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A Systematic Mapping Study of Geopolymers for Radioactive Waste Management

Jerome Ignatius T. Garces[a], April Anne S. Tigue[a], Michael Angelo B. Promentilla[b,*]

[a]Chemical Engineering Department, College of Engineering, De La Salle University, Manila 1004, Philippines
[b]Center for Engineering and Sustainable Development Research, De La Salle University, Manila 1004, Philippines
michael.promentilla@dlsu.edu.ph

Proper radioactive waste management is one of the many critical factors for a successful and safe nuclear power program. Technologies to contain such waste must be available to reduce the environmental burden attributed to the nuclear power plant and ensure public safety. Engineered or technical barriers for nuclear waste disposal have commonly employed cementitious materials for long-term containment in surface and underground geological radioactive waste repositories. One of the promising emerging materials that can be used as an alternative is the geopolymer. This is an emerging cementitious material with lower embodied energy and carbon footprint compared to conventional Portland cement-based materials. This study aims to assess the scope of the extent, range, and nature of research activity around geopolymer and radioactive waste management. The results revealed geopolymer can be an alternative material for immobilization and encapsulation of radioactive waste. Moreover, understanding the chemistry and the mechanism of how radioactive wastes are bounded to the matrix is necessary to aid in evaluating the long-term performance of such an alternative.

1. Introduction

One of the major obstacles being faced by nuclear power plants and the application of nuclear-based technologies, in general, is the issue of radioactive waste management. Radioactive waste is a type of hazardous waste that contains radioactive material. Common sources of radioactive wastes are nuclear fuel cycle, nuclear weapons reprocessing, medical wastes, industrial wastes, and naturally occurring radioactive materials. The nuclear fuel cycle produces most of these wastes (Khelurkar et al., 2015). These types of waste are produced from the front- and back-end of the nuclear energy production process. Front-end wastes are usually alpha-emitting wastes from the extraction of uranium, which includes uranium purification, conversion, enrichment, and fuel fabrication facilities (Liu et al., 2015). On the other hand, back-end wastes mostly contain spent fuel rods. On the other hand, nuclear weapon reprocessing wastes contain alpha-emitting actinides such as Pu-239 and very small amounts of beta or gamma-emitting Tritium and Americium. Medical wastes generally contain beta particles and gamma-ray emitters (Khelurkar et al., 2015). Naturally occurring radioactive materials include all radioactive elements found in the environment. Since radioactive materials lose their radioactivity over time, the most common way of handling radioactive wastes is to store them until they become harmless. However, different types of radioactive wastes require different storage times before they become harmless to people and the surrounding environment. Understanding the nature and the proper management technique in handling radioactive waste is one of the key aspects of promoting a safe and sustainable nuclear power program.

Some of the techniques used in radioactive management include storage in a deep geological repository, spent fuel pool, dry cask, and depleted uranium concrete. Commonly used to engineer this storage and repository are cementitious materials, typically employing Ordinary Portland Cement (OPC) concrete. The storage and management of radioactive wastes have made use of cement-based concrete as protective barriers for storage vaults of wastes as early as 1952 (Powell and Andrews, 1952). From then on, cementitious barriers have been proven to be sufficient for radioactive storage purposes due to cement's many favorable physical and chemical properties. The chemical properties of cement favor sorption and ion substitution which are necessary for binding and immobilizing radionuclides (IAEA, 2013). On the other hand, the physical properties of cement such

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as its durability and low permeability help in protecting the radioactive waste from leaching and contributes to its safe transportation and storage. Additionally, cementitious materials are stable when irradiated and can act as radiation shielding. These types of materials can shield radiations such as alpha rays, beta rays, gamma rays, X-rays, and neutrons owing to their high density and the presence of large amounts of crystalline water within them (Han et al., 2017). Additionally, cementitious barriers and shielding are easier to mold and are economical. However, there are environmental issues related to cement production such as its energy-intensiveness and extensive greenhouse gas emissions. Recent estimates have shown that cement production contributes to 8 % of the total volume of global CO_2 emissions (Andrew, 2019). As such, there are undergoing efforts to develop alternative materials which substitute partially or fully the use of ordinary Portland cement (OPC) for concrete production.
One of the most promising alternative materials for OPC is the geopolymer. It is an emerging class of material composed of tetrahedral silica and alumina and has properties that are comparable to ordinary Portland cement. It has exceptional and excellent properties in terms of waste immobilization and encapsulation. As proposed by Davidovits (1991), geopolymer has various applications based on Si/Al ratio such as for solidification and stabilization. Geopolymers are also being researched as an alternative to conventional cementitious barriers for encapsulating and immobilizing radioactive wastes. As such, to determine the status of the potential of geopolymers for radioactive waste management, a systematic mapping study is performed. This study aims to scope the extent, range, trend, and nature of research activity around geopolymer and radioactive waste management. This is achieved through the application of the systematic mapping study by Petersen et al. (2008) on the topic of radioactive waste management through geopolymers. To the researcher's knowledge, this study is the first instance of the application of systematic mapping method to analyze the trends and present status of this technology.

2. Materials and method

A systematic mapping study by Petersen et al. (2008) depicted in Figure 1 was employed in this study to evaluate the trend of geopolymer research in terms of radioactive waste management using Scopus as a search engine. Systematic mapping was chosen as the research method to explore the existing studies related to the trend of utilizing geopolymers in radioactive waste management. The results of the mapping study would help identify and map research areas that could serve as a reference for further investigations.

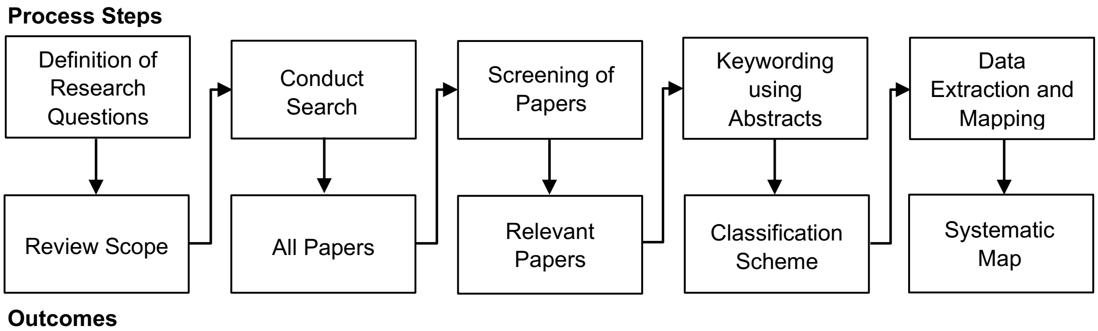

Figure 1: Systematic mapping process (Petersen et al., 2008)

2.1 Definition of research questions

The first step in a systematic mapping study is to define the research question. In this study, the concepts such as the trend, the level of waste, the type of waste, and the geopolymer precursor in the context of radioactive waste management were explored. The defined questions are tabulated in Table 1.

2.2 Conduct search

The second stage of mapping is to search for all relevant papers related to the study. To gather these papers, a search protocol was defined. The return search result that contains the defined keywords in the abstract published as of December 2021 was considered in this study. Boolean operators were used to limiting the return of results. This study is limited to the search results from the Scopus database only. The dataset for the identified questions was obtained using the search string ABS (geopolymer and ((nuclear waste) OR (radioactive waste))). The string "ABS" was used to limit the search only to the abstracts of the papers in the Scopus database.

2.3 Screening of papers

In screening the relevant papers, the return result of the conduct search method was used as the dataset. The papers were screened by reading the abstract, title, and keyword. With the aid of Mendeley software, the papers were inspected for the following: duplicated references, references without authors, and references addressing topics unrelated to the topic of interest.

2.4 Keywording

The relevant papers were classified by the established research questions and based on the keywords in the abstract.

2.5 Data extraction and mapping

Extracting the information relevant to address the research questions was performed. The returned search results based on the search method, screening of relevant papers, and keywording were quantified to have an overview of the research trend. For the context of radioactive waste management and geopolymer research, relevant information such as the trend, the level of waste, the type of radionuclide, and the geopolymer precursor reported in the study was assessed.

Table 1: Research questions on the use of geopolymer for radioactive waste management

Research Questions (RQ)	
1	What is the research trend on the use of geopolymer for radioactive waste management?
2	What levels of radioactive wastes are treated with geopolymer?
3	What types of radionuclides are treated with geopolymer?
4	What geopolymer precursor materials are explored for radioactive waste management?

3. Results and discussion

3.1 RQ1: What is the research trend on the use of geopolymer for radioactive waste management?

The defined search protocol has returned 130 papers search results as preview shown in Figure 2. Upon screening of papers by title and abstract, 78 out of the 130 publications were found to be relevant. The papers were then assessed based on the level of waste, type of radionuclide immobilized, and the geopolymer precursor considered in various studies and was discussed in the subsequent research questions below.

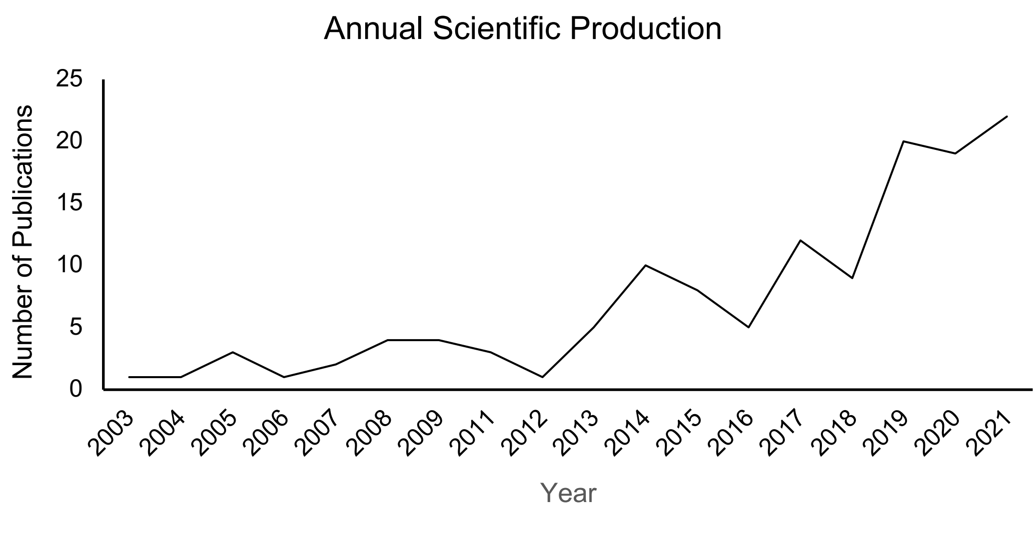

Figure 2: Trend on using geopolymer in radioactive waste management

3.2 RQ2: What level of radioactive wastes are currently treated with geopolymer?

Radioactive wastes can be classified into five namely exempt waste, very short-lived waste, very low-level waste, low-level waste, intermediate level waste, and high-level waste. Most of the wastes reported to be treated with geopolymer are under low-intermediate-high level waste.

Low-level wastes are usually generated from hospitals and industry, as well as the nuclear fuel cycle. This accounts for a much larger volume but a few percent of radioactivity (Vance and Perera, 2011). It usually does not require a shield during handling and transport and is suitable for shallow land burial. Compaction and incineration are usually performed to reduce its volume. It ranges from a level just above very low-level waste to a level upon which shielding is necessary for periods up to several hundred years. On the other hand, wastes containing long-lived radionuclides and in quantities that need more isolation from the environment and require shielding are classified under intermediate. Disposal of intermediate-level waste is carried out at a depth ranging from a few tens to hundreds of meters. Intermediate level waste contributes to 7 % of the total volume of radioactive wastes, and 4 % of the total volume of the radioactivity. Lastly, high-level waste volume accounts for only 3 % of the total radioactive waste but is responsible for 95 % of the radioactivity. It is usually from the uranium fuel and other elements present in the core of a nuclear reactor core.

The use of cement to contain these types of wastes has been the traditional radioactive waste management technique for years. The use of geopolymer as an alternative has been promising due to its excellent properties in terms of immobilizing and encapsulating radioactive wastes as revealed by various studies. For instance, a study conducted by Perera et al. (2003) showed that geopolymers can be used to immobilize low to intermediate levels of radioactive waste. More so, Vance et al. (2006) revealed that geopolymer is a potential material that can immobilize uranium-rich waste which is under the high-level waste category.

3.3 RQ3: What are the types of radionuclides currently treated with geopolymer?

Based on the scoping review, the most widely studied radionuclide in terms of radioactive waste and geopolymer research is Cesium and Strontium. The chemistry of cesium and the ordinary Portland cement in immobilization has been known to be problematic as Cesium tends not to bond strongly with the alkali component of the cement. Consequently, the use of geopolymer in the immobilization of cesium has gained a lot of interest. Various studies have successfully immobilized cesium in the geopolymer matrix. Characterization of cesium immobilized in geopolymer matrix showed the possibility of it being chemically bonded to geopolymer structure (Perera et al., 2003). A study by Berger et al. (2009) noted that the efficiency of Cesium being incorporated in the matrix heavily depends on the composition of activating solution, mainly the silica content and the alkali used. Another study revealed that cesium immobilization using geopolymer comparatively performs better than ordinary Portland cement (Li et al., 2013). The result of the study revealed that lower concentration has been leached out with geopolymer as an encapsulation matrix. The successful immobilization of cesium in the matrix of geopolymer may be attributed to it being chemically bounded as a charge balancing ion in the matrix which behaves similarly to sodium or potassium (Provis, 2009).

While the behavior and chemistry of strontium and ordinary Portland cement have already been established to be well immobilized, the use of geopolymer as an alternative for a low-cost immobilization technique is explored. Tan et al. (2019) reported that the geopolymer-based matrix has immobilized strontium better than that of an ordinary Portland-based matrix. The strontium is encapsulated in the amorphous component of both matrices. In aggressive conditions, particularly in sulfuric acid, magnesium sulfuric, and acetic acid buffer solutions, better leaching resistance has been observed for samples encapsulated with a geopolymer matrix. This may be attributed to the strontium being physically sealed in the geopolymeric gel. Moreover, Xu et al. (2017) reported that strontium partly replaces the sodium ions in the matrices of geopolymer which is similar to the behavior of cesium that has been observed. A similar finding has been revealed by Walkley et al. (2020) in which the strontium displaces some of the sodium and potassium ions from the charge balancing sites.

3.4 RQ4: What are the raw materials being explored as a geopolymer precursor for radioactive waste management?

Several studies have dealt with metakaolin and fly ash as the main precursor in developing geopolymer for radioactive waste management. Metakaolin is composed of alternating layers of silicate and aluminate which is derived from kaolin clay calcination at a temperature between 600 - 800 °C. It is also one of the most widely studied precursors for geopolymerization. Some countries have successfully used metakaolin-based geopolymer in encapsulating radionuclide on an industrial scale as it decreases the rate of diffusion of radionuclide samples (Vehmas et al., 2020). The study also revealed that some of the concentrations of leachates were measured to be below the detection limit. Several studies have demonstrated success in encapsulation radionuclides. For instance, Ofer-Rozovsky et al. (2019) have explored the use of nitrate-bearing metakaolin as a geopolymer precursor to encapsulate cesium. The results showed that the formulation of precursor can be correlated with the enhanced immobilization of Cesium. He et al. (2020) evaluated the use of Na-based and K-based metakaolin geopolymer in radioactive immobilization, specifically Cs and Sr. The study reported that it has exhibited better performance than that ordinary Portland cement. The chemistry of metakaolin which contains a significant amount of Si and Al and has a highly disordered structure makes it interesting to be a viable source for geopolymerization. When mixed with an alkali activator, a sodium aluminosilicate hydrate gel is formed with the presence of alkali cations as charge-balancing ions.

Another precursor which gains a lot of interest is fly ash which is a by-product of thermal power plants that contains a significant amount of Si and Al. Tian et al. (2019) reported that geopolymer synthesized with fly ash has considerably reduced the leaching of Strontium and Cesium after solidification. Similar findings were observed by Luo et al. (2017) in which the recorded leaching rate was far below the national standard limit. Moreover, it was also revealed that the solidified geopolymer containing Strontium and Cesium with fly ash as a precursor has an exceptional curing effect.

The properties of aluminosilicate sources and the mix formulation in developing geopolymer are among the key factors that greatly affect the immobilization and encapsulation efficiency. As such, investigating the properties of locally available materials which can be tailored for the management of radioactive waste can aid countries around the world in improving the sustainability of their nuclear power programs.

4. Conclusions

The initial mapping review using the search engine Scopus on published papers showed that there is an increase in interest in utilizing geopolymers in radioactive waste management. The results from this mapping analysis suggest the potential of geopolymer research in radioactive waste immobilization and a need for more research on understanding the long-term durability of such cementitious barriers including their self-healing capability. Moreover, knowing the mechanism and related chemistry behavior of radionuclides in the geopolymer matrix system will aid the researchers in better understanding the potential of this technology in nuclear waste management. Future iterations of this mapping study can widen its scope to include traditional cementitious barriers made from OPC and compare their reported performances against those for geopolymers. The steady build-up of interest on this topic can potentially lead to increased research agenda on alternative cementitious engineered barriers using geopolymers for radioactive waste management. Additionally, the results of this mapping study can potentially guide interested researchers on which areas they can focus on to systematically contribute towards the development of the technology.

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References

Andrew R. M., 2019, Global CO_2 emissions from cement production, 1928-2018, Earth System Science Data, 11, 1675–1710.

Berger S., Frizon F., Joussot-Dubien C., 2009, Formulation of caesium based and caesium containing geopolymers, Advances in Applied Ceramics, 108(7), 412–417.

Davidovits J., 1991, Geopolymers - inorganic polymeric new materials, Journal of Thermal Analysis 37(8), 1633–56.

Han B., Zhang L., Ou J., 2017, Chapter 19 Radiation shielding concrete, In Smart and Multifunctional Concrete Toward Sustainable Infrastructures, Springer Singapore, 329–337.

He P., Cui J., Wang M., Fu S., Yang H., Sun C., Duan X., Yang Z., Jia D., Zhou Y., 2020, Interplay between storage temperature, medium and leaching kinetics of hazardous wastes in metakaolin-based geopolymer, Journal of Hazardous Materials, 384, 121377.

IAEA, 2013, The behaviours of cementitious materials in long term storage and disposal of radioactive waste results of a coordinated research project <https://www-pub.iaea.org/MTCD/Publications/PDF/TE-1701_web.pdf> accessed 01.12.2020.

Khelurkar N., Shah S., Jeswani H., 2015, A review of radioactive waste management, 2015 International Conference on Technologies for Sustainable Development (ICTSD), September, 1–6.

Li Q., Sun Z., Tao D., Xu Y., Li P., Cui H., Zhai J., 2013, Immobilization of simulated radionuclide $133Cs^+$ by fly ash-based geopolymer, Journal of Hazardous Materials, 262, 325–31.

Liu X., Wei F., Xu C., Liao Y., Jiang J., 2015, Characteristics and classification of solid radioactive waste from the front-end of the uranium fuel cycle, Health Physics, 109(3), 183–186.

Ofer-Rozovsky E, Arbel Haddad M., Bar-Nes G., Borojovich E.J.C., Binyamini A., Nikolski A., Katz A., 2019, Cesium immobilization in nitrate-bearing metakaolin-based geopolymers, Journal of Nuclear Materials, 514, 247–54.

Perera D. S., Vance E. R., Aly Z., Finnie K. S., Hanna J. V., Nicholson C. L., Trautman R. L., Stewart M. W. A., 2003, Characterisation of geopolymers for the immobilisation of intermediate-level waste, Proceedings of the International Conference on Radioactive Waste Management and Environmental Remediation, ICEM, 3, 1807–1814.

Petersen K., Feldt R., Mujtaba S., Mattsson M., 2008, Systematic mapping studies in software engineering, 12th International Conference on Evaluation and Assessment in Software Engineering, EASE.

Powell C. C., Andrews H. L., 1952, Radioactive waste disposal, Public Health Reports (1896-1970), 67(12), 1214.

Provis J. L., 2009, Immobilisation of toxic wastes in geopolymers, In Geopolymers: Structures, Processing, Properties and Industrial Applications, 421–440.

Tan Q., Li N., Xu Z., Chen X., Peng X., Shuai Q., Yao Z., 2019, Comparative performance of cement and metakaolin based-geopolymer blocks for strontium immobilization, Journal of the Ceramic Society of Japan, 44–49.

Vance E. R., Perera D. S., 2011, Development of geopolymers for nuclear waste immobilisation, In Handbook of Advanced Radioactive Waste Conditioning Technologies, Elsevier, 207–229.

Vance Eric R., Perera D. S., Aly Z., 2006, Feasibility of immobilizing tank wastes in geopolymers, In Ceramic Transactions, 176.

Vehmas T., Myllykylä E., Nieminen M., Laatikainen-Luntama J., Leivo M., Olin M., 2020, Geopolymerisation of gasified on-exchange resins, mechanical properties and short-term leaching studies, In IOP Conference Sieries: Materials Science and Engineering, Institute of Physics Publishing.

Walkley B., Ke X., Hussein O. H., Bernal S. A., Provis J. L., 2020, Incorporation of strontium and calcium in geopolymer gels, Journal of Hazardous Materials, 382, 121015.

Xu Z., Jiang Z., Wu D., Peng X., Xu Y., Li N., Qi Y., Li P., 2017, Immobilization of strontium-loaded zeolite A by metakaolin based-geopolymer, Ceramics International, 43(5), 4434–4439.