# **PREDIS**

# Deliverable D6.8. Submitted scientific journal papers about the RSOW stability and durability tests after reconditioning Date version 28.08.2024 Version Final

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### Abstract

grant agreement No 945098.

With the objective of disseminating PREDIS activities, and in particular the results of experimental work on Radioactive Solid Organic Waste management carried out in the WP6, a minimum of five articles published in scientific journals has been targeted by the end of the PREDIS project.

This report summarized the list of articles published in scientific journals (01.09.2020 - 30.08.2024).

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# 1 Introduction

The PREDIS project targets the development and implementation of activities for pre-disposal treatment of radioactive waste streams, in particular the Low- and Intermediate-Level Radioactive Waste. The work package (WP6) focused on innovation in the management of Radioactive Solid Organic Waste (RSOW), in which 14 research entities were involved. Several thermal processes were considered for the treatment of RSOW, usually followed with an immobilization of the treated materials using geopolymer or cement-based matrices, but also to the production of glassy / ceramics materials. During the PREDIS project, numerous experiments were carried out, and it is absolutely essential that these results are disseminated through participation in international conferences and publications. A target of five papers published in scientific journals was set in the PREDIS Grant Agreement.

This deliverable (D6.8) lists the paper published or under submission to date.

# 2 List of publications in scientific journals

### 2.1 Published articles

**"Fenton-like treatment for reduction of simulated carbon-14 spent resin",** Hafeez, M.A., Jeon, J., Hong, S., Hyatt, N., Heo, J., Um, W. Journal of Environmental Chemical Engineering, 2021, 9:104740, 9 p.

### https://doi.org/10.1016/j.jece.2020.104740

<u>Abstract:</u> Spent resins with <sup>14</sup>C are enlisted among the most problematic radioactive wastes. In current study, we successfully prepared the simulated 14C-loaded spent resins using HCO<sub>3</sub><sup>-</sup>, and for the first time the Fentonlike treatment of simulated 14C-loaded spent resins were compared with fresh Amberlite IRN-150 resin (without simulated <sup>14</sup>C loading) by optimizing the influential reaction parameters. The experimental results revealed that 91.26 % weight loss and 85.47 % in TOC removal were achieved for fresh Amberlite IRN-150 resin (without simulated <sup>14</sup>C loading) using 0.35 M Cu(II) catalyst, 30 % H<sub>2</sub>O<sub>2</sub> as oxidant at pH 2, and 90 ± 3 °C temperature for 4 h reaction time. However, the simulated <sup>14</sup>C-loaded spent resin using HCO<sub>3</sub><sup>-</sup> exhibited 87.34 % weight loss under the similar reaction conditions. The reduction in weight loss (%) of simulated <sup>14</sup>C-loaded spent resin was attributed to the presence of HCO<sub>3</sub><sup>-</sup> on the resins, which could produce the carbonate radical (CO3<sup>-°</sup>) of lower oxidation capability as compared to hydroxyl radical and in turn affect the efficacy of Fentonlike treatment for simulated <sup>14</sup>C-loaded spent resin. Scanning electron microscopy and energy dispersive spectroscopy (SEM-EDS) and Fourier transform infrared spectroscopy (FT-IR) analyses were used to characterize and the results also supported the resin's degradation.

"Stability of SrCO<sub>3</sub> within composite Portland-slag cement blends Walling", S. A., Gardner, L.J., Prentice, D.P., Dixon Wilkins, D.P., Hammad, A.A., Um, W., Corkhill, C.L., Hyatt, N.C., Cement and Concrete Composites, 2023,135:104823, 12 p.

### https://doi.org/10.1016/j.cemconcomp.2022.104823

<u>Abstract:</u> The stability and reactivity of SrCO<sub>3</sub> within a blended Portland-slag cement at both 20 °C and 60 °C (to simulate an indicative waste form for disposal) was determined via XRD, TG-MS, SEM-EDX and thermodynamic modelling. Sr<sup>14</sup>CO<sub>3</sub> is a potential long-term sink for trapping radioactive <sup>14</sup>C, produced through the nuclear fuel cycle, therefore understanding its stability in potential cementitious waste forms is of interest and importance. Incorporation of 30 wt% SrCO<sub>3</sub> in blended Portland-slag cement caused minor reactions to occur, resulting in increased formation of carbonated AFm phases, along with stabilisation of ettringite at 20 °C, precluded at 60 °C due to the reduced stability to ettringite at this temperature. Thermodynamic modelling predicted only minor SrCO<sub>3</sub> reactivity up to 360 days, with carbonate remaining stable over this timeframe, validated by our experimental results. Thus, thermodynamic simulations predict that SrCO<sub>3</sub> is an effective immobilisation matrix for <sup>14</sup>C, within a blended Portland-slag cement waste form, suitable for long-term geological disposal.



"Design of sustainable geopolymeric matrices for encapsulation of treated radioactive solid organic waste", Santi, A., Mossini, E., Magugliani, G., Galluccio, F., Macerata, E., Lotti, P., Gatta, G.D., Vadivel, D., Dondi, D., Cori, D., Nonnet, H., Mariani, M., Frontiers in Materials, 2022,17 p.

### https://doi.org/10.3389/fmats.2022.1005864

Abstract: Among radioactive by-products generated by nuclear technologies, solid organic waste is drawing attention because of difficult management and incompatibility with the disposal strategies traditionally adopted. Recently, geopolymers have been proposed as valid and green alternatives to cement-based matrices. In this work, novel geopolymeric formulations have been studied at laboratory scale to encapsulate ashes from incineration of surrogate solid organic waste and to further pursue sustainability and circular economy goals. Indeed, the most widely used precursor of literature geopolymers, calcined kaolin, has been totally replaced by natural raw materials and recycled industrial by-products. In addition, a highly zeolitized volcanic tuff has been chosen to further improve the intrinsic cation-exchange capacity of the geopolymer, hence enhancing waste-matrix interaction. The alkaline activation of the precursors, achieved without silicates of any metal, resulted in a promisingly durable geopolymeric matrix, whose chemical composition has been optimised to provide compressive strength above 10 MPa after 28 days of curing. A water-saturated sealed chamber provided the optimal curing condition to limit the efflorescence and improve the mechanical properties. At least 20 wt% loading of treated surrogate waste was achieved, without compromising workability, setting time, and compressive strength, the latter remaining within acceptable values. In order to demonstrate matrix durability, leaching behaviour and thermal stability were preliminarily assessed by immersion tests and thermogravimetric analyses, respectively. The leachability indices of constituent elements resulted far above 6, which is the generally agreed requirement for cement-based matrices. Moreover, the mechanical resistance was not worsened by the water immersion. The preliminarily obtained results confirm the promising properties of the new matrix for the immobilization of nuclear waste.

"Effect of incorporating a molten salt waste from nuclear power plants on the properties of geopolymers and Portland cement wasteforms", Perez-Cortes, P., Garcia-Lodeiro, I., Puertas, F., Alonso, M.C., Cement and Concrete Composites, 2023, 142:105210, 12 p. ISSN 0958-9465,

### https://doi.org/10.1016/j.cemconcomp.2023.105210

<u>Abstract:</u> This research studies the effect of incorporating a surrogate molten salt radioactive waste (labelled as MS, composed of a mixture of carbonates, chlorides and sulphates) on the mechanical, mineralogical and microstructural features of two types of cementitious systems: i) Portland cementitious systems and ii) a novel "one-part geopolymer". As Portland cementitious systems, a CEM I/42.5 SR and a CEM III/B 32.5, were selected. The "one-part" geopolymer was prepared with mixtures of metakaolin and blast furnace slag as precursors, and NaOH and Na<sub>2</sub>SiO<sub>3</sub> powders, as solid activators. Results shown that the MS interacts with both cementitious matrixes, affecting the hydration/activation and promoting the crystallisation of sodium/calcium carbonate hydrated phases. Mechanical strengths substantially declined and the microstructure was clearly affected, especially in samples containing 30% of the MS. Some lines of action are suggested to improve the cementation treatment of MS minimising its effect in the development of the different cementitious materials.

"Cementation of Spent Radioactive Ion-Exchange Resin Ashes Using Alkali-Activated Cements: Physicochemical and Structural Changes", Perez-Cortes, P., Garcia-Lodeiro, I., Alonso, M.C., Puertas, F., Cement and Concrete Composites, 2023, 149: 105517, 15 p.

### https://doi.org/10.1016/j.cemconcomp.2024.105517

<u>Abstract:</u> Spent ion-exchange resins (IERs) constitute the major volume of solid radioactive wastes generated by the nuclear industry and Portland cement wasteforms exhibit limitations for their conditioning and storage. In this paper, an alternative approach is conducted involving the solidification of IER ashes (IERAs) through



the use of "one-part" alkali-activated cements (AACs). Systems with different chemistry based on blast furnace slag (BFS) and metakaolin (MK) were formulated and the physicochemical and structural changes of the gel products after incorporating IERAS were investigated. The incorporation of the IERAs reduced the amount of C-A-S-H and (N,C)-A-S-H products while changing the pore structure and microstructure, reducing the mechanical strengths of the cement matrices; these effects were more remarkable in the high BFS systems. New insights are elucidated on the microstructural changes of the gel phases and suggest that AACs with BFS/MK ratios of 25/75 and 50/50 are promising candidates for conditioning IERA wastes.

### 2.2 Submitted articles

**"Solidification of molten salt from Molten Salt Oxidation technology",** S V. Galek, A. Černá, P. Pražák, T. Černoušek, M. Vacek, V. Berger, J. Hadrava

In preparation in "Frontiers in Materials"

"Explorative scale-up of Fenton Oxidation and Geopolymer Encapsulation for the management of spent mixed bed ion exchange resins", F. Galluccio, E. Mossini, A. Santi, G. Magugliani, M. Giola, E. Macerata, G. D. Gatta, P. Lotti, D. Cori, G. Bilancia, P. Peerani, and M. Mariani

In preparation in "Nuclear Engineering and Technology Journal"

## 3 Conclusion

The research activities in the workpackage 6, dealing with innovation in the management of Radioactive Solid Organic Waste, have been published in scientific journals, with the target of a minimum of five published articles as agreed in the PREDIS grant agreement. This target has been met, with six papers published and two submitted. Some experimental work is still in progress, or requires further investigation to validate hypotheses. They will be published after the end of the PREDIS, mentioning they were obtained in the framework of the Euratom research and training programme 2019-2020.

