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GAMMA IRRADIATION STABILITY OF NOVEL BINDERS FOR RADIOACTIVE LIQUID ORGANIC WASTE IMMOBILIZATION

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INTRODUCTION

Management of Radioactive Liquid Organic Waste (RLOW) is challenging due to the combination of radiological and physico-chemical properties. The adoption of Geopolymers (GP) and Alkali Activated Materials (AAM) has introduced a new route for the direct conditioning of RLOW. These binders are less affected by organic waste and have proven effective in incorporating up to 30%v of low-viscosity oils [1]. The aim of this work is to evaluate the stability under gamma irradiation of these waste forms, to assess how they will withstand the dose from real waste.

METHODOLOGY

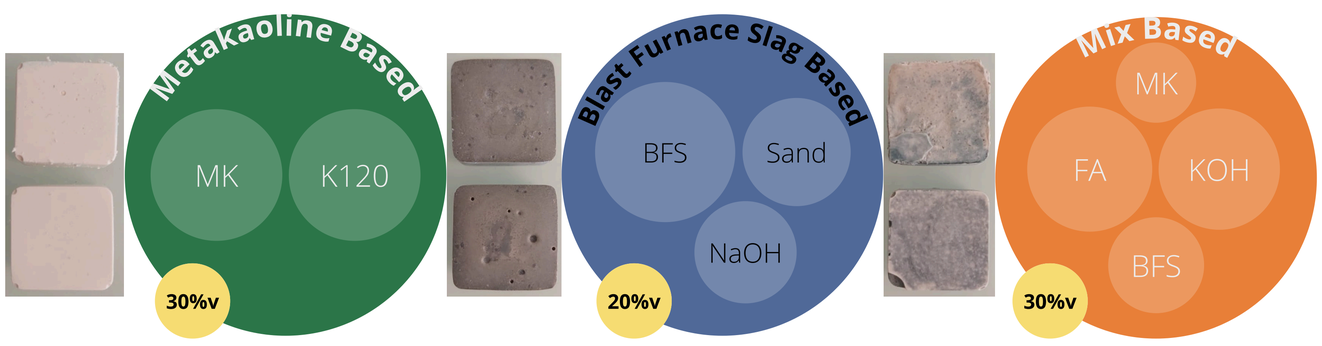


Figure 1 - Composition of different GP studied and picture of the real samples. In the yellow circle the amount in volume of oil added.

Samples were prepared from different partners following protocols [2]. Samples were analysed both **with** and **without surrogate wastes** (Nevastane oil). Irradiation was perfromed with **Cs-137 gamma source** with a dose rate of **0.45 kGy/h**.
Irradiated samples without waste were evaluated to identify the most reliable formulations through measurements of **hydrogen production**, with μGC, and **leaching rates**, with ANSI ANS 16.1 procedure.
Samples containing surrogate waste were examined using **SEM** and **micro-computed tomography** (μ-CT) to assess the interaction and distribution of the waste in the conditioning matrix.

RESULTS

NO OIL

Hydrogen quantification results are expressed as radiolysis gas yields ($G(H_2)$) [3].

$$G(gas)_{material} = \frac{n\,(gas)}{D \times m_{sample}} \qquad G(gas)_{norm} = \frac{G(gas)_{material}}{Water}$$

Table 1 - Hydrogen yield evaluated for the different GP. The normalised $G(H_2)$ is calculate as linear regression from H_2 measurement from 0 to 500 kGy and as if nothing was evaporated.

Sample	Water content (w%)	G(H) (material) [x10^{-7} mol/J]	G(H) (normalised) [x10^{-7}mol/J]
MK GP	17%	0.23 ± 0.01	1.34 ± 0.01
BFS GP	17.7%	0.0.7 ± 0.01	0.42 ± 0.01
MIX GP	9.8%	0.06 ± 0.01	0.57 ± 0.01

MK GP is the **highest in H_2** production probably due to its **open porosity** structure in comparison to BFS and MIX GP.
The results are in agreement with previous analysis on GPs that measure a value around 0.75 x10^{-7}mol/J.

The **leaching** solution, ultrapure water, was renewed at time intervals defined by the protocol, and analyzed to **monitor changes** in matrix **elements release**.
The Leachability Index (LI) was calculated [3].
MK GP showed a slightly **higher release,** probably due to its porous structure.

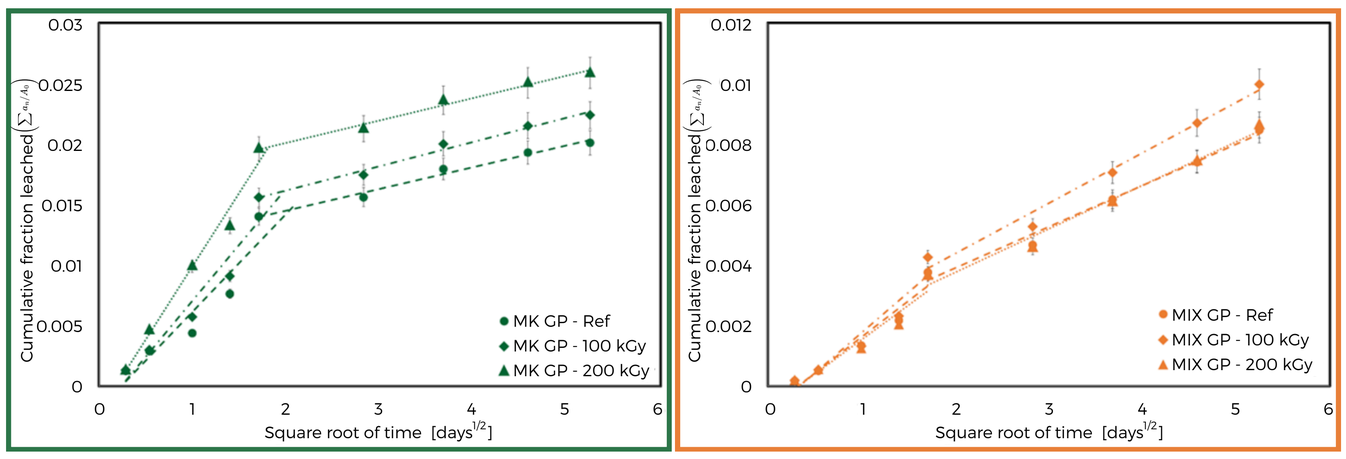


Figure 2 - Normalised cumulative trend of Si release in leaching solution of samples without surrogate waste.

OIL

SEM and **μ-CT** analyses show the **distribution of the oil** and the **complexity** of BFS and MIX GPs in comparison to MK GP.

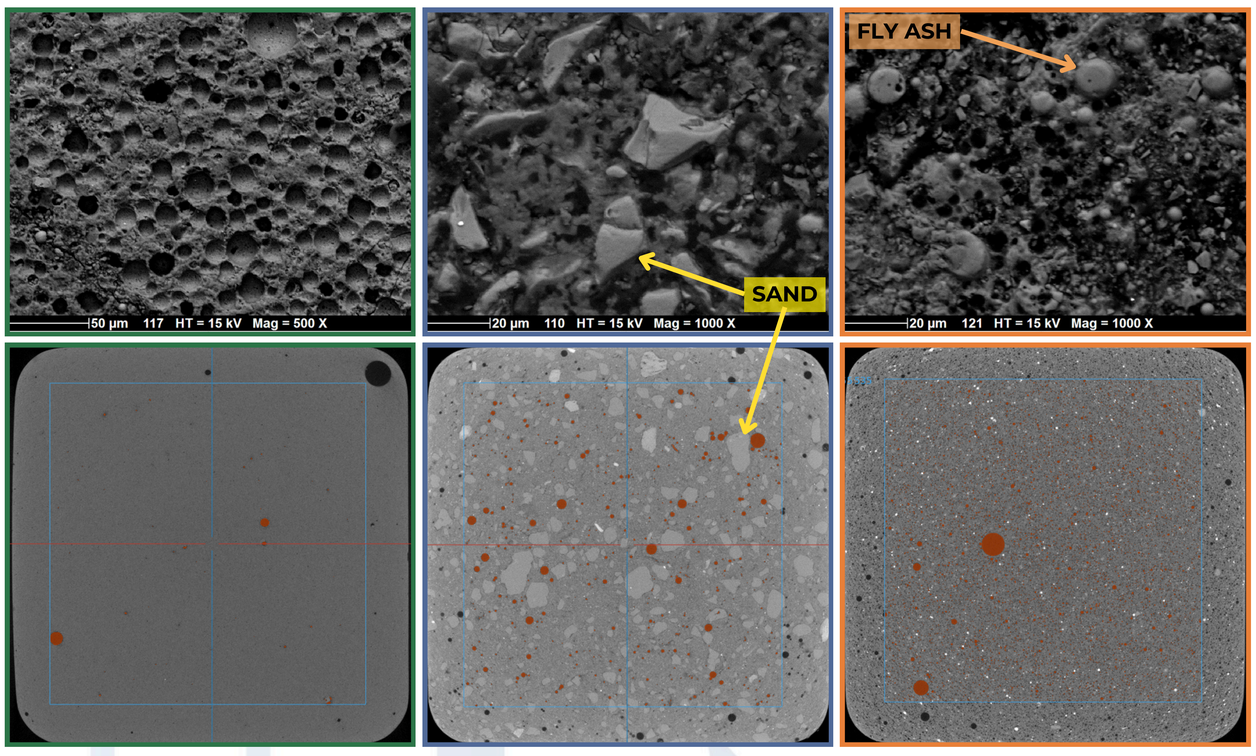


Figure 3 - SEM (top) performed on GPs with surrogate waste. Oil pore measured less than 3 μm.
Micro-CT (bottom) performed on GPs with surrogate waste. Resolution of 15 μm. Oil highlighted in orange.

The **droplet shape factor** (SF), derived from μ-CT, was used to evaluate the **emulsification quality**. The SF varies from 0 to 1, where 1 indicating a spherical shape of the droplet. A better emulsification is obtained when the SF is far from 1 [3].
The **worst** result is obtained with BFS GP, probably due to the **large grain size** of the **sand**.

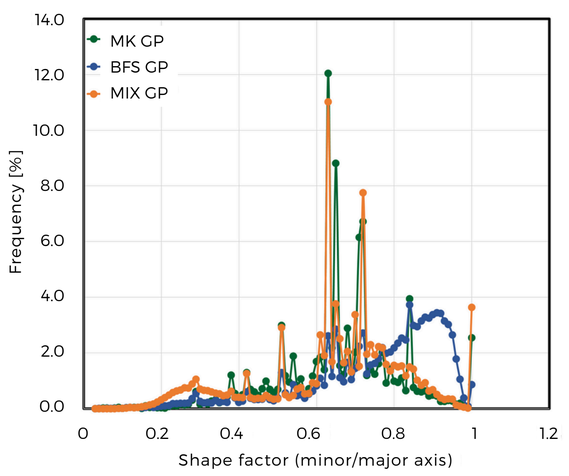
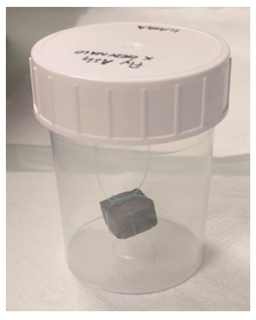


Figure 4 - Shape factor evaluated with μ-CT.



Leaching test was also performed to evaluate the **oil release**. A small quantity of oil was released from all three GP formulations investigated but could not be measured due to low volumes.

Figure 5 - Leaching test system

CONCLUSIONS

The results show how the different formulations yield varying structures and robustness, particularly in the formation of open and closed pore structures, depending on the raw materials used. Although the rate of hydrogen production and leaching is higher for the metakaolin-based GP, these values are still compliant with regulatory limits [4]. The choice of raw materials also affect oil encapsulation; however, the variation between the most and least emulsified samples does not result in significant oil release during leaching. Barring additional interventions, these promising findings are helpful for demonstrating the practical applicability of RLOW direct conditioning.

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