20 (TA instrument)
Microhardness/ OM	Micro-Vickers hardness tester. Atmosphere/ temperature	MTR3 optical microscope: OLYMPUS image system
Climate Chamber	Temperature and humidity	VCL 0003 (Vötsch)
Furnace	1700°C, atmosphere	Termolab
Polisher		Vector LC250 (Buehler)
Mixer Mill	Max. frequency of 30 Hz	MM 400 (Retsch)
Uniaxial Press	25 T	Power Team



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 22 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

Technique	Parameter	Equipment
Dry oxidation	Reactor with O ₂ /temperature control	Termya, Berghof


3.4. CHRONOGRAM

The project has a strong multidisciplinary/interdisciplinary character. The expected chronogram is included in Table III.

Table III Chronogram of IONMAT project.

Task	Year 1				Year 2				Year 3			
	T1	T2	T3	T4	T5	T6	T7	T8	T9	T10	T11	T12
Task 1	<i>Fabrication of irradiated nuclear fuel analogues as pellets. Study and development of actinide conversion by wet route. Influence of fabrication method on the stability of the final fuel pin.</i>											
ST 1.1												
ST 1.2												
ST 1.3												
ST 1.4												
Task 2	<i>Fabrication of Advanced Tolerant Fuel pellets and degradation studies</i>											
ST 2.1												
ST 2.2												
ST 2.3												
Task 3	<i>Characterization and examination of the properties of irradiated nuclear fuel analogues.</i>											



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 23 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023


4.SCIENTIFIC-TECHNICAL IMPACT, SOCIAL AND/OR ECONOMIC IMPACT FROM THE RESULTS. INTERNATIONALIZATION.

The application of new processes and methods in nuclear fuel fabrication for P&T cycles, for advanced fuels and for characterizing SNF behaviour has a great interest in the nuclear research area since they are one of the key points for the demonstrations of an improved nuclear energy. Until now, the most developed approaches are related to industrial methods for UO₂ or MOX manufacturing based on classical powder–pellet route. However, due to the requirements of the possible nuclear waste scenarios, it is necessary to further develop new compatible radiotoxic dust-free processes with technologic requirements: to minimize the volume/radiotoxicity of SNF; to design fuels for current NPP which increase safety for accident management and; to understand irradiated fuel response in different environments. These results will be transferred to interested third parties in Spain with which we have a professional relationship like ENRESA and/or ENUSA. In addition, the results will be published in international, indexed scientific journals and will be shared with the scientific community.

At mid-term, the expected knowledge gathered within the IONMAT project will certainly produce positive results that will allow to:

- 1) Provide industry and decision makers (public agencies) with the best possible information for deciding on the best strategy for optimizing natural resources and HLW management; and public opinion could be convinced that a better management of SNF is now technologically feasible.
- 2) Keep Spain in contact with the latest developments worldwide in the field of Nuclear fuel technologies and HLW management. Compare the viability of different routes proposed in the project.
- 3) Develop alternative technologies that allow reducing the requirements for long-term storage of high level waste.
- 4) Have a complete overview of the efficiency and resistance of different fabrication routes for conversion from separation streams to final solid fuels, to make decision about its real applicability.
- 5) Implement advanced methods of manufacturing ceramic nuclear fuels, avoiding handling radiotoxic dust and minimizing process steps. IONMAT would be the first step by means of surrogates toward the additive manufacturing of fuels containing MA or FP, which requires automation and remote fabrication for minimizing personnel exposure to radiation, guaranteeing the non-dissemination of hazardous radionuclides within the biosphere.
- 6) Prepare and characterize of large grain UO₂ for ATF by studying the influence of preparation parameters and additives concentration.
- 7) Improve the reliability of un-irradiated analogues of irradiated SNF by doping natural UO₂ matrix with a series of non-radioactive elements and porosity, that can replicate the chemical and microstructural effects of irradiation on fuel at various burnups degrees . Thus, samples can provide a proper way to study different environment effects on fuel properties such as degradation behaviour (oxidation and dissolution) without the problems of intense radiation fields.



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 24 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

- 8) To evaluate the social impact in terms of waste reduction and/or recycling of different base scenarios.

Positive results of this IONMAT project will play a constructive role in climate change issues (GHG emission) for administrative institutions and third parties. There will be key in the safety of nuclear fuel production and in ensuring proliferation –resistance. With respect to radionuclides (Pu and MA) recycling, this impact could be even higher since to demonstrate any of the possible scenarios under study it is necessary to develop all steps of the cycle, including the conversion and fabrication routes suitable with the high actinide required. The application of new manufacturing technologies methods field would allow a real demonstration of the fully closed cycle. Therefore, with the technical advances expected within the IONMAT project, closed ANFC would include the recycling of An that could be proposed to industry and Technology Providers.

The results will be made public through publication in international scientific journals and national/international congresses.

The application of the new manufacturing strategy proposed, and its derived results, are expected to provide multiple environmental, social and economic benefits, in terms of reducing radioactive exposure risks. Another relevant aspect of the possible benefits for the expected results arises from the close relationship of both research groups with ENRESA agency and ENUSA (nuclear fuel supplier). They are interested, or participating, in the coordinated research program for new fabrication routes and also to translate the basic experimental data using the new methods to pilot plant and industrial production. In consequence, the benefits of the project may gain broader social diffusion and, possibly, commercial exploitations. Separation of MA and FP and subsequently conversion into solid matrices can potentially lead to significant gains in affordable economics and repository performance due, not only to improved management of the HLW but also, to the fact that the remaining FP wastes can be solidified as medium or low-active wastes. In addition, public acceptability of nuclear energy use is expected to increase in the near future. Anyhow, new technologies require process efficiency and technical maturity to deal with intrinsic difficulties due to the high radiation fields present.

HLWU activities are circumscribed in collaboration with national waste agencies fulfilling the strategic functions of maintaining acquired knowledge as a support for the evaluation of scenarios and national decision-making, as well as to support the R&D&I framework and current priorities. The HLWU is formed for multidisciplinary team coming from different research institutions and universities that support an international formative context due to the high number of collaboration and participation in European Projects. We have collaboration with KIT-INE, GER; Chalmers University of Technology, SWE; FZK Jülich, GER; Idaho National Laboratory (INL), USA; Nuclear National Laboratory, UK; French Atomic Energy Commission (CEA), FR; JRC-Karlsruhe, GER to share information and resources about systems and characterization, and eventually PhD students may do some work in these labs. All this framework allows to the team members a global overview of the international scenarios related to nuclear energy research. Therefore, the collaborations and external projection of the group have increased as consequence of the implication and work carried out within the international research programs, consolidating a successful network of collaborations and alliances.




Ciemat DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 25 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

5. ACKNOWLEDGEMENTS

This project is supported by the State Research Agency (SRA) as PID2021-124913OA-I00, IONMAT project.




 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 26 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

6. REFERENCES


- [1] EGRM U. "Application of the United Nations Framework Classification for Resources and the United Nations Resource Management System: Use of Nuclear Fuel Resources for Sustainable Development" – Entry Pathways. Geneva: 2021.
- [2] UNECE-EGRM. "The Role of Nuclear Energy in Sustainable Development - Entry Pathways" (Draft Report). UN, 2020.
- [3] Nelson, AT. "Features that Further Performance Limits of Nuclear Fuel Fabrication: Opportunities for Additive Manufacturing of Nuclear Fuels". Oak Ridge National Laboratory, 2019.
- [4] BOE. Ley 25/1964, de 29 de abril, sobre energía nuclear. 1964.
- [5] Commission E. The strategic energy technology (SET) plan2019.
- [6] Fernandez, S., Nieto, M.I., Cobos, J., Moreno, R. "CeO₂ pellet fabrication as spent fuel matrix analogue". J. European Ceramic Society. 2016; 36:3505–12.
- [7] Bonales, L.J., Elorrieta, J.M., Lobato, A., Cobos, J. "Raman Spectroscopy, a Useful Tool to Study Nuclear Materials, Applications of Molecular Spectroscopy to Current Research in the Chemical and Biological Sciences". In: Stauffer DM, editor. Applications of Molecular Spectroscopy to Current Research in the Chemical and Biological Sciences. 1: INTECH open science; 2016.
- [8] Colmenero, F., Bonales, L.J., Cobos, J., Timón, V. "Density Functional Theory Study of the Thermodynamic and Raman Vibrational Properties of γ -UO₃ Polymorph". J. Phys. Chem. C. 2017;121(27):14507-16.
- [9] Naji, M., Magnani, N., Bonales, L.J., Mastromarino, S., Colle, J.Y., Cobos, J., et al. "Raman spectrum of plutonium dioxide: Vibrational and crystal field modes". Physical Review B. 2017;95(10):104307.
- [10] Manara, D., Naji, M., Mastromarino, S., Elorrieta, J.M., Magnani, N., Martel, L., et al. "The Raman fingerprint of plutonium dioxide: Some example applications for the detection of PuO₂ in host matrices". J. Nuclear Materials. 2018; 499:268-71.
- [11] Elorrieta, J.M. Bonales, L.J., Fernandez, S., Rodríguez-Villagra, N., Gutiérrez-Nebot, L., Baonza, V.G., Cobos, J. "Pre-and post-oxidation Raman analysis of (U, Ce)O₂ oxides". J. Nuclear Materials. 2018;508:116-22.
- [12] Milena-Pérez, A., Bonales, L.J., Rodríguez-Villagra, N., Fernández, S., Baonza, V.G., Cobos, J. "Raman spectroscopy coupled to principal component analysis for studying UO₂ nuclear fuels with different grain sizes due to the chromia addition". J. Nuclear Materials. 2021; 543:152581.
- [13] Palomo, C., Rodríguez-Villagra, N., Iglesias, E., Nieto, J., Cobos, J., Quiñones, J. "Incorporation of REE into Secondary Phase Studtite". MRS Online Proceedings Library. 2014;1665(1):71-7.
- [14] Bonales, L.J., Menor-Salván, C., Cobos, J. "Study of the alteration products of a natural uraninite by Raman spectroscopy". J. Nuclear Materials. 2015;462:296-303.



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 27 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023


- [15] Bonales, L.J., Colmenero, F., Cobos, J., Timon, V. "Spectroscopic Raman characterization of rutherfordine: a combined DFT and experimental study". *Physical Chemistry Chemical Physics*. 2016;18(24):16575-84.
- [16] Bonales, L.J., Elorrieta, J.M., Menor-Salván, C., Cobos, J. "The behavior of unirradiated UO₂ and uraninite under repository conditions characterized by Raman". *MRS Advances*. 2017;1(62):4157-62.
- [17] Elorrieta, J.M., Manara, D., Bonales, L.J., Vigier, J.F., Dieste, O., Naji, M., et al. "Raman study of the oxidation in (U,Pu)O₂ as a function of Pu content". *J. Nuclear Materials*. 2017;495:484-91.
- [18] Colmenero, F., Bonales, L.J., Cobos, J., Timón, V. "Study of the thermal stability of studtite by in situ Raman spectroscopy and DFT calculations". *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*. 2017;174:245-53.
- [19] Colmenero, F., Bonales, L.J., Cobos, J., Timón, V. "Thermodynamic and Mechanical Properties of the Rutherfordine Mineral Based on Density Functional Theory". *J. of Physical Chemistry C*. 2017;121(11):5994-6001.
- [20] Elorrieta, J.M., J.Bonales, L., Rodríguez-Villagra, N., Baonza, V., Cobos, J. "Spent fuel matrix oxidation studies under dry storage conditions". *MRS Advances*. 2017;2(12):675-80.
- [21] Colmenero, F., Timón, V., Bonales, L.J., Cobos, J. "Structural, mechanical and Raman spectroscopic characterization of the layered uranyl silicate mineral, uranophane- α , by density functional theory methods". *Clay Minerals*. 2018;53(3):377-92.
- [22] Elorrieta, J.M., Bonales, L.J., Baonza, V.G., Cobos, J. "Temperature dependence of the Raman spectrum of UO₂". *J. Nuclear Materials*. 2018;503:191-4.
- [23] Elorrieta JM, Bonales LJ, Naji M, Manara D, Baonza VG, Cobos J. Laser-induced oxidation of UO₂: A Raman study. *J. Raman Spectroscopy*. 2018;49(5):878-84.
- [24] Sánchez-García, I., Núñez, A., Bonales, L.J., Perlado, J.M., Cobos, J. "Study of the adsorption capacity of graphene oxide under gamma radiation in different media". *Radiation Physics and Chemistry*. 2019;165:108395.
- [25] Colmenero, F., Plášil, J., Cobos, J., Sejkora, J., Timón, V., Čejka, J., et al. "Crystal structure, hydrogen bonding, mechanical properties and Raman spectrum of the lead uranyl silicate monohydrate mineral kasolite". *RSC Advances*. 2019;9(27):15323-34.
- [26] Rodríguez-Villagra, N., Bonales, L.J., Cobos, J. "New insight studies of the secondary phase formation under repository conditions". *MRS Advances*. 2020;5(11):539-47.
- [27] Elorrieta, J.M., Bonales, L.J., Rodríguez-Villagra, N., Baonza, V.G., Cobos, J. "A detailed Raman and X-ray study of UO_{2+x} oxides and related structure transitions". *Physical Chemistry Chemical Physics*. 2016;18:28209-16.
- [28] Evins, L.Z., Valls, A., Duro L. "DISCO 1st Annual Meeting Proceedings". Grant Agreement: 755443 DELIVERABLE D1.10. 2018.
- [29] Galán, H., Murillo, M.T., Sedano, R., Núñez A., de Mendoza J., González-Espartero, A., et al. "Hydrolysis and Radiation Stability of m-Xylylene Bis-diglycolamide: Synthesis and Quantitative



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 28 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

- Study of Degradation Products by HPLC–APCI+”. *European J. Organic Chemistry*. 2011 (20-21):3959-69.
- [30] Galán, H., Núñez, A., Espartero, A.G., Sedano, R., Durana, A., de Mendoza, J. “Radiolytic Stability of TODGA: Characterization of Degraded Samples under Different Experimental Conditions”. *Procedia Chemistry*. 2012; 7: 195-201.
- [31] Núñez, A., Galán, H., Espartero, A.G. “Optimization Studies of An(III) and Ln(III) Extraction and Back-Extraction from a PUREX Raffinate by BisDGA Compounds”. *Procedia Chemistry*. 2012; 7: 309-14.
- [32] Galán, H., Zarzana, C.A., Wilden, A., Núñez, A., Schmidt, H., Egberink R.J.M., et al. “Gamma-radiolytic stability of new methylated TODGA derivatives for minor actinide recycling”. *Dalton Transactions*. 2015;44(41):18049-56.
- [33] Hubscher-Bruder, V., Mogilireddy, V., Michel, S., Leoncini, A., Huskens, J., Verboom, W., et al. “Behaviour of the extractant Me-TODGA upon gamma irradiation: quantification of degradation compounds and individual influences on complexation and extraction”. *New J. of Chemistry*. 2017;41(22):13700-11.
- [34] Sánchez-García, I., Bonales, L.J., Galán, H., Perlado, J.M., Cobos, J. “Spectroscopic study of acetohydroxamic acid (AHA) hydrolysis in the presence of europium. Implications in the extraction system studies for lanthanide and actinide separation”. *New J. of Chemistry*. 2019;43(39):15714-22.
- [35] Sánchez-García, I., Galán, H., Perlado, J.M., Cobos, J. “Stability studies of GANEX system under different irradiation conditions”. *EPJ Nuclear Sci Technol*. 2019;5:19.
- [36] Sánchez-García, I., Galán, H., Perlado, J.M., Cobos, J. “Development of experimental irradiation strategies to evaluate the robustness of TODGA and water-soluble BTP extraction systems for advanced nuclear fuel recycling”. *Radiation Physics and Chemistry*. 2020;177:109094.
- [37] Sánchez-García, I., Bonales, L.J., Galán, H., Perlado, J.M., Cobos, J. “Advanced direct method to quantify the kinetics of acetohydroxamic acid (AHA) by Raman spectroscopy”. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*. 2020;229:117877.
- [38] Sánchez-García, I., Galán, H., Núñez, A., Perlado, J.M., Cobos, J. “Development of a gamma irradiation loop to evaluate the performance of a EURO-GANEX process”. *Nuc. Eng. and Tech*. 2021.
- [39] Geist, A., Adnet, J-M., Bourg, S., Ekberg, C., Galán, H., Guilbaud, P., et al. “An overview of solvent extraction processes developed in Europe for advanced nuclear fuel recycling, part 1 — heterogeneous recycling”. *Separation Science and Technology*. 2021;56(11):1866-81.
- [40] MITYC. Sexto plan general de residuos radiactivos. Madrid: Ministerio de industria, turismo y comercio. Gobierno de España, 2006 23 de Junio. Report No.
- [41] OECD-NEA. “State-of-the-art Report on Innovative Fuels for Advanced Nuclear Systems”. Paris: 2014.
- [42] Bunn, M., Holdren, J.P. “Managing Military Uranium and Plutonium in the United States and the Former Soviet Union”. *Annual Review of Energy and the Environment* 1997;22:403-86.



 DIVISIÓN DE FISIÓN NUCLEAR	INFORME DFN/RA-02/SP-23	HOJA 29 DE 29
	REF. EXTERNA	REVISIÓN 0 FECHA 10/04/2023

- [43] Ewing, R.C. "Ceramic matrices for plutonium disposition". Progress in Nuc. Energy. 2007;49(8):635-43.
- [44] IAEA. "Status of Minor Actinide Fuel Development". Vienna International Atomic Energy Agency, 2010.
- [45] "Viability of inert matrix fuel in reducing plutonium amounts in reactors 2006".
- [46] SNETP. SNETP Project Portfolio. Available from: <https://snetp.eu/project-portfolio/>.
- [47] SNETP. Strategic Research And Innovation Agenda. Strategic research and innovation agenda (SRIA) . Brussels, Belgium2021.
- [48] Ministerio de Ciencia e Innovación. Secretaría General Técnica del Ministerio de Ciencia e Innovación. "Plan Estatal de Investigación Científica y Técnica y de Innovación (PEICTI) 2021-2023.", (2021).



<u>FIRMANTE</u>	<u>NOMBRE</u>	<u>FECHA</u>	<u>NOTAS</u>
FIRMANTE[1]	MARIA NIEVES RODRIGUEZ VILLAGRA	06/07/2023 10:58 Sin acción específica	
FIRMANTE[2]	MARIA DE LOS HITOS GALAN MONTANO	06/07/2023 15:27 Sin acción específica	
FIRMANTE[3]	M.SOLEDAD FERNANDEZ FERNANDEZ	20/07/2023 09:52 Sin acción específica	
FIRMANTE[4]	ENRIQUE MIGUEL GONZALEZ ROMERO	20/07/2023 09:54 Sin acción específica	

AMBITO
GEN

CÓDIGO SEGURO DE VERIFICACIÓN
GEN-b5ea-ae89-1a43-4f71-c9fb-bee1-a341-084f

DIRECCIÓN DE VALIDACIÓN
<https://sara.ciemat.es:8443/csv/CsvRecoverService?csv=b5eaae891a434f71c9fbbee1a341084f>

