

Deliverable 5.4 Disposability Assessment Report for Direct Conditioning 28 June 2024 Version Final

Dissemination level: Public

Delhia Alby, Georges Daval

Galson Sciences Ltd. 5 Grosvenor House Melton Road, Oakham Rutland, LE15 6AX United Kingdom

da@galson-sciences.co.uk



Project acronym	Project title Grant agree			ement No.	
PREDIS	PRE-DIS	posal management of radioactive waste		945098	
Deliverable No.	Deliverat	Deliverable title			Version
D5.4	Disposab	ility assessment report for direct condition	oning		Final
Туре	Dissemin	Dissemination level			Due date
Report	Public			M46	
Lead beneficiary			WP No.		
Galson Sciences Limited (GSL), SOGIN 5			5		
Main author Reviewed by Accepted by		•			
Delhia Alby, GSL Liz Harvey and Steve Wickham, GSL Maria Oksa, VTT, Coord			dinator		
Contributing author			Pages		
Georges Daval, GSL			44		

Abstract

This deliverable provides information to assess the disposal implications of the direct conditioning routes for Radioactive Liquid Organic Waste (RLOW) studied in PREDIS (liquid organics, oils and solvents).

This deliverable uses all the data gathered from the information request sent in January 2024 to the partners.

This deliverable provides a methodology to assess the disposability of the formulations developed within PREDIS.

After presenting a series of generic waste acceptance criteria, the selected criteria for assessing the formulations are discussed. An assessment is then presented on the disposability of the different geopolymer formulations studied within PREDIS following a Red-Amber-Green (RAG) method.

Due to the absence of specific packaging information, only a qualitative assessment has been realised, but the results are promising, particularly regarding the oils. However, not all the criteria were assessed, and criteria related to activity and those dependent on waste package design were excluded because of the absence of data. These criteria will need to be assessed in future disposability assessments.

Keywords

Assessment, disposability, radioactive liquid organic waste

Coordinator contact Maria Oksa VTT Technical Research Centre of Finland Ltd Kivimiehentie 3, Espoo / P.O. Box 1000, 02044 VTT, Finland E-mail: <u>maria.oksa.@vtt.fi</u> Tel: +358 50 5365 844

Notification

The use of the name of any authors or organisation in advertising or publication in part of this report is only permissible with written authorisation from the VTT Technical Research Centre of Finland Ltd.

Acknowledgement

This project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 945098.

TABLE OF CONTENTS

1	INT	RODUC	TION	5
	1.1	Aims	and Objectives	5
	1.2	Scope	e, Interfaces and Exclusions	6
	1.3	Repor	t Structure	6
2	DIS	POSAB	ILITY ASSESSMENT APPROACH AND METHODOLOGY	7
	2.1	Dispo	sal concepts	8
	2.2	Asses	ssment Scenarios	11
		2.2.1	Screened out criteria due to lack of relevance to geopolymer treatment	11
		2.2.2	Criteria not assessed due to the lack of data	12
3	INP	UT DAT	Α	15
4	DIS	POSAB	ILITY ASSESSMENT CRITERIA	15
	4.1	Selec	ted Criteria	15
		4.1.1	Physical form	15
		4.1.2	Mechanical stability	15
		4.1.3	Homogeneity	16
		4.1.4	Void space	16
		4.1.5	Free liquids (before disposal)	16
		4.1.6	Chelating/complexing agents	16
		4.1.7	Leaching	17
5	DIS	POSAB	ILITY ASSESSMENT EVALUATION	17
	5.1	0ils		17
		5.1.1	Metakaolin	17
		5.1.2	MIX (MK+BFS+FA)	18
		5.1.3	Blast Furnace Slag + sand	19
	5.2	Solve	nts	20
		5.2.1	Metakaolin	20
	5.3	Scinti	llation cocktails	21
		5.3.1	Metakaolin	21
6	CON	IPARIS	ON TO THE BASELINE (VA)	23
	6.1	0ils		25
	6.2	Solve	nts	26
	6.3	Scinti	llation cocktails	27
7	CON	ICLUSI	ONS	28

LIST OF ACRONYMS

ALARA	As Low As Reasonably Achievable
BFS	Blast Furnace Slag
%vol	Volume fraction
%wt	Weight fraction
CEA	Commissariat à l'Énergie Atomique et aux Énergies Alternatives
ULA	(France)
CIEMAT	Centre for Energy, Environmental and Technological Research
	(Spain)
DGR	Deep Geological Disposal
EC	European Commission
EUG	End User Group
FA	Fly Ash
GBP	Great British Pound
GD	Geological Disposal
GDF	Geological Disposal Facility
GWAC	Generic Waste Acceptance Criteria
H&S	Health and Safety
ID	Intermediate Depth
ILW	Intermediate Level Waste
KIPT	Kharkov Institute of Physics and Technology (Ukraine)
LLW	Low Level Waste
MK	Metakaolin
MS	Milestone
NNL	National Nuclear Laboratory
NS	Near-surface
NUCLECO	Radiological Services Branch of SOGIN (Italy)
POLIMI	Politecnico di Milano (Italy)
PREDIS	PREDISposal management of radioactive waste
RAG	Red Amber Green
RATEN	Romanian Autonomous Company for Nuclear Energy
RLOW	Radioactive Liquid Organic Waste
RSOW	Radioactive Solid Organic Waste
SC	Scintillation Cocktail
SCK CEN	Belgium Nuclear Research Centre
SOGIN	Italian Waste Management Organisation
TBP	TriButyl Phosphate
TRL	Technology Readiness Level
VA	Value Assessment
WMO	Waste Management Organisation
WAC	Waste Acceptance Criteria
WP	Work Package



1 Introduction

The PREDIS project (PREDISposal management of radioactive waste) is a research and innovation action granted by the European Commission's (EC) Euratom Research Programme. PREDIS targets the development and improvement of activities for the characterisation, processing, storage, and acceptance of radioactive intermediate- and low-level waste (ILW/LLW) streams. The focus is on the treatment and conditioning of metallic materials, Radioactive Liquid Organic Wastes (RLOW) and Radioactive Solid Organic Wastes (RSOW) arising from nuclear plant operations, monitoring of encapsulated waste packages and storage facilities, decommissioning and other industrial processes.

Work Package Five (WP5) 'Innovations in liquid organic waste treatment and conditioning' of the PREDIS project is concerned with the treatment and conditioning of RLOW. WP5 is investigating and developing options for direct conditioning of RLOW using innovative geopolymers and related alkali-activated materials, resulting in less secondary waste and a safer management route of treatment and conditioning. Direct conditioning consists of encapsulating the RLOW in a solid matrix (geopolymer in this case) to obtain a composite material (conditioning and stabilising the organic waste), which can constitute the wasteform of a radioactive waste package that could fulfil the requirements of disposal facilities [1]. Figure 1.1Error! Reference source not found. illustrates the direct conditioning route developed in PREDIS WP5.



Figure 1.1: Treatment and conditioning route based on direct conditioning

WP5 is divided into tasks focused on studying the direct conditioning process (Task 5.3, T5.3), the performance of conditioned matrices, durability and their disposability (T5.4), and assessing the overall technical, economic, and environmental performance of the direct conditioning route (T5.5). This report, deliverable D5.4, falls under T5.4 (conditioning matrix performance), sub-task T5.4.9 (disposability assessment).

1.1 Aims and Objectives

Demonstration of the compatibility of the final waste package properties and performance with safety and technical requirements related to disposal is a key issue and challenge of WP5 [1]. Activities carried out under Task T5.4.9, the resulting data and disposability considerations are presented in this report and address this challenge. Deliverable D5.4 has been written with the aim of:

- Discussing and identifying considerations key to the disposability assessment of geopolymer wasteforms developed in PREDIS WP5.
- Linking these disposability considerations to wasteforms and waste package characteristics, exploiting experimental results from T5.4 [2] to assess and discuss the suitability of these wasteforms for final disposal.
- Providing, through a colour-coded Red-Amber-Green (RAG) analysis, an indication of disposability considerations that are seen as non-challenging (green), slightly challenging (amber), or extremely challenging (red).

Detailed guidance on the application of the RAG rating is provided in Section 2.

The overarching objective of this deliverable is to provide technology developers and end-users with an objective assessment of the likely performance of waste packages produced via the direct conditioning route in terms of disposability. Due to the relative novelty of geopolymers as waste matrices, and uncertainties regarding their application to RLOW, such an assessment is not definitive; rather, it provides an easy-to-read dashboard of wasteform characteristics that requires improvement or substantiation if final disposability is to be sufficiently demonstrated. Conversely, it also highlights areas assessed as already meeting disposability requirements.

1.2 Scope, Interfaces and Exclusions

The scope of this report is limited to the following three sub-groups of RLOW, which are the types of RLOW studied in PREDIS WP5:

- Oils.
- Solvents.
- Scintillation cocktails.

Other RLOW types are excluded from the scope of this report.

The three geopolymer formulations selected for further research in the course of the project (see milestone MS34 [3] and deliverable D5.2 [4]) are included in the scope of this report, namely:

- The Metakaolin (MK)-based formulation developed by NNL.
- The Blast Furnace Slag (BFS)-based formulation developed by SCK CEN; and
- The MIX formulation (based on MK, BFS, and Fly Ash (FA)), developed by KIPT.

Wasteform disposability was assessed according to the following radiological waste categories (VLLW, LLW/ILW-SL (Short Lived), and ILW-LL (Long Lived)) and according to disposal facility types, ranging from near-surface to intermediate-depth facilities, and Deep Geological Repositories (DGR). These facilities are described in Section 2.

The method for Generic Waste Acceptance Criteria (GWAC) identification derived in PREDIS WP2 [5] was used to define a set of disposability considerations. They are presented in Section 2.

1.3 Report Structure

This rest of this report is structured as follows:

- The approach and methodology used to carry out this disposability assessment are presented in Section 2. The different disposal concepts and the excluded criteria are discussed in this section.
- The input data used for the assessment are presented in Section 3.
- The criteria assessed and their definitions are presented in Section 4
- Some further criteria are detailed in Section **Error! Reference source not found.** These criteria have not been applied in this report for various reasons but would need to be considered in a later phase of disposability assessment for a particular application.
- A comparison to the baseline studied in the Value Assessment conducted under T5.5 is made in Section 6 to support the study on disposability assessment.
- Section 7 presents the conclusions of the report.
- Annexe 1 presents the data collected from the partners via the data request form

2 Disposability Assessment Approach and Methodology

The approach taken to disposability assessment was first to define the types of facilities that would be considered as potential disposal options. This was motivated by the identification of "near-surface or intermediate-depth (50-100 m) disposal facilities or geological disposal facilities (GDFs)" as a topic area of interest at the start of the project [1]. A broad set of generic facilities was defined, consistent with the range of planned and operating LLW and ILW disposal facilities in Europe spanning from surface facilities through to DGRs. These facilities are described in Section 2.1. The concepts are also studied in [6].

Guidance on formulating Generic Waste Acceptance Criteria (GWAC) has been developed under PREDIS WP2 and is made available in Deliverable 2.7 [5]. Many aspects of this guidance informed the selection of assessment areas and the definition of GWAC. Such work is particularly relevant to assessments such as this one, which are wasteform-specific, but facility-generic. Therefore, this report presents a disposability assessment against GWAC, defined as:

"WAC put in place to facilitate predisposal waste management planning or processing activities in the face of ongoing uncertainty over downstream management strategies (such as storage or disposal routes) and, hence, over detailed site- or facility-specific requirements. [5]"

The relevant waste management scenarios (e.g. disposal to near-surface or geological disposal facilities), assessment areas and criteria were identified, based on a review of existing literature and D2.7 [5]. Justification for the inclusion or exclusion of assessment areas is provided in Section 2.2. This section also aims to establish, when possible, the range of acceptability for a given waste package characteristic or acceptance criterion. However, this is not always possible due to:

- Variability in country-specific requirements. This has been mostly overcome by broadening the range of acceptability for acceptance criteria.
- Risk-informed approach to disposal: for some criteria the waste producer may need to demonstrate that dose rates or risk has been reduced to levels as low as reasonably achievable (ALARA). Where this applies it is explicitly noted in the relevant section.

Based on these GWAC, the suitability of geopolymer wasteforms for disposal to various types of facility was assessed, using the data listed in Annexe 1.

Disposability was assessed using a mixture of qualitative and quantitative arguments. Due to the generic nature of this assessment, a traffic light system was established (see Table 2.1) and, for each assessment area, a rating was attributed.

Rating	Risk to disposability
Green	No foreseeable risk to disposability.
Grey	No, limited or partially applicable experimental data or industrial experience to substantiate disposability.
Amber	Limited risk to disposability, which may be addressed through further development work. Waste product behaviour falls at the frontier of what is typically considered acceptable for a given parameter.

Table 2.1: Assessment	outcome	definitions
-----------------------	---------	-------------



Rating	Risk to disposability
Red	Significant risk to disposability. Waste product behaviour falls well outside what is typically considered acceptable for a given parameter.

2.1 Disposal concepts

To evaluate the disposability considerations applicable to the range of proposed or operating LLW and ILW disposal facilities across Europe, a set of five generic disposal facilities were defined at depths spanning from surface to deep geological disposal [6]. These generic facilities are not necessarily consistent with any one facility, but instead defined to capture the broad characteristics of a single 'class' of facilities.

Each of the considered disposal concepts was given an identifier consisting of two letters and a number, with the letters indicating whether it is a Near-Surface (NS), Intermediate Depth (ID) or Geological Disposal (GD) concept, and the number differentiating concepts in the same category. The considered disposal concepts are illustrated in Figure 2.1 and described in further detail in the subsections below, including assumptions about each concept that have relevance to disposability.

In reality, there is a continuum of possible facility depths which may be considered for underground facilities. In this report, geological disposal concepts are considered to include both DGRs (typically deeper than 200 m) and shallower DGR-like facilities. Geological disposal concepts are distinguished from intermediate-depth facilities in this report by their increased reliance on geological barriers.

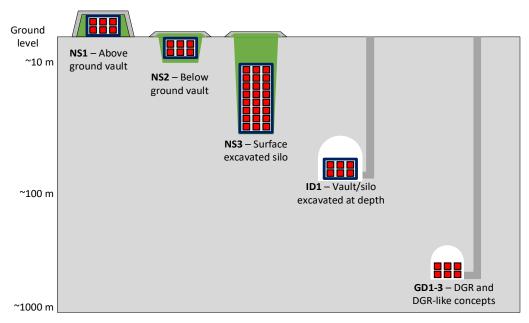


Figure 2.1: Illustration of the generic disposal concepts considered in this work. Illustration is based on [6] [7]

NS1 – Above-ground vault



- **Description:** Waste is emplaced in vaults at ground level over an operational period spanning several decades, and the facility is mounded over with an engineered cover. Positioning above the water table combined with engineered barriers and water management systems means that the waste is unsaturated (and potentially entirely dry). The position at the surface means there is a risk of large-scale human intrusion following the end of institutional control.
 - **Barriers:** Durable concrete or steel packages with a concrete or gravel backfill inside concrete vaults. The facility is covered by an engineered cap consisting of layers of polyethylene, clay, gravel and soil.
 - **Depth:** 0 m (natural ground level)

Wastes: LLW¹

- **Examples:** Category A facility, Dessel, Belgium
 - L/ILW facility, El Cabril, Spain
 - CSA, Aube, France

NS2 – Below ground vault

- **Description:** Waste is emplaced in engineered vaults excavated from the surface which are then filled to ground level or mounded over. The waste becomes saturated after closure, with engineered barriers aimed at preventing or reducing water flow. Positioning at the surface means there is a risk of large-scale human intrusion following the end of institutional control.
 - **Barriers:** Durable concrete or steel packages with a concrete backfill inside concrete vaults. The facility is covered by an engineered cap consisting of layers of polyethylene, clay, gravel and soil.
 - Depth: 0-20 m
 - Wastes: LLW
 - **Examples:** LLWR, Drigg, UK
 - RÚ RAO L/ILW facility, Mochovce, Slovakia

NS3 – Surface excavated shaft or silo

- **Description:** Waste is emplaced in a surface excavated silo consisting of a vertical excavation from the surface to several tens of metres depth. The top of the emplaced waste is significantly (~10 m) below ground level, with the remainder of the silo containing the engineered cap. Emplacements at greater depths than other surface excavated concepts reduce the risk of human intrusion. Waste becomes saturated after closure, with engineered barriers aimed at preventing or reducing water flow.
 - **Barriers:** Durable concrete or steel packages with a concrete backfill inside the concrete lined silo. The silo is capped with concrete, clay and soil.
 - Depth: 10-70 m
 - Wastes: LLW
 - Examples: LILW disposal facility, Vrbina-Krško, Slovenia

¹ The waste classifications used are in accordance with the IAEA General Safety Guide GSG-1

ID1 – Silo or vault-type facility excavated at depth

- **Description:** Excavation of a vault or silo at intermediate depth and accessed by a shaft or draft which will be subsequently backfilled. The increased disposal depth provides a higher degree of isolation from surface processes. Significant reliance is still placed on engineered barriers.
 - **Barriers:** Durable concrete or steel packages in a concrete-lined vault which is backfilled with concrete. Plugging of access tunnels by concrete, crushed rock, clay or a mixture of these.
 - **Depth:** 50-100 m
 - Waste: Up to ILW

Examples: • VLJ L/ILW facility, Olkiluoto, Finland

- SFR L/ILW facility, Forsmark, Sweden
- National Radioactive Waste Repository, Bátaapáti, Hungary

GD1 – Geological disposal in High Strength Rock

- **Description:** Disposal to a DGR (typically >200m) or a shallower DGR-like concept (200m) located in a low permeability, high strength rock (e.g. granite). Distinguished from intermediate depth disposal based on an increased reliance on geological barriers to provide isolation and prevent release, and on the increased need to manage potentially heat-generating waste. Expect radionuclide transport in the host rock to be dominated by advection resulting from groundwater flow through fractures.
 - **Barriers:** Concrete, steel, or other metallic packages in a vault backfilled with a cementitious grout or bentonite. Host rock provides a significant barrier to radionuclide migration.
 - Depth: 200-800 m
 - Wastes: Up to HLW
 - **Examples:** ONKALO® DGR facility, Olkiluoto, Finland
 - SFL ILW facility, Sweden (Proposed)

GD2 – Geological disposal in Low Strength Sedimentary Rock

- **Description:** Disposal to a DGR (typically >200 m depth) or a shallower DGR-like concept (200 m depth) located in a low permeability, low strength sedimentary rock (e.g. clays, shales, mudstones). The low strength of the host rock means that fractures cannot be maintained and will self-seal. Expect radionuclide transport in the host rock to be dominated by diffusion through the rock matrix.
 - Barriers: Reinforced concrete disposal container in a tunnel backfilled with a cementitious grout and presence of metallic overpack or sleeve. Host rock provides a significant barrier to radionuclide migration.
 Depth: 200-800 m
 - Wastes: Up to HLW
 - **Examples:** Cigeo DGR, Meuse, France (proposed)
 - DGR facility, Nördlich Lägern, Switzerland (proposed)



GD3 – Geological disposal in Evaporite

Description: Disposal to a DGR located in an evaporite (salt) formation (typically halite). Evaporites exhibit significant plastic flow (creep) which will tend to close any open fissures and excavations over time. Evaporite formations provide a dry geological environment such that there is expected to be effectively no transport of radionuclides outside of the gas phase.

Barriers: Vaults are backfilled with crushed host rock. The evaporite host rock is the primary barrier to radionuclide migration.

Depth: 200-800 m

Wastes: Up to HLW

- **Examples:** WIPP, New Mexico, USA
 - ERAM, Morsleben, Germany

2.2 Assessment Scenarios

2.2.1 Screened out criteria due to lack of relevance to geopolymer treatment

Several factors can influence disposability, they vary between the different facilities and jurisdictions. PREDIS Deliverable 2.4 presents a review of international waste acceptance systems for radioactive waste [8]; the assessment areas adopted here are based on the areas identified in that review ([8], Table 3). Identified assessment areas were screened to remove assessment areas considered not relevant or not differentiating the considered technologies and geopolymer waste forms; the screened out assessment areas are listed in **Error! Not a valid bookmark self-reference.**

Assessment	Description	Reason for screening out
area		
Chemo-toxic waste	Presence of chemically toxic species, typically defined by legislation (EU water framework directive, EU REACH regulations, etc.). Examples include mercury, lead, cadmium and per- and polyfluorinated substances (PFAS).	Considered that the geopolymer treatment approaches will not introduce chemically toxic species. The presence of chemo-toxic species and the resulting suitability of the treatment approach will be waste-stream specific, and not related to the geopolymer treatment process.
Reactive metals	Typically, mass or reactive metals are limited or requirement that issues with reactive metals (gas generation, expansive corrosion) are mitigated.	Considered that the geopolymer treatment approaches will not introduce reactive metals. The presence of reactive metals and their behaviour during treatment and disposal will therefore be waste-stream specific, and not related to the geopolymer treatment process.
Heat generation	Radiological heat generation, dependent on the activity of the waste.	It is assumed that waste will be LLW or ILW (including after thermal treatment), therefore, heat generation will not be a specific concern.
Management and data recording	Ability to add a durable and readable waste package label/identifier and other	It is expected that suitable management and data recording

Table 2.2: Disposability assessment areas, which were screened out of consideration.



Assessment area	Description	Reason for screening out
	documentation and tracking considerations.	processes may be developed for any of the considered treatment approaches.
Putrescible, fermenting, or infectious material	Of concern for biodegradable organic materials. Limits may be specific or derived from gas, void space, or chemical stability requirements. Infectious materials (such as carcasses) are typically required to be excluded.	Considered that treatment approaches will not introduce putrescible, fermenting, or infectious material. The presence of putrescible, fermenting or infectious material and their behaviour during treatment and disposal will therefore be waste-stream specific, and not related to the geopolymer treatment process.

2.2.2 Criteria not assessed due to the lack of data

Some assessment areas are of interest but cannot be assessed at this stage of the project because of the absence of data related to the package or the radioactivity of the wasteform. They are detailed in **Error! Not a valid bookmark self-reference.**

Table 2.3: Disposability assessment areas to account for disposability but not be assessed due to
the absence of data

Assessment area	Description	Reason for screening out
Dose-rate	The dose rate (measured as contact dose or at nominal stand- off) impacts handling requirements for the package. It will be impacted by the waste activity and the shielding by the package and wasteform.	NS1-3: Near-surface facilities typically have handling processes designed with minimal shielding. High-dose-rate packages are more likely to be unacceptable. This criterion is not assessed because of the absence of data regarding the radiological classification of the waste.
Surface contamination	Surface contamination is typically required to be very low to minimise particle dissemination hazard, and may consider both radioactive and non-radioactive contamination (e.g. salt deposits).	This criterion is not assessed because of the absence of data regarding the radiological classification of the waste and the package.
Activity content	There is typically a maximum acceptable activity content for waste packages disposed to near- surface and intermediate-depth facilities. It may be defined in terms of specific activity, activity concentration or total activity. Limits may be set on the activity content of individual radionuclides or based on total alpha and beta/gamma activities.	Waste packages with higher total activities and containing more long- lived radionuclides typically require a greater degree of containment and isolation from the surface environment. This criterion is not assessed because of the absence of data regarding the radiological fingerprint of the waste.
Radiation stability	Stability of wasteform under ambient radiation and under extreme conditions.	Post-irradiation strength tests carried out at POLIMI on the MK formulation show a marked reduction in compressive strength, without it being



Assessment area	Description	Reason for screening out
		possible to dissociate the effect of irradiation and the effect of drying, which as we saw in the first chapters is important for this matrix [9].
		New experiments are needed to provide further information on radiation stability of geopolymer wasteforms.
Package	Use of standard/approved packages Maximums on package weight and size. Package performance under impact accidents. Package stacking	NS3/ID2: Stacking packages to significant heights requires silo or vault concepts. Stacking typically requires consideration of larger drop heights than other concepts. This criterion is not assessed due to the absence of data on specific packaging
Radiological gas generation	Generation of radiological gases. Typically, facilities place an upper limit on the production of radiological gases. Gases of concern include radon (uranium decay series) and tritium or C-14.	concepts. This criterion is not assessed because of the absence of data regarding the radiological composition of the waste.
Void space in the package	Presence of void space in the package. Need to consider void space that will evolve within a package due to compression or degradation. The type of facility (near-surface, geological disposal), facility design and geological context will have limits on acceptable voidage and the processes considered.	 NS1/NS2: Tolerance to void space depends on the engineering of the cap and vault. GD1: Potentially very tolerant of void space GD2: Potentially not tolerant of void space GD3: Moderate tolerance of void space This criterion is not assessed because of the absence of data on the package types.
Non- radiological gas generation	Generation of gases due to radiological and chemical degradation. Concerns include the over-pressurisation of the container and the creation of pathways for radionuclide migration.	This criterion cannot be assessed due to the absence of waste-specific data.
Swelling/ shrinkage after packaging	Excessive swelling or shrinkage (over time or due to exposure to, for example, water) is typically prohibited	NS1/NS2: higher potential for seasonal dehydration/rehydration and thermal cycling. This criterion is not assessed due to the absence of waste-specific data.
Free liquid after disposal	The presence of significant free liquids in a waste package is typically unacceptable in any type of repository.	This criterion corresponds to the free liquid which could be produced after packaging and disposal. It is unrelated to the bleed that may occur after mixing RLOW with organic liquids and which is typically fed back into the geopolymer encapsulation process.



Assessment area	Description	Reason for screening out
Criticality	Criticality risk is impacted by fissile element mass and the presence and configuration of neutron moderators, poison and reflectors. This criterion has only an impact where significant fissile mass is disposed, and therefore only on a GDF facility. It includes specific consideration as to how the encapsulant material interacts with the fissile material, along with the additional aspect of long-term behaviour for post-closure. Criticality safety for all GDF lifecycle phases and post-closure timescales and scenario evolution will need to be demonstrated [18].	Criticality will be of concern for wasteforms that have a large loading of fissile material. For geopolymer encapsulation of RLOW this could only ever be relevant to oils containing significant fissile mass in suspension.
Wasteform Interactions (Container and Engineered Barriers)	Issues could arise regarding the evolution of the pH within the near- field environment with implications for the longevity of the container and engineered barriers [10].	This criterion is not assessed because it concerns the evolution of the container and the engineered barriers, not the geopolymer wasteform.
Evaluation of Performance under Accident Conditions	Evaluation of impact and fire performance	No data was given regarding the impact or fire accident performance. The products are likely to offer adequate impact performance, but it is recommended that small-scale testing, supported by modelling, be undertaken to establish the likely behaviour and release fractions from an impact accident. Moreover, the brittle nature of the encapsulants (notably Metamax) will need to be considered further for impact accident performance [10]. Regarding the fire performance, the criteria will have to be experimentally evaluated. However, the first results show that the formulations tested (MK and MIX ones) show microcracking of the matrices under thermal cycling. Moreover, the MIX formulation seems to offer a higher resistance to temperature in the range between 200°C and 300°C but more experiments need to be carried out [9].

3 Input Data

In the first year of the PREDIS project, in order to identify priority waste streams for WP5, an inventory questionnaire was distributed to all WP5 partners to identify the main wastes present in the partner countries. The responses to this questionnaire identified the following main types of RLOW (also noting the organisations that identified them):

- Oils: KIPT, CVRez, SOGIN, UJV, RATEN, CEA
- Solvents: KIPT, Sellafield, NNL, CVRez, SOGIN, UJV, CEA
- Scintillation cocktails: CVRez, UJV, RATEN
- Cleaning / decontamination liquids: UJV, CVRez
- Organic effluents: CEA

An input data synthesis report was produced in 2021 to summarise this data [11]. The wastes include materials that have already been produced, and some that will arise from future operations and decommissioning.

Following discussion between the partners it was decided that the priority waste streams to consider in WP5 would be oils, solvents and scintillation cocktails, as these were present in most countries. This was explained in PREDIS MS30. The oils considered include low-viscosity oils (such as Nevastane) that may be operational wastes (pump oils) or legacy wastes; and an organic solvent (TBP-dodecane, 30:70 volume ratio). These are the two largest waste groups (by volume) identified in Europe.

The data used in this report were obtained after sending a data request to the partners of the project in January 2024 [12] [13] [14] [15] [16] [17] and summarised in Annexe 1 and from the Value Assessment workshop realised in February 2024 and whose conclusions are presented in Deliverable 5.5 [18].

4 Disposability Assessment Criteria

4.1 Selected Criteria

4.1.1 Physical form

Typically, to be disposable, there is a requirement for the wasteform to be physically solid and compact. In the following assessment, if a wasteform has a monolithic form, it would be given a green rating, whereas a non-monolithic wasteform would be given a red rating.

NS1-2: The presence of discrete objects with a high dose rate will be of concern due to the significant risk dose impact in human intrusion scenarios.

4.1.2 Mechanical stability

The compressive strength of the material is an important property and disposability consideration. Indeed, all facilities propose a minimum compressive strength value for the waste to be accepted for disposal. From the WAC studied [19] [20] [21] [22], the value differs in each country. All countries consider that a wasteform with a compressive strength of at least 10 MPa can be accepted. In this case, no risk to disposability is foreseen and in the following assessment such a value is given a green rating. Between 5 MPa and 10 MPa, limited risk to disposability is foreseen, allowing an amber rating to be assigned. From the WAC studied, no disposal facilities accept a wasteform with a



compressive strength under 5 MPa, such a value would be given a red rating for a significant risk to disposability.

NS1-3: Potential for freeze/thaw cycling.

NS3/ID2: Silo-type concepts typically require the ability to stack packages to larger heights than vault concepts.

4.1.3 Homogeneity

The immobilisation matrix is typically required to be homogeneous, requiring that there is no segregation of waste/matrix. In this case, a homogeneous wasteform would be given a green rating and a heterogeneous wasteform would be given a red rating.

4.1.4 Void space

Void space in the WAC for disposal facilities is usually attributed to the total voiding in the package. In Italy for example [19] it is evaluated to be 10% maximum. It is important to consider void space that may evolve within a package due to thermo-mechanical stress and/or other degradation mechanisms. The type of disposal facility (near surface, geological disposal), facility design and geological context will impact the allowable voidage and the processes considered. This parameter cannot be assessed due to the absence of data on the packaging. However, some partners evaluated the porosity of the wasteform. As an example, CIEMAT estimated in the case of oils encapsulated in geopolymer, a porosity of 29% in aerated conditions and 41% in a closed vessel (plastic film) with the use of metakaolin. For the case of solvents encapsulated in geopolymer, POLIMI estimated a porosity of 33 to 38% with the use of metakaolin. For scintillation cocktails, with metakaolin, POLIMI estimated a porosity of 33 to 38% and CIEMAT of 50% in aerated conditions.

NS1/NS2: Tolerance to void space depends on engineering of cap and vault.
GD1: Potentially very tolerant of void space
GD2: Potentially not tolerant of void space
GD3: Moderate tolerance of void space

4.1.5 Free liquids (before disposal)

The presence of significant free liquids within a waste package is typically unacceptable. However, during the Value Assessment workshop [18], it was decided that the bleed produced by geopolymer wasteforms before disposal should not be taken into account as it will be captured and reintroduced into the geopolymer encapsulation process for the next batch. Therefore, a green rating was given to all the formulations studied.

4.1.6 Chelating/complexing agents

It is important to highlight the presence of chelating/complexing agents (such as superplasticisers) that may evolve within a package, and which could increase the mobility of radionuclides in the near-field and geosphere. These agents are typically required to be excluded or of limited mass, since they have the potential to increase the mobility of radionuclides and contaminants. Therefore, the absence of chelating/complexing agents results in a green rating and the presence of chelating/complexing agents results in a red rating.

A surfactant may behave as a complexant in some cases. The potential detrimental impacts of surfactants include the following [23]:

- Functional groups in the hydrophilic 'heads' of surfactant molecules can act as ligands, forming complexes that help to promote solution of radionuclides.
- Surfactant degradation products can generate non-aqueous phase liquids (NAPLs), potentially mobilising radionuclides via a non-aqueous pathway.
- Surfactants can help to stabilise mobile colloid species, facilitating colloid-mediated radionuclide transport.



All of these effects will depend on the nature of the surfactant and its degradation products, as well as factors such as dissolved ions in the water (particularly Ca, Mg, Na) and other environmental factors. There will not always be a significant effect from the presence of surfactants. Nevertheless, their presence indicates a potentially detrimental series of indirect effects in the disposal environment that would need to be evaluated on a case-by-case basis.

The use of a surfactant in this study is usually in order to promote [4]:

- A decrease in bleeding
- A homogeneous material even if the workability decreases
- Improved miscibility
- An increase in the stability of the product

The surfactant is usually added at the first stage of the process.

NS1: Unsaturated conditions reduce the impact of complexing agents. Timescales of concern for the evolution of complexing agents are short (~300 years).

NS2-3: Timescales of concern for the evolution of complexing agents are short (~300-10,000 years).

ID1: Moderate timescales of concern for the evolution of complexing agents (100,000 years). **GD1-2:** Timescales of concern for the evolution of complexing agents are very long (up to 1,000,000 years).

GD3: Dry conditions mean that chelating/complexing agents are not a concern.

4.1.7 Leaching

The rate of leaching of radionuclides and hazardous chemicals from the wasteform into groundwater is typically required to be low. Water-soluble species such as chlorides/sulphates may be excluded. The partners used different methods to estimate leaching over different timescales. It is therefore difficult to make a direct comparison. As the limit specified in the WAC can be different from one country to another, it is proposed that only leaching tests following the ANSI/ANS-16.1-2019 protocol would be considered. For the other tests, comparison to an international norm or value is difficult and these are not given a colour rating and will stay grey.

NS1: unsaturated conditions reduce the impact of leaching. **DG3:** Dry conditions mean that leaching is not a concern

For information, the waste loadings are presented in the tables below but are not considered for disposability assessment.

5 Disposability Assessment Evaluation

5.1 Oils

5.1.1 Metakaolin

5.1.1.1 CIEMAT

Data from CIEMAT	
Assessment	Identified disposability risks
area	
Waste loading	Up to 40% vol
Physical form	Monolithic product

Data from CIEMAT	
Assessment	Identified disposability risks
area	
Mechanical	Oil 40%vol.:
stability	 28day-curing in a closed vessel (plastic film) conditions: 10.9MPa
	 28 day-curing in aerated conditions: 11.6MPa
Homogeneity	Homogeneous product
Void space	Porosity is between 29 to 41% in function of the curing conditions.
Free liquids	No free liquids were observed
Chelating/ complexing agents/surfactant	No chelating/complexing agents used
Leaching	Oil 40%vol:
	- Aerated: 0,4%vol. leached (worst case: 1,65%vol.)
	- In a closed vessel (plastic film): 0.15%vol. leached

5.1.1.2 NNL

Data from NNL	
Assessment	Identified disposability risks
area	
Waste loading	Up to 50% vol
Physical form	Monolithic product
Mechanical stability	The mechanical strength is higher than 10 MPa
Homogeneity	Homogeneous product
Void space	No void observed
Free liquids	Bleed was recorded at a maximum of 1.25 vol% of product for all formulations at 48 h. It was assumed that all the bleed is recycled within the same geopolymer process.
Chelating/ complexing agents/surfactant	No chelating/complexing agents used
Leaching	No leaching test conducted

5.1.2 MIX (MK+BFS+FA)

Data from NCS KIPT	
Assessment	Identified disposability risks
area	
Waste loading	30% vol
Physical form	Monolithic product



Data from NCS K	Data from NCS KIPT	
Assessment	Identified disposability risks	
area		
Mechanical stability	Compres. strength = 32 MPa (28 day of curing) for geopolymer without waste Compres. strength = 15 MPa (28 days of curing) for geopolymer with 30% of	
	oils	
Homogeneity	The product is homogeneous	
Void space	Dense homogeneous structure of hardened samples without large pores	
	No macroscopic void spaces and cracks	
Free liquids	No water observed to be exuded	
Chelating/ complexing agents/surfactant	No chelating/complexing agents	
Leaching	Without any oil, a leaching rate of 3.75 mg/cm^2 is measured at its maximum for Si and 0.75 for Al before decreasing. The maximum leaching rate is similar for the geopolymers with 30% of oil at pH = 12.7.	
	No AI is leached after 30 days without oil while an amount of 0.2 mg/cm ² is present after 30 days with 30% of oil. For AI, the leaching rate of Si is similar for 90 days.	

5.1.3 Blast Furnace Slag + sand

Data from CVRez/SCK CEN	
Assessment	Identified disposability risks
area	
Waste loading	30 wt. %
Physical form	Monolithic product
Mechanical stability	The mechanical strength depends on the amount of oil added (5 wt. % Nevastane incorporated >10 MPa, more than 5 wt.% of Nev. < 10 MPa, Mogul oil both 5 and 10 wt.% >10 MPa)
Homogeneity	It depends on the amount of surfactant added
Void space	Porosity (mercury method, 0.1-400 MPa) varied between samples from 13–21 %.
Free liquids	Laboratory samples did not exude water
Chelating/ complexing	Surfactant used
agents/surfactant	Tween 80 was used as a surfactant to enhance the mixing between the oil and BFS [2]. The impact of this surfactant still needs to be assessed.
Leaching	Leaching experiments: demineralised water, 90 days, laboratory temperature, leachate replacement and analysis days 2, 7, 14, 28, 56 and 90. The median of leached oil (cumulative) is 1.1 %.

5.2 Solvents

5.2.1 Metakaolin

Data from NNL	
Assessment area	Identified disposability risks
Waste loading	Up to 30% vol
Physical form	Monolithic product
Mechanical stability	Compressive strength of products ranged from 10.9 MPa – 40.3 MPa at 90 d at 90 days over a range of molar ratio formulations for MetaMax MK
Homogeneity	Homogeneous product
Void space	Visual assessment of the external surfaces showed no significant macroscopic voidage. Additionally, some samples were split in half and these also showed no voidage.
Free liquids	Potential for free liquid in the form of TBP, dodecane (70:30) and 1-3 vol % of Tween 80 (surfactant) to TBP/dodecane in the pores of the geopolymer matrix Assumed to be reinjected into the process.
Chelating/ complexing agents/surfactant	The waste was emulsified using a surfactant of 1-3 vol% of the volume of waste (10-30 vol%). The impact of this surfactant still needs to be assessed.
Leaching	None conducted

Data from POLIMI	
Assessment	Identified disposability risks
area	
Waste loading	30% vol
Physical form	Monolithic product
Mechanical stability	Compressive strength without any waste: 18,6 MPa, with waste: 10 MPa
Homogeneity	Homogeneous
Void space	If samples are stored in a humid environment (>90% relative humidity), nothing significant happens, and samples are durable (no cracks or defects after immersion). On the contrary, if samples are stored in <90% relative humidity, i.e. even
	under normal ambient conditions, they tend to dry out and loose about 25% of their initial weight. This becomes a problem when samples then get in contact with water: they reabsorb the water they have lost and they crack dramatically
Free liquids	Wasteforms bleed an amount of water <1% of the total wasteform volume during curing. Afterwards, no additional bleeding was observed (observation time of 6 months after casting). It is assumed that the bleed is reinjected into the process
Chelating/ complexing agents/surfactant	The waste was emulsified using a surfactant. Tween 80 was used as a surfactant at 3 %w. before adding the activator solution and the metakaolin [9]. The impact of this surfactant still needs to be assessed.



Data from POLIMI		
Assessment	sment Identified disposability risks	
area		
Leaching	The leachability indices are all superior to 8.	

Data from UJV	
Assessment	Identified disposability risks
area	
Waste loading	No information
Physical form	Monolithic product
Mechanical stability	21.93 MPa (Ni-63), 16.51 MPa (C-14)
Homogeneity	Homogeneous
Void space	No significant cracking or significant intergranular porosity.
Free liquids	No tendency for the final product to exude water.
Chelating/ complexing agents/surfactant	Use of a surfactant. The impact of using a surfactant still needs to be assessed.
Leaching	The required minimum value of the leachability index "Li" 8 was attained.

5.3 Scintillation cocktails

5.3.1 Metakaolin

Data from POLIMI	
Assessment area	Identified disposability risks
Waste loading	30% vol
Physical form	Monolithic product
Mechanical stability	Compressive strength was studied (without any waste: 18.6 MPa, with waste: 8.9 MPa)
Homogeneity	Homogeneous product
Void space	If samples are stored in a humid environment (>90% relative humidity), nothing significant happens, and samples are durable (no cracks or defects after immersion).
	On the contrary, if samples are stored in <90% relative humidity, i.e. even under normal ambient conditions, they tend to dry out and lose about 25% of their initial weight. This becomes a problem when samples then get in contact with water: they reabsorb the water they have lost and they crack dramatically



Data from POLIMI	
Assessment	Identified disposability risks
area	
Free liquids	Wasteforms bleed an amount of water <1% of the total wasteform volume during curing. Afterwards, no additional bleeding was observed (observation time of 6 months after casting). The bleed is supposed to be reinjected to the process.
Chelating/ complexing agents/surfactant	Most LSC cocktails contain surfactants (usually around 2%w).
Leaching	The leachability indices are all superior to 8.

Data from UJV	
Assessment	Identified disposability risks
area	
Waste loading	No information
Physical form	Monolithic product
Mechanical stability	25.71 MPa (Ni-63), 18.6 MPa (C-14)
Homogeneity	Homogeneous
Void space	No significant cracking or significant intergranular porosity.
Free liquids	No tendency for the final product to exude water.
Chelating/	Most LSC cocktails contain surfactants (usually around 2%w). The impact of
complexing	this surfactant still needs to be assessed.
agents/surfactant	
Leaching	The required minimum value of the leachability index "Li" 8 was attained.

Data from CIEMA	Data from CIEMAT							
Assessment	Identified disposability risks							
area								
Waste loading	30% vol							
Physical form	Monolithic product							
Mechanical stability	28day-curing in a closed vessel (plastic film) 25.5MPa							
Homogeneity	Homogeneity not assessed.							
	Precursors and activator not fully reacted: detected in first leaching steps							
Void space	Total porosity (%):							
	LSC 30%vol:							
	Aerated: 50%							
Free liquids	The amount of water, a priori not significant, but cannot be assessed or extrapolated to larger volumes. Scaling-up required							
Chelating/ complexing agents/surfactant	Most LSC cocktails contain surfactants (usually around 2%w). The impact of this surfactant still needs to be assessed.							
Leaching	Regarding the leaching of LSC, estimations based on 1-year leaching tests in deionised water are:							



Data from CIEMAT								
Assessment	Identified disposability risks							
area								
	LSC 30%vol. aerated: 15%vol. leached							

6 Comparison to the baseline (VA)

For Value Assessment, the different technologies were compared to a baseline [18]. The comparison is presented in Table 6.1. The disposal route is assumed to be near-surface disposal. Not all the formulations considered in disposability assessment were evaluated in value assessment.

Waste Type	Scenario ID	Formulation/process description	Waste	Radiological classification
Oil	5.1.1	Encapsulation in metakaolin (Metamax) geopolymer	Nevastane oil	LLW
	5.1.2	Encapsulation in composite metakaolin (Metamax), blast furnace slag (Ecocem), fly ash (Italy) geopolymer	Nevastane oil	
	5.1.3	Encapsulation in blast furnace slag geopolymer	Nevastane oil	
	5.1.B	Two-step process: Step 1: absorption onto Experlite and transfer to a 115 L drum. The sorbent is then encapsulated with cement. Step 2: 115 L is placed into a 216 L drum. Cement is used to fill the void between the two drums. Cement is assumed to be ordinary Portland cement.	Nevastane oil	
Solvents (TBP- Dodecane)	5.2.1	Encapsulation in metakaolin (Metamax) geopolymer	TBP-Dodecane (30/70)	LLW and ILW suitable for near- surface disposal ²
	5.2.B	Step 1: transport to, and incineration at an incinerator using the IRIS process (assumption: at the CEA in France) Step 2: cement encapsulation of ashes in 200 L drum (assumption: collocated with incinerator).	Solvents (incl. TBP-dodecane 30/70) used in the PUREX process (spent fuel reprocessing).	
Scintillation Cocktails	5.3.1	Encapsulation in metakaolin (Metamax) geopolymer	INSTAGEL Plus	LLW

Table 6	6.1: B	aseline	scenarios
---------	--------	---------	-----------

² Based on the activity values used in active experiments, as reported in D5.2 (38 GBq/t Ni-63/C-14).

D5.4: Disposability Assessment of Waste Packages Produced via the Direct Conditioning Route

Waste Type	Scenario ID	Formulation/process description	Waste	Radiological classification
	5.3.B	 Two-step process: Step 1: absorption onto Experlite and transfer to a 115 L drum. The sorbent is then encapsulated with cement. Step 2: 115 L is placed into a 216 L drum. Cement is used to fill the void between the two drums. Cement is assumed to be ordinary Portland cement. 	Scintillation cocktails in drums with or without stabilisation, conditioned or unconditioned, modelled for value assessment by INSTAGEL Plus	

For the oils and scintillation cocktails, the baseline is adsorption on Nochar and cementation. The data provided for this baseline by Sogin are presented in Annexe 1. For solvents, the baseline is incineration and cementation of the ashes. No data pertaining to disposability are available on the waste produced by the solvent baseline.

The comparison is presented below.



6.1 Oils

5.1.x vs 5.1.B				Input metri	Input metric values		aseline	Weaknesses vs. baseline	
	Criterion	Metric examples	Boundaries and exclusions	Baseline	Variant	Treatment and conditioning	Disposal	Treatment and conditioning	Disposal
	Disposability of final waste product	Existing disposability assessments. Known or anticipated issues with waste product characteristics.	Exclude packaging Exclude radioactivity	For discussion only.	There is the potential for some of the oil to exist as a free organic liquid in pores within the geopolymer matrix. Due to the polycondensation reaction of the geopolymer system, it expels water over time. Bleed was recorded at a maximum of 1.25 vol% of product for all formulations at 48	NA		The criteria evaluated are promi to the baseline, however, for the formulation, a few criteria were r such as the mechanical strength homogeneity and the use of surf could potentially be problematic	CIEMAT not conclusive n, the factant, which



6.2 Solvents

5.2.1 vs 5.2.B					Input metric values	Strengths vs	baseline	Weaknesses vs. baseline	
	Criterion	Metric examples	Boundaries and exclusions	Baseline	Variant	Treatment and conditioning	Disposal	Treatment and conditioning	Disposal
	Disposability of final waste product	Existing disposability assessments. Known or anticipated issues with waste product characteristics.	Exclude packaging Exclude radioactivity	For discussion only.	Due to the polycondensation reaction of the geopolymer system, it expels water over time. Bleed was recorded at a maximum of 1.75 vol% of product for all formulations at 48 h.	NA		Potential for free liquid in the dodecane (70:30) and 1-3 vo 80 (surfactant) to TBP/dodec pores of the geopolymer ma TBP and dodecane are toxic Dodecane is flammable, with limits of 0.5 vol% - 4 vol% No evidence to indicate that materials are destroyed in the this has not been assessed. Incorporation of TBP/dodeca geopolymer formulation, it has assessed if the TBP is held v pores or encapsulated into the Use of a surfactant which can complexing agent, which is up prohibited in disposal facility acceptance criteria (WAC). A	ol % of Tween cane in the trix. c chemicals. n explosive these re process, but ane in as not been within the ne matrix. n behave as a usually waste



6.3 Scintillation cocktails

5.3.1 vs 5.3.B				Input metric values		Strengths vs baseline		Weaknesses vs. baseline	
	Criterion	Metric examples	Boundaries and exclusions	Baseline	Variant	Treatment and conditioning	Disposal	Treatment and conditioning	Disposal
	Disposability of final waste product	Existing disposability assessments. Known or anticipated issues with waste product characteristics.	Exclude packaging Exclude radioactivity	For discussion only.	No additional bleeding was observed after the initial curing stage.	NA		The criteria evaluated are prom function of the formulation the c all conclusive (mechanical stren homogeneity)	riteria are not



7 Conclusions

Due to the absence of data on real packaged waste streams, a qualitative disposability assessment was implemented on oils, solvent and scintillation cocktails encapsulated in a geopolymer matrix, based on assessment criteria pertaining to the geopolymer wasteforms with the different RLOW. A summary results table is presented in Table 7.1.

Table 7.1: Summary table showing the disposability evaluation of RLOW encapsulated in various
geopolymer types.

Assessment	Oil -	Oil -	Oil -	Oil -	Solvent	Solvent	Solvent	SC -	SC -	SC - MK
area	МК	MK	Mix	BFS	- MK	- MK	- MK	MK	MK	
	CIEMAT	NNL	KIPT	CVRez	NNL	POLIMI	VIV	POLIMI	VIU	CIEMAT
Physical form										
Mechanical stability										
Homogeneity										
Void space										
Free liquids										
Chelating										
agents										
Leaching										

It is understood that the disposability assessment is country-dependent as the criteria and their limits differ from country to country. The experiments were mainly realised at a laboratory scale, meaning that many criteria cannot be assessed, because they are waste package and/or radioactivity dependant. This study is the first step in evaluating the disposal of the three RLOW geopolymer wasteforms. However, the scale-up from desk scale to full scale can also result in different properties of the final package. It is recommended that after providing the results for all the criteria, the analysis should be repeated at a larger scale to see if the results are comparable.

However, the analysis shows promising results for the encapsulation of oils, solvent and scintillation cocktails, but many criteria still need to be assessed or the rating refined further, mainly for the amber-rating criteria.

Other criteria related to the package design and the radioactivity in the wasteform also need to be assessed. The next phase of work must investigate this aspect. The need for further research and development to bring the process towards a TRL of nine is acknowledged and is reflected in the EURAD-2 proposals.

However, note that Slovakia and the Czech Republic are already using geopolymer technology at TRL = 9 (SiAL and Alusil respectively) to encapsulate problematic waste such as sludges and spent ion exchange resins for disposal [24] [25] [26] [27] [28] [29]. The waste forms created via the SiAL and Alusil processes are accepted for disposal at the national near-surface disposal facilities in these two countries, showing that geopolymers have a promising future as encapsulation matrices.

REFERENCES

- [1] European Commission, "PREDIS: Pre-disposal Management of Radioactive Waste, Annex 3 (detailed implementation plan associated with Section 3).," September 2019.
- [2] M. Briffaut et al. (ECL), "Deliverable 5.3: Technical Report "Synthesis of Conditioning Matrix Performance Studies"," January 2024.
- [3] F. Pancotti et al. (SOGIN, RATEN), "Milestone 34: Optimised Formulations for Reference Formulations," MS34, November 2022.
- [4] F. Pancotti et al. (SOGIN, RATEN), "Deliverable 5.2: Report on Synthesis of Formulation and Process Studies Results," Deliverable 5.2 Version final, 09/02/2024.
- [5] A. Baksay (TS.ENERCON), "Deliverable 2.7: Guidance on Formulating Generic Waste Acceptance Criteria," April 2024.
- [6] C. Eldridge, "Deliverable 6.3: Economic, Environmental and Disposability Impacts of Novel Treatment Technologies for Low-Level and Intermediate-Level Solid Organic Wastes," May 2024.
- [7] International Atomic Energy Agency, "Design Principles and Approaches for Radioactive Waste Repositories," IAEA Nuclear Energy Series No. NW-T-1.27, IAEA, Vienna, 2020.
- [8] L. K.-N. Lumir Nachmilner, "PREDIS Deliverable 2.4: International approaches to establishing a waste acceptance system," 2021.
- [9] F. Pancotti and C. Bucur, "Deliverable 5.3: Report on Synthesis of formulation & process studies results," 9 February 2024.
- [10] M. Dowson (Sellafield Limited), "Expert View on use of Alternative Encapsulants," WMIDA-2042652762-8039, 06 December 2023.
- [11] A. Fuller, Galson Sciences Ltd., "Deliverable 5.1: Input Data Synthesis Report," D5.1 Version 2.0, August 2021.
- [12] "Personal communication, "E. Mossini (POLIMI) to D.Alby (GSL)"," 25 January 2024.
- [13] "Personal communication, "A.Sears (CVRez) to D. Alby (GSL)," 29 January 2024.
- [14] "Personal communication, "M. Kiselovà (UJV) to D. Alby (GSL)"," 17 January 2024.
- [15] "Personal communication, "S. Irving (NNL) to D.Alby (GSL)"," 30 January 2024.
- [16] "Personal communication, "F. Pancotti (Sogin) to D. Alby (GSL)"," 23 January 2024.
- [17] "Personal communication, "Svitlychnyi Yevhenii (KIPT) to D. Alby (GSL)"," 30 January 2024.
- [18] G. Daval, T. Harrison, D. Alby, "Deliverable 5.5: Report on Direct Conditionning of Liquid Organic Waste Route," 2024.
- [19] Ispettorato Nazionale per la Securezza Nucleare e la Radioprotezione, "GUIDA TECNICA N.33 Criteri di sicurezza nucleare e radioprotezione per la gestione dei rifiuti radioattivi," 2023.
- [20] Rozhodnutí, "LIMITY A PODMÍNKY BEZPEČNÉHO PROVOZU ÚRAO DUKOVANY PŘÍLOHA 1. PODMÍNKY PŘIJATELNOSTI," 18.09.2012.
- [21] Rozhodnuti, "Samostatná příloha směrnice S.13 Evidenční označení: S.13p1LAP ÚRAO RICHARD, PODMÍNKY PŘIJATELNOSTI K UKLÁDÁNÍ," 30.08.2016.
- [22] ANDRA, "Spécification technique générale Spécification d'acceptation des colis de déchets radioactifs au Centre de Stockage de l'Aube – INB n°149," ACO.SP.ASRE.99-0001/E, 23.06.2023.
- [23] Radioactive Waste Management, "Geological Disposal: Guidance on the disposability of waste packages containing chemical decontamination agents," WPSGD no. WPS/928/01, February 2017.



- [24] Jacobs, "Retrieving and treatment of radioactive sludges and spent ion exchange resins in ED," 2021.
- [25] Jacobs, "SIAL® solidification technology," 2020.
- [26] D. Majersky, S. Sekely, D. Zavodska and M. Breza, "Application of Inorganic SIAL Matrix and Movable Technology in Solidification of the TRU Sludges and Sludge/Resin Mixtures," Conference: Waste Management 2006 Symposium - WM'06 - Global Accomplishments in Environmental and Radioactive Waste Management: Education and Opportunity for the Next Generation of Waste Management Professionals, Tucson, AZ (United States), WM Symposia, Inc., PO Box 13023, Tucson, AZ, 85732-3023 (United States), 26 Feb - 2 Mar 2006.
- [27] Jacobs, "SIAL® makes waste encapsulation better for the environment".
- [28] V. Havlova, R. Trtílek (UJV), "Use of Geopolymers in the Disposal of LLW/ILW in the Czech Republic," Geopolymers in Radioactive Waste Management webinar, 26.10.2021.
- [29] State Office for Nuclear Safety, "The Czech Republic report under the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management," Prague, 2020.
- [30] Personal communication, "Briefing pack for PREDIS WP5 Value Assessment Workshop," S. Wickham (GSL) to PREDIS WP5 Group, 02 February 2024.
- [31] Radioactive Waste Management Ltd. (RWM), "Waste Package Specification and Guidance Documentation: Guidance on the Production of Encapsulated Wasteforms," WPS/502/01, August 2015.
- [32] Radioactive Waste Management Ltd. (RWM), "Waste Package Specification and Guidance Documentation: Specification for Waste Packages Containing Low Heat Generating Waste. Part C: Fundamental Requirements," WPS/220/01, March 2020.
- [33] D. Alby (GSL), "Annexe 1_Data obtained by the partners," April 2024.
- [34] D. Alby et al. (GSL), "Deliverable 5.4: Disposability Assessment Report for Direct Conditioning," April 2024.
- [35] Personal communication, "A. Sears (CVRez) to G. Daval (GSL)," 23 January 2024.
- [36] Personal communication, "A.Sears (CVRez) to G. Daval (GSL)," 01 February 2024.
- [37] Personal communication, "H. Nonnet (CEA) to C. Eldridge (GSL)," 14 February 2024.
- [38] Personal communication, "H. Nonnet (CEA) to G. Daval (GSL)," 16 February 2024.



ANNEXE 1: DATA OBTAINED FROM THE REQUEST FORM

• Oils

<u>Metakaolin</u>

Criteria	NNL	CIEMAT
Homogeneity	Homogeneous No additional materials/ precursors added.	Homogeneous Precursors and activator not fully reacted: detected in first leaching steps
Integrity	Monolithic product Compressive strength of products ranged from 0.9 MPa – 37.5 MPa at 90 d over a range of molar ratio formulations for MetaMax MK Due to the polycondensation reaction of the geopolymer system it expels water over time. Bleed was recorded at maximum of 1.25 vol% of product for all formulations at 48 h.	 Monolithic product Oil 40%vol.: 28 day-curing in a closed vessel (plastic film) conditions: 10.9 MPa 28 day-curing in aerated conditions: 11.6MPa Amount of water, a priori not significant, but cannot be assessed or extrapolated to larger volumes. Scaling-up required
Waste loading	Up to 50% vol	40% vol.
Radiological gas generation	None conducted	None conducted
Non radiological gas generation	None conducted	None conducted.
Chemical content	Potential for free organic liquid in the form of Nevastane (CAS:192268-65-8) to exist in pores within the geopolymer matrix.	Oil seems to be saponified by highly-alkaline geopolymer environment, and therefore, according to leaching tests, immobilised. However, in the last leaching steps (~1 year), increasing TOC values are observed. This could indicate a declining retention capacity with time. Long-term



Criteria	NNL CIEMAT		
	No flammables, explosives, pyrophoric materials, chemotoxic or putrescible materials in Nevastane and therefore final wasteforms.	performance of the waste form may be jeopardised. So, durability should be assessed by means of longer duration tests.	
Chemical durability	None conducted	In case, of structural elements, Si & Al in the range of tens of ppm in the first leaching steps: possibly due to unreacted precursors. At longer times, leaching seems to stabilise close to 0.1 ppm. After 1-year leaching tests, specimens, in general, preserve their structural integrity, though some debris/chippings were observed in the renewal of leaching solution. Oil 40%vol: - Aerated: 0.4%vol. leached (worst case: 1.65%vol.) - In a closed vessel (plastic film): 0.15%vol. leached	
Fire performance	None conducted	Not assessed. In Spain, waste form performance in a fire event is not mandatory, only required for the concrete container where drums are backfilled	
Voidage	Monolithic product which did not exhibit cracking No macroscopic void spaces were observed Intergranular porosity not assessed	Large cracks observed in specimens cured in aerated conditions. After visual inspection, specimens with large cracks discarded for mechanical and leaching tests. Total porosity (%): Oil 40%vol.: Aerated curing:29% In a closed vessel (plastic film): 41% Average pore diameter (nm): Oil 40%vol.: Aerated curing:42.6 nm In a closed vessel (plastic film): 43.0 nm	



Criteria	NNL	CIEMAT	
		(values from MIP measurements)	
Secondary waste	Technological waste.	Technological waste.	

MIX (MK+BFS+FA) (NCS KIPT)

Criteria	Data
Homogeneity	Homogeneous
Integrity	The solidified product is monolithic
	Compres. strength = 32 MPa (28 day of curing) for geopolymer without waste
	Compres. strength = 15 MPa (28 day of curing) for geopolymer with 30% of oils
	No exuded water in samples
Waste loading	Waste (30 % vol) encapsulated in geopolymer in a 200 L drum.
Radiological gas generation	None conducted
Non radiological gas generation	None conducted
Chemical content	No chemically reactive species
Chemical durability	Without any oil, a leaching rate of 3.75 mg/cm^2 is measured at its maximum for Si and 0.75 for AI before decreasing. The maximum leaching rate is similar for the geopolymers with 30% of oil at pH = 12.7. No AI is leached after 30 days without oil while an amount of 0.2 mg/cm ² is present after 30 days with 30% of oil. For AI, the leaching rate of Si is similar for 90 days.
Fire performance	None conducted

Criteria	Data
Voidage	Dense homogeneous structure of hardened samples without large pores No macroscopic void spaces and cracks
Secondary waste	Technological waste.

BFS + sand (CVRez/SCK CEN)

Criteria	Data
Homogeneity	Depending on the amount of oil/surfactant added. In general, on a laboratory scale, we observed separate phases when using less than 1 wt. % of surfactant. No precursors were used. The lower surfactant amount was insufficient (the samples showed bleeding and heterogeneity - the separate oil phase on the top of the sample). When the amount of surfactant was sufficient, and the added oil was not too high (up to 30 wt.%), it was homogenous.
Integrity	Wasteform before solidification – paste with sand, monolithic after hardening. The compression strength varied depending on the amount of oil incorporated. (5 wt. % Nevastane incorporated >10 MPa, more than 5 wt.% of Nev. < 10 MPa, Mogul oil both 5 and 10 wt.% >10 MPa) Laboratory samples did not exude water.
Waste loading	Waste loading up to 30 wt. % Nevastane on the lab. scale – Necessity to find a compromise between waste load and WAC (compression strength).
Radiological gas generation	None conducted
Non radiological gas generation	None conducted



Criteria	Data
Chemical content	No chemically reactive species
Chemical durability	Leaching experiments: demineralised water, 90 days, laboratory temperature, leachate replacement and analysis days 2, 7, 14, 28, 56 and 90. The median of leached oil (cumulative) is 1.1 %.
Fire performance	The material should not be flammable.
Voidage	Cracking was not observed. Porosity (mercury method, 0.1-400 MPa) varied between samples from 13 – 21 %.
Secondary waste	Dust particles (in ppm) Technological waste

Nochar absorption and cementation (SOGIN) - Baseline

Criteria	Data
Homogeneity	The solidification process (Nochar will react with the oily phase and cement with the water) to produce a homogeneous matrix in a 220 I drum. The 220 I drum is then conditioned in heterogeneous form with cement in a bigger drum (380 I) - the 220 I drums are overpacked in 380 I drums, with cement poured into the annular space
Integrity	The solidified product is monolithic The requirement for the compression strength is referred to the conditioning cement matrix in 380 I drum (> 10 MPa)
	The measured compressive strength of the cement matrix is 60 MPa No free liquids have been observed in the solidified waste and in the final cemented package



Criteria	Data	
Waste loading	14 wt %	
	Solidification: max 100 kg waste / 220 l drum	
	Max 34 kg oil + 66 kg water + 17 kg Nochar (Nochar/oil 0.5) + 133 kg Cem (water/cement 0.5)	
	Conditioning: same amount of 380I drums (220 I drum goes into each 380 I drum)	
	No concentration of activity but dilution	
Radiological gas generation	The gas generation within the conditioned waste (380 litre package) was analytically evaluated by calculation and it was demonstrated that the contribution of gas produced by the metallic corrosion of the embedded 220 I metallic drum was about 3 orders of magnitude higher than the contribution given by the radiolysis of water and organic material contained in the waste. The maximum overpressure generated inside the package, acting on the solidified cement matrix, was evaluated and compared with the tensile strength of the embedding cement matrix. Analysis confirms the absence of any detrimental mechanical impact linked to gas generation.	
Non radiological gas generation	None conducted	
Chemical content	No chemically reactive species	
	If yes, the treatment process will not destroy such materials except free liquids that would be solidified with the process	
Chemical durability	The leachability index (Li) has been calculated according to the procedure set out in the ANSI/ANS-16.1 Standard. This is a dimensionless quantity.	
	The requirement for durability is referred to the conditioning cement matrix in 380 l drum (Li > 6 for Cs-137 as determined via ANSI ANS 16.1). A Li of 8.92 for Cs-137 has been calculated for the cement matrix via the same method.	
	The leachability index is inversely proportional to the log of the effective diffusivity of nuclide calculated from the test data. Thus, higher index means less diffusivity	



Criteria	Data
Fire performance	The fire and high temperatures resistance checks were performed using the Finite Element software Comsol Multiphysics (Heat Transfer module version 5.3) and a 2D transient axisymmetric model.
	The data were used to verify that the temperature inside the package did not raise up to value that could generate any degradation on the solidified waste: an upper limit for the temperature close to the waste was fixed to 200°C. It was demonstrated that an increase of 5 cm of the cement filler in the bottom of the package (the total thickness was raised to 9.5 cm) would overcome any anomalous behaviours of the package that may compromise its stability and therefore the qualification of the conditioning process.
Voidage	No macroscopic void spaces and cracks - Any voids are filled during the overpacking process: the 220 I drum is inserted into the overpack, the inner lid is removed and the empty space in the upper part of the inner drum (220 I) is filled with cement matrix. After the closure of the 220 I lid, the cement matrix is poured in the annular space of the overpack.
Secondary waste	For the solidification, overpacking and conditioning of the total volume of waste we produced:
	2 packages with no secondary waste except technological waste (suits, gloves, etc.) + empty tanks
	1 package with secondary solidified liquid waste produced during the decontamination of the reusable paddle

• Solvents - Metakaolin

Criteria	NNL	POLIMI	UJV
Homogeneity	Homogeneous	Homogeneous	Homogeneous



Criteria	NNL	POLIMI	UJV
	The waste was emulsified using a surfactant of 1-3 vol% of the volume of waste (10-30 vol%).		
Integrity	Monolithic product Compressive strength of products ranged from 10.9 MPa – 40.3 MPa at 90 d at 90 d over a range of molar ratio formulations for MetaMax MK Due to the polycondensation reaction of the geopolymer system it expels water over time. Bleed was recorded at maximum of 1.75 vol% of product for all formulations at 48 h. All material incorporated and a homogeneous grout. No segregation of samples. Visual assessment.	 (without any waste: 18.6 MPa, with waste: 10 MPa) also following post-curing ageing induced by: immersion in water for of 28 days (without waste: 16.1 MPa, with waste: 6.3 MPa) irradiation up to 200 kGy (without waste: 7.9 MPa, with waste: no data). 	().
Waste loading	Waste loadings of up to 30 vol% - this was achieved at 4 L scale. To be assessed with active trials to be performed by others.	Loading of 30% waste by volume.	Depending on the type of loaded waste, using cementation or geopolymerisation, the volume change compared to the raw waste could be from 1:2 to 1:8.
Radiological gas generation	None conducted	None conducted	No significant radiological gas generation.

Criteria	NNL	POLIMI	UJV
Non radiological gas generation	None conducted	None conducted	No significant non radiological gas generation.
Chemical content	Potential for free liquid in the form of TBP, dodecane (70:30) and 1-3 vol % of Tween 80 (surfactant) to TBP/dodecane in the pores of the geopolymer matrix TBP and dodecane are toxic chemicals Dodecane is flammable, with explosive limits of 0.5 vol%- 4 vol% No evidence to indicate that these materials are destroyed in the process, but this has not been assessed. Incorporation of TBP/dodecane in geopolymer formulation, it has not been assessed if held within the pores or encapsulated into the matrix	Wastes encapsulated retain their reactivity (flammability and potentially toxicity) as they are not degraded by any pre-treatment. TBP/Dodecane waste are mixed with the geopolymer thanks to surfactants. TBP/Dodecane waste is encapsulated by pre-emulsification using a surfactant (5%w with respect to the waste).	Does not affect disposability. The only hazard is radioactivity. No flammables, explosives, pyrophoric materials, chemotoxic or putrescible materials ionic solution and final products.
Chemical durability	None conducted	The leaching experiments have been conducted following the ANSI/ANS-16.1-2019 protocol. The leachant was ultrapure water and was periodically renewed. The test was conducted at room temperature ($20 \ ^{\circ}C \pm 1 \ ^{\circ}C$). The leachability indices are all superior of 8. The compressive strength values of wasteform immersed in water for	The leaching experiments followed the ANSI method. The volume of the leaching solution (DEMI water) was calculated as: V(L)/S=10 +/- 0.2 cm. A constant temperature 17.5 - 27.5 °C was maintained during the testing procedure. The specified volume of the leaching solution (1 ml) was taken at the set intervals: 1, 2, 3, 4, 5, 19, 47 and 90d. The required minimum



Criteria	NNL	POLIMI	UJV
		1 month are reported above ("Integrity of the wasteform").	value of the leachability index "Li" 8 was attained.
Fire performance	None conducted	None conducted	None conducted
Voidage	Monolithic product which did not exhibit cracking macroscopic void spaces not assessed visually Intergranular porosity not assessed Visual assessment of the external surfaces showed no significant macroscopic voidage. Additionally some samples were split in half and these also showed no voidage.	No visible cracking or macroscopic porosity after curing and storage in humid environment. For MK-based samples we measured porosities between 33- 38%. If samples are stored in a humid environment (>90% relative humidity), nothing significant happens, and samples are durable (no cracks or defects after immersion). On the contrary, if samples are stored in <90% relative humidity, i.e. even under normal ambient conditions, they tend to dry out and loose about 25% of their initial weight. This becomes a problem when samples then get in contact with water: they reabsorb the water they have lost and they crack dramatically	No significant cracking or significant intergranular porosity.
Secondary waste	Technological waste	Technological waste	Technological waste

• Scintillation cocktails – Metakaolin

Criteria	Polimi	UJV	CIEMAT
Homogeneity	Homogeneous	Homogeneous	Homogeneity not assessed. Light density and organic nature hinders the characterization by means of SEM-EDX or μ-CT Precursors and activator not fully reacted: detected in first leching steps
Integrity	 Monolithic. Compressive strength was studied (without any waste: 18.6 MPa, with waste: 8.9 MPa) also following post-curing ageing induced by: immersion in water for of 28 days (without waste: 16.1 MPa, with waste: 7.1 MPa), or irradiation up to 200 kGy (without waste: 7.9 MPa, with waste: 4.9 MPa). Wasteforms bleed an amount of water <1% of the total wasteform volume during curing. Afterwards, no additional bleeding was observed (observation time of 6 months after casting). 	exude water. Compression strength (separate samples with Ni-63 and C-14): MK matrix + scintillation cocktail: 25.71 MPa (Ni-63), 18.6 MPa (C-	Monolithic specimens. LSC 30%vol: 28day-curing in a closed vessel (plastic film) 25.5 MPa Specimens cured in a closed vessel (plastic film) show a tendency to exude water and LSC? (not sure if LSC is exuded or remains in the surface of specimen during curing) Amount of water, a priori not significant, but cannot be assessed or extrapolated to larger volumes. Scaling-up required
Waste loading	Loading of 30% waste by volume.	Depending on the type of loaded waste, using cementation or geopolymerisation, the volume change compared to the raw waste could be from 1:2 to 1:8.	In the case of LSC, incineration is the accepted national management strategy. Direct immobilisation in geopolymer does not seem technically or economically feasible: volume reduction



Criteria	Polimi	UJV	CIEMAT
			achieved by incineration (and later, cementation in drums) is much greater: 1 drum of 220 I for incineration+cementation (conventional treatment): 50 drums of 220I for immobilisation of 30%vol. in geopolymer (PREDIS). Besides, significant LSC leached fractions found in tests make unsuitable the immobilisation in the geopolymer formulation tested. Concern not just for radionuclides leached, but by the release of organics that may act as mobilisers or complexants of radionuclides in the disposal medium
Radiological gas generation	None conducted	No significant radiological gas generation.	No significant radiological gas generation.
Non radiological gas generation	None conducted	No significant radiological gas generation.	No significant radiological gas generation, but H_2S discharge observed in some LSC- tests during the renewal of the leachant. This seems to point to microbial activity.
Chemical content	Wastes encapsulated retain their reactivity (flammability and potentially toxicity) as they are not degraded by any pre-treatment. LSC wastes are mixed with the geopolymer thanks to surfactants.	No affection of its. Does not affect disposability. The only hazard is radioactivity. Scintillation cocktail complies with the Regulation 2003/53/EC of the European Parliament/Council. ROTISZINT.	LSC is not retained by the geopolymer matrix. High leaching rates calculated based on the TOC values measured in leachant. Not acceptable for disposal.



Criteria	Polimi	UJV	CIEMAT
	Most of LSC cocktails contain surfactants (usually around 2%w).		
Chemical durability	The leaching experiments have been conducted following the ANSI/ANS- 16.1-2019 protocol. The leachant was ultrapure water and was periodically renewed. The test was conducted at room temperature (20 °C ± 1°C). The leachability indices are all superior of 8. The compressive strength values of wasteform immersed in water for 1 month are reported above ("Integrity of the wasteform").	The leaching experiments followed the ANSI method. The volume of the leaching solution (DEMI water) was calculated as: V(L)/S=10 +/- 0.2 cm. A constant temperature 17.5 - 27.5 °C was maintained during the testing procedure. The specified volume of the leaching solution (1 ml) was taken at the set intervals: 1d, 2d, 3d, 4d, 5d, 19d, 47d and 90d. The required minimum value of the leachability index "Li" 8 was attained.	In case, of structural elements, Si & Al in the range of tens of ppm in the first leaching steps: possibly due to unreacted precursors. At longer times, leaching seems to stabilize close to 0.1 ppm. After 1-year leaching tests, specimens, in general, preserve their structural integrity, though some debris/chippings were observed in the renewal of leaching solution. Regarding the leaching of LSC, estimations based on 1-year leaching tests in deionized water are: LSC 30%vol. aerated: 15%vol. leached
Fire performance	None conducted	None conducted	None conducted
Voidage	No visible cracking or macroscopic porosity after curing and storage in humid environment. For MK-based samples we measured porosities between 33-38%. If samples are stored in a humid environment (>90% relative humidity), nothing significant happens, and	No significant cracking or significant intergranular porosity.	Large cracks observed in specimens cured in aerated conditions. After visual inspection, specimens with large cracks discarded for mechanical and leaching tests. Total porosity (%): LSC 30%vol:

Criteria	Polimi	UJV	CIEMAT
	samples are durable (no cracks or defects after immersion). On the contrary, if samples are stored in <90% relative humidity, i.e. even under normal ambient conditions, they tend to dry out and loose about 25% of their initial weight. This becomes a problem when samples then get in contact with water: they reabsorb the water they have lost and they crack dramatically		Aerated: 50% Average pore diameter (nm): LSC 30%vol: Aerated: 34.7 nm (values from MIP measurements)
Secondary waste	Technological waste	Technological waste	Technological waste

