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DIRECT IMMOBILIZATION OF REAL RADIOACTIVE LIQUID ORGANIC WASTE IN NOVEL GEOPOLYMERS: OUTCOMES FROM THE PREDIS PROJECT

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Introduction

The safe and cost-effective management of Radioactive Liquid Organic Wastes (RLOWs) poses many challenges. The physico-chemical properties of RLOW severely hinder their immobilization and solidification with conventional conditioning matrices such as Ordinary Porland Cement (OPC), with which only modest waste loading factors can be achieved. To better exploit the potential of the direct conditioning approach for the management of RLOW, new matrices with better waste compatibility are necessary.

Scope of the work

The research aimed at developing a geopolymeric (GP) matrix suitable for the immobilization and solidification of RLOW at high loading factors via the direct conditioning approach, *i.e.* without waste pre-treatment. The work was subdivided into two main parts: firstly, a matrix formulation with enhanced compatibility with organic liquids was developed. Then, waste forms were produced by incorporation of both surrogate and real RLOW samples. Obtained specimens were tested for durability after accelerated degradation and evaluated in terms of mechanical strength and radionuclide immobilization effectiveness. Validation of the results was performed by comparison with Waste Acceptance Criteria (WAC).

PREDIS

Materials and Methods

Formulation development

GP formulation based on metakaolin, K-silicate, KOH, water, surfactant.

Surrogate RLOW: tributyl phosphate/dodecane, 30/70 vol%
Real RLOW: ligand in kerosene/1-octanol, 95/5 vol%,
spent partitioning solvent w/ actinides, 0.2 M HNO_3

Exploration of molar ratios in the range:

SiO_2/K_2O	1.0 – 1.4	**H_2O/K_2O**	11 – 15
K_2O/Al_2O_3	1.0 – 1.5	**Surfactant**	1 – 3 vol%
		RLOW	0 – 50 vol%

Preparation through high-shear mixing protocol:

RLOW + surfactant + activator sol. → High-shear mixing → **Add metakaoiln** → High-shear mixing → **Casting & curing**

Samples cured at 20 °C, >90% relative humidity

Testing protocols

Physical properties:
- viscosity, reaction heat, setting time, porosity.

Compressive strength R_c:
- Between 2 – 90 d curing time to monitor strength evolution
- After degradation by
  - 28-day water immersion
  - Co-60 irradiation up to 200 kGy

DAY 0	DAY 28	DAY 56	DAY 84	TIME
CURING	STORAGE	STORAGE	COMPRESSION	
CURING	STORAGE	IMMERSION	COMPRESSION	
CURING	IRRADIATION	STORAGE	COMPRESSION	

Radionuclides leaching: ANSI/ANS-16.1-2019 protocol, water immersion + leachate analysis to determine leachability index L of several contaminants

Validation: comparison with WAC: **R_c > 5 MPa, L > 6**

Results

Formulation development – main observations

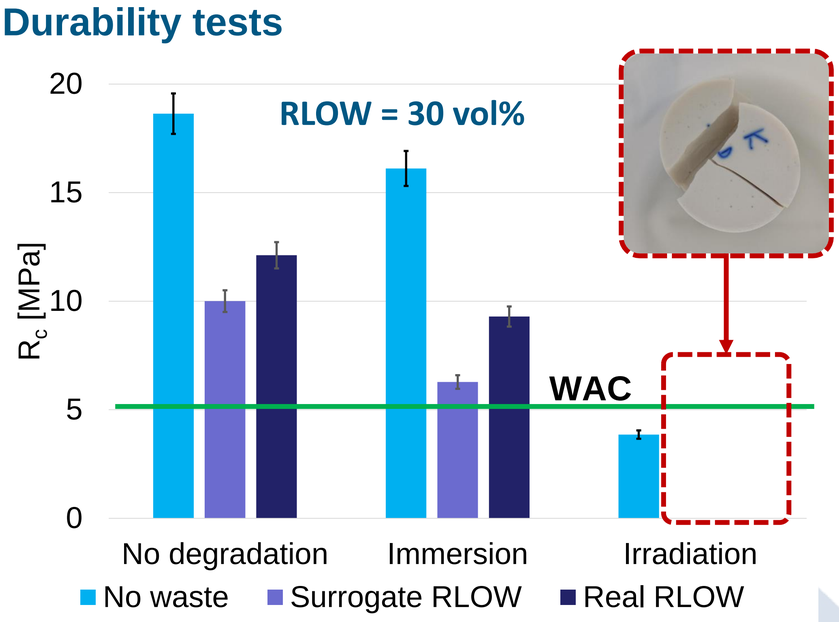
- Lowering H_2O/K_2O and K_2O/Al_2O_3 ratios results in difficult waste incorporation, especially at high loading factors
- Increasing H_2O/K_2O and K_2O/Al_2O_3 ratios results in lower compressive strength
- 3 vol% surfactant allows highest waste loading, with minimal mechanical drawbacks
- Higher waste loadings imply higher viscosity and reduced compressive strength. 30 vol% reliably achievable

Take-home message: mid-point formulation (**1.2 – 1.2 – 13 molar ratios**) as best compromise. High workability, reaction heat similar to OPC. Rapid strength gain (<10 d) with only slight bleeding (<1%), for RLOW up to 30 vol%.

Similar porosity values for all samples, regardless of waste

Porosity	No waste	30 vol% RLOW
No degradation	33%	35%
Immersion	33%	33%
Irradiation	32%	38%

Durability tests



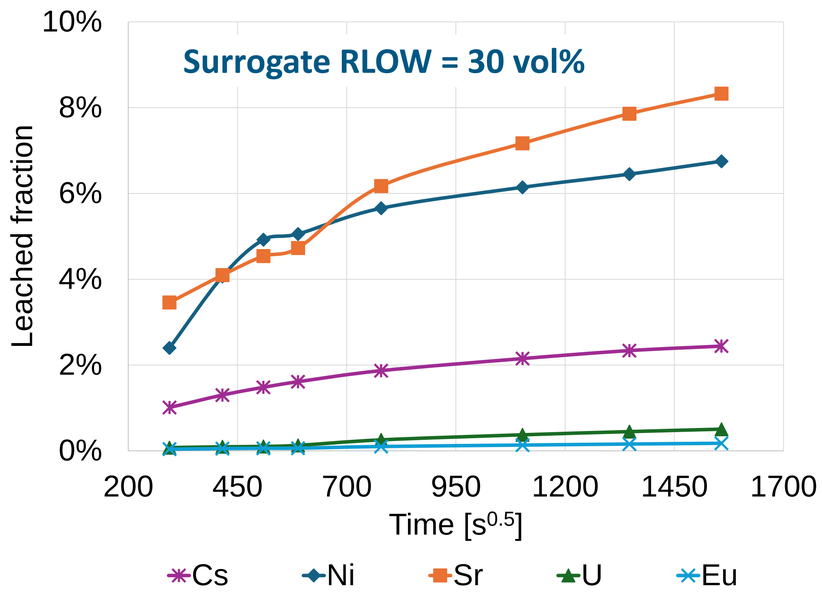
Decrease in R_c with waste, but **> WAC**

WAC satisfied also after degradation by 28 days water immersion

Missing data for samples degraded by irradiation – samples cracked due to air drying. Problem enhances in case of subsequent contact with water

No significant difference between surrogate and real waste samples = **robustness**

All leaching indexes **L > 8,** conservative with respect to WAC



Conclusions

The feasibility of direct conditioning of RLOW via a new GP formulation was assessed. The developed conditioning matrix showed good waste compatibility also at high loading factors and adequate mechanical resistance. It also demonstrated good robustness, having been applied successfully for the conditioning of RLOWs of different nature. The durability of waste forms was verified and found compliant with WAC. The matrix becomes prone to cracking in case of storage in sub-optimal conditions. This behavior should be addressed to allow its use for a safe, effective and efficient implementation of the direct conditioning approach of RLOW.

Acknowledgment

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