



HAL
open science

Geopolymers in nuclear waste storage and immobilization: mechanisms, applications, and challenges

M. Houhou, N. Leklou, H. Ranaivomanana, Jd. Penot, Silvio DE BARROS

► **To cite this version:**

M. Houhou, N. Leklou, H. Ranaivomanana, Jd. Penot, Silvio DE BARROS. Geopolymers in nuclear waste storage and immobilization: mechanisms, applications, and challenges. *Discover Applied Sciences*, 2025, 7 (2), pp.126. 10.1007/s42452-025-06536-x . hal-04938863

HAL Id: hal-04938863

<https://hal.science/hal-04938863v1>

Submitted on 10 Feb 2025

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial - NoDerivatives 4.0 International License


Review

Geopolymers in nuclear waste storage and immobilization: mechanisms, applications, and challenges

M. Houhou¹  · N. Leklou²  · H. Ranaivomanana²  · JD. Penot¹  · S. de Barros^{1,3} 

Received: 10 October 2024 / Accepted: 29 January 2025

Published online: 08 February 2025

© The Author(s) 2025 

Abstract

According to the latest report by the International Atomic Energy Agency (IAEA), nuclear facilities generate over 30 tons of high-level radioactive waste and 300,000 tons of medium-level waste annually, highlighting the need for secure immobilization methods to safeguard environmental and public health. Cementitious materials such as ordinary Portland cement (OPC) and other materials are commonly employed as engineered barriers for the long-term containment of nuclear waste in both surface and underground geological repositories. However, recent studies show that geopolymer (GP), a novel class of cementitious materials, exhibit superior performance in immobilizing radioactive contaminants with reported compressive strength exceeding 50 MPa and leach rates for cesium and strontium ions reduced by over 95% compared to OPC based barriers. This paper provides a comprehensive review of the application of GPs in nuclear waste immobilization. First, the influence of various parameters on geopolymerization and the performance of GPs in immobilizing nuclear waste is analyzed. Examples are then provided to highlight the differences between GPs and ordinary Portland cement in immobilizing both solid and liquid nuclear waste. Additionally, the mechanisms involved in stabilizing cations and anions are described. The paper also discusses early developments in the use of GP-based materials for tunnel linings in underground nuclear waste storage cells. Despite their promising advantages, challenges associated with GPs, such as standardization difficulties due to the variability of raw material sources and inconsistencies in compressive strength, are explored. This review is particularly significant as it provides valuable insights into better understanding the use of GPs for nuclear waste immobilization, explores the challenges they face, and uncovers some of the gaps in current research.

Article highlights

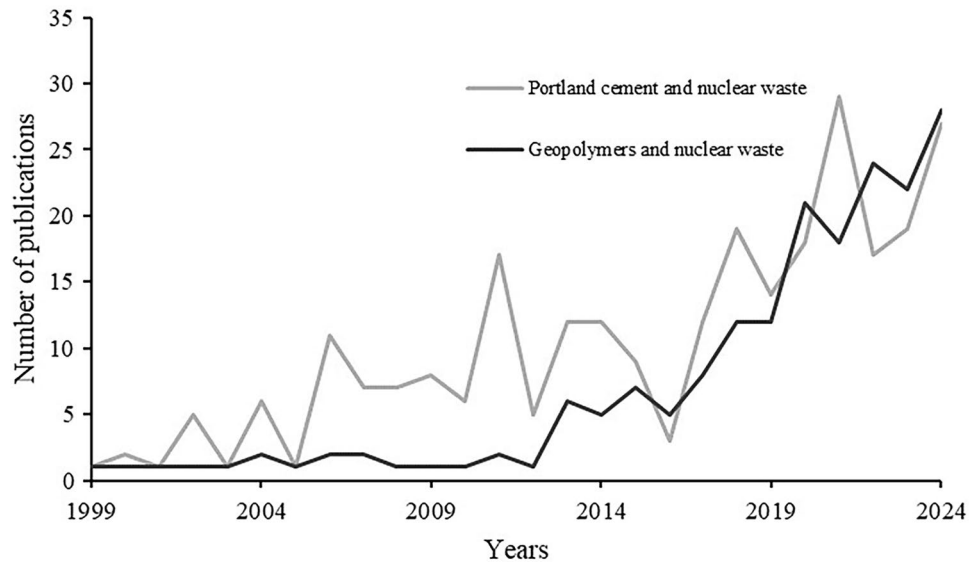
- GPs are a promising material for nuclear waste immobilization.
- GP matrices are effective in encapsulating and stabilizing heavy metals.
- The main challenge is the understanding of long-term properties, which requires further research.

Keywords GP · Radioactive waste · Radionuclide · Solidification · Stabilization · Encapsulation · Heavy metals · Gamma irradiation

✉ S. de Barros, silvio.debarros@gmail.com | ¹CESI LINEACT, UR 7527, Saint-Nazaire, France. ²Nantes Université, École Centrale de Nantes, CNRS, GeM, UMR 6183, 44600 Saint-Nazaire, France. ³Federal Center for Technological Education CEFET/RJ, Rio de Janeiro 20271-110, Brazil.



Fig. 1 Evolution of the number of publications studying GPs and OPC for radioactive waste management (Scopus 1999–2024)



1 Introduction

Nuclear energy is a potential solution for addressing the global energy crisis and reducing CO₂ emissions [1]. In 2021, it contributed to nearly 15% of global electricity production, with 437 reactors in operation worldwide. However, this industry, along with others, generates radioactive waste [2]. Radioactive waste is generated from chemical sludges, reactor decommissioning, fission products and spent fuel; these wastes contain hazardous radioactive materials that can negatively impact the environment and all living organisms for hundreds of years. Nuclear waste can be characterized based on their level of activity as follows: low-level radioactive waste (LLW), intermediate-level radioactive waste (ILW) and high-level radioactive waste (HLW). The management of radioactive waste poses a significant challenge for nuclear power plants and the nuclear-based technologies. Each type of radioactive wastes requires different method of management, and should be safely and properly disposed to promote a safe and a sustainable nuclear power program. Depending on the category of radioactive waste, the disposal can be carried out in landfill disposal, shallow level disposal and deep geological repositories. The multi-barrier system is composed of several layers of protection. The first layer is the waste form itself, followed by the second layer, which is the HLW container. The third layer consists of an engineered barrier, and the fourth layer is a geological barrier. Immobilization is one of the waste management treatment techniques that involves transforming radioactive waste into a specific waste-form through solidification, embedding, or encapsulation [2]. The objective is to dilute the radioactivity and minimize the risk of radionuclide migration or dispersion into the environment during activities such as handling, transportation, storage, or disposal [3]. Different types of materials are commonly used in radioactive waste management: cementitious materials typically Ordinary Portland cement (OPC), bitumen and glass are widely used to solidify LLW and ILW liquids [3–5], steel is used for HLW management and underground tunnel lining [6]. OPC was proven successful for this application and is considered a mature ready to be used technology. However, cement solidification is still facing some challenges regarding the durability of the material that is prone to degradation in contact with acids and at extreme temperature conditions [7]; radionuclides in solidified cement matrices are primarily retained through chemisorption and physical encapsulation. However, durability concerns regarding the material itself can lead to issues, such as high porosity, which may result in elevated leaching rates during the solidification process and subsequent disposal [8]. As for steel, the corrosion of metals in anaerobic conditions is likely to degrade the mechanical properties of the lining and produce explosive hydrogen [6]. In this context, geopolymer (GP) has emerged as a potential material for radioactive waste management, garnering increasing interest in recent years [9]. Fig. 1 shows the number of publications per year according to Scopus keyword analysis. There is a slower start in the number of publications for “Geopolymer and nuclear waste”, with a significant rise post-2010 and accelerating interest closer to 2024. In contrast, “Portland cement and nuclear waste” shows a steady increase with some fluctuations, particularly spiking around 2015 and towards 2024. These trends suggest that while Portland cement has been more widely studied historically, interest in GPs is rapidly growing, especially in recent years. This growing number of publications has been driven by technological

advancements, increased funding, growing environmental awareness, policy shifts regarding nuclear energy. Geopolymers named by Davidovits in the 1970s, are inorganic binders that include materials of inorganic or mineral origin [10]. GPs have tetrahedral silica and alumina structure and are known to their properties that are comparable to ordinary Portland cement, and their cationic binding sites [11–15]. GPs are recognized as promising alternative materials to OPC due to their exceptional durability properties. Specifically, GPs exhibit significant advantages over traditional cementitious materials, including enhanced durability, outstanding thermal resistance, and superior mechanical characteristics [7, 16–19]. Comprising zeolite-like structures, GPs demonstrate greater resilience to extreme environmental conditions compared to OPC. They maintain impressive thermal stability, retaining good compressive strength even after exposure to temperatures as high as 1000 °C, exhibiting only a 25% reduction in strength; in contrast, OPC faces a dramatic decline in strength between 400 and 600 °C due to the degradation of portlandite phase ($\text{Ca}(\text{OH})_2$) [19]. Furthermore, GP's three-dimensional $[\text{Si}-\text{O}-\text{Al}-\text{O}]_n$ framework imparts greater strength, while OPC's vulnerable chemical bonds, such as van der Waals and hydrogen bonds, increase its susceptibility to cracking under pressure. The unique microstructure of GPs, characterized by silicon and aluminum tetrahedral units forming a closed cage cavity, enhances their ability to immobilize nuclear waste and trap radionuclides effectively. Additionally, GPs have lower CO_2 emissions; for instance, metakaolin (MK), a key material in GP production, is produced at much lower temperatures (500–800 °C) compared to 1500 °C used in the Portland cement industry, saving up to 80% of the energy during production [20, 21]. While numerous reviews have explored the use of geopolymers for various environmental applications, such as heavy metal and nuclear waste stabilization [22–24], this paper provides a concise review for those new to the field. Offering a clear overview of the current advancements and potential of geopolymer-based solutions for nuclear waste immobilization. The paper begins by examining the factors influencing geopolymerization and evaluates their effectiveness in immobilizing nuclear waste. Comparative examples are provided to illustrate the differences between GPs and OPC for both solid and liquid nuclear waste. The stabilization mechanisms and examples for cations and anions heavy metal immobilization are also explained. Furthermore, the paper highlights early advances in using GP-based materials for tunnel linings in underground nuclear waste storage facilities. Finally, it addresses the challenges facing GPs, including standardization issues arising from raw material variability and inconsistencies in compressive strength. In addition, some potential areas for future studies are highlighted in several sections.

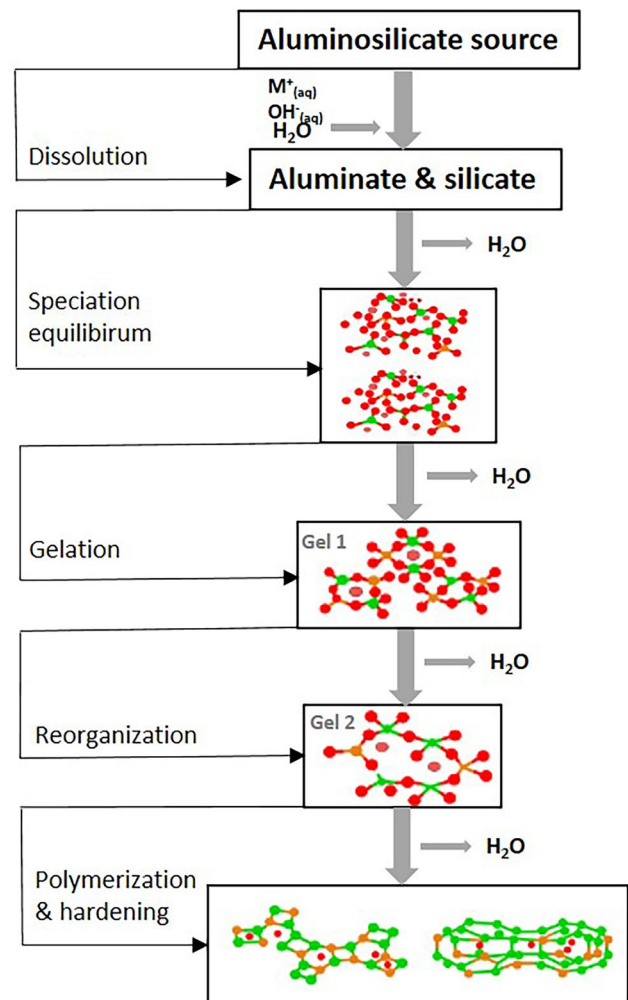
2 Geopolymers

The term 'GP' appeared in the 1970's by the French scientist Joseph Davidovits, 'geo' is used to refer to the aluminosilicate product as an inorganic equivalent of polymers [10]. The GP is an aluminosilicate material that is formed by the condensation polymerization of minerals. It is consisted of three-dimensional network arrangement that alternates connections between silicon tetrahedral ($\text{Si}(\text{OH})_4^-$) and aluminum tetrahedral ($\text{Al}(\text{OH})_4^-$), linked by a shared oxygen atom. The empirical chemical formula of a GP is $\text{Mn}[-(\text{SiO}_2)_z\text{AlO}_2]_n\text{wH}_2\text{O}$, where M represents an alkali cation like K^+ and Na^+ and Cs^+ , n indicates the polymerization degree, z denotes the Si/Al molar ratio and w for the water content [25]. The Geopolymerization mechanism is not fully understood; in 1956 Glukhovskiy [26] was the first researcher who proposed a general mechanism of the alkali activation of aluminosilicate materials by dividing it into 3 main stages: the first stage is destruction/coagulation, the second is coagulation/condensation and the third is condensation-crystallization. Later on, different researchers [27, 28], have extended the theories provided by Glukhovskiy [26], that what lead to the publish of a simplified reaction mechanism for the process [29]. This model as described in Fig. 2 consists of different stages: The dissolution of the aluminosilicate source to form the reactive precursors $\text{Si}(\text{OH})_4$ and $\text{Al}(\text{OH})_4^-$. Followed by the restructuring of the aluminosilicate precursor to a more stable state. Then, polycondensation and gelation of the system, that occur through the condensation of mono-silicates and mono-aluminates to form $\text{Si}-\text{O}-\text{Si}$ and $\text{Si}-\text{O}-\text{Al}$.

2.1 Factors that influence the geopolymer properties

Numerous factors influence the properties of GP products, significantly affecting the outcome. It is essential to carefully manage these factors to achieve the desired materials, particularly in the context of immobilizing radioactive waste. For instance, current studies indicate that the effectiveness of GPs is influenced by several factors, including the type of precursor, the type of activator, the Si/Al ratio, the water content and curing conditions. However, there remains a lack of comprehensive studies investigating the impact of these variables on the immobilization of nuclear waste within GPs.

Fig. 2 Simplified reaction mechanism of geopolymerization delivered by Duxson, Fernandez-Jimenez, Provis, Lukey, Palomo and Deventer-Modified from [29]



This section aims to summarize some of the findings from existing literature to highlight the current understanding and gaps in research.

2.1.1 Influence of precursor

Despite the similarities on the molecular level for aluminosilicate sources or precursors, they can have different final properties. For example, the mechanical properties of the metakaolin-based GPs are usually superior compared to fly ash-based GPs. The explanation is given by the microstructural difference of metakaolin-based GP having more isolated pores, whereas in the case of fly ash-based GP the pores are bigger and more interconnected [28, 30]. This is shown by the SEM images taken for both types as seen in Fig. 3. Image (a) corresponds to the metakaolin based GP, it represents a more homogeneous aspect with traces of unreacted materials [28]. In comparison the image (b), the fly ash based GP has an important amount of non-reacted material. However, these variations may also depend on the specific composition of each GP according to the initial mix design [28, 31]. The variability of the properties and reactivity between sources, and even between batches from the same source, necessitates a quantitative study to understand the effect of different compositional, synthesis, and post-synthesis parameters on the performance of the geopolymeric products [28].

Iron rich precursors have been thoroughly investigated in the literature [32–35]. However, few studies have explored the use of this class of GPs in nuclear waste applications. In a recent study, Xu et al. [36] investigated laterite–phosphoric acid– Fe_3O_4 -based GPs that have shown considerable promise for the immobilization of radioactive borate liquid waste (RBLW). The inclusion of Fe_3O_4 enhanced not only the compressive strength of the GP but also mitigated the retardation effects that RBLW can impose on the geopolymerization process. The reactions between Fe_3O_4 and phosphoric acid played an important role by generating heat and forming additional GP gel, which resulted in the creation of amorphous

Fig. 3 SEM micrographs of **a** metakaolin-based and **b** fly ash-based GPs activated with sodium silicate solution with $\text{SiO}_2/\text{Na}_2\text{O}=2$ —Modified from [28]

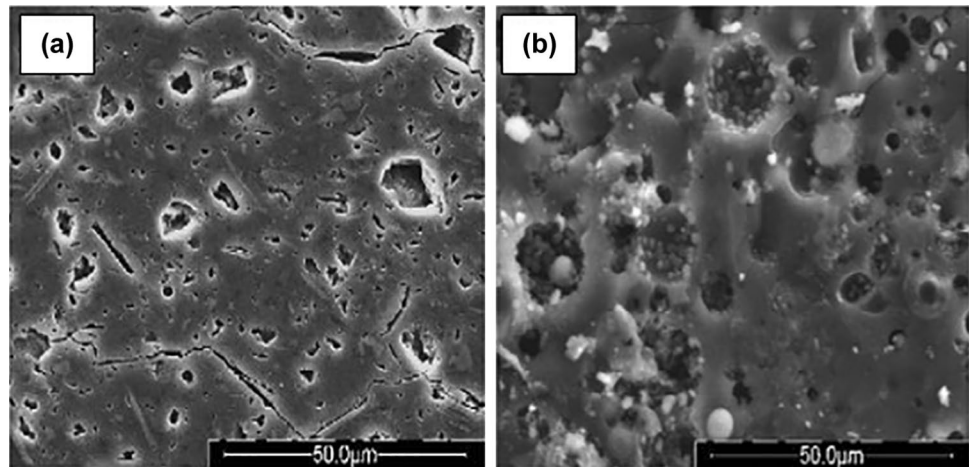
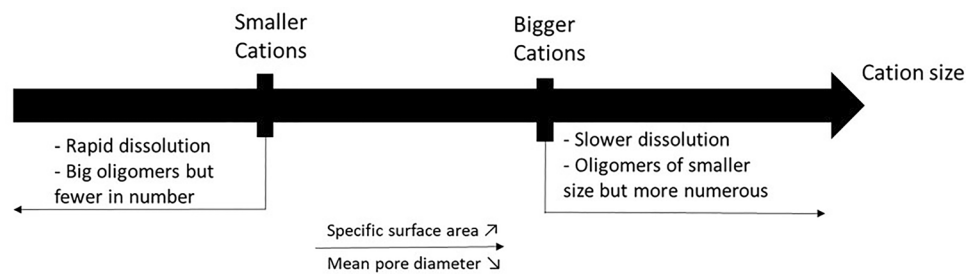


Fig. 4 Effect Cation size on the GP



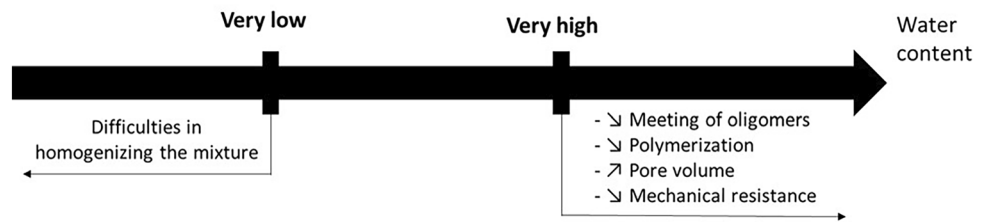
iron–phosphorus phases, further contributing to the mechanical integrity of the waste forms. Additionally, the structural modification achieved through the partial substitution of $[\text{AlO}_4]$ tetrahedra with $[\text{FeO}_6]$ octahedra lead to the formation of stable $-\text{Fe}-\text{O}-\text{P}-\text{O}-\text{Al}-\text{O}-\text{Si}-$ network structures. These structural enhancements have been correlated with improved leaching resistance, attributed to a reduction in open porosity, which facilitates more effective radionuclide immobilization through physical encapsulation. This study underscores the potential use of iron rich precursors as a promising material for radioactive liquid borate treatment. Further studies are still needed to achieve a better understand the effectiveness of this class of GPs in the immobilization of nuclear waste and heavy metals.

2.1.2 Influence of alkali cation and activating solution

In the process of geopolymerization, the alkaline metal plays a major role in forming the GP structure. It balances the negative charge of aluminates, ensuring their stability, and affects every stage of the chemical reaction from the setting time, to mechanical and chemical properties [31, 37]. The alkaline metal is provided by the activating solution that is also a silicate source. Typically, this cation is either Na^+ , K^+ , and to a lesser extent Cs^+ , Li^+ . A study on the dissolution of metakaolin by alkaline hydroxide solutions revealed that the dissolution using different alkali ions (Na, K, and Cs) occurs more rapidly for smaller alkali ions [38]. Therefore, it is faster for Na than for K or Cs. On the other hand, silicate species are more polymerized, and the formed alumino-silicate oligomers are more numerous and smaller in size for larger cations, polymerization is then more effective Fig. 4 summarizes the effect of cation size on the geopolymerization process where an increase in the cation size leads to a decrease in the dissolution kinetics and to the formation of smaller but more numerous oligomers. For instance, studies has shown that potassium based GPs exhibit higher pore volume and specific area than sodium based GPs [39, 40]; Steins et al. [39] observed a finer and more homogeneously distributed porosity with potassium activation compared to sodium activation. This was also validated by a study that investigated borate immobilization in K-based and Na-based GPs [41].

Chupin et al. [42] also investigated the effect of the porous structure of Na-based GP, K-based GP, and Cs-based GP on the hydrogen production yield after exposure to gamma irradiation. They found that as the cation size increased, the hydrogen production yield also increased. This was explained by the larger surface area resulting from the formation of a finer porous structure, which leads to the production of more Compton electrons at the solid/water interface under

Fig. 5 Effect of water content on the GP



irradiation. A second explanation provided is that with smaller cluster sizes (as in the case of Cs-based GP), electron trapping is less efficient [42].

Cation type and ratio is also a factor that affects the immobilization of heavy metals by the ions exchange mechanism. Numerous studies have shown that when using NaOH and KOH activation solutions to immobilize cesium, sodium cesium GPs showed the best immobilization efficiency of 98.03% before heating with an optimum ratio of Na/Cs=9 [43, 44]. The enhanced efficiency of sodium based solution was attributed to the increase in gel formation in which Cs⁺ radionuclides are secured through the interaction of elastic force and negative charge sites [24].

In addition, some previous studies have demonstrated that phosphate-based GPs, known as aluminosilicate phosphate (ASP) GPs, which use phosphoric acid as an activating solution, exhibit remarkable adsorption and solidification properties for heavy metals and radioactive nuclear waste [45, 46]. This can be explained by the fact that phosphate-based GPs can securely trap heavy metal ions within the intricate three-dimensional network of their structure, effectively minimizing the leaching of heavy metal ions and radioactive elements. Research by Pu and Njimou et al. [47, 48] indicated that ASP GPs exhibit strong stabilization of Pb²⁺ ions, outperforming both alkali-aluminosilicate GPs and OPC, particularly in acidic conditions. This might open the door to the use of ASP GPs in the stabilization of anionic heavy metal ions, a promising area of research that remains relatively underexplored in the literature. This interest is largely driven by the potential of GPs to achieve enhanced fixation of negative ions, particularly in acidic environments, thereby limiting the leaching of heavy metals from the GP matrix.

2.1.3 Influence of water content

The amount of water originally introduced into the mix has a huge impact on the microstructure [49]. The effect of the H₂O/M₂O (M: alkali cation) ratio is significant on both pore volume and pore size. Increasing water in the mix design could lead to the appearance of larger pores and causes a loss in the mechanical resistance [49]. Figure 5 shows that the water content should not be too low, as it is necessary for mixing the reactants and allowing ionic transfer. However, it should not be too high either, to avoid diluting the system, which would hinder the encounter of oligomers and slow down the polymerization process [50].

Geddes et al. [51] investigated the impact of irradiation on two GP formulations with H₂O/K₂O ratios of 11 and 13, both cured for 168 h. Following exposure to a total irradiation dose of 1 MGy, thermogravimetric analysis (TGA) indicated that the post-irradiation water loss in the formulation with a higher water content (H₂O/K₂O ratio of 13) was comparable to that of the lower water content formulation (ratio of 11). Additionally, the study revealed that the formulation with lower water content exhibited a higher degree of carbonation under irradiation than the formulation with higher water content.

2.1.4 Influence of Si/Al ratio

The variation of Si/Al molar ratio affects the setting time, compressive strength as well as the porosity of the material. Duxson et al. have found that ranging Si/Al ratio from 1.15 to 2.15, the pore volume decreases from 0.206 to 0.082 cm³/g. Furthermore, the size distribution of these pores is also altered, as presented in Fig. 6 [52]. Figure 7 also shows that the compressive strength increases with an increasing Si/Al ratio, with the optimum mechanical resistance reached at a Si/Al ratio of 1.9. Further increase in the ratio lead to a decrease in the resistance [53]. This drop in compressive strength can be due to the rapid setting and the limited mobility in the system [31] or due to the amount of unreacted material in the specimens, which act as defect sites [11]. The effect of Si/Al ratio on the GP is summarized in Fig. 8. When the Si/Al ratio is lower than 1.4, large interconnected pores are formed. Between 1.4 and 1.9, smaller, isolated pores appear, leading to a more homogeneous structure. The optimum performance is observed at an Si/Al ratio of 1.9. However, when the ratio exceeds 1.9, unreacted phases start to accumulate, causing a decrease in mechanical performance.

Fig. 6 Pore volume distribution of sodium based GP with different Si/Al ratios—Modified from [52]

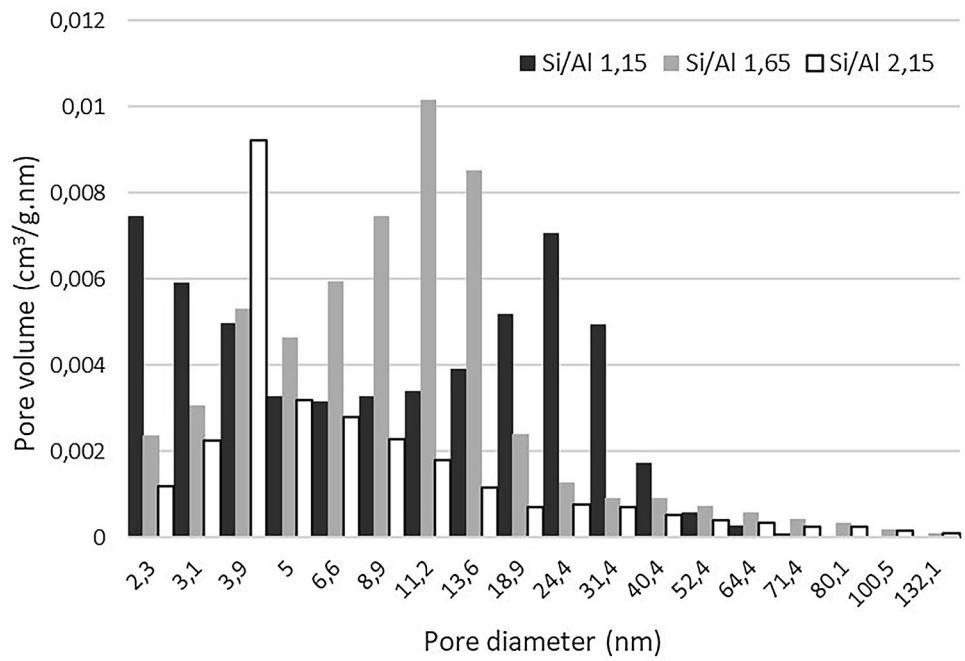


Fig. 7 Compressive strength evolution of metakaolin-based GP with Si/Al ratio variation from 1.15 to 2.15—Modified from [53]

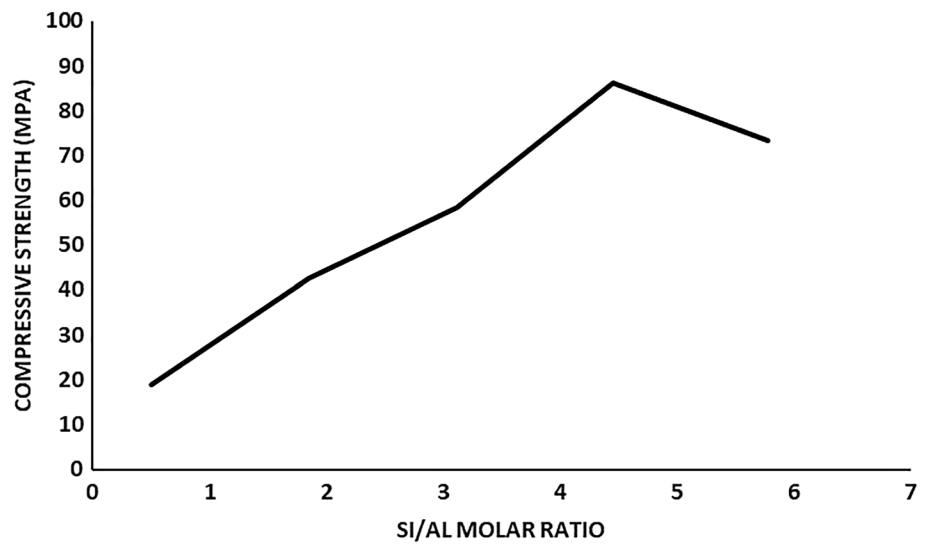


Fig. 8 Effect of the Si/Al ratio on the GP

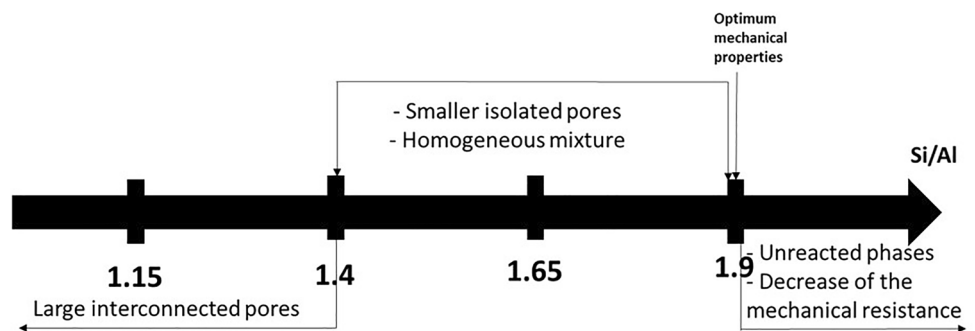
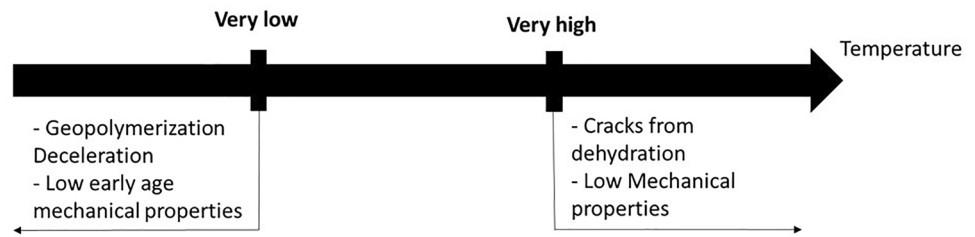


Fig. 9 Effect of the curing temperature on the GP



At the present stage of investigation, it has been found that low Si/Al ratio results in more tetrahedrons distributed in small rings having stronger locking effects for cationic heavy metal nuclides because of the adsorption density, whereas a higher Si/Al ratio leads to the distribution of tetrahedrons in larger rings, and this can increase the leaching effect of the heavy metal [54, 55]. The influence of the Si/Al ratio on the immobilization of anionic nuclides remains poorly understood, representing a gap in current knowledge.

To study the effect of this ratio on the radiolytic yield of hydrogen production under gamma irradiation, Chupin et al. [31] irradiated sodium-based GPs with different Si/Al ratios ranging from 1.8 to 2.2. It was found that this yield increased linearly but not significantly, from 0.12×10^{-7} mol/J at Si/Al = 1.8 to 0.14×10^{-7} mol/J at Si/Al = 2.2 [31] whereas a more significant difference was noted with the pore size distribution measurement where an increase in Si/Al ratio led to a decrease in the average pore diameter [31, 56].

2.1.5 Influence of cure parameters

It was found that the curing of GP under temperatures between (50 and 80 °C) after demolding increases the geopolymerization kinetics [57]. However, this acceleration can affect the mechanical properties of the final product this why the GP should be sealed under controlled relative humidity conditions. Figure 9 summarizes the effect of curing temperature on the GP: at low temperatures such as 10 °C, the reaction is slow and the mechanical properties are less important, whereas at a very high temperature (80 °C and more), micro cracks can occur due to dehydration and therefore affect the mechanical properties [34, 40, 58, 59]. Rovnanik [59] shows that curing at high temperatures for only 1 h does not increase the mechanical properties, whereas curing for a longer time (more than 4 h) results in good mechanical properties. However, the GP network is not completely formed even one week after the setting time, and the GP continues to densify [39, 40].

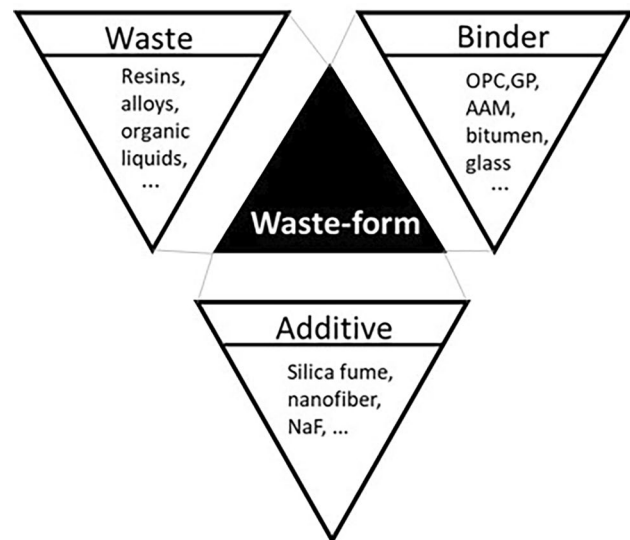
In nuclear waste immobilization application, Chupin et al. [42] found a strong correlation between the water content measured with thermogravimetric analysis in GP resulting from curing under various relative humidity and temperature conditions, and the radiolytic yield of hydrogen production. This yield exhibited an increase with the increase of the final water content in the GP. For instance, the radiolytic hydrogen production yield showed a linear increase from 0.02×10^{-7} mol/J at a water mass fraction of 0.01 to 0.6×10^{-7} mol/J at a water mass fraction of 0.33 for sodium based GP [42].

3 Radioactive waste types

Radioactive waste can be classified through different criteria, including its source, physical state (solid, liquid, gas), level of radioactivity, half-life, ultimate disposal method, or radiotoxicity. The key parameters for waste categorization typically are (i) half-lives and (ii) activity concentrations. Contaminated waste is then sorted based on its activation level, considering both the quantity and type of radiation emitted.

- 1- HLW: High-level radioactive waste—refers to waste with a significant amount of highly active materials, releasing heat or containing long-lived radionuclides. This type of waste requires careful consideration in the design of disposal facilities. The anticipated disposal method for HLW involves placing it in deep, stable geological formations.
- 2- ILW: Intermediate level radioactive waste—refers to waste with lower levels of radioactivity than HLW, containing long-lived radionuclides. This type of waste also necessitates a disposal in a geological facility far from the surface.
- 3- LLW: Low level radioactive waste- refers to waste that emits low radiation dose. However, this type of waste requires to be isolated to hundreds of years.

Fig. 10 Waste-form components



- 4- VLLW: Very low-level radioactive waste—refers to the waste, which does not need robust isolation and can be disposed near surface landfill.

The majority of waste materials treated with GPs fall within the categories of low, intermediate, and high-level waste. Very low-level waste primarily originates from medical facilities, industries, and various stages of the nuclear fuel cycle. Despite of its large volume, it contains only a small percentage of radioactivity. Typically, it does not necessitate shielding during handling and transportation and is suitable for shallow land burial. Techniques like compaction and incineration are often employed to reduce its volume. Low-level waste ranging from just above the category of very low-level waste to a level where shielding becomes necessary for periods extending up to several hundred years. In contrast, waste materials containing long-lived radionuclides, requiring greater isolation from the environment and enhanced shielding, fall into the intermediate-level waste category. Disposal of intermediate-level waste occurs at depths ranging from a few tens to hundreds of meters [2]. High-level waste represents only 3% of the total radioactive waste volume but contributes to a staggering 95% of the total radioactivity [60]. This type of waste typically originates from uranium fuel and other elements present in the core of a nuclear reactor. Materials like OPC, steel, glass, etc has traditionally been used to contain these types of wastes as part of radioactive waste management practices. However, the use of GPs as an alternative has shown promise due to their excellent properties in immobilizing and encapsulating radioactive waste, as confirmed by various studies [61, 62]. Vance et al. [62] uncovered the potential of GPs to immobilize uranium-rich waste falling into the high-level waste category.

Nuclear waste management requires understanding specific technical, scientific, and regulatory terminology. Some of the key terms used in the context of nuclear waste management includes: Waste-form; that is defined by the International Atomic Energy Agency (IAEA) [3] as the physical and chemical state of waste after treatment and conditioning, prior to packaging. The waste-form is produced by chemically incorporating the waste into a suitable matrix, such as glass, GP concrete, or ceramics for high-level radioactive waste. Materials commonly used for radioactive waste treatment include OPC, GPs, bitumen, homogeneous and heterogeneous glasses, single-phase and multiphase ceramics/minerals, and metals the wasteform as presented in Fig. 10 is composed of the waste, the binder and the additive. Conditioning refers to processes that transform waste into a form suitable for handling, transportation, storage, and disposal [3, 63]. This includes converting the waste into a solid waste-form, enclosing it in containers, and overpacking, which involves adding an outer layer for enhanced protection. Immobilization involves transforming the waste into a waste-form through solidification, embedding, or encapsulation to reduce the migration of radionuclides [13, 16, 64]. Encapsulation and embedding refer to surrounding the waste with a flowable material like cement to isolate waste particles and retain radionuclides, possibly with chemical incorporation. Encapsulation involves immobilizing powdered solids by mixing them with a matrix, while embedding refers to surrounding solid waste, such as metallic materials, with a matrix to form a waste-form. If no chemical interaction occurs between the waste and the encapsulation material, embedding is essentially the same as encapsulation [3, 63]. Solidification is typically used to refer to the immobilization of liquid and semi-liquid wastes by a chemical integration of the waste components within an appropriate matrix.

Fig. 11 Evolution of compressive strength of GP and OPC after extreme thermal exposure [20, 21]

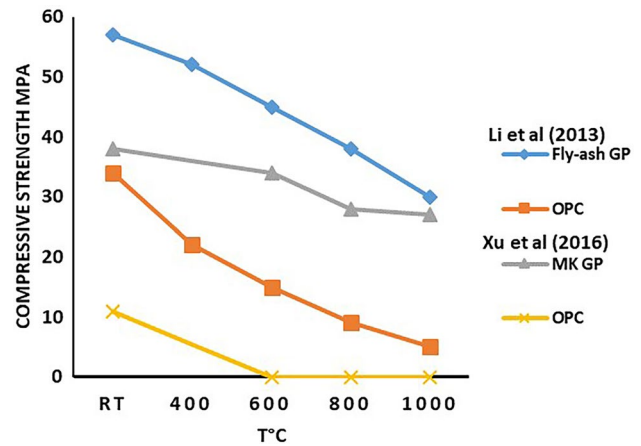


Table 1 Effect of gamma irradiation on the compressive strength of the cementitious material

Base Material	Dose	Compressive strength variation	Refs
GGBFS	1000 kGy	$\Delta\sigma = +35\%$	[68]
MK	9.5 MGy	$\Delta\sigma = 0$	[42]
MK	750 kGy	$\Delta\sigma = +10\%$	[20]
MK	50–1000 kGy	$\Delta\sigma = +10\%$	[69]
OPC	300–550 kGy	$\Delta\sigma = -10\%$	[67]

4 Geopolymer for nuclear waste immobilization

GPs have emerged as a promising solution for the immobilization of radioactive waste, wherein radio-contaminant elements are integrated into a GP-based matrix. This matrix functions as a binding agent, converting both solid and liquid waste into a cohesive and stable solid, thereby effectively immobilizing hazardous materials [27, 54, 61, 62]. This process, seeks to generate a waste form that adheres to regulatory criteria for radionuclide retention, water intrusion, and structural stability in near-surface disposal sites. As mentioned above, immobilization is achieved through methods such as solidification, embedding, or encapsulation. Such immobilization aims at creating a waste form that meets stringent regulatory criteria concerning radionuclide retention, resistance to water intrusion, and structural integrity, particularly in near-surface disposal environments. Among these factors are the composition of the radio-contaminant elements, the presence of free water, porosity, density, thermal and radiation stability, compressive strength, and leaching resistance. The introduction of GP materials as a substitution for OPC in the management of nuclear waste began in 1993, driven by their exceptional durability and long-term durability [65]. Numerous studies have demonstrated that GPs exhibit commendable stability under extreme conditions, including high temperatures, freeze–thaw cycles, and exposure to fire [21, 66]. For instance, comparative investigations into the mechanical stability of GP and OPC after extreme thermal exposure revealed that fly ash- and metakaolin-based GPs retained substantial mechanical performance even after exposure to temperatures reaching 1000 °C. In contrast, OPC displayed a significant decline in mechanical strength, reducing to 5 MPa after exposure to 100 °C and exhibiting cracking after reaching 400 °C (see Fig. 11). Additionally, both studies indicated that GP materials exhibited superior performance in resisting freeze–thaw cycles compared to OPC [20, 21]. Further research examining the effect of gamma irradiation on the compressive strength of GP and OPC materials illustrated notable differences in performance. Data presented in Table 1 reveal that alkali-activated materials, such as ground granulated blast-furnace slag (GGBFS) and metakaolin-based GPs, maintain impressive mechanical resistance even after receiving high doses of gamma irradiation. Conversely, a reduction in initial compressive strength of 10% was observed on OPC after irradiation with a dose of 550 kGy [67].

These findings highlight the effectiveness of GPs as an interesting material for the immobilization of radioactive waste, especially in situations that require resistance to thermal and radioactive conditions.

4.1 Effect of irradiation on geopolymers

Due to the presence of water within their pores, GPs are expected to produce hydrogen under gamma radiation through the radiolysis of the interstitial water. Several factors, including dose rate, radiation type, porosity, and water content, influence the radiolytic hydrogen yield in these materials [31]. Assessing these factors is critical for evaluating GPs as potential binders for nuclear waste treatment.

Research on gamma irradiation effects on GPs has largely focused on parameters like radiation chemical yield and compressive strength changes, as well as cumulative leach fraction (CLF) to measure radionuclide stabilization effectiveness [69–73]. Despite these investigations, the hydrogen radiolytic yield in GPs under wide range of gamma radiation exposure dose remains less studied, creating a gap in understanding the broader implications of irradiation on these materials.

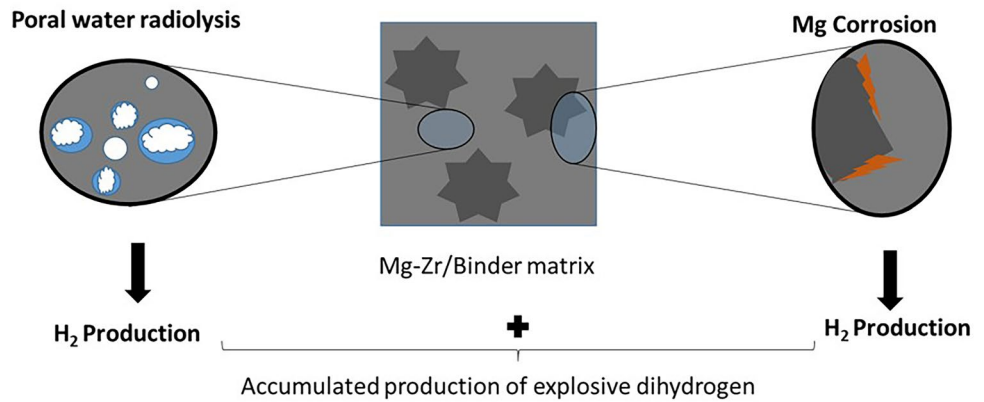
These modifications in the GP structure, including changes in mechanical strength, network densification, dehydration, precipitation, phase modification, porosity, leaching behavior, and radiolytic yield, are discussed below:

- **Mechanical strength:** The first paper that studied the effect of gamma irradiation on the GP was published by Roose et al. [14] and Lambertin et al. [69] in 2013 and 2014 respectively. The study was conducted on GPs with the formulations $1 \text{ Al}_2\text{O}_3$; 3.8 SiO_2 ; $1 \text{ Na}_2\text{O}$; $x \text{ H}_2\text{O}$, with $x = 11, 12$ or 13 . In the initial study, irradiation was conducted on a sample with a water molar ratio of 11, which had been stored in ambient air before analysis. Subsequent gamma irradiation at a rate of 600 kGy/h under argon revealed low hydrogen radiolytic yields, approximately $6 \times 10^{-9} \text{ mol/J}$, following a dose of 50 kGy. In the second study, the impact of gamma irradiation on the GP structure was examined. Regarding compressive strength, an enhancement of approximately 10% was noted after exposure to a dose of 1 MGy [69]. Other studies investigated the impact of beneficial equilibration resulting from γ irradiation on compressive strength. After an exposure to gamma irradiation for 2 months at a dosage of 1574 kGy, they found that the compressive strength increased by approximately 45%, from around 57 MPa to about 83 MPa. This enhanced strength surpassed the expected value in ambient curing alone at any stage. Remarkably, even after 6 months of gamma irradiation at a dosage of 4822 kGy, the compressive strength remained high at approximately 64 MPa, exceeding the initial unirradiated compressive strength of around 58 MPa. Further investigation revealed that with increasing aging times (up to 12 months) and dosages (up to 10,214 kGy), there was a decrease in compressive strength [70].
- **Network densification:** Brunet et al. [74] studied the effect of electrons irradiation with a dose of 1 MGy on glass silicate network with controlled porosity. Two modifications were detected: a decrease of the number of Q^2 sites and an increase in the number of Q^3 and Q^4 sites. This indicates that silica atoms became linked to more tetrahedral resulting in a densification of the structure. Lambertin et al. [69] also studied the effect of gamma irradiation on the structure of GP and they concluded a slight densification of the GP.
- **Porosity:** It was also found that the porosity size distribution changes from monomodal to bimodal after gamma irradiation [69, 75]. The same studies found that the total porosity percentage tends to increase after irradiation and no modifications in the morphology of the GPs were detected by scanning electron microscopy. Lambertin et al. [69] also investigated the evolution of the porosity of GP under gamma irradiation, revealing an expansion in porosity after reaching a dose of 1 MGy. Daniel et al. [71] also reported this in a more recent study, after testing the response of various GP formulations, they concluded that there was a significant loss of free water and thus an increase in the porosity.

4.2 Immobilization of metallic alloys

Silico-calcic materials such as OPC are generally used for medium radioactive waste immobilization. They offer numerous advantages, including low cost, good mechanical properties and long-term stability under irradiation. However, the use of these materials presents challenges, particularly when dealing with metallic waste, such as magnesium and zirconium alloys (Mg–Zr). Over time, silico-calcic materials exhibit vulnerability to corrosion, which consequently leads to an increased production of hydrogen (H_2) gas [31]. This hydrogen accumulation, combined with the hydrogen resulting from pore water radiolysis, raises the risk of explosion as represented in Fig. 12. In the search for a viable alternative to silico-calcic materials, GPs were investigated by Roose et al. [14]. They conducted a

Fig. 12 Dihydrogen production from pore water radiolysis and Mg corrosion

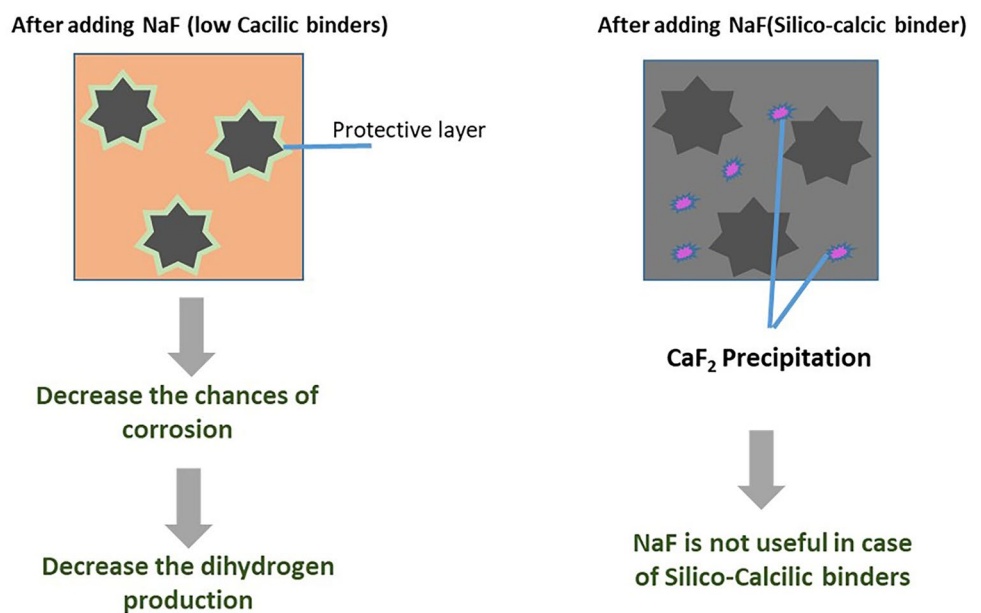


comprehensive study by incorporating Mg–Zr alloys within a metakaolin GP matrix. A comparison of the rates of galvanic corrosion exhibited by these alloys when embedded in both GP and OPC with the addition of NaF (sodium fluoride) was carried out. Their findings concluded that NaF addition to metakaolin-based GPs are more suitable when aiming for enhanced resistance to magnesium corrosion and limiting the dihydrogen production from the corrosion Mg–Zr alloys. The reason behind this result is that the absence of calcium in metakaolin-based GP offers the advantage of using NaF to form a protective passive layer around the metallic waste [76]. However, in the case of OPC or binders based on blast furnace slag, the addition of NaF would favor the precipitation of calcium fluoride (CaF_2) due to its limited solubility in water, which can inhibit the creation of an effective passivating layer on the metal (see Fig. 13). [77]. Hence, the variations in corrosion resistance and passivation efficacy between the two binder types further substantiate the potential of GPs as a preferable material for the immobilization of metallic radioactive waste.

4.3 Liquid waste immobilization

Figure 14 details the immobilization by solidification process of organic oil waste in the GP matrix; that involves the emulsification of the organic liquid within an activating solution prior to the addition of precursor materials. Within this waste-form structure, organic liquid is dispersed as spherical droplets, each having a radius ranging from 5 to 15 μm . These droplets are effectively isolated from each other and from the surrounding environment by the mesoporous network of the GP. A pertinent study by Cantarel et al. [11], explored the suitability of metakaolin-based GPs for this application, focusing on the composite structure which included chemical composition, GP porosity, and oil dispersion

Fig. 13 Difference between low silico-calcic and high silico-calcic binders in the presence of NaF



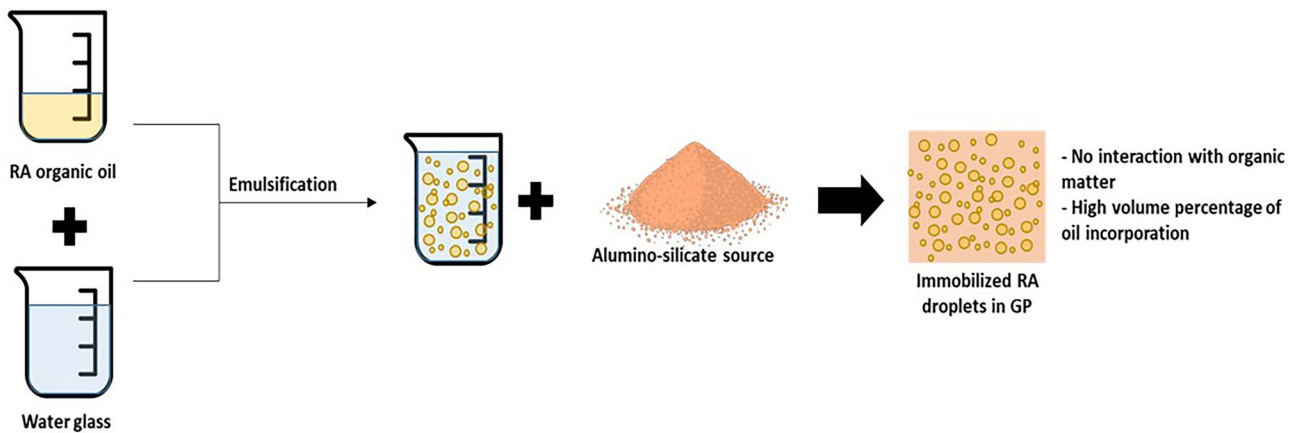


Fig. 14 Radioactive organic oil immobilization process

characteristics relevant to the immobilization of radioactive waste in oil form. In this comparative analysis, the authors evaluated the effectiveness of GP matrices against traditional calcium silicate-based cementitious matrices. Notably, the study revealed that the structural integrity of the GP remained unaffected by the presence of organic liquids, indicating a favorable compatibility for immobilization purposes in contrast to OPC matrices that reacted with the oil droplets.

In a recent study, Hasnaoui et al. [78] explored the management of radioactive liquid organic waste through the relationship between viscosity ratios of GP grouts and organic liquids. By incorporating three types of mineral oils at a constant rate of 30% volume, the research investigated how these viscosity differences affect the rheological, mechanical, and microstructural properties of GP/organic liquid composites. The results indicated that organic liquids with viscosities above 0.05 Pa.s can be effectively encapsulated in MK-based GP grouts without surfactants, highlighting viscosity ratio as a key factor in shaping the emulsions characteristics. Increasing GP grout viscosity enhanced the encapsulation of organic liquid droplets, and strong correlations between rheological parameters and the viscosities of both phases enabled the creation of empirical models with over 90% accuracy for estimating final rheological properties. The findings of this study [78] confirmed the suitability of MK-based GPs for solidifying high volumes of organic liquid waste.

4.4 Immobilization of heavy metals

Heavy metal contaminants from anthropogenic activities stands as a highly dangerous form of environmental pollution. Normally, heavy metals can be divided into cationic and anionic metals and both of them can cause serious pollution to the environment. Substantial advancements have been made in the research on utilizing GPs for the immobilization of cesium and strontium. Many studies demonstrate that GPs exceed OPC in terms of immobilization effectiveness, resistance to high temperatures, corrosion resistance, durability against freeze–thaw cycles and leaching resistance [22]. Despite the widespread use of OPC similar applications, it has several disadvantages, such as high permeability, elevated leaching concentrations, and incompatibility with certain contaminants like copper, lead, zinc, and tin. [79].

The process of immobilization of heavy metals involves the mixing of contaminants with a binder through a sequence of physical and chemical actions: physical encapsulation, chemical bonding, adsorption effect, etc. This transformation results into an environmentally acceptable waste-form that is suitable for handling, transport, and disposal [80, 81].

In this immobilization process, heavy metals are predominantly physically encapsulated by GP, forming a water-insoluble state. Conversely, when the metal is in a dissolved ion form, the main effects are chemical bonding and adsorption, where ions bond or adhere to the surface or pore structure [82]. Below, heavy ion stabilization through physical encapsulation, chemical bonding, and adsorption effects have been briefly described:

4.4.1 Physical encapsulation

During the process of geopolymerization, aluminosilicate dissolves under the influence of an alkaline solution. Subsequently, silicon (Si) and aluminum (Al) combine to create a binder through polycondensation reactions in the forms of $[\text{SiO}(\text{OH})_3]^-$, $[\text{SiO}_2(\text{OH})_2]^{2-}$, $[\text{SiO}_3(\text{OH})]^{3-}$, and $[\text{Al}(\text{OH})_4]^-$, leading to the formation of an amorphous three-dimensional network structure. Within this reaction, heavy metals are encapsulated within the matrix structure of GP through the

coating effect of oligomeric gel [83]. One of the materials that is used to immobilize heavy metal by physical encapsulation is the fly ash-based GP [79, 80].

4.4.2 Chemical bonding

The GP structure is an alkaline aluminosilicate hydrate characterized by a three-dimensional framework. This configuration consists of tetrahedral silicate and aluminate units interconnected by covalent bonds. Negative charges associated with tetrahedral Al^{3+} are balanced by alkali cations (Na^+ , K^+). The chemical bonding of heavy metals in GP predominantly occurs through the substitution of alkali cations, whereby the heavy metal cations bond to aluminate tetrahedral units and become immobilized within the GP structure [80].

4.4.3 Adsorption effect

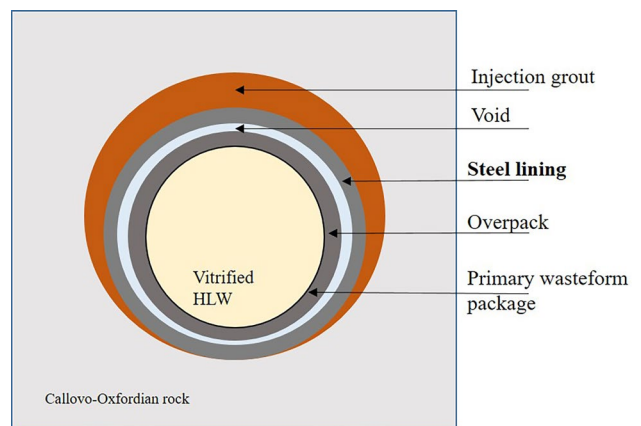
The adsorption behavior of metakaolin based GP for heavy metals has been characterized using the Langmuir model, revealing the presence of multiple types of binding sites on the metakaolin based GP surface capable of immobilizing various heavy metal ions [82]. Another study that investigated metakaolin based geopolymer, with a composition ratio of $\text{K}_2\text{O}:\text{SiO}_2:\text{H}_2\text{O}$ at 1:1:13, demonstrated a significant ability to adsorb cationic radionuclides like Cs^+ , Sr^{2+} , and Co^{2+} [84]. GP can also be manufactured in a way to have rough and irregular surface, providing it with a higher specific surface, consequently high adsorption capacity. Heavy metals can be adsorbed onto the surface of the GP [49]. In a study conducted on the fly ash, it was found that after using nano- Al_2O_3 particles, the immobilization was significantly improved. It was concluded that this improvement is due to the increase in surface area after nano particles addition [80]. Jin et al. [85] explored the application of 3D printed GP adsorption sieve for removal of methylene blue and adsorption mechanism. This novel method demonstrated notable adsorption rates for methylene blue (MB), achieving 83.6% in dynamic conditions after 200 min and 97.1% in static conditions over seven days. Furthermore, it was concluded that the interaction of GP adsorption sieve with MB was influenced by electrostatic interactions, hydrogen bonding, and electron transfer processes. This novel research [85] offers a promising framework for the design and preparation of environmentally friendly adsorbent systems using 3D printing technology.

4.4.4 Cations immobilization

The radioactive isotopes of cesium and strontium are typically found in the cooling water of nuclear reactors and storage ponds in cationic form (Cs^+ and Sr^{2+}). Considerable advancements have been achieved in research on using GPs to immobilize Cs and Sr. Research has shown that GPs outperform OPC in terms of immobilization effectiveness, resistance to high temperatures, corrosion, and freeze–thaw cycles [22]. This makes them strong candidates to replace cement as the next-generation solidification matrix for Cs and Sr. Initially, it was thought that Cs^+ immobilization relied mainly on ion exchange with cations (like K^+ and Na^+) within the GP structure, allowing Cs^+ to penetrate the matrix and balance the negative charge of $[\text{AlO}_4]^-$ groups, yet its high solubility presents challenges, particularly in both acidic and alkaline conditions. However, recent studies suggest that Cs^+ can also form chemical bonds with geopolymerization products. Arbel-Haddad et al. [86] found that using low-Si metakaolin-based GPs to immobilize Cs resulted in the formation of a small amount of crystalline zeolite F phase. It was found that Cs showed a tendency to bind with zeolite F, even at relatively low concentrations. In examining the effects of ash-based and slag-based GP on Cs^+ and Sr^{2+} leaching, a study [87] have shown a significant variance among materials. Cs^+ exhibited the GP highest leaching rates in OPC, followed by slag-based GP, while fly ash-based GP demonstrated the lowest leaching. This enhanced immobilization in fly ash-based GP is attributed to its smaller critical pore size, which effectively reduces nuclide diffusion. Compared to OPC, the diffusivity of cesium and strontium in fly ash-based GP was lower, underscoring its superior containment performance. Additionally, zeolitic phases within GPs, particularly those formed in fly ash-based GPs, contribute to immobilization through selective ion exchange mechanisms. Research [88] has confirmed that zeolite structures, like mordenite and zeolite A, show high Cs^+ adsorption capacities reaching 99%, even when competing ions like Na^+ and K^+ are present. The adsorption kinetics for Cs^+ and Sr^{2+} in these materials follow pseudo-second-order models, indicating that both physisorption and chemisorption play roles in the sorption process [89]. Moreover, ion exchange with Na^+ and other activators within the GP matrix further supports Cs^+ immobilization, as shown in metakaolin/slag-based zeolite GP microspheres.

Furthermore, the type of activator in GPs affects Cs^+ immobilization efficiency, with Na-based GPs generally showing better performance than K-based GPs, in alignment with soft acid–base theory [44]. In an additional study [90], Cs^+

Fig. 15 Cross section view of the underground tunnel for radioactive waste storage



ions were observed to preferentially bind with aluminate phases in Na-activated GPs, effectively replacing Na^+ within the gel structure. Functionalized GPs, such as those incorporating $\text{K}_2\text{CuFe}(\text{CN})_6$, also show increased selectivity for Cs^+ in competitive environments, enhancing the material's retention capacity in nuclear waste applications. The effect of Cs^+ incorporation on the mechanical properties of GPs depends on both the method of addition and the type of activator used [24]. In sodium hydroxide-activated fly ash, Cs^+ reduces tensile strength due to the lower pH when forming $\text{CsOH}\cdot\text{H}_2\text{O}$ [88]. Replacing sodium hydroxide with cesium hydroxide in metakaolin-based GPs also negatively impacts compressive strength. However, in sodium silicate-activated slag GPs, adding Cs^+ increases compressive strength by about 20 MPa, likely due to enhanced slag reactions and the formation of more N–A–S–H gels [91].

4.4.5 Anions immobilization

In GPs, the $(\text{Al}(\text{OH})_4^-)$ groups need cations to balance out their negative charges, which gives GPs a special advantage in trapping positively charged nuclear waste. However, selenium (Se) is one of the fission products in nuclear reactors and it possesses an extremely long half-life. It is present in nuclear waste as SeO_3^{2-} and SeO_4^{2-} . These anionic forms are repelled by the negative charges in the $(\text{Al}(\text{OH})_4^-)$ groups, reducing the effectiveness of GPs in immobilizing anionic pollutants like selenium [84]. This limitation restricts the use of GPs in containing anionic contaminants. There are only a few studies about the stabilization of Se in GPs. Tian et al. [92] research has investigated the potential of GPs activated by different alkaline solutions (NaOH and Na_2SiO_3) for immobilizing selenium in the form of SeO_3^{2-} and SeO_4^{2-} , the study found that Na_2SiO_3 -activated GPs generally had superior immobilization effectiveness, with lower leaching rates for SeO_3^{2-} and SeO_4^{2-} (10% and 18%, respectively) compared to NaOH-activated GPs (58% and 74%). This enhanced performance was attributed to electrostatic interactions within the denser $\text{Na}_2\text{SiO}_3^-$ activated structures. Furthermore, the inclusion of calcined hydrotalcite (CHT) as an additive affected the compactness of the GP matrix, decreasing its density and impacting leaching behaviors. In NaOH-activated systems, CHT appeared beneficial, aiding Se immobilization likely through hydrotalcite formation, whereas in $\text{Na}_2\text{SiO}_3^-$ activated GPs, CHT addition unexpectedly increased leaching. These findings highlight the complexities of GP-alkaline activator interactions in immobilizing anionic pollutants and suggest that alkaline solution and additive compatibility are crucial for optimizing immobilization performance. Tian and Sasaki [93] also developed alkali-activated GPs aimed at immobilizing Se^{4-} , but the results showed limited success, with over 60% of Se leaching out. When MgO was added, however, it reduced Se leaching to below 10%, which was attributed to a decrease in the specific surface area and pore volume of the solidified matrix. Further studies by Tian et al. [55] examined how Si/Al ratios (2, 3, 4, and 5) impacted the ability of GPs to contain SeO_3^{2-} and SeO_4^{2-} . Results showed that increasing the Si/Al ratio led to a rise in Se leaching. This immobilization relies mainly on electrostatic forces, yet due to charge repulsion, the efficiency remains less than ideal. Using acidic based GP to enhance anions fixation remains a possible yet underexplored research area.

4.5 Alternative to steel for the nuclear waste cells lining

The use of Callovo-Oxfordian argillites in the production of GPs to potentially replace steel components, such as the tunnel lining of HLW storage cells that may be subjected to anaerobic corrosion under extreme exposure conditions [6, 94] was investigated by several researches [95, 96]. The cross-sectional view of the underground tunnel, including the

lining, is represented in Fig. 15; the geological system is formed of: an overpack containing solidified waste from placed in a steel lining cell, and grout injected between the steel and the excavated Callovo-Oxfordian rock. This use case addresses practical challenges, including the development of materials that are neutral in storage processes and the reduction of environmental impact by integrating excavated argillite waste from the site into the facility construction. To identify optimal formulations meeting the specifications for lining HLW storage cells and understand the underlying mechanisms, various additions and mass fractions of metakaolin, glass fibers, and wollastonite were tested as part of a study conducted by Andra (French national agency for radioactive wastes management) [95]. The mechanical properties of the resulting composites varied in compression from 22 to 101 MPa and in flexion from 3 to 20 MPa. These properties remained stable over time under different humidity levels and temperature for some formulations [97]. These findings, coupled with structural and microstructural characteristics, demonstrate that adding wollastonite enhances polycondensation reactions, increases the reactive mixture's viscosity, and improves mechanical properties, while glass fibers hinder crack formation. However, Archez [95] did not conduct any studies to assess the response of tested GP mix design to irradiation. Therefore, there is a need to further investigate the durability of GP composite under the corresponding extreme coupled conditions: chemical attacks under saturated conditions from the geological environment, irradiations resulting from encapsulated HLW and ILW.

5 Challenges and outlook

GPs are increasingly recognized for their potential in nuclear waste management, offering significant advantages over traditional materials such as OPC [7, 81]. However, several challenges hinder their broader application and effectiveness in this field [24, 81]. Standardization issues arise due to the variability in chemical composition and reactivity of different aluminosilicate sources (e.g., metakaolin, fly ash, slag), complicating the standardization of GP production processes and potentially affecting scalability for nuclear waste containment. Additionally, inconsistent mechanical performance is a concern, as the compressive strength of fly ash-based GPs can decrease sharply beyond certain thresholds when mixed with heavy materials [80]. Current research on the leaching behavior of GPs mainly emphasizes binder-only formulations, necessitating further evaluation of GP-based mortars and concretes to understand their long-term stability in radioactive waste containment. Long-term durability, especially under the influence of radioactive decay, remains underexplored and requires further investigation to comprehend how such conditions might alter GP microstructure over time. Issues like efflorescence and increased porosity can compromise waste containment, and while chemical additives have been examined to mitigate these effects, more research is needed on their impact on radionuclide immobilization. The drying shrinkage of GP binders, which is typically five to six times greater than OPC binders, also poses risks of cracking and increased permeability; although incorporating fibers like glass fibers shows promise in reducing shrinkage [98], optimization for nuclear waste applications is still needed. Additionally, Metakaolin GP does not have the ability to incorporate anionic radionuclides but it has a high capacity to immobilize cationic radionuclides [84]. Hence, GPs are until now more suitable for the removal of cationic substances from wastewater due to negative charge of GPs from charge deficiency of Al. Lastly, while GPs effectively manage low to moderate activity wastes, the challenges in handling higher activity wastes are more complex, necessitating additional studies to accurately predict interactions and assess associated risks.

6 Conclusion

GPs, as innovative inorganic polymers, demonstrate exceptional properties, including high mechanical strength, durability, and chemical resistance, making them promising materials for immobilizing hazardous pollutants. Their unique advantages have spurred significant research interest in their application for nuclear waste containment. However, several challenges remain, particularly concerning standardization, scalability, and environmental impact, which require further investigation to unlock their full potential. This study identifies the advancements and key research gaps in the geopolymer application for nuclear waste immobilization, such as durability and leaching behavior in complex environments, and their effectiveness in immobilizing high-level radioactive waste. Additionally, the influence of high temperatures, radiation exposure, and additive incorporation on GP performance remains underexplored. Expanding research on the immobilization of organic pollutants and improving anion containment through novel methods to enhance leaching resistance are also critical areas for future work. By addressing these challenges and gaps, this review provides a foundation for advancing the application of GPs as effective materials for immobilizing a wide range of nuclear wastes.

Acknowledgements The authors acknowledge financial support from CESI Engineering School.

Author contributions M.H. was responsible for the conceptualization, literature review, methodology, data curation and writing of the original draft preparation. S.dB., N.L., H.R. and J-D.P. were on the supervision, methodology of review, review and editing. Each author has reviewed and approved the final version of the manuscript.

Data availability No datasets were generated or analysed during the current study.

Declarations

Competing interests The authors declare no competing interests.

Open Access This article is licensed under a Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International License, which permits any non-commercial use, sharing, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if you modified the licensed material. You do not have permission under this licence to share adapted material derived from this article or parts of it. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by-nc-nd/4.0/>.

References

1. Nathaniel SP, Alam MdS, Murshed M, et al. The roles of nuclear energy, renewable energy, and economic growth in the abatement of carbon dioxide emissions in the G7 countries. *Environ Sci Pollut Res*. 2021;28:47957–72. <https://doi.org/10.1007/s11356-021-13728-6>.
2. Alwaeli M, Mannheim V. Investigation into the current state of nuclear energy and nuclear waste management—a state-of-the-art review. *Energies*. 2022;15:4275. <https://doi.org/10.3390/en15124275>.
3. Internationale Atomenergie-Organisation. Radioactive waste management glossary. 2003rd ed. Vienna: International Atomic Energy Agency; 2003.
4. The behaviours of cementitious materials in long term storage and disposal of radioactive waste : results of a coordinated research project. – Vienna : International Atomic Energy Agency, 2013. p. ; IAEA-TECDOC series, ISSN1011-4289 ; no. 1701. <https://www.iaea.org/publications/10439/the-behaviours-of-cementitious-materials-in-longterm-storage-and-disposal-of-radioactive-waste>
5. Zheng Z, Li Y, Zhang Z, Ma X. The impacts of sodium nitrate on hydration and microstructure of Portland cement and the leaching behavior of Sr²⁺. *J Hazard Mater*. 2020;388:121805. <https://doi.org/10.1016/j.jhazmat.2019.121805>.
6. Crusset D, Deydier V, Necib S, et al. Corrosion of carbon steel components in the French high-level waste programme: evolution of disposal concept and selection of materials. *Corros Eng, Sci Technol*. 2017;52:17–24. <https://doi.org/10.1080/1478422X.2017.1344416>.
7. Wang A, Zheng Y, Zhang Z, et al. The durability of alkali-activated materials in comparison with ordinary Portland cements and concretes: a review. *Engineering*. 2020;6:695–706. <https://doi.org/10.1016/j.eng.2019.08.019>.
8. Liu X, Ding Y, Lu X. Immobilization of simulated radionuclide 90Sr by Fly Ash-Slag-Metakaolin-Based geopolymer. *Nucl Technol*. 2017;198:64–9. <https://doi.org/10.1080/00295450.2017.1292810>.
9. Garces Jerome Ignatius T, Tigue April Anne S, Promentilla Michael Angelo B. A systematic mapping study of geopolymers for radioactive waste management. *Chem Eng Trans*. 2022;94:1345–50. <https://doi.org/10.3303/CET2294224>.
10. Davidovits J. Geopolymers: inorganic polymeric new materials. *J Therm Anal*. 1991;37:1633–56. <https://doi.org/10.1007/BF01912193>.
11. Cantarel V, Nouaille F, Rooses A, et al. Solidification/stabilisation of liquid oil waste in metakaolin-based geopolymer. *J Nucl Mater*. 2015;464:16–9. <https://doi.org/10.1016/j.jnucmat.2015.04.036>.
12. Li L, Xu Z, Li H, et al. Immobilization of strontium and cesium by aluminosilicate ceramics derived from metakaolin geopolymer-zeolite a composites via 1100°C heating treatment. *Ceram Int*. 2022;48:15236–42. <https://doi.org/10.1016/j.ceramint.2022.02.054>.
13. Jiang Z, Xu Z, Shuai Q, et al. Thermal stability of geopolymer—Sr contaminated zeolite a blends. *Key Eng Mater*. 2017;727:1089–97. <https://doi.org/10.4028/www.scientific.net/KEM.727.1089>.
14. Rooses A, Steins P, Dannoux-Papin A, et al. Encapsulation of Mg–Zr alloy in metakaolin-based geopolymer. *Appl Clay Sci*. 2013;73:86–92. <https://doi.org/10.1016/j.clay.2012.09.023>.
15. Arbel-Haddad M, Ofer-Rozovsky E, Goldbourt A. Facile formation of pollucite in geopolymers: implications for radioactive Cs immobilization. *Ceram Int*. 2023;49:30881–5. <https://doi.org/10.1016/j.ceramint.2023.07.006>.
16. Oliveira L, Azevedo A, Marvila M, et al. Durability of geopolymers with industrial waste. *Case Stud Constr Mater*. 2021;16:e00839. <https://doi.org/10.1016/j.cscm.2021.e00839>.
17. Işıkdağ B, Yalghuz MR. Strength development and durability of metakaolin geopolymer mortars containing pozzolans under different curing conditions. *Minerals*. 2023;13:857. <https://doi.org/10.3390/min13070857>.
18. Chindapasirt P, Lao-un J, Zaetang Y, et al. Thermal insulating and fire resistance performances of geopolymer mortar containing auto glass waste as fine aggregate. *J Build Eng*. 2022;60:105178. <https://doi.org/10.1016/j.jobe.2022.105178>.
19. Duan P, Yan C, Zhou W, et al. An investigation of the microstructure and durability of a fluidized bed fly ash–metakaolin geopolymer after heat and acid exposure. *Mater Des*. 2015;74:125–37. <https://doi.org/10.1016/j.matdes.2015.03.009>.

20. Li Q, Sun Z, Tao D, et al. Immobilization of simulated radionuclide $^{133}\text{Cs}^+$ by fly ash-based geopolymer. *J Hazard Mater*. 2013;262:325–31. <https://doi.org/10.1016/j.jhazmat.2013.08.049>.
21. Xu Z, Jiang Z, Wu D, et al. Immobilization of strontium-loaded zeolite a by metakaolin based-geopolymer. *Ceram Int*. 2016. <https://doi.org/10.1016/j.ceramint.2016.12.092>.
22. Liu J, Xu Y, Zhang W, et al. Solidification performance and mechanism of typical radioactive nuclear waste by geopolymers and geopolymer ceramics: a review. *Prog Nucl Energy*. 2024;169:105106. <https://doi.org/10.1016/j.pnucene.2024.105106>.
23. Phillip E, Choo TF, Khairuddin NWA, Abdel Rahman RO. On the sustainable utilization of geopolymers for safe management of radioactive waste: a review. *Sustainability*. 2023;15:1117. <https://doi.org/10.3390/su15021117>.
24. Zhu Y, Zheng Z, Deng Y, et al. Advances in immobilization of radionuclide wastes by alkali activated cement and related materials. *Cement Concr Compos*. 2022;126:104377. <https://doi.org/10.1016/j.cemconcomp.2021.104377>.
25. Zhang ZH, Zhu HJ, Zhou CH, Wang H. Geopolymer from kaolin in China: an overview. *Appl Clay Sci*. 2015. <https://doi.org/10.1016/j.clay.2015.04.023>.
26. Glukhovskiy D (1959) *Soil Silicates*. Gosstroyizdat USSR, Kiev
27. Fernandez-Jimenez A, Macphee DE, Lachowski EE, Palomo A. Immobilization of cesium in alkaline activated fly ash matrix. *J Nucl Mater*. 2005;346:185–93. <https://doi.org/10.1016/j.jnucmat.2005.06.006>.
28. van Deventer JSJ, Provis JL, Duxson P, Lukey GC. Reaction mechanisms in the geopolymeric conversion of inorganic waste to useful products. *J Hazard Mater*. 2007;139:506–13. <https://doi.org/10.1016/j.jhazmat.2006.02.044>.
29. Duxson P, Lukey GC, Van Deventer JSJ. The thermal evolution of metakaolin geopolymers: part 2—phase stability and structural development. *J Non-Cryst Solids*. 2007;353:2186–200. <https://doi.org/10.1016/j.jnoncrysol.2007.02.050>.
30. Duxson P, Lukey GC, Van De Venter JSJ, et al. Microstructural characterisation of metakaolin-based geopolymers: 106th annual Meeting of the American ceramic society. *Ceram Trans*. 2005;165:71–85.
31. Chupin F (2017) Caractérisation de l'effet des irradiations sur les géopolymères
32. Cyriaque Kaze R, Naghizadeh A, Tchadjie L, et al. Lateritic soils based geopolymer materials: a review. *Constr Build Mater*. 2022;344:128157. <https://doi.org/10.1016/j.conbuildmat.2022.128157>.
33. Kaze CR, Venyite P, Nana A, et al. Meta-halloysite to improve compactness in iron-rich laterite-based alkali activated materials. *Mater Chem Phys*. 2020;239:122268. <https://doi.org/10.1016/j.matchemphys.2019.122268>.
34. Rodrigue Kaze C, Ninla Lemougna P, Alomayri T, et al. Characterization and performance evaluation of laterite based geopolymer binder cured at different temperatures. *Constr Build Mater*. 2021;270:121443. <https://doi.org/10.1016/j.conbuildmat.2020.121443>.
35. Gomes KC, Lima GST, Torres SM, et al. Iron distribution in geopolymer with ferromagnetic rich precursor. *MSF*. 2010;643:131–8. <https://doi.org/10.4028/www.scientific.net/MSF.643.131>.
36. Xu Z, Li C, Peng X. Immobilization of radioactive borate liquid waste using calcined laterite–phosphoric acid– Fe_3O_4 -based geopolymer waste forms. *Ceram Int*. 2024;50:48164–73. <https://doi.org/10.1016/j.ceramint.2024.09.166>.
37. Alves L, Leklou N, De Barros S. A comparative study on the effect of different activating solutions and formulations on the early stage geopolymerization process. *MATEC Web Conf*. 2020;322:01039. <https://doi.org/10.1051/mateconf/202032201039>.
38. Steins P, Poulesquen A, Diat O, Frizon F. Structural evolution during geopolymerization from an early age to consolidated material. *Langmuir*. 2012;28:8502–10. <https://doi.org/10.1021/la300868v>.
39. Steins P, Poulesquen A, Frizon F, et al. Effect of aging and alkali activator on the porous structure of a geopolymer. *J Appl Crystallogr*. 2014;47:316–24. <https://doi.org/10.1107/S160057671303197X>.
40. Benavent V, Frizon F, Poulesquen A. Effect of composition and aging on the porous structure of metakaolin-based geopolymers. *J Appl Crystallogr*. 2016;49:2116–28. <https://doi.org/10.1107/S1600576716014618>.
41. Kim B, Lee J, Kang J, Um W. Development of geopolymer waste form for immobilization of radioactive borate waste. *J Hazard Mater*. 2021;419:126402. <https://doi.org/10.1016/j.jhazmat.2021.126402>.
42. Chupin F, Dannoux-Papin A, Ngono Ravache Y, d'Espinose de Lacaillerie J-B. Water content and porosity effect on hydrogen radiolytic yields of geopolymers. *J Nucl Mater*. 2017;494:138–46. <https://doi.org/10.1016/j.jnucmat.2017.07.005>.
43. He P, Wang R, Fu S, et al. Safe trapping of cesium into doping-enhanced pollucite structure by geopolymer precursor technique. *J Hazard Mater*. 2019;367:577–88. <https://doi.org/10.1016/j.jhazmat.2019.01.013>.
44. Kuenzel C, Cisneros JF, Neville TP, et al. Encapsulation of Cs/Sr contaminated clinoptilolite in geopolymers produced from metakaolin. *J Nucl Mater*. 2015;466:94–9. <https://doi.org/10.1016/j.jnucmat.2015.07.034>.
45. Tchakouté HK, Fotio D, Rüscher CH, et al. The effects of synthesized calcium phosphate compounds on the mechanical and microstructural properties of metakaolin-based geopolymer cements. *Constr Build Mater*. 2018;163:776–92. <https://doi.org/10.1016/j.conbuildmat.2017.12.162>.
46. Ma S, Zhang Z, Liu X. Comprehensive understanding of aluminosilicate phosphate geopolymers: a critical review. *Materials*. 2022;15:5961. <https://doi.org/10.3390/ma15175961>.
47. Pu S, Zhu Z, Song W, et al. A novel acidic phosphoric-based geopolymer binder for lead solidification/stabilization. *J Hazard Mater*. 2021;415:125659. <https://doi.org/10.1016/j.jhazmat.2021.125659>.
48. Liu Q, Feng L, Sun Y, et al. Effects of phosphate glass on Cs+ immobilization in geopolymer glass-ceramics. *Ceram Int*. 2023;49:6545–53. <https://doi.org/10.1016/j.ceramint.2022.10.113>.
49. Steveson M, Sagoe-Crentsil K. Relationships between composition, structure and strength of inorganic polymers. *J Mater Sci*. 2005;40:4247–4259. <https://doi.org/10.1007/s10853-005-2794-x>.
50. Barbosa VFF, MacKenzie KJD, Thaumaturgo C. Synthesis and characterisation of materials based on inorganic polymers of alumina and silica: sodium polysialate polymers. *Int J Inorg Mater*. 2000;2:309–17. [https://doi.org/10.1016/S1466-6049\(00\)00041-6](https://doi.org/10.1016/S1466-6049(00)00041-6).
51. Geddes DA, Walkley B, Galliard CL, et al Effect of exposure of metakaolin-based geopolymer cements to gamma radiation. *Journal of the American Ceramic Society*. <https://doi.org/10.1111/jace.19747>.
52. Duxson P, Provis JL, Lukey GC, et al. Understanding the relationship between geopolymer composition, microstructure and mechanical properties. *Colloids Surf, A*. 2005;269:47–58. <https://doi.org/10.1016/j.colsurfa.2005.06.060>.

53. Duxson P, Mallicoat SW, Lukey GC, et al. The effect of alkali and Si/Al ratio on the development of mechanical properties of metakaolin-based geopolymers. *Colloids Surf, A*. 2007;292:8–20. <https://doi.org/10.1016/j.colsurfa.2006.05.044>.
54. Tian Q, Nakama S, Sasaki K. Immobilization of cesium in fly ash-silica fume based geopolymers with different Si/Al molar ratios. *Sci Total Environ*. 2019;687:1127–37. <https://doi.org/10.1016/j.scitotenv.2019.06.095>.
55. Tian Q, Changshuai C, Wang M, et al. Effect of Si/Al molar ratio on the immobilization of selenium and arsenic oxyanions in geopolymer. *Environ Pollut*. 2021;274:116509. <https://doi.org/10.1016/j.envpol.2021.116509>.
56. Steins P Influence des paramètres de formulation sur la texturation et la structuration des géopolymères. PhDthesis, Université de Limoges, 2014. <https://theses.hal.science/tel-01083951>.
57. Palomo A, Grutzeck MW, Blanco MT. Alkali-activated fly ashes: A cement for the future. *Cem Concr Res*. 1999;29:1323–9. [https://doi.org/10.1016/S0008-8846\(98\)00243-9](https://doi.org/10.1016/S0008-8846(98)00243-9).
58. Perera DS, Uchida O, Vance ER, Finnie KS. Influence of curing schedule on the integrity of geopolymers. *J Mater Sci*. 2007;42:3099–106. <https://doi.org/10.1007/s10853-006-0533-6>.
59. Rovnaník P. Effect of curing temperature on the development of hard structure of metakaolin-based geopolymer. *Constr Build Mater*. 2010;24:1176–83. <https://doi.org/10.1016/j.conbuildmat.2009.12.023>.
60. Radioactive Waste Management - World Nuclear Association. <https://world-nuclear.org/information-library/nuclear-fuel-cycle/nuclear-waste/radioactive-waste-management>. Accessed 5 Dec 2024
61. Khalil MY, Merz E. Immobilization of intermediate-level wastes in geopolymers. *J Nucl Mater*. 1994;211:141–8. [https://doi.org/10.1016/0022-3115\(94\)90364-6](https://doi.org/10.1016/0022-3115(94)90364-6).
62. Vance E, Vance E, Kiyama S, et al. Geopolymers as candidates for the immobilisation of low- and intermediate-level waste. *MRS Proc*. 2006. <https://doi.org/10.1557/PROC-985-0985-NN10-01>.
63. Jantzen CM, Lee WE, Ojovan MI. 6 - Radioactive waste (RAW) conditioning, immobilization, and encapsulation processes and technologies: overview and advances. In: Lee WE, Ojovan MI, Jantzen CM (eds) *Radioactive waste management and contaminated site clean-up*. Woodhead Publishing, 2013;171–272. <https://doi.org/10.1533/9780857097446.1.171>.
64. Künzel C, Metakaolin based geopolymers to encapsulate nuclear waste. Imperial College London; 2013. <https://core.ac.uk/download/17294539.pdf>.
65. Davidovits J (2020) *Geopolymer: chemistry & applications*, 5th ed. Institut Géopolymère, Saint-Quentin
66. Zhang H, Kodur V, Cao L, Qi S. Fiber reinforced geopolymers for fire resistance applications. *Proced Eng*. 2014;71:153–8. <https://doi.org/10.1016/j.proeng.2014.04.022>.
67. Vodák F, Trtík K, Sopko V, et al. Effect of γ -irradiation on strength of concrete for nuclear-safety structures. *Cem Concr Res*. 2005;35:1447–51. <https://doi.org/10.1016/j.cemconres.2004.10.016>.
68. Lee W-H, Cheng T-W, Ding Y-C, et al. Geopolymer technology for the solidification of simulated ion exchange resins with radionuclides. *J Environ Manage*. 2019;235:19–27. <https://doi.org/10.1016/j.jenvman.2019.01.027>.
69. Lambertin D, Boher C, Dannoux-Papin A, et al. Influence of gamma ray irradiation on metakaolin based sodium geopolymer. *J Nucl Mater*. 2013;443:311–5. <https://doi.org/10.1016/j.jnucmat.2013.06.044>.
70. Yeoh MLY, Ukritnukun S, Rawal A, et al. Mechanistic impacts of long-term gamma irradiation on physicochemical, structural, and mechanical stabilities of radiation-responsive geopolymer pastes. *J Hazard Mater*. 2021;407:124805. <https://doi.org/10.1016/j.jhazmat.2020.124805>.
71. Geddes DA, Walkley B, Le Galliard C, et al. Effect of exposure of metakaolin-based geopolymer cements to gamma radiation. *J Am Ceram Soc*. 2024;107(7):4621–30. <https://doi.org/10.1111/jace.19747>.
72. Cantarel V, Lambertin D, Labed V, Yamagishi I. Online measurement of the atmosphere around geopolymers under gamma irradiation. *J Nucl Sci Technol*. 2021;58:62–71. <https://doi.org/10.1080/00223131.2020.1801531>.
73. Leay L, Potts A, Donocli T. Geopolymers from fly ash and their gamma irradiation. *Mater Lett*. 2018;227:240–2. <https://doi.org/10.1016/j.matlet.2018.05.088>.
74. Brunet F, Charpentier T, Le Caer S, Renault J-P. Solid-state NMR characterization of a controlled-pore glass and of the effects of electron irradiation. *Solid State Nucl Magn Reson*. 2008;33:1–11. <https://doi.org/10.1016/j.ssnmr.2007.12.001>.
75. Deng N, An H, Cui H, et al. Effects of gamma-ray irradiation on leaching of simulated 133 Cs + radionuclides from geopolymer wastefoms. *J Nucl Mater*. 2015;459:270–5. <https://doi.org/10.1016/j.jnucmat.2015.01.052>.
76. Gulbrandsen E, Taftø J, Olsen A. The passive behaviour of Mg in alkaline fluoride solutions. Electrochemical and electron microscopical investigations. *Corros Sci*. 1993;34:1423–40. [https://doi.org/10.1016/0010-938X\(93\)90238-C](https://doi.org/10.1016/0010-938X(93)90238-C).
77. Macías A, Escudero ML. The effect of fluoride on corrosion of reinforcing steel in alkaline solutions. *Corros Sci*. 1994;36:2169–80. [https://doi.org/10.1016/0010-938X\(94\)90015-9](https://doi.org/10.1016/0010-938X(94)90015-9).
78. Hasnaoui A, Reeb C, De Campos M, et al. Controlling the properties of metakaolin-based geopolymer/liquid organic waste emulsions: a rheological approach. *Appl Clay Sci*. 2023;245:107160. <https://doi.org/10.1016/j.clay.2023.107160>.
79. Wang Y, Han F, Mu J. Solidification/stabilization mechanism of Pb (II), Cd (II), Mn (II) and Cr (III) in fly ash based geopolymers. *Constr Build Mater*. 2018;30(160):818–27.
80. Guo X, Zhang L, Jiabao H, Shi H. Detoxification and solidification of heavy metal of chromium using fly ash-based geopolymer with chemical agents. *Constr Build Mater*. 2017;151:394–404. <https://doi.org/10.1016/j.conbuildmat.2017.05.199>.
81. Ji Z, Pei Y. Bibliographic and visualized analysis of geopolymer research and its application in heavy metal immobilization: a review. *J Environ Manage*. 2019;231:256–67. <https://doi.org/10.1016/j.jenvman.2018.10.041>.
82. Siyal AA, Shamsuddin MR, Khan MI, et al. A review on geopolymers as emerging materials for the adsorption of heavy metals and dyes. *J Environ Manage*. 2018;224:327–39. <https://doi.org/10.1016/j.jenvman.2018.07.046>.
83. Phair JW, Van Deventer JSJ. Effect of the silicate activator pH on the microstructural characteristics of waste-based geopolymers. *Int J Miner Process*. 2002;66:121–43. [https://doi.org/10.1016/S0301-7516\(02\)00013-3](https://doi.org/10.1016/S0301-7516(02)00013-3).
84. Niu X, Elakneswaran Y, Islam CR, et al. Adsorption behaviour of simulant radionuclide cations and anions in metakaolin-based geopolymer. *J Hazard Mater*. 2022;429:128373. <https://doi.org/10.1016/j.jhazmat.2022.128373>.
85. Jin H, Zhang Y, Zhang X, et al. 3D printed geopolymer adsorption sieve for removal of methylene blue and adsorption mechanism. *Colloids Surf, A*. 2022;648:129235. <https://doi.org/10.1016/j.colsurfa.2022.129235>.

86. Arbel-Haddad M, Harnik Y, Schlosser Y, Goldbourt A. Cesium immobilization in metakaolin-based geopolymers elucidated by ^{133}Cs solid state NMR spectroscopy. *J Nucl Mater.* 2022;562:153570. <https://doi.org/10.1016/j.jnucmat.2022.153570>.
87. Jang JG, Park SM, Lee HK. Physical barrier effect of geopolymeric waste form on diffusivity of cesium and strontium. *J Hazard Mater.* 2016;318:339–46. <https://doi.org/10.1016/j.jhazmat.2016.07.003>.
88. Munthali MW, Johan E, Aono H, Matsue N. Cs^+ and Sr^{2+} adsorption selectivity of zeolites in relation to radioactive decontamination. *J Asian Ceram Soc.* 2015;3:245–50. <https://doi.org/10.1016/j.jascer.2015.04.002>.
89. El Alouani M, Saufi H, Moutaoukil G, et al. Application of geopolymers for treatment of water contaminated with organic and inorganic pollutants: State-of-the-art review. *J Environ Chem Eng.* 2021;9:105095. <https://doi.org/10.1016/j.jece.2021.105095>.
90. Hou D, Zhang J, Pan W, et al. Nanoscale mechanism of ions immobilized by the geopolymer: A molecular dynamics study. *J Nucl Mater.* 2020;528:151841. <https://doi.org/10.1016/j.jnucmat.2019.151841>.
91. Komljenović M, Tanasijević G, Džunuzović N, Provis JL. Immobilization of cesium with alkali-activated blast furnace slag. *J Hazard Mater.* 2020;388:121765. <https://doi.org/10.1016/j.jhazmat.2019.121765>.
92. Tian Q, Guo B, Sasaki K. Immobilization mechanism of Se oxyanions in geopolymer: effects of alkaline activators and calcined hydrotalcite additive. *J Hazard Mater.* 2020;387:121994. <https://doi.org/10.1016/j.jhazmat.2019.121994>.
93. Tian Q, Sasaki K. Effect of Additives on Immobilization of Se(VI) and Cr(VI) in Geopolymer Kuei Suan Jen Hsueh Pao. *J Chin Ceram Soc.* 2022;50:1335–46.
94. Kursten B, Smart NR, Senior NA, et al. Overview of anaerobic corrosion of carbon steel radioactive waste packages in alkaline media in support of the Belgian supercontainer concept. *Mater Corros.* 2021;72:32–51. <https://doi.org/10.1002/maco.202011794>.
95. Archez J, Formulations de composites à base de liants basse température type géopolymère à base d'argilite et de différents renforts : réalisation d'une pièce par fabrication additive. Thèse de doctorat, Limoges, 2020. <https://hal.science/tel-03115499v1>.
96. Gharzouni A, Contrôle de l'attaque des sources aluminosilicates par la compréhension des solutions alcalines. Thèse de doctorat, Limoges, 2016. <https://theses.hal.science/tel-01413354>
97. Archez J, Gharzouni A, Sobrados I, et al. Geopolymer local network evolution under time and temperature. *J Non-Cryst Solids.* 2021;566:120870. <https://doi.org/10.1016/j.jnoncrysol.2021.120870>.
98. Alves L, Leklou N, De Souza F, De Barros S. Assessment of the effect of fiber percentage in glass fiber reinforced slag-based geopolymer. *J Asian Ceram Soc.* 2021;9:1265–74. <https://doi.org/10.1080/21870764.2021.1966977>.

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.