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Review Article

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Predisposal conditioning, treatment, and performance assessment of radioactive waste streams

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Abstract. Before the final disposal of radioactive wastes, various processes can be implemented to optimise the waste form. This can include different chemical and physical treatments, such as thermal treatment for waste reduction, waste conditioning for homogenisation and waste immobilisation for stabilisation prior to packaging and interim storage. Ensuring the durability and safety of the waste matrices and packages through performance and condition assessment is important for waste owners, waste management organisations, regulators and wider stakeholder communities. Technical achievements and lessons learned from the THERAMIN and PREDIS projects focused on low- and intermediate-level waste handling is shared here. The recently completed project on Thermal Treatment for Radioactive Waste Minimization and Hazard Reduction (THERAMIN) made advances in demonstrating the feasibility of different thermal treatment techniques to reduce volume and immobilise different streams of radioactive waste (LILW) prior to disposal. The Pre-Disposal Management of Radioactive Waste (PREDIS) project addresses innovations in the treatment of metallic materials, liquid organic waste and solid organic waste, which can result from nuclear power plant operation, decommissioning and other industrial processes. The project also addresses digitalisation solutions for improved safety and efficiency in handling and assessing cemented-waste packages in extended interim surface storage.

1 Introduction

To secure the interim storage and final disposal of radioactive waste, the different wastes must go through safe and efficient predisposal processes to minimise the waste and condition and stabilise the reactive components. The conditioning and treatment of radioactive wastes before their final disposal can be performed in many ways for various waste streams. These methods include decontamination, thermal treatment methods, metal melting processes, innovative encapsulation techniques for, e.g., different reactive metals, using geopolymers and related alkali-activated materials as mineral binders both for liquid and solid organic wastes, and finally, verifying the matrix performance of conditioned waste according to waste acceptance criteria. The THERAMIN and PREDIS projects have been and are focusing on these issues to develop and implement the optimised technologies and processes in safety and economy for predisposal.

The "THERAMIN: Thermal Treatment for Radioactive Waste Minimization and Hazard Reduction" project [1] concluded in 2020 and made advances in demonstrating the feasibility of different thermal treatment techniques to reduce volume and immobilise different streams of radioactive waste (LILW) prior to disposal. Six different thermal treatment techniques were demonstrated, treated products were characterised, and the impact of treatment on disposability was evaluated. In addition, generic disposability criteria were developed to evaluate products from any type of thermal treatment for disposal in various disposal facilities. Results of the project clearly showed the benefits of thermal treatment for pre-processing low and intermediate-level waste (LILW) in order to reduce the volume of waste to be disposed of and improve overall safety.

The "PREDIS: Pre-Disposal Management of Radioactive Waste" project [2] started in 2020 to address

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innovations in the treatment of metallic materials, liquid organic waste and solid organic waste, which can result from nuclear power plant operation, decommissioning and other industrial processes. The project also addresses digitalisation solutions for improved safety and efficiency in handling and assessing cemented-waste packages in extended interim surface storage. PREDIS includes 47 partners from 17 countries and an additional 25 companies as End User Group members. The PREDIS project focuses on predisposal waste management, particularly for metallic materials, liquid and solid organic wastes and cemented waste packages. The PREDIS project develops in parallel innovations of conditioning and treatment methods for these three waste streams and monitoring and digital technologies for storage and package quality assurance.

Both projects, through all of the predisposal treatment activities, recognised that waste acceptance criteria, environmental impact metrics and economic metrics are critical parameters to optimise the safe and efficient handling and minimisation of waste over the whole life cycle.

2 Physical conditioning of waste

2.1 Thermal treatment of waste

Thermal treatment of different types of radioactive waste can offer several benefits, however, the technology has not been applied widely to date. The primary objective of the previous THERAMIN project was to increase awareness, knowledge and confidence in thermal treatment processes, which can be used to improve safe long-term storage and disposal of intermediate- and low-level radioactive waste streams (ILW and LLW). The THErmal treatment for RAdioactive waste MINimisation and hazard reduction (THERAMIN) project was a European Commission (EC) programme of work jointly funded by the Horizon 2020 Euratom research and innovation programme and European nuclear waste management and research organisations, ran from June 2017 to May 2020.

The THERAMIN project was targeted to provide tools for coordinated EU-wide research and technology demonstration. The project was designed to provide an improved understanding and optimisation of the application of thermal treatment in radioactive waste management programmes across Europe and to improve the technology readiness level to accelerate the industrial implementation of thermal waste treatment.

The THERAMIN project provided an EU-wide strategic review and assessment of the value of thermal treatment technologies, which apply to a broad range of radioactive waste streams (including ion exchange media, soft operational wastes, sludge, organics and liquids). The project compiled an EU-wide database of wastes suitable for thermal treatment, evaluated and documented the strategic benefits of thermal treatment, and identified the opportunities, synergies, challenges, timescales and cost impacts to improve radioactive waste management. The largest and perhaps the most essential part of the project was a demonstration of six different thermal treatment technologies (such as in-container vitrification, plasma treatment, Hot Isostatic Pressing (HIP) and gasification). Demonstration trials were carried out to evaluate the applicability and achievable volume reduction achieved by the thermal treatment of waste. Demonstration trials were carried out with 'at-scale' rig tests with active and non-active wastes. The processed wastes were characterised, and their disposability was assessed. This demonstration program benefited from the previous large investments made by project partners in the thermal treatment R&D facilities, which were used by the project to maximise the benefit. The End User Group (waste producers and management organisations) gave the project very valuable support through their active involvement. The THERAMIN project also enabled mobility and training for the development of engineers and scientists and organised both a technical training workshop and a scientific conference [3].

The THERAMIN project achieved its objectives, and the results of the project confirmed perceptions of the ability of thermal treatment technologies to effectively reduce the volume of waste while improving its properties for safe long-term disposal. All the demonstrated treatment technologies required some further development to be ready for industrial application; therefore, it is essential to continue developing thermal treatment technologies. In addition, it is necessary to build on the success of THERAMIN and increase the awareness of thermal radioactive waste treatment technologies across the sector to improve the safety of radioactive waste disposal and achieve savings in the overall waste management chain. The ongoing PREDIS project has an important role in this work.

3 Chemical treatments of waste

3.1 Conditioning of metallic waste streams

The objectives of one area of PREDIS entitled 'Innovations in metallic material treatment and conditioning (Work Package 4)' are to develop decontamination processes for radioactive metallic waste, including all aspects of characterisation, effluent management as well reactive metallic waste encapsulation.

For decontamination activities, the existing process 'Chemical Oxidation Reduction Decontamination (CORD)' is being optimised for pre-oxidised steel and Inconel alloy treatments (Fig. 1). Optimisation includes particularly reactant concentrations and contact time. Post-treatment characterisations show good decontamination results. Furthermore, advances in the treatment technique using inorganic gels have been achieved with optimisation of gel viscosity and adherence properties on the metallic waste.

The metallic waste classification and characterisation achieved good progress, including radionuclides generation and distribution in metallic radioactive waste at the different nuclear reactors (pressurised water reactor PWR, boiling water reactor BWR, high-power channel reactor RBMK, water-water energetic reactor VVER). Possible management routes for metallic waste and each route's

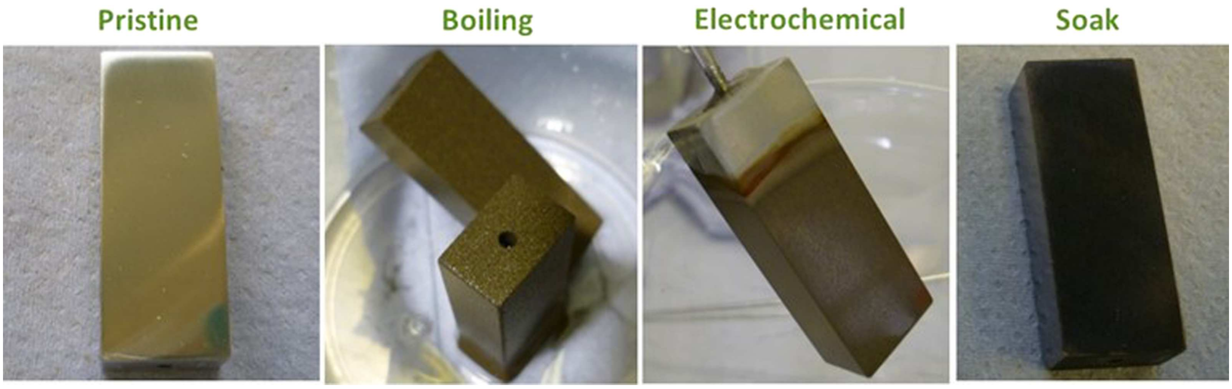

Fig. 1. Pristine and oxidised stainless steel ingots prepared using different techniques.

requirements for the material characterisation are proposed as well as a method for validation of scaling factors. For DTM (difficult-to-measure), measurement optimisation of experimental procedures has been achieved for the measurement of Zr-93.

With regards to the encapsulation of reactive metallic waste in magnesium phosphate cement (MPC) matrices, new MPC pastes and mortar formulations were defined, and physicochemical properties depend on the MgO/phosphate ratio and the nature of the filler. The cost optimisation of MPC compared to traditional materials could be achieved by careful review of the MgO sourcing as well as by decreasing the ratio of cement/filler. The benefit of the system is that the passivation of Al and Al-Mg is much faster in MPC than in traditional Portland cement-based matrices.

4 Immobilisation of waste

4.1 Direct conditioning of liquid organic wastes using geopolymers

For radioactive liquid organic waste (RLOW), the PREDIS Work Package aims to investigate and develop innovative direct conditioning solutions by implementing geopolymers and related alkali-activated materials as mineral binders. Such binders will fulfil technical and economic requirements related to RLOW, leading to final waste with properties and performance compatible with the safety and technical requirements related to disposal but also suitable for prolonged storage and transport.

An inventory of the RLOWs of interest to the Partners and End Users of the PREDIS project identified and categorised 1200 m^3 of RLOW – together with the best possible knowledge of their characteristics. This inventory was divided between oils (35%), solvents (25%), scintillation cocktails (20%) and decontamination liquids (20%). This information was particularly important in order to ensure that the technical developments of PREDIS effectively responded to European concerns.

An important experimental screening work was then carried out to identify binder formulations guaranteeing the best trade-off between high waste loading and good material properties, such as mechanical resistance or workability. Dozens of innovative formulations were tested at a laboratory scale (Fig. 2), spanning a wide range of raw materials: commercial products, locally sourced supplies or from recycled and recovered resources. The proposed formulations fall into three families of interest: (i) metakaolin-based, (ii) blast furnace slag-based, and (iii) those based on original blends together with recycled polymers. Some formulations resulting from these scoping tests achieve very high waste loadings of 30 to 50 vol.% and promising physicochemical properties. The course of the collaborative approach will now target formulation optimisation and verification of the robustness of the products.

The three most promising formulations, belonging to the three families of interest, will be the subject of more in-depth characterisations. This includes evaluating their durability under various conditions, their resistance to irradiation and their thermal behaviour. This work will logically lead to disposability assessment and demonstration, a key issue and challenge of great interest to all European waste management organisations.

4.2 Conditioning solid organic wastes

The PREDIS project also tackles the vast problem of radioactive solid organic waste (RSOW) management by considering thermal treatment prior to their immobilisation. The aim is to change their physicochemical characteristics, which are currently incompatible with long-term waste management solutions. Amongst the RSOW, the ion-exchange resins (operational waste) and other organic resins, common cemented wastes containing organic materials such as consumables or wood, and polymerised/bituminised wastes are the types of wastes in which the End User Group (EUG) expressed the most interest.

To meet the Waste Acceptance Criteria (WAC), the RSOW have to be treated, and several processes (each at different maturity levels) are considered in the PREDIS Work Package 6, which includes: Plasma incineration, Incineration/Gasification, Molten Salt Oxidation (MSO) and Wet oxidation. With the exception of plasma incineration, the immobilisation of the treated waste is required

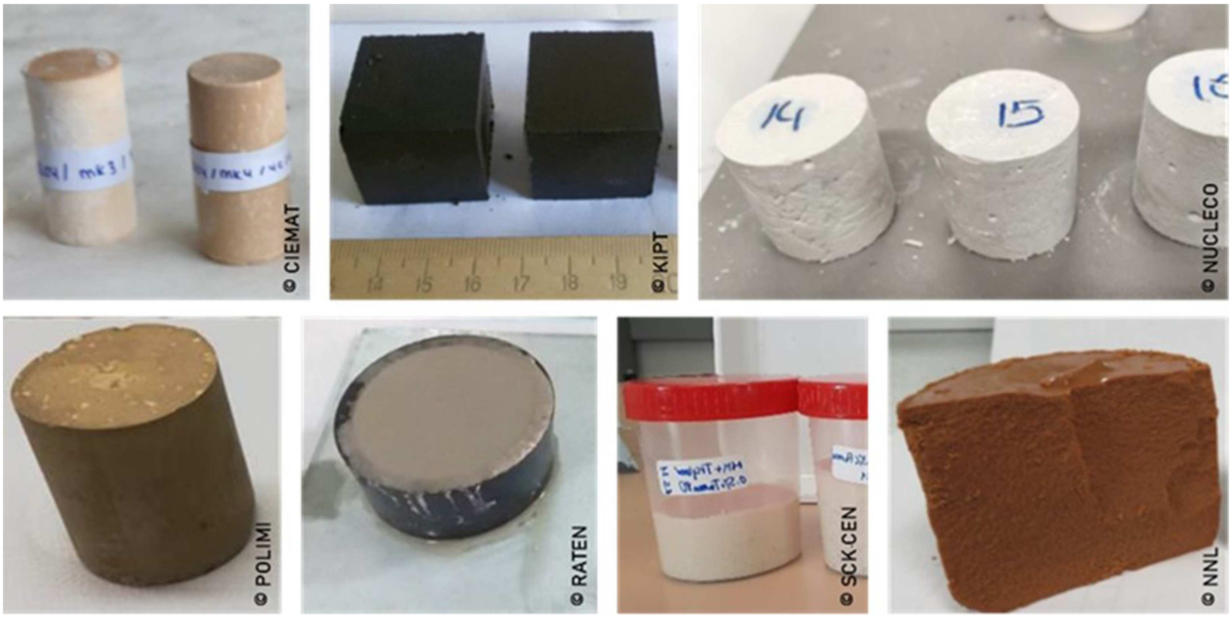

Fig. 2. Examples of mineral binders for RLOW from PREDIS scoping tests.

using binder materials or by Hot Isostatic Pressing (HIP). Two binders are tested and compared to assess the most appropriate solution between classical cementitious materials and novel matrices such as geopolymers.

A broad variety of geopolymer formulations exists, but the focus is on blast furnace slag and metakaolin-based formulations, which are also used for conditioning the RSOW. Optimisation of the formulations is the main issue, but innovative formulations are also tested using natural resources such as volcanic tuff to replace metakaolin to gain economic and environmental impact while improving some properties.

The collaborative work between the PREDIS partners developing geopolymers and cement-based materials led to a parametric study for the immobilisation of the MSO residue to identify the key parameters to achieve the highest waste loading and the best geopolymer design. After that, the representability of the samples will be increased by adding a radiotracer or irradiating a selection of samples to evaluate the stability and durability of the reconditioned wastes. This will utilise an experimental protocol based on the feedback from the EUG and the ongoing PREDIS work, which is representative of the expected conditions prevailing in a long-term storage facility.

5 Monitoring concrete package and repository integrity

According to information collected within PREDIS from end users, only a limited set of sensors and parameters are currently monitored by the end users in their respective facilities. In most cases, just general information (e.g., temperature, radiation) is measured on a facility level. For individual packages, other than compliance with waste acceptance criteria before entering the storage facility, no other inspection and monitoring are typically performed besides occasional visual inspection. On the other hand, there are reports on the degradation of various types of packages in storage or disposal, e.g., by corrosion of the outer metal skin or by expansion effects in the package content caused again by corrosion or Alkali Silica Reaction (ASR). In most cases, this degradation is detected at a late stage when signs of damage are visible (or increased radiation is measurable) on the outside of a package.

In PREDIS Work Package 7, titled "Innovations in cemented waste handling and pre-disposal storage", several instruments and digital tools are evaluated and improved to provide better means for safe and effective monitoring of cemented waste packages. Safety enhancement (e.g., less exposure of testing personnel) and cost-effectiveness are part of the intended impact, as well as an improved prediction to assess future integrity development during pre-disposal activities. The work includes but is not limited to inspection methods such as muon imaging to detect inclusion in heritage packages. Wireless sensors integrated into waste packages are under development to measure proxy parameters such as temperature, humidity and pressure. External sensors, again wireless, are applied for radiation monitoring. First prototypes are meanwhile available and are under test using mock-ups. Sensors will also be made cost-effective to allow the installation of many more sensors compared to current practice. The measured data are to be collected at a joint data platform, where they can be used in digital twins of the waste packages for specific simulations (geochemical, integrity), providing a prediction of future behaviour. Machine Learning techniques trained by characterising older waste packages will help connect the models to the actual data. All data (measured and simulated) will be used in a decision framework to provide means for a more reliable and faster management of individual packages. All findings will be demonstrated at the end of the project at an actual end-user facility.

6 Impacts of the innovations and future steps

All of the work described has been intended to directly impact the safety and efficiency of waste handling processes for waste generators, including nuclear power plant operators and waste owners, such as those responsible for managing legacy waste storages. The intention has been to provide economically feasible solutions that are also demonstrated for technical reliability so that they can be taken into practice. It is acknowledged that not all of the processes are mature, yet gains have been made to help understand the limitations and opportunities with these new and/or improved techniques developed in both THERAMIN and PREDIS. Actions have also been taken during the development to consider how the improved processes can impact the disposability, with respect to alignment with Waste Acceptance Criteria and stakeholder views of the environmental and economic impact of the new solutions. There are many cases where localised guidelines and national radiation safety regulations may need to be adjusted before the solutions can be fully implemented, but the target is that through these research and development projects, more trust can be gained in the solutions.

The challenges of predisposal waste management (and disposal) of operational and decommissioning wastes are many and varied. Some programmes extend many decades into the future and have considerable financial implications. The THERAMIN and PREDIS projects have made progress on specific classes of waste and some applicable technologies, however, there remain many opportunities to enhance current predisposal activities.

For this reason, the PREDIS project, through Work Package 2 (Strategic Implementation), is working together with its End User Group and Stakeholders to build a comprehensive Strategic Research Agenda (SRA) for Predisposal Waste Management. The first stage was completed with the publication of the PREDIS Gap Analysis Report, focusing on R&D needs in the areas of PREDIS technical Work Packages 4–7 [4]. Similarly, the Work Package 2 team produced a consolidated baseline SRA [5], bringing together knowledge from recognised SRAs from other entities, including SNETP [6]. This consolidated baseline, now published, was the starting point for a period of ongoing end-user engagement running from March 2022 through to the production of the Predisposal SRA in 2023. The engagement with end users and industry groups will utilise mixed methods to identify the key challenges and priorities. The final PREDIS SRA will be a valuable tool to focus collaborative research efforts to address the challenges facing the national radioactive waste management missions.

7 Conclusions

Both of the projects, THERMIN and PREDIS, have been demonstrating innovations in the handling of radioactive wastes prior to disposal. Viable methods and materials have been developed for thermal treatment, chemical treatment, conditioning and monitoring waste streams to enhance their disposability while maintaining safety. The waste acceptance criteria, economics and environmental impact of the new solutions are also evaluated to help accelerate their acceptance by end users from the industry, and regulators take the techniques into practice.

Conflict of interests

The authors declare that they have no competing interests to report.

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Data availability statement

Data associated with this article is described within the project web pages at http://www.theramin-h2020.eu/ and https://predis-h2020.eu/. Some data associated with this article cannot be disclosed due to confidential information from industry or will be disclosed at the conclusion of the (PREDIS) project.

Author contribution statement

The allocation of contributions per author to this article includes Holt and Oksa as the primary authors responsible for the abstract, Chapters 1 and 6–8; Nieminen responsible for Chapter 2; Abdelouas responsible for Chapter 3; Fournier responsible for Chapter 4.1; Mennecart responsible for Chapter 4.2; Niederleithinger responsible for Chapter 5; Banford contributed to Chapter 6.

References

1. THERAMIN project web page http://www.theramin-h2020.eu/
2. PREDIS project web page https://predis-h2020.eu/
3. IoP conference proceedings – materials science and engineering, Vol. 818 (2020), https://iopscience.iop.org/issue/1757-899X/818/1 (open access)

4. T. Schatz, Gap analysis, deliverable 2, PREDIS project (2021), https://predis-h2020.eu/wp-content/uploads/2021/06/PREDIS-D2.2-Gap-Analysis_Final_2021-05-31.pdf
5. A. Wareing, Draft strategic research agenda on pre-disposal, milestone 6, PREDIS project (2021), https://predis-h2020.eu/wp-content/uploads/2021/08/PREDIS-T2.2_Baseline-SRA_M2.3_August-2021.pdf
6. O. Martin, in *NUGENIA Global Vision*, edited by A. Al Mazouzi, G. Bruna, S. Reese (Nugenia, JRC96613, Brussels, Belgium, 2015)

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