Pre-impregnation approach to encapsulate radioactive liquid organic waste in geopolymer

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HIGHLIGHTS

- RLOW disposal by pre-impregnation and subsequent encapsulation in geopolymers is feasible.
- Recycled polyurethane impregnant is incompatible with the geopolymer activator.
- NOCHAR N910 polymer promisingly absorbs and stabilizes RLOW in a geopolymer.
- Surfactants should be avoided as they would compromise stability towards water immersion.

GRAPHICAL ABSTRACT
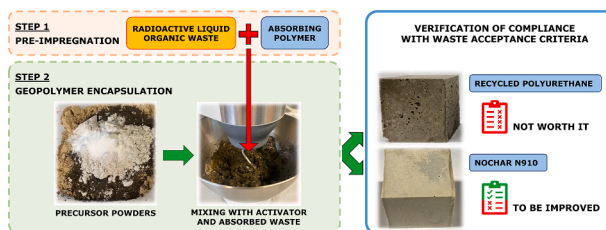
ABSTRACT

The pre-disposal management of Radioactive Liquid Organic Waste (RLOW) is hampered by its challenging physico-chemical properties. In this work, a straightforward conditioning option based on RLOW impregnation on absorbing materials and followed by encapsulation in a stable geopolymeric matrix is proposed, avoiding onerous pre-treatments and the use of surfactants. Recycled materials have been investigated as adsorbent and geopolymer precursors to foster process sustainability. Relevant properties have been studied to ascertain the waste acceptance criteria accomplishment: materials compatibility, RLOW loading factor and bleeding, microstructure, compressive strength, leaching and thermal stability. This approach is promising, although some criticalities remain unsolved.

1. Introduction

The public acceptance of nuclear technologies and applications greatly relies on the implementation of safe and robust management of the radioactive waste, which is unavoidably generated [1,2]. First of all,

identification, classification, and estimation of the global inventory of the radioactive materials should be accomplished [3]. This piece of information is pivotal for the selection of the optimal waste route, which in turn depends on societal and economic factors, as well as technological and regulatory aspects, often specific of each waste and country [4].

Hence, the identification of the best available technologies for waste pre-disposal – retrieval, characterization, treatment, conditioning, interim storage, and transport – is just part of the problem. Moreover, the implementation of the selected waste route requires the availability of temporary storage or disposal facilities [5]. According to the International Atomic Energy Agency, besides spent nuclear fuel, the management of radioactive waste may be challenging for countries with limited human and financial resources or underdeveloped infrastructures [6]. Further difficulties may be posed by the waste endowed with challenging physico-chemical-radiological properties, produced in rather small amounts, or for which there is not yet an accredited management route. Indeed, development and implementation of dedicated processes for the disposal of such radioactive waste is often difficult to justify from economical and strategical points of view [4].

According to these considerations, Radioactive Liquid Organic Waste (RLOW) belongs to this class of challenging waste. It includes a diverse range of materials, such as lubricating greases and oils, hydraulic and mechanical oils, liquid scintillation cocktails (LSC) from radiochemistry laboratories, solvents, such as tributyl phosphate (TBP) and kerosene mixtures, and decontamination liquids [7]. Their pre-disposal management is hampered by poor physico-chemical stability and high mobility [8]. In fact, most of them are volatile, combustible, or explosive. Besides the low volumes, the high diversity of physico-chemical and radiological properties of RLOW generated so far by nuclear technologies contribute to the challenging nature of this waste. If only short-lived radionuclides are concerned, the "delay and decay" approach could be pursued. During the interim storage, the activity of these radionuclides decays below clearance levels. Therefore, the waste can be released from regulatory control and treated following conventional procedures. However, in most of the cases, the "concentrate and contain" strategy entailing treatment processes for volume reduction and subsequent confinement of the residues in a suitable matrix for long-term waste stabilization is unavoidably required to prevent, or at least retard, the release of radionuclides and toxic compounds in the environment. Indeed, the low density and immiscibility of RLOW with water would ease their migration in the environment, e.g., by floating on water surface [8]. Although several treatments to stabilize RLOW before conditioning – incineration, wet oxidation, electrochemical oxidation, acid digestion, alkaline hydrolysis, and distillation – have been proposed and investigated so far, several drawbacks still hamper their implementation [7]. For instance, corrosive or even radioactive off-gases may be produced and released during mineralization, thus requiring proper management countermeasures, including the use of expensive and corrosion-resistant materials [7–10]. Moreover, the high diversity between each RLOW requires the adoption of precise characterization and segregation practices and the optimization of waste-specific treatment process conditions, which often become too expensive and complex, and necessitate experienced and qualified operators [11,12]. Conversely, the conditioning of RLOW in a cementitious matrix, without laborious pre-treatment operations, would be a cheaper and more straightforward approach [13,14]. Unfortunately, the organic waste is known to strongly interfere with the hydration of the binder, leading to setting delays [15–17]. In addition, the scarce waste-matrix compatibility causes low stability of the mixture, poor loading factors, substantial bleeding and leachability of RLOW [18,19]. A possible breakthrough consists in the use of alkali-activated materials (AAM), especially geopolymers, as they are known to be less affected by the organic content [20–22]. AAM and geopolymers are promisingly durable and environmentally friendly matrices obtained from the dissolution, and subsequent reaction, of precursor's reactive silicate and aluminate species by an alkaline activator [23–26]. Several research works have proven that AAM are less suited than geopolymers to achieve the stable encapsulation of RLOW [27,28]. Indeed, the precursors of AAM, rich in Ca and silicates, follow a hydration mechanism similar to Portland cements [29]. On the other hand, the geopolymers are produced through a different reaction,

usually called poly-condensation [30,31].

Several approaches have been developed to pursue the stable incorporation of RLOW in geopolymers [32]. The most straightforward way is the conditioning of the organic liquid by direct mixing with the fresh grout. This process is successful if the higher viscosity of the cementitious slurry hinders coalescence of the dispersed organic liquid during setting and curing stages. Nevertheless, when the organic liquid viscosity is too different from that of the grout, namely it is too low, the formation of small droplets is impeded, while their coalescence is favored [33]. For these reasons, this route has mainly shown to promisingly incorporate oils [34–36]. On the other hand, other more challenging organic liquids require an additional step of pre-emulsification with the alkaline activation solution, often by means of proper surfactants [32,34]. Indeed, surfactants help to avoid the organic liquid coalescence by lowering the interfacial tension or by improving the quality and the stability of the emulsion [36,37]. Although the implementation of the pre-emulsification step is practically simple and only requires the adoption of easily manageable devices, the high diversity of RLOW composition and physico-chemical properties requires the validation of waste-specific protocols [7]. Moreover, despite improving the miscibility between activation solution and organic liquids, surfactants may as well enhance their leachability during water immersion, as they promote diffusion at the organic/aqueous interface [38]. To cope with these issues, another approach has been proposed. It entails a preliminary impregnation step to absorb the organic liquid on a suitable solid material before its immobilization in a durable matrix. Several versatile and less waste-specific materials have been proposed to this aim, such as natural or synthetic fibers or clays [7,39]. In this way, the process would be more general and robust, as no surfactants would be required and the coalescence of the RLOW should not represent a problem. Although promising, the addition of the pre-impregnation step would be a slight complication of the direct conditioning approach and could potentially lead to unsatisfactory waste loading [7].

The aim of this work is the development of a straightforward and general conditioning option which relies on challenging RLOW pre-impregnation on absorbing materials followed by encapsulation in a stable geopolymeric matrix. This approach would allow to avoid dangerous, complex, and expensive pre-treatments and the use of surfactants to emulsify the organic liquid within the matrix. Lubricating, hydraulic, and mechanical oils have not been considered in this work, because demonstrations of their direct conditioning feasibility are already available in the literature, in some cases without recurring to surfactants [34]. Hence, the attention has been focused on organic liquids for which the direct incorporation resulted to be more problematic. In particular, the class of LSC and solvents typical of reprocessing plants were considered for their scarce compatibility with inorganic binders. In the last decades, a class of high technological polymers, e.g. NOCHAR N910, has proven high efficacy in capturing organic liquids to allow their stable immobilization in a cementitious matrix [40,41]. As a cheaper and more environmentally sustainable alternative, recycled polyurethane (rPU) powder has been proposed to effectively absorb organic liquids [42]. In a previous work, the interaction between rPU and RLOW surrogates was studied, and the optimal loading capacity and protocol were identified [43]. The need for subsequent encapsulation of the absorbed organic liquid in a stable conditioning matrix to accomplish the waste acceptance criteria (WAC) was evidenced [44]. In order to pursue environmental sustainability, the geopolymer binder employed in this work is obtained by alkali activation of a mix of natural and recycled aluminosilicate raw materials: zeolitic volcanic tuff (VT), fly ash (FA), and ground-granulated blast furnace slag (BFS). Previously, this matrix was promisingly proposed for the encapsulation of treated radioactive solid organic waste [45]. The occurrence of organic liquid bleeding during mixing and curing was monitored. Preliminary tests were performed to assess the loading factors of surrogate RLOW. Furthermore, the compressive strength and stability toward water immersion were studied to assess compliance of the obtained waste forms

with WAC. Finally, phase composition and thermal behavior of the waste forms were investigated by X-ray Diffraction (XRD) and Thermogravimetric Analysis (TGA).

2. Materials and methods

2.1. Chemicals

In this work, representative surrogate materials were employed to mimic real RLOW. As a typical LSC widely employed by radiochemistry laboratories, Ultima Gold Low Level Tritium cocktail (UG LLT, Perkin Elmer) was used. Concerning organic solvents, a mixture of kerosene (Carlo Erba Reagents) and 30%v. of TBP (Thermo Fisher Scientific) was considered. Since the fifties, with occasional slight composition modifications, this has been the reference solvent employed by the nuclear industry for the purification of uranium in front-end activities and for the reprocessing of the spent nuclear fuel by original and modified versions of the Plutonium Uranium Reduction Extraction (PUREX) process [46–48]. As absorbing material, rPU (Oko-Pur®, provided by Airbank S.r.l.) was chosen for its low cost and low environmental footprint. Indeed, it is obtained from the recycling of refrigerator insulation panels. Its use as absorber of organic liquids is already known [42,49]. Some additional preliminary tests were performed with the N910 (NOCHAR® Inc.) styrene block co-polymer, a thermoplastic elastomer [50].

The employed geopolymer matrix is based on the use of natural and recycled aluminosilicate precursor materials: micronized volcanic tuff (VT, Zeolite Fertenia™), fly ash (FA) and ground-granulated blast-furnace slag (BFS), both provided by Buzzi Unicem and used without any pre-treatment. Their properties and compositions have been described in a previous work by the same authors [45]. The alkaline activation solution was prepared by dissolving sodium hydroxide pellets (Marten S. r.l., technical grade) in demineralized water, to reach the final NaOH concentration of 4.8 M [45].

2.2. Pre-absorption and encapsulation

The experimental protocol consisted of a preliminary impregnation step to absorb weighed amounts of surrogate RLOW on the polymeric material to obtain the desired loading factor, namely 100% or 200% by weight for rPU and N910. In agreement with previous works, this condition should ensure a sufficiently stable absorption of the organic liquid [40,43]. In parallel, the aluminosilicate precursors were weighed and mixed with the sodium hydroxide activation solution following the optimized protocol to obtain the final composition: 28.5%wt. SiO_2, 10.3%wt. Al_2O_3, 12.7%wt. CaO, 6.9%wt. Na_2O, 29.7%wt. H_2O and 0.40 water-to-solid ratio [45]. This geopolymer has been identified as formulation "C" in a previous work by the same authors [45]. Afterwards, weighed amounts of the loaded materials were mixed by a planetary blender at mid-low speed for ten minutes with the geopolymeric fresh grout to obtain a visually homogeneous mixture to be cast. Cubic (side of 5 cm) and cylindric (diameter of 2.8 cm, height of 3.1 cm) molds were respectively employed to prepare samples for compressive strength and other characterizations: XRD, TGA, and water immersion tests. Both cubic and cylindrical molds were closed on the top side to obtain planar casting faces. Reference samples containing the same weight amounts of polymer, but without liquid organic waste, were prepared following the same procedure. All samples were cured in a climatic chamber for 28 days in endogenous conditions (22 ± 1 °C).

2.3. Characterizations

Compression tests were carried out on cubic samples after 28 days of curing. The protocol is consistent with international standards [45,51]. According to the recently published document by the Italian regulator, the minimum required compressive strength for homogeneously

conditioned low and intermediate level waste is 10 MPa [52].

In order to preliminary ascertain the water immersion stability, the obtained cylindrical samples were completely immersed in about 400 mL of ultrapure water for the overall duration of four weeks. The amount of water was determined by the reference standard protocol [53]. Given the qualitative aim of this leaching experiment, no periodic renewal of the leachant was performed. At the end of the static immersion period, the leachant pH and conductivity were measured just for the most promising samples, i.e., in absence of visible organic liquid release. Moreover, the release of geopolymer principal constituents – namely Ca, Al, Si, Na, and K – was directly determined on duly acidified leachates by Inductively Coupled Plasma - Optical Emission Spectroscopy (ICP-OES, Avio 550, Perkin Elmer). The calibration of ICP-OES was performed by using a set of Inorganic Ventures CMS-x standard solutions suitably diluted at different analytes concentrations with ultrapure nitric acid. Internal standard and standard addition methods were used to identify and correct possible matrix effects. In addition, the retention of surrogate RLOW was estimated by analyzing the release of P (coming from surfactant present in LSC and TBP) and organic matter by ICP-OES and Chemical Oxygen Demand (COD) measurements, respectively. The COD analyses were performed with COD cuvette kits (LCI400, Hach-Lange) in the 0–1000 mg/L O_2 range, performing the optical measure with DR1900 spectrophotometer (Hach-Lange) following a standardized method [54,55]. To confirm the COD values measured with respect to the blank (i.e., ultrapure water), the standard addition method of standard solution of potassium phthalate was employed. The results obtained with the specimens loaded with organic liquids were compared with those in absence of the waste, but just containing the same amount of polymer.

XRD analyses were conducted on hardened specimens for a full description of their phase composition. After grinding in an agate mortar, the diffraction patterns of the powder samples were collected by an automated Panalytical X'Pert Pro-diffractometer, equipped with a X'Celerator-type detector. The experimental conditions were the following: monochromatized Cu-$K\alpha$ radiation, 40 kV and 40 mA, 2θ range between 4 and 80° with step size of 0.017° and counting time of 240 s per step. Phase identification was performed using the X'pert HighScore suite.

TGA was performed to investigate the thermal behavior of the waste forms. These experiments were carried out using a Mettler Toledo thermogravimetric analyzer, heating the samples with a rate of 20 °C/min, under air atmosphere (mass flow: 4 L/h), from 25 to 1000 °C. The samples for this analysis, 5 mg sized, were prepared by grinding hardened specimens down to sub-millimetric powders in an agate mortar.

3. Results

3.1. Geopolymers with rPU as absorbing material

The above-described procedures were used to pre-impregnate UG LLT or the mixture of kerosene + 30%v. of TBP on rPU absorbing material. Afterwards, cubic and cylindrical geopolymeric samples were prepared for further analyses. The list of prepared specimens is reported in Table 1, with the information about polymer and RLOW surrogate content. The tested geopolymeric samples contained 10%wt. and 20% wt. of organic liquid pre-absorbed on 5%wt. and 10%wt. of rPU. For reference, geopolymeric samples containing the same weight fraction of rPU, but without organic liquid, were prepared. The pre-impregnation step was performed the day before the encapsulation. As expected by previous results, total absorption of the added RLOW surrogate was obtained at the tested conditions [43]. Although no appreciable bleeding occurred during the pre-impregnation step, slight organic liquid release was evidenced once the absorbed material and the geopolymeric grout were mixed. This loss continued during the first hours of setting, until the complete hardening of the matrix. As noticeable in Figs. 1, 2, and 3, the prepared specimens exhibited evident cavities. This

Table 1

Compressive strength of geopolymer samples encapsulating recycled polyurethane polymer alone or previously pre-impregnated with kerosene + 30%v. TBP or Ultima Gold LSC.

Samples ID	Label	GP [%wt.]	rPU [%wt.]	k-TBP [%wt.]	UG-LLT [%wt.]	R_c [MPa]
1	GP	100	0	0	0	17.3 ± 1.7
2	GP+rPU5	95	5	0	0	7.7 ± 0.8
3	GP+rPU10	90	10	0	0	1.1 ± 0.2
4	GP+rPU5+TBP10	85	5	10*	0	4.3 ± 0.5
5	GP+rPU10+TBP10	80	10	10*	0	1.5 ± 0.2
6	GP+rPU10+TBP20	70	10	20*	0	1.0 ± 0.2
7	GP+rPU5+UG10	85	5	0	10*	3.3 ± 0.5
8	GP+rPU10+UG10	80	10	0	10*	1.9 ± 0.3
9	GP+rPU10+UG20	70	10	0	20*	1.0 ± 0.2

* Non-negligible RLOW bleeding upon swelling of rPU during curing.

outcome is coherent with partial loss of organic liquid and is further explained below. A similar behavior was previously observed trying to incorporate the pre-impregnated rPU in a cement-based matrix [42]. Albeit evident, the released amount of organic liquid could not be accurately quantified for practical reasons. This likely amounts to 10-20%wt. fraction of the total organic content.

Although the obtained specimens presented evident defects and had already released a non-negligible amount of organic liquid, monoaxial compression tests were performed with a view to determine if the mechanical requirements were met. The results of the compressive strength are reported in Table 1. Anyway, other attempts are required to identify an alternative absorbing material. Indeed, rPU should be disregarded and a more stable material should be identified to solve the key issue of organic liquid bleeding.

The cylindrical samples with the same compositions reported in Table 1 were immersed in ultrapure water to ascertain their stability. As it can be clearly evidenced from Fig. 4, only the leaching solutions corresponding to the pure geopolymer (sample 1) and to the specimens containing just rPU (samples 2–3) were limpid after 4 weeks of immersion. On the contrary, the leachants corresponding to samples 4–9 undeniably showed the release of the organic liquid. Indeed, in the case of samples 4–6, the leachates became colored with floating drops of TBP and kerosene. For the samples 7–9, the leachates became milky due to

the likely release of surfactant, with floating heterogeneous agglomerates. Given the inhomogeneous aspect of the leachates, physicochemical characterization was not carried out because the results would not be reliable. After water immersion, the specimens preserved their integrity, without any evident surface alteration, neither dimensional changes nor crack formations.

In order to better explain the scarce stability of rPU in the geopolymer, XRD and TGA investigations were performed. In Fig. 5, selected diffractograms are reported.

The thermogravimetric patterns of geopolymers with and without rPU and organic liquid are drawn in Fig. 6.

3.2. Geopolymers with N910 as absorbing material

As reported in Table 2, a new series of geopolymer specimens containing 5%wt. of N910 was prepared.

When dealing with organic liquids, the polymer was pre-impregnated with 10%wt. of LSC (UG LLT) or the PUREX solvent before being encapsulated in the geopolymer. In agreement with the literature, the added RLOW surrogates were totally absorbed in the N910 polymer [40]. Unlike the experiments with rPU, no organic liquid bleeding was observed after mixing with the geopolymeric grout nor during curing, meaning that the impregnated N910 polymer is stable in


Fig. 1. Encapsulation of rPU in cubic geopolymer samples (side of 5 cm). From left to right, the recycled polyurethane polymer loading factor is 0, 5, and 10%wt.
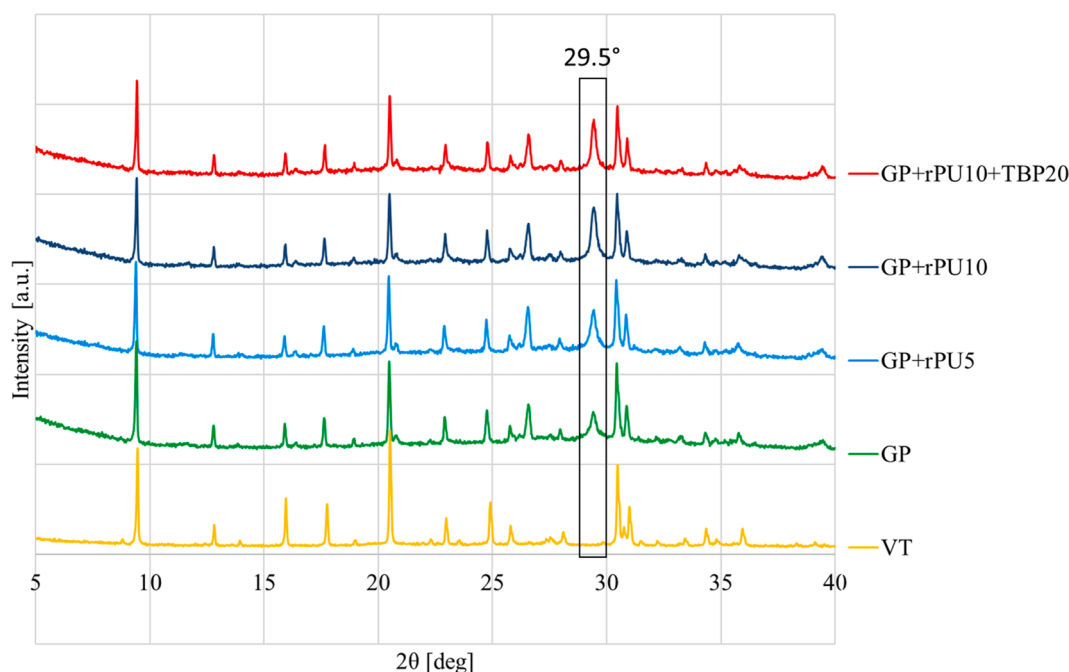

Fig. 2. Encapsulation of rPU previously pre-impregnated with kerosene + 30%v. TBP in cubic geopolymer samples (side of 5 cm). From left to right, the rPU polymer loading factor is 5, 10, and 10%wt., the kerosene + 30%v. TBP loading factor is 10, 10, and 20%wt.

Fig. 3. Encapsulation of rPU previously pre-impregnated with UG LLT in cubic geopolymer samples (side of 5 cm). From left to right, the rPU loading factor is 5, 10, and 10%wt., the UG LLT loading factor is 10, 10, and 20%wt.
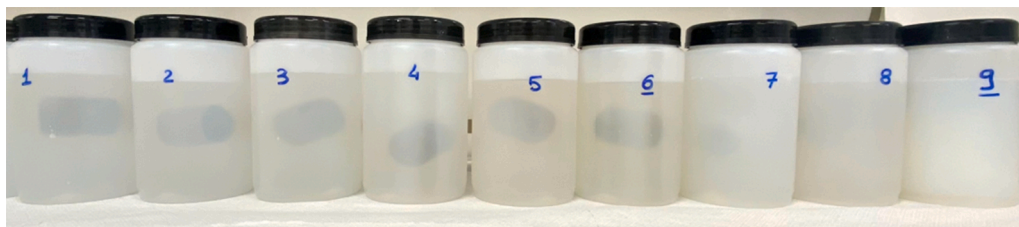
Fig. 4. Aspect of water leaching solutions after 4 weeks of immersion experiments. The numbers on the side of the jars identifies the immersed samples, which is the same as reported in Table 1. The immersed specimens are cylindrical samples (diameter of 2.8 cm, height of 3.1 cm). The bottles contain about 400 g of ultrapure water, according to the reference standard protocol [53].

Fig. 5. X-ray diffraction patterns of VT precursor and geopolymer samples encapsulating rPU polymer alone or previously pre-impregnated with RLOW. As commented in the discussion section, the frame identifies the carbonate Bragg peak at $2\theta \sim 29.5°$

the alkaline environment. As shown in Fig. 7, the obtained cubic specimens did not show visible cavities nor inhomogeneities. The results of the monoaxial compression tests are reported in Table 2.

The leachates deriving from the water immersion experiments on the cylindrical samples containing N910 were limpid and homogeneous, without apparent signs of organic liquid release. In addition, the specimens seemed to preserve their integrity after water immersion.

Hence, some relevant physico-chemical properties of the leachates were characterized. The results are reported in Table 3 and compared with those of the pristine ultrapure water.

4. Discussion

The outcomes of the experimental campaign carried out on geopolymer specimens containing rPU are herein discussed. As reported in Table 1, at increasing rPU content, the compression resistance falls below the limit imposed by the Italian authority. Even if the trend is not surprising, given the increased porosity and the reduced density of the specimens caused by rPU addition, the magnitude of the loss of compressive strength is severe [56,57]. Moreover, once the rPU polymer is pre-impregnated with the organic liquid, the compression resistance is
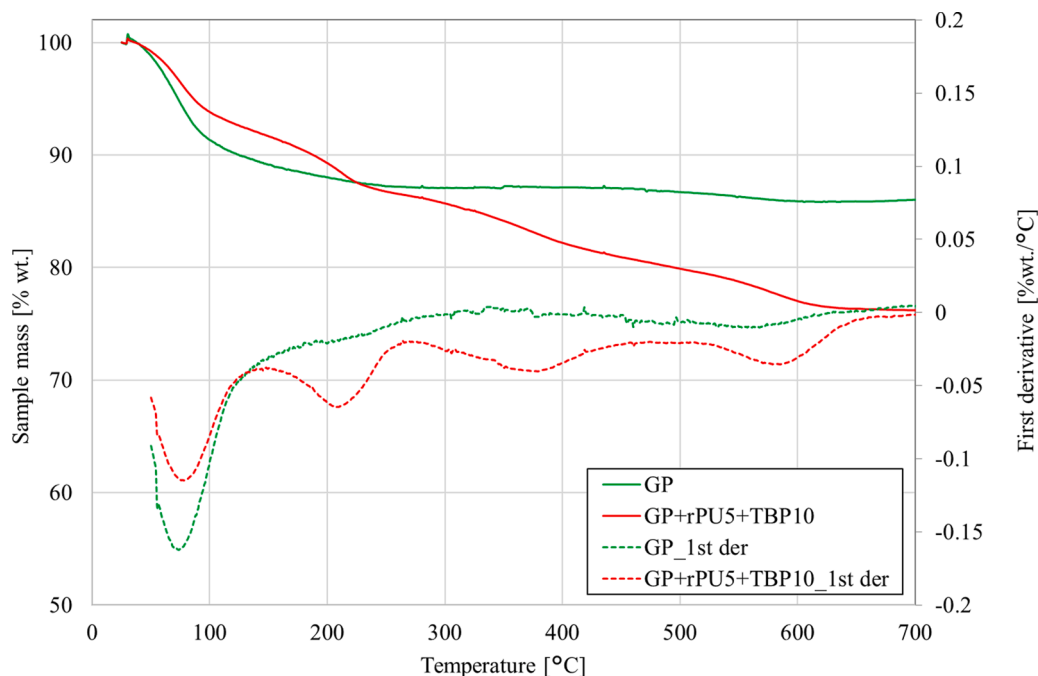
Fig. 6. Thermogravimetric patterns (continuous lines) and first derivatives (dotted lines) of the pure geopolymer sample (green) and of a geopolymer sample encapsulating 5%wt. of rPU previously pre-impregnated with 10%wt. of kerosene + 30%v. of TBP (red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 2
Compressive strength of geopolymer samples encapsulating N910 polymer (here called N) alone or previously pre-impregnated with UG LLT or kerosene + 30%v. TBP.

Samples ID	Label	GP [%wt.]	N910 [%wt.]	k-TBP [%wt.]	UG-LLT [%wt.]	R_c [MPa]
1	GP	100	0	0	0	17.3 ± 1.7
10	GP+N5	95	5	0	0	13.4 ± 1.4
11	GP+N5+UG10	85	5	0	10	10.4 ± 1.1
12	GP+N5+TBP10	85	5	10	0	5.9 ± 0.7

further reduced. In the case of geopolymers containing 5%wt. of rPU loaded with 10%wt. of organic liquid, the worsening of the mechanical strength is evidenced by approximately halving the compression resistance. The same comparison cannot be made for the samples containing 10%wt. of rPU and the organic liquid. Indeed, their compressive strength values are too close to the validity limit of the press (about 1 MPa), hence the relative uncertainties are too high. The loss of mechanical properties at increasing organic liquid waste loading factor is coherent with the previous findings reported in the literature [34,58].

Another important WAC concerns water immersion stability. As shown in Fig. 4, the specimens containing the PUREX solvent (samples 4–6) have failed the immersion test, as the leachants have become slightly colored, with evident release of organic liquid floating on the top. Moreover, the leaching solutions deriving from the geopolymer samples loaded with LSC (samples 7–9) are milky. This apparently negative result is enlightening instead. Indeed, the surfactants intrinsically present in LSC specimens are released during water immersion and could favor the loss of the organic liquid. This observation suggests that the use of surfactants to favor the emulsification of the organic liquid may be discouraged as it could ease the release of the mentioned waste during water immersion. Nevertheless, the integrity of the immersed samples seems to be preserved. Probably, the scarce capacity of the matrix to retain the RLOW surrogate is strongly affected by the high porosity of the geopolymer containing the rPU as absorbing material.

The collection and study of XRD (Fig. 5) and TGA (Fig. 6) data are useful to characterize and explain the observed behavior of the rPU-
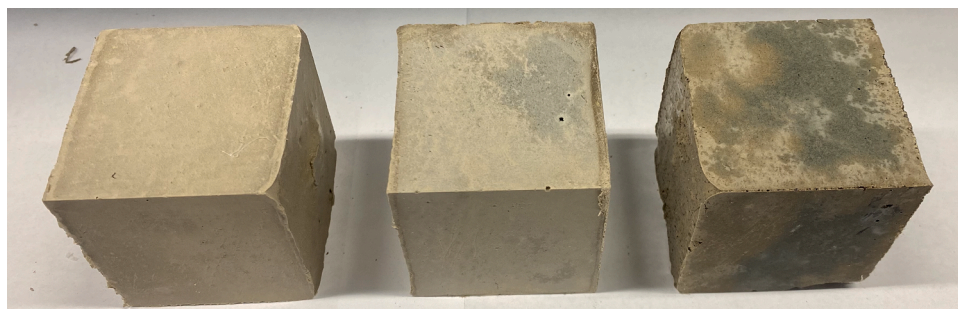
Fig. 7. Encapsulation of 5%wt. of N910 polymer in cubic geopolymer samples (side of 5 cm). The polymer is encapsulated alone (left) or after pre-impregnation with 10%wt. of UG LLT (middle) or kerosene + 30%v. TBP (right).

Table 3

Physico-chemical analyses of the leaching solutions before and after the immersion tests of the geopolymer samples encapsulating N910 polymer alone (sample 10) or previously pre-impregnated with UG LLT (sample 11) or kerosene + 30%v. TBP (sample 12).

Samples ID	Ultrapure water (Type I)	10	11	12
pH	5.5	11.5	11.5	11.5
Conductivity [mS]	0.001	4.21	4.28	4.76
Net COD [mg/L]	0	19 ± 5	894 ± 5	699 ± 5
Na [mg/L]	< 0.040	$473.8 \pm 0.5\%$	$530.8 \pm 1.2\%$	$492.7 \pm 0.8\%$
K [mg/L]	< 0.474	$25.72 \pm 1.0\%$	$29.39 \pm 1.8\%$	$27.11 \pm 1.2\%$
Ca [mg/L]	< 0.047	$5.56 \pm 1.0\%$	$6.09 \pm 1.2\%$	$5.35 \pm 1.5\%$
Si [mg/L]	< 0.085	$29.02 \pm 0.9\%$	$25.67 \pm 1.7\%$	$25.02 \pm 1.3\%$
Al [mg/L]	< 0.009	$8.84 \pm 0.4\%$	$11.49 \pm 1.7\%$	$10.52 \pm 1.4\%$
P [mg/L]	< 0.077	$0.68 \pm 8.1\%$	$9.60 \pm 2.1\%$	$18.84 \pm 3.3\%$

The pH is measured with litmus paper, the accuracy is ± 0.5. The conductivity uncertainty is equal to ± 1%.

containing geopolymers. The X-ray diffractogram labelled "VT" belongs to pristine zeolitic VT, which is a highly crystalline material. The main species are ascribable to chabazite (about 65%wt.) and phillipsite (5% wt.) zeolites, in agreement with supplier's data and a previous work by the same authors [45]. The mineralogical composition of the reference geopolymer (not loaded with the waste, labelled "GP") preserves most of the crystalline species present in the precursor materials, especially in the zeolitic VT, as already reported in a previous work [45]. Nevertheless, a relevant amorphous phase is present, which is encouraging evidence that poly-condensation reactions have occurred. Consistently, the TGA pattern of the reference geopolymer reported in Fig. 6 shows the losses of physically adsorbed and structural water molecules at about 100 °C and 200 °C, respectively [26].

Moreover, a new Bragg peak at $2\theta \sim 29.5°$ appears in the X-ray diffractogram. It is unequivocally attributable to sodium and calcium carbonates. The larger width of this peak with respect to other crystalline species is typical of micro-crystalline compounds. This observation may suggest that carbonates could have been formed during the geopolymer preparation by reaction between excess NaOH or $Ca(OH)_2$ and the environmental CO_2. Consistently, at increasing amounts of rPU and organic liquids, the X-ray diffraction patterns present higher amorphous fraction with respect to the reference geopolymer. Interestingly, as noticeable in the XRD profiles labelled "GP+rPU5" and "GP+rPU10" belonging to the geopolymers containing 5 and 10%wt. of rPU, the relative intensities of the carbonate peak increase, with respect to the reference geopolymer, at increasing rPU content. Coherently, regardless of the organic liquid content, the XRD pattern of the geopolymer containing 10%wt. of rPU loaded with 20%wt. of kerosene and TBP mixture (labelled "GP+rPU10+TBP20") presents a relative intensity of the carbonate peak comparable with that of the specimen "GP+rPU10". This outcome suggests that additional formation of carbonates may be caused by rPU. In fact, polyurethanes are not stable towards hydrolysis, and under the conditions found in the geopolymeric matrix can undergo base-catalyzed degradation with release of CO_2 [59,60]. The subsequent reaction of carbon dioxide with the alkaline environment of the geopolymeric grout leads to the formation of sodium and calcium carbonates as determined by XRD. Another evidence of sodium and calcium carbonates formation comes from the TGA patterns of "GP" and "GP+rPU5+TBP10" samples reported in Fig. 6. Indeed, the carbonates decomposition is visible at around 600 °C [61]. The geopolymer containing 5%wt. of rPU presents a much more evident thermal decarbonation loss if compared to the reference geopolymer sample. In addition, new weight losses are visible in the TGA of the loaded specimen with respect to the pristine geopolymer. The loss that occurs between 150 °C and 250 °C belongs to kerosene and TBP, while the one between 300 °C and 450 °C belongs to rPU, as already pointed out in a previous work where loaded rPU was encapsulated in concrete [43]. It could be inferred that both the rPU polymer and the organic liquids present the same thermal degradation patterns, whether they are tested alone or embedded in the matrix. This result suggests that a physical immobilization of the waste within the geopolymer occurs, without any

chemical interaction besides the already mentioned rPU hydrolysis followed by carbonation. The above-described outcomes of XRD and TGA investigations may be exploited by researchers engaged in the study of lightweight concrete. This would allow to better explain the high porosity and low density of these materials, and possibly to reach the desired properties of their composite materials [56,62].

Although rPU has proven to be a valid material for the absorption of organic liquids and has found extensive use as filler in lightweight concrete, its incompatibility with the highly basic environment of the geopolymeric grout discourages its usage as RLOW absorbing material to be then immobilized in a geopolymer. Further efforts were made to test another polymer for organic liquids absorption. Indeed, the well-established N910 polymer showed promise in absorbing RLOW and being then stabilized in a cementitious matrix [63]. In this perspective, a few preliminary experiments were performed following the same above-described protocols. In fact, the use of the recently developed geopolymer as conditioning matrix would be more sustainable than concrete. As reported in Table 2, the addition of 5%wt. of N910 polymer causes a more tolerable reduction of the compression resistance than the same weight amount of rPU. The mechanical strength of specimens containing 5%wt. of N910 and 10%wt. of organic liquid is further reduced closer to the limit imposed by the Italian regulator. Hence, besides presenting better mechanical properties than the corresponding rPU analogues, the N910-containing geopolymers provide stable retention of pre-impregnated organic liquids. This better behavior is proof that N910 is more compatible than rPU with the geopolymeric grout. These observations are confirmed by the water immersion tests, which promisingly provide limpid leachates. Regardless of the RLOW surrogate content, the geopolymer constituents are released from all specimens following water immersion, leading to evident pH and conductivity increase, as reported in Table 3. Coherently, the measured concentrations of Na, K, Ca, Si, and Al in the leachates are higher than in the pristine water solution, but without any discernible effect attributable to the presence of the organic waste [45]. This outcome further confirms that geopolymers are suitable materials for the encapsulation of RLOW, as their setting is poorly affected by the organic matter content [27]. On average, the cumulative leached fractions of Na and K are approximately equal to 2.3%wt. and 0.3%wt., respectively. Instead, Ca, Si, and Al are less leached (around 0.02%wt. each). Contrariwise, the evident increase of P concentrations proves that release of nonylphenyl polyoxyethylene ether phosphate – a surfactant presents in UG LLT– and TBP occurs from the waste-containing specimens. According to these data, it was estimated that 14%wt. of the surfactant was cumulatively released. Again, this result suggests that surfactants may be problematic during the leaching experiments. If not already present in the organic liquid, their addition should be avoided. On the other hand, the cumulative TBP release was less than 1%wt.. It was estimated as the ratio between the released amount of phosphorus to the total content, given that the initial amount of TBP is known and that phosphorus is present just in TBP. Since the leachate was limpid, with no observable organic liquid droplets on top, this value is representative of the overall TBP release.

Further evidence of the organic liquid release is provided by the net COD data reported in Table 3. The higher value is measured for the LSC-containing specimen, coherently with the previous observations about the potential role of the surfactant in affecting the leaching stability.

5. Conclusions

The outcomes of this work could drive the ongoing and future efforts of researchers in the development of a valid pre-disposal solution for the management of this important class of radioactive waste. If no pre-treatment is implemented to decompose the organic matter, the pre-impregnation strategy could be promisingly pursued to cope with the immobilization of the most challenging RLOW, such as the PUREX solvent and LSC. This work demonstrates that rPU is not a valid impregnation material to achieve a stable encapsulation of the organic liquid. Indeed, it is unstable in contact with the highly basic activation solution. Instead, N910 polymer seems to successfully meet the requirement of stable organic liquid retention without excessively compromising the mechanical and water immersion stability of the resulting waste form. The sustainability goals would be achieved by using natural or recycled materials for the preparation of the geopolymer matrix. The preliminary results of this work also suggest that surfactants would probably hinder the water immersion stability and favor the release of organic liquids, although this aspect should be further investigated.

Additional efforts should be aimed at confirming the promising preliminary results of this work and supporting the applicability of the pre-impregnation approach. Despite the first results on N910 are promising, further efforts should be made, especially to identify a more suitable and sustainable absorbing material to effectively optimize the stabilization and solidification approach. An important parameter to be optimized is the RLOW loading factor, which should be maximized to reduce the resulting waste volumes and the footprint on the repository, without compromising compliance with WAC. Then, a complete characterization campaign should be performed to validate the proposed approach with respect to WAC. Hence, mechanical, thermal, fire, radiolytic, leaching, and biodegradability resistance should be demonstrated to verify the long-term durability of the waste form and ascertain its safe disposability.

CRediT authorship contribution statement

Eros Mossini: Conceptualization, Methodology, Validation, Writing – original draft. **Andrea Santi:** Methodology, Investigation, Data curation, Writing – review & editing. **Gabriele Magugliani:** Methodology, Investigation, Data curation, Writing – review & editing. **Francesco Galluccio:** Validation, Writing – review & editing. **Elena Macerata:** Validation, Writing – review & editing. **Marco Giola:** Validation, Writing – review & editing. **Dhanalakshmi Vadivel:** Investigation, Writing – review & editing. **Daniele Dondi:** Investigation, Writing – review & editing. **Davide Cori:** Investigation, Writing – review & editing. **Paolo Lotti:** Investigation, Data curation, Writing – review & editing. **Giacomo Diego Gatta:** Investigation, Data curation, Writing – review & editing. **Mario Mariani:** Funding acquisition, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships with other people or organizations that could have inappropriately influenced the work presented in this paper.

Data availability

Data will be made available on request.

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