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Editorial

Challenges in the Long-Term Behaviour of Highly Radioactive Materials

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Highly radioactive materials are at the core in many useful applications ranging from operating nuclear reactors (including fast breeder reactors) to vitrified high-level radioactive waste, which is currently stored and awaiting final disposal into dedicated facilities within deep geological formations. The stability and durability of highly radioactive materials are greatly affected by both continuous irradiation and adverse action of the environment. The crucial question in all useful applications stands, therefore, with the behaviour of materials in the conditions of intense irradiation combined with adverse and often highly corrosive environments. The effect of self-irradiation has especially emerged in nuclear waste immobilisation with the importance of predictable long-term behaviour of materials which extends for time periods, exceeding many hundred and thousand years. Indeed, the nuclear waste shall withstand immobilised in a geological repository for many millennia if not much longer, depending on the content of long-lived radionuclides [1,2]. Even small changes in material performance, which are negligible from a short-term standpoint, can gradually lead to structural and functional changes and consequently cause materials failure in the long-term perspective. The accidents in nuclear installations, including that at Three Mile Island, Chernobyl, and Fukushima, have focused the attention of researchers on the highly radioactive materials in the form of nuclear fuel debris and hot particles generated within or after the accident [3,4]. Moreover, the analysis of these materials can indicate the nature of processes that have caused the accident [4]. Irreversible transformations, such as swelling and phase separation, acceleration of material ageing, and corrosion, have already been reported for highly radioactive crystalline and vitreous materials, which are of practical importance, e.g., within the safety assessment [5–8]. One of the aspects herewith is the upper limit of wasteform loading with radioactive species, where an incentive is to increase it without, however, compromising the performance during the storage period and within the disposal environment. The IAEA has recently launched a dedicated coordinated research project INWARD to combine efforts of researchers to utilise accelerators aiming to simulate and analyse the effect of the radiation of materials [9].

Although radiation effects have been comprehensively overviewed in two fundamental publications, both for crystalline [5] and glassy [6] materials, some updates published more recently indicate additional unexpected effects, i.e., the so-called "unknown unknowns" within the science of highly radioactive materials, see, e.g., [8,10,11]. A typical limitation of the content of radionuclides in a durable matrix material is related to the content of the fissile element in the wasteform aiming to avoid any potential criticality in the nuclear waste facility both currently and in the future, accounting for any potential scenario of events in a repository or a disposal facility. The content of both fissile and non-fissile radionuclides is also limited by the detrimental effects caused by radiation damage. For crystalline materials, this is typically related to the amorphisation of materials which results in material swelling, mechanical damage, and, overall, leads to a loss of radionuclide retention performance. However, long-term experiments with materials that

Citation: Ojovan, M.I. Challenges in the Long-Term Behaviour of Highly Radioactive Materials. *Sustainability* **2022**, *14*, 2445. https://doi.org/10.3390/su14042445

Received: 14 February 2022
Accepted: 18 February 2022
Published: 21 February 2022

contain radionuclides indicated that detrimental effects are evidently revealed only after quite long time periods, which can last from years to decades, and that the mechanisms of the slowly occurring processes in highly radioactive materials are not fully understood [11]. An example of such a type of unexpected effect is demonstrated in Figure 1.

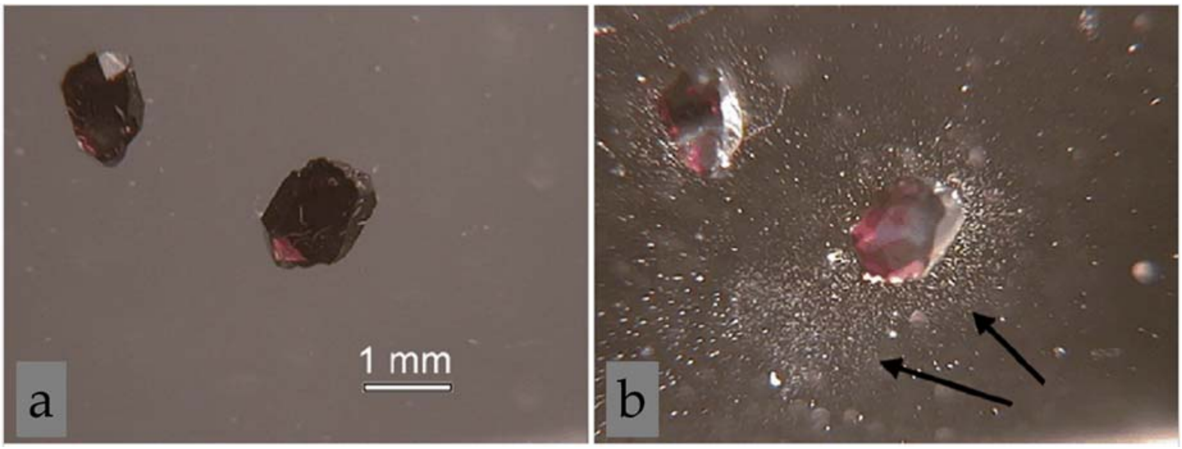

Figure 1. Example of experimentally observed radiation-induced destruction with generation (ejection) of hot particles by synthetic radioactive (4.9 wt.% of ^{238}Pu) monazite crystals: (**a**)—The radioactive crystals after synthesis; (**b**)—The radioactive crystals after 14 months of storage along with dispersed tiny particles around them indicated by black arrows. Reproduced with permission after [11]. Copyright 2010 Elsevier.

Oxide glasses also demonstrated an unexpected behaviour in intensive radiation fields: radiation-induced fluidisation (or quasi-melting [12,13]) when the temperature remained practically unchanged, although the viscosity dropped to that of a liquid (Figure 2).

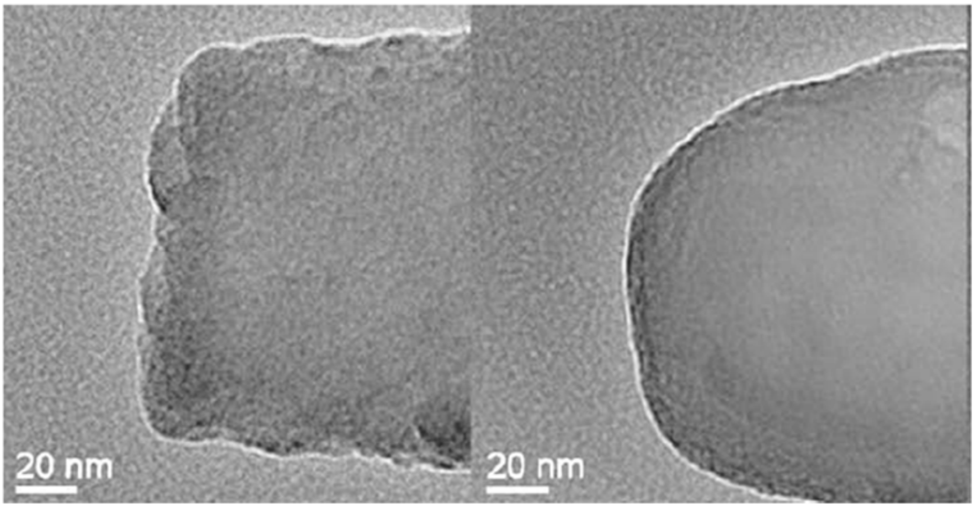

Figure 2. Fluidisation (quasi-melting) of glasses inside of the electron beam of a transmission electron microscope: transformation of a glass fibre end from the solid state to the liquid state. Reproduced with permission after [13]. Copyright 2018 Elsevier.

Fludisation is associated with a drastic decrease in the glass transition temperature at a high enough intensity of radiation [12,13] and was independently confirmed by another research group as well [14].

The behaviour of highly radioactive materials presents, therefore, a challenge to science and technology, particularly in the aspect of durability and long-term stability of materials. Henceforth, this Special Issue is an attempt to invite researchers to analyse the challenging aspects of highly radioactive materials' behaviour in systems and activities related to the peaceful utilisation of nuclear energy. It aims to support environmentally safe and sustainable utilisation of nuclear energy, assessing effects resulting from self-irradiation on crystalline, vitreous, and glass crystalline materials used in nuclear applications, including nuclear reactor and waste immobilisation materials. Although this Special Issue has only published three papers [15–17], it has emphasised the problem and will attract the attention of experts in the future.

Funding: This research received no external funding.

Data Availability Statement: Not applicable.

Acknowledgments: The support by co-editors of this Special Issue B.E. Burakov and B.Yu. Zubekhina is highly appreciated.

Conflicts of Interest: The author declares no conflict of interest.

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