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Editorial

Material Aspect of Sustainable Nuclear Waste Management

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Citation: Petrov, V.A.; Ojovan, M.I.; Yudintsev, S.V. Material Aspect of Sustainable Nuclear Waste Management. *Sustainability* **2023**, *15*, 11934. https://doi.org/10.3390/su151511934

Received: 21 July 2023
Accepted: 1 August 2023
Published: 3 August 2023

1. Introduction

Effectively managing nuclear waste is crucial to ensure the safe sustainable usage of nuclear energy, which ranges from large-scale applications in power generation to numerous smaller-scale applications in medicine, industry, and agriculture, and scientific research is needed at the current state of development. The waste, which is generated as a by-product of nuclear energy use, is termed nuclear or radioactive waste since it contains levels of hazardous radionuclides that are above the clearance/exemption levels set by national authorities in each country. Depending on the radionuclide contents, the nuclear waste is classified as very-low-level, low-level, intermediate-level, or high-level waste (VLLW, LLW, ILW, and HLW, correspondingly). The lifecycle of nuclear waste management (NWM) contains the following stages: pretreatment, treatment, conditioning, and disposal. The characterization of these stages is mandatory and often crucial, and transportation is also a necessary linkage between the different players of NWM. Nuclear waste is first treated and conditioned for its safe handling, transportation, storage, and ultimate placement into a disposal facility. Finally, the conditioned nuclear waste is disposed at the back end of its lifecycle with the aim of permanently protecting humans and the biosphere from dangerous radioactive materials [1].

The largest source of nuclear waste by volume is the nuclear fuel cycle (NFC), which has nuclear power plants that produce electricity at its core. Figure 1 schematically shows the NFC activities and associated nuclear waste streams, emphasizing that hazardous radioactive materials (uranium ores) are initially mined from deep crystalline or shallow sedimentary formations; these materials, along with newly formed radiotoxic and artificial nuclides, are finally taken to geological disposal facilities (GDF) that are located deep underground within geological formations (crystalline rocks, salt, clays, etc.) that have been stable for millions of years (see, e.g., [2]).

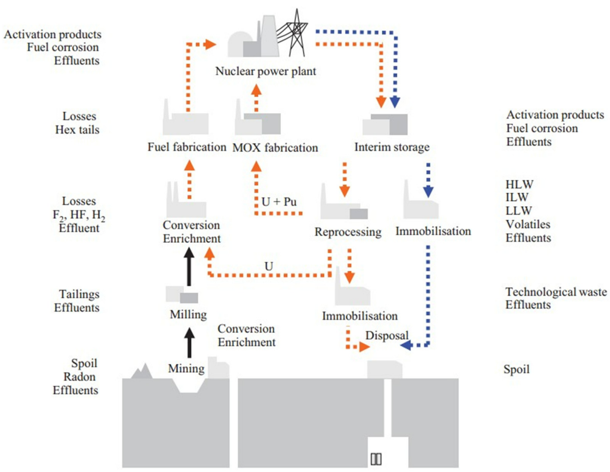

Figure 1. Closed (shown in orange) and open NFC activities with associated nuclear wastes (see [2], publicly available).

2. Materials for Nuclear Waste Management

The safety of nuclear waste relies on the materials that are used, and this aspect is especially important in its disposal. Nuclear waste processing, storage, and disposal activities encompass the utilization of advanced technologies and materials, aiming to ensure the reliability of long-term waste isolation. Considering the material aspect is hence one of the keys to ensure sustainable and safe nuclear waste management. Cements, geopolymers, glasses, glass composite materials, ceramics, and metals are generically the basic materials that are used and systematically analyzed for their expected long-term performance in the disposal environment. The selection of materials within the nuclear waste management problem is a complex task accounting for technology availability and waste characteristics, and typically involves a compromise between various constraints including flexibility, ease of processing, waste loading, characteristics of the waste, required lifetime performance affected by self-irradiation from decaying radionuclides, and water alteration accounting for the mineralogical–geochemical aspects [3–6]. In addition to laboratory tests, the available data on natural analogue materials that have been proven for their long-term stability and durability are used to ensure confidence in the multi-scale approaches that are currently used to predict the behavior of waste disposal systems on geological timescales. The generic rule is that the higher the nuclear waste hazard, the more durable materials are being used to immobilize and ultimately dispose of the radionuclides (Figure 2).

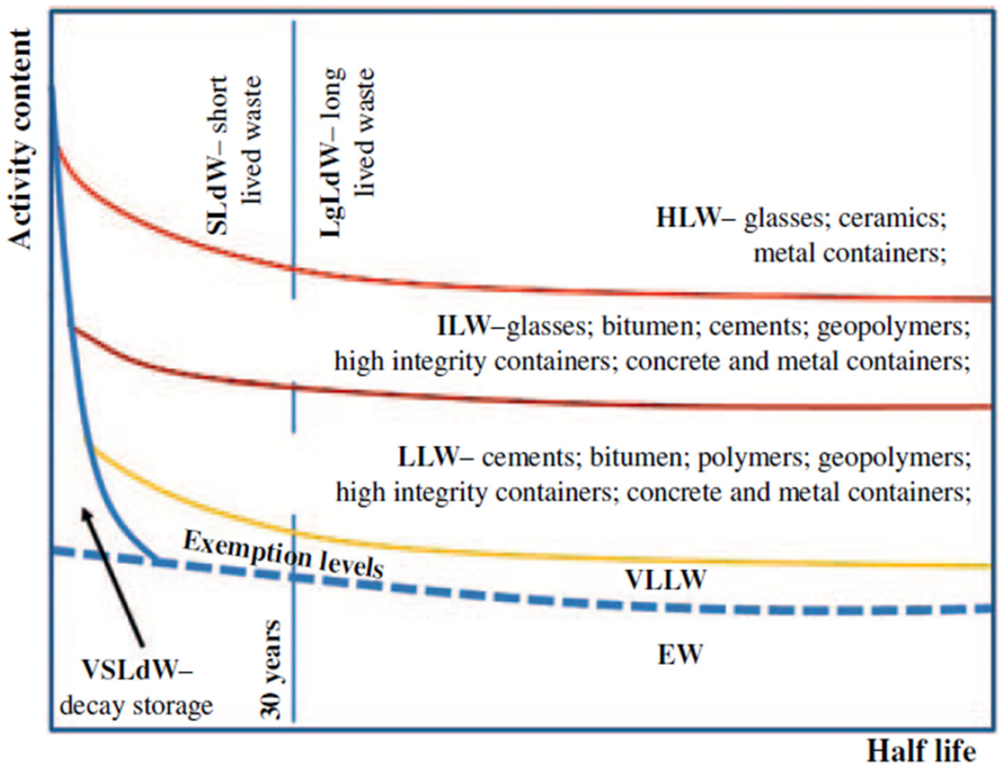

Figure 2. Materials and disposal routes of nuclear waste following IAEA classification scheme. Reproduced from [3] with permission from Elsevier. EW—waste exempt of regulatory control.

At the core of the multibarrier system preventing radionuclide migration into the biosphere is the wasteform [4]—the product of immobilizing waste into a matrix (Figure 3).

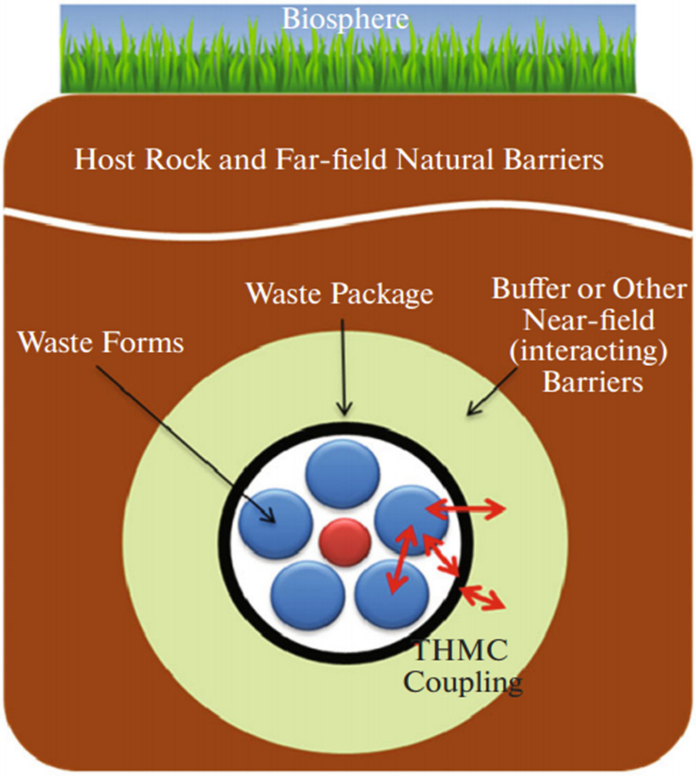

Figure 3. Schematic representation a of multibarrier system (see [4], publicly available). THMC—coupling of thermo–hydro–mechanical and chemical processes.

Immobilization is defined as the conversion of waste into a wasteform via solidification, embedding, or encapsulation aiming to facilitate handling, transportation, storage, and disposal. The immobilization of waste is achieved by its chemical incorporation into the structure of a suitable matrix (e.g., cement, glass, or ceramic) so it is captured and unable to escape. The management of HLW is the most challenging because the times required for radionuclides to decay to background levels are of geological timescales. Chemical immobilization (binding of radionuclides at atomic/molecular level) is typically applied for HLW, while encapsulation is used for ILW, and the immobilization of LLW is achieved by physically surrounding the waste constituents with materials such as bitumen or cement so it is isolated and so the radionuclides are retained. Physical encapsulation is often applied to ILW and can also be used for HLW, especially where the chemical incorporation of radionuclides in the surrounding matrix is also possible. Choosing the wasteforms and packages for higher activity radioactive wastes such as ILW and HLW is challenging, as the crucial decisions largely depend on the nature of the waste streams and are based on the proven technologies used worldwide [4–6].

VLLW does not require immobilization, although some forming packaging is used to assist with handling and transportation. LLW is typically immobilized using cement, although more durable materials such as bitumen or glass can provide a much better retention of contaminants and reduce the final waste disposal volume. Since ILW is more hazardous than LLW, it can be immobilized using glasses, bitumen, cements, and geopolymers. LLW and ILW can be packed in high-integrity metal and concrete containers. The most hazardous waste is HLW, typically resulting from spent fuel reprocessing, which requires the most durable and stable wasteforms. These are represented by durable crystalline ceramics that are currently under the deployment stage, and by glasses that have been used industrially for many decades. The vitrified HLW is typically encased in stainless steel canisters, while corrosion-resistant copper containers are used to dispose of spent nuclear fuel. Table 1 indicates the various nuclear waste classes from the papers that are published within this Special Issue (i.e., A, B, C. . .P are chapters of this book).

Table 1. Relevant nuclear waste classes within publications of current *Sustainability* Special Issue.

Chapter	Title of Contribution, Web Reference	Waste Class
A	"Material Aspect of Sustainable Nuclear Waste Management" (Editorial)	All
B	"Removal of Cs-137 from Liquid Alkaline High-Level Radwaste Simulate Solution by Sorbents of Various Classes" (Article), https://www.mdpi.com/2071-1050/15/11/8734 (accessed on 20 July 2023).	All
C	"Long-Term Chemical Alteration of ^{238}Pu-Doped Borosilicate Glass in a Simulated Geological Environment with Bentonite Buffer" (Article), https://www.mdpi.com/2071-1050/15/7/6306 (accessed on 20 July 2023).	HLW
D	"An Introduction to Nuclear Industrial Archaeology" (Article), https://www.mdpi.com/2071-1050/15/7/6178 (accessed on 20 July 2023).	All
E	"Influence of Radioactive Sludge Content on Vitrification of High-Level Liquid Waste" (Article), https://www.mdpi.com/2071-1050/15/6/4937 (accessed on 20 July 2023).	HLW
F	"Toward Deep Decontamination of Intermediate-Level-Activity Spent Ion-Exchange Resins Containing Poorly Soluble Inorganic Deposits" (Article), https://www.mdpi.com/2071-1050/15/5/3990 (accessed on 20 July 2023).	ILW
G	"Influence of Rock Structure on Migration of Radioactive Colloids from an Underground Repository of High-Level Radioactive Waste" (Article), https://www.mdpi.com/2071-1050/15/1/882 (accessed on 20 July 2023).	HLW
H	"Evaluation of a Long-Term Thermal Load on the Sealing Characteristics of Potential Sediments for a Deep Radioactive Waste Disposal" (Article), https://www.mdpi.com/2071-1050/14/21/14004 (accessed on 20 July 2023).	HLW
I	"Natural Clay Minerals as a Starting Material for Matrices for the Immobilization of Radioactive Waste from Pyrochemical Processing of SNF" (Article), https://www.mdpi.com/2071-1050/13/19/10780 (accessed on 20 July 2023).	HLW, ILW
J	"The Influence of Liquid Low-Radioactive Waste Repositories on the Mineral Composition of Surrounding Soils" (Article), https://www.mdpi.com/2071-1050/12/19/8259 (accessed on 20 July 2023).	LLW
K	"Calculation of Potential Radiation Doses Associated with Predisposal Management of Dismantled Steam Generators from Nuclear Power Plants" (Article), https://www.mdpi.com/2071-1050/12/12/5149 (accessed on 20 July 2023).	All
L	"Effect of Gamma Irradiation on Structural Features and Dissolution of Nuclear Waste Na–Al–P Glasses in Water" (Article), https://www.mdpi.com/2071-1050/12/10/4137 (accessed on 20 July 2023).	HLW
M	"On the Sustainable Utilization of Geopolymers for Safe Management of Radioactive Waste: A Review" (Review), https://www.mdpi.com/2071-1050/15/2/1117 (accessed on 20 July 2023).	LLW, ILW
N	"Recent Advances in Alternative Cementitious Materials for Nuclear Waste Immobilization: A Review" (Review), https://www.mdpi.com/2071-1050/15/1/689 (accessed on 20 July 2023).	LLW, ILW
O	"Toward Sustainable Cementitious Radioactive Waste Forms: Immobilization of Problematic Operational Wastes" (Review), https://www.mdpi.com/2071-1050/13/21/11992 (accessed on 20 July 2023).	LLW, ILW
P	"Glass Crystalline Materials as Advanced Nuclear Wasteforms" (Review), https://www.mdpi.com/2071-1050/13/8/4117 (accessed on 20 July 2023).	HLW, ILW

Cements are widely used within NWM starting from its inception stage and are used at an incomparable larger scale compared to other materials [7–9]. The cementation of both solid and liquid nuclear waste has become an important and developing part of the waste management system owing to its simplicity and versatility. Cements are inorganic materials that set and harden because of hydration reactions between its constituents and water, forming a composite material containing both crystalline (i.e., portlandite, ettringite, monosulfate, hydrogarnet) and amorphous (the tobermorite gel termed CSH phase) constituents. Geopolymers are used to a lesser extent in NWM. They are materials often termed alkali-activated cements and are made by mixing a reactive source of alumina and silica, such as fly ash, or by mixing metakaolin with an activator, typically in the form of concentrated aqueous solutions of NaOH or KOH, resulting in the formation of a solid material comprising a hydrous aluminosilicate gel, which binds much of the added alkali [7]. Cements and geopolymers allow for the microencapsulation of waste, although some physicochemical binding of waste cations can also occur. Modern cementitious wasteforms include Portland-based cements, calcium aluminate, calcium sulfoaluminate, phosphate, ceramicrete (hydrous potassium magnesium phosphate), magnesium silicate, and alkali-activated cement (also termed geopolymers). The overviews (N, O, P) provide data on the advances within the uses of cements and geopolymers including their application to so-called problematic waste. The continued development of the cementation technique is driven by the improvement and expansion of cementitious materials that are suitable and efficient for NWM, with advances that have significantly improved nuclear waste cementation technology and the quality of cementitious nuclear wasteforms.

Crystalline ceramics containing one or more crystalline phases are the most durable and reliable host materials for the retention of long-lived radionuclides [10–12]. Single-phase crystalline ceramics can be used to immobilize separated radionuclides such as weapon grade ^{239}Pu, minor actinides after nuclear fuel reprocessing, as well as more chemically complex waste streams including HLW from fuel reprocessing. The atomic structure of the ceramic phase must have multiple cations and sites that can accommodate the variety of radionuclides in the waste stream; therefore, multiphase crystalline ceramics are preferred for complex chemical compositions of waste. Chapter (I) investigates the use of ceramic materials obtained from natural bentonite clay as immobilizing matrices for radioactive waste in the form of a LiCl–KCl eutectic resulting from the pyrochemical reprocessing of used nuclear fuel.

Glasses are amorphous solid materials below the glass transition temperature T_g, which are typically produced via vitrification (liquid–glass transition) by quickly cooling them from a molten to a solid state to avoid crystallization [13]. Durable glasses of borosilicate and alumina phosphate families are the worldwide choice for HLW immobilization [3–6,10,14]. The vitrification of LLW and ILW is also deployed by including legacy and nuclear power plant operational waste [3,10,14]. Chapter (E) provides information on the HLW vitrification program in China, while Chapter (L) provides information on the radiation effects in glasses used in Russia. The results presented in Chapter (C) provide evidence on the enhanced retention properties of glasses in natural waters compared to the deionized water of testing protocols, whereas Chapter (P) supports the recent trend of using inhomogeneous glass crystalline materials (GCMs), which provide superior waste loading and durability compared to homogeneous vitreous materials, as illustrated by Figure 4.

Homogeneous vitreous materials are shown in the lower left corner of the triangle. If durable crystals such as spinel are allowed to form in the glass, then GCMs (left leg of the triangle) that have superior waste loading and durability relative to homogeneous glass can be produced. Fully crystalline materials (ceramics) are shown in the upper corner of the triangle and are the most durable wasteforms with high waste loadings. However, the incorporation of certain species (e.g., Mo, Cr, S, and Cl) into glass creates non-durable secondary phases (lower right corner or triangle) that may have unacceptable durability.

The multibarrier system preventing radionuclide migration into the biosphere includes natural barriers, which are key factors in the case of HLW disposal, accounting for the geological timescales needed to retain the long-lived radionuclides (see Figure 3). The disposal is always the end point of NWM, although it is its integral part [3,15,16]. The natural barrier materials were analyzed with a focus on the rock structure and migration of radioactive colloids in Chapter (G), the chemical and thermal impact of nuclear waste on the minerals in the repository near field in Chapter (J), and the effect of the HLW thermal load on the sealing characteristics of surrounding geological formations in Chapter (H).

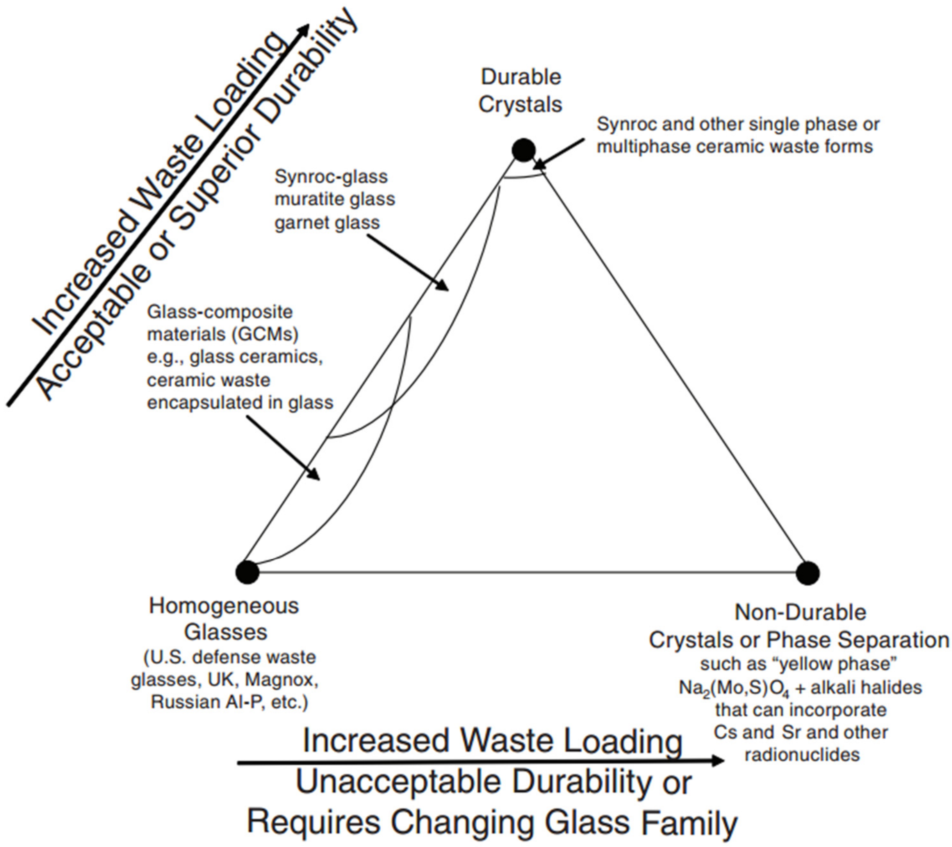

Figure 4. Schematic diagram illustrating the durability and waste loading of GCM wasteforms relative to homogeneous glass. Reproduced from [6], publicly available.

The technological aspect of NWM at both its predisposal and disposal stages [3] is analyzed within Chapter (K), focusing on the potential radiation doses associated with predisposal activities; Chapter (B) provides details on the removal of Cs-137 from alkaline HLW solutions; and Chapter (O) describes the decontamination of ILW in the form of spent ion-exchange resins.

Chapter (N) of this Special Issue introduces a novel discipline—nuclear industrial archaeology—which specializes in finding and characterizing abandoned nuclear sites.

Altogether, the papers published within this Special Issue (Table 1) present evidence on the progress achieved within NWM, aiming to support the sustainable and safe utilization of nuclear energy.

3. Conclusions

The *Sustainability* Special Issue "Nuclear Waste Management and Sustainability" provides a substantial addition to the literature on nuclear waste management, focusing on the material aspect and the technologies used.

Author Contributions: Conceptualization, V.A.P., M.I.O. and S.V.Y.; methodology, V.A.P., M.I.O. and S.V.Y.; validation, V.A.P., M.I.O. and S.V.Y.; formal analysis, V.A.P., M.I.O. and S.V.Y.; investigation, V.A.P., M.I.O. and S.V.Y.; resources, V.A.P.; data curation, V.A.P., M.I.O. and S.V.Y.; writing—original draft preparation, M.I.O.; writing—review and editing, V.A.P., M.I.O. and S.V.Y.; visualization, V.A.P., M.I.O. and S.V.Y.; supervision, V.A.P. and S.V.Y.; project administration, V.A.P. and S.V.Y.; funding acquisition, V.A.P. All authors have read and agreed to the published version of the manuscript.

Funding: This work was carried out within the framework of a state assignment for the Institute of Geology of Ore Deposits, Petrography, Mineralogy, and Geochemistry at the Russian Academy of Sciences.

Acknowledgments: The editors are grateful to all contributing authors.

Conflicts of Interest: The authors declare no conflict of interest.

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