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SPECIALTY SECTION

This article was submitted to Radioactive Waste Management, a section of the journal Frontiers in Nuclear Engineering

RECEIVED 23 September 2022 ACCEPTED 24 October 2022 PUBLISHED 17 November 2022

CITATION

Grambow B (2022), Mini review of research requirements for radioactive waste management including disposal. *Front. Nucl. Eng.* 1:1052428. doi: 10.3389/fnuen.2022.1052428

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Mini review of research requirements for radioactive waste management including disposal

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This brief overview of research needs related to radioactive waste management and disposal is intended to provide a framework for addressing some important research questions related to historical developments and some large industrial mega-projects for geological disposal of radioactive waste that are currently being developed. Considering that the level of industrial development varies considerably between countries, research in this field of knowledge has a strong international and long-term orientation, encompassing both basic and applied research, including optimisation.

KEYWORDS

Radioactive Waste, disposal program, Management, Research, Science, Technology, optimisation, long term safety

History, current state and future challenges in radioactive waste management including disposal

Radioactive material from nuclear research, the nuclear arms race and nuclear energy production was buried in trenches in the United States under the Manhattan Programme before 1945 and dumped in the sea in worldwide programmes from 1946 until it was completely banned in 1994 (IAEA, 1999). In the meantime, waste volumes continued to rise. Spent nuclear fuel has accumulated around the world. The total inventory of non-reprocessed fuel has reached some 250,000 metric tonnes at hundreds of different sites. In addition some 120,000 metric tonnes have been reprocessed1 (IAEA, 2018), creating large volumes of high-level radioactive waste.

Worldwide research on radioactive waste disposal in geological formations has been initiated by the Report "The Disposal of Radioactive Wastes on Land" of the Committee of Waste Disposal of the Division of Earth Sciences of the United States National Academy of Science—National Research Council of 1957 (Hess et al., 1957).

In more than 60 years, repository concepts have been developed with the necessary basic and conceptual research.

In Germany, initial research on final disposal "on land" in a former potash and rock salt mine, Asse II, was started as early as 1967. Around the same time, specific characterisations for potential repository sites for low and high-level radioactive waste were established in many other countries such as the United Kingdom, Switzerland,

France, Sweden, Finland, and Canada, supported by more fundamental generic research of interest to many repository programmes. The potential release of radionuclides from a repository site and their subsequent fate and transport in the environment is a public health concern and has stimulated research into waste management techniques and the potential long-term migration of safety-relevant radionuclides. The European Commission has funded such research on waste management, including repositories, since the 1973 Declaration (Council of the European Communities, 1973). More than 1000 reports have been published, focusing in the early days more on practical issues such as inventory, transport, interim storage, disposal, solidification of high-level radioactive waste, definition of responsibilities and management principles. Sites for surface or near-surface disposal of short-lived low-level radioactive waste and very low-level radioactive waste now exist in many countries (France, Sweden, Finland, Spain, United Kingdom, United States). For long-lived transuranic waste in the United States, the safety case was accepted in 1998 and the WIPP was approved in 1999. Costly technical drawbacks of disposal programmes are sometimes encountered, linked to technical issues as well as to societal acceptance.

Projects for the final disposal of highly radioactive waste in deep geological formations have taken more time due to the long half-lives and high radioactivity of this type of waste. Both general and site-specific research has been carried out for more than 40 years. Particular attention has been paid to the long-term development of the geological barrier system at a depth of 500 m, which is designed to protect humans and the environment from a potentially dangerous release of radionuclides into the environment and the food chain for hundreds of thousands of years. It is the first time in human history that risk and safety management has covered such long periods of time.

High-level waste is immobilised in very low solubility matrices (the fuel matrix or glass for HLW from reprocessing) under final disposal conditions and confined in a suitable highly stable waste container. Waste products such as radioactive glasses were developed as early as 1950 to confine high-level waste from reprocessing, such as the nepheline syenite glass in Canada (the only radioactive glass ever buried). The composition of R7T7 glass, produced in France for global baseload customers, has changed little since 1980. On the other hand, research to understand the behaviour of this glass under the conditions of final disposal in deep geological formations has continued to this day and has always confirmed the suitability of the glass for final disposal. Immobilization strategies for separated actinide stockpiles still need to be developed in some countries. While separated PuO2 can be recycled at least once in MOX fuel, actinides remain in reused MOX and multirecycling schemes are still far from being the state of the art. In the event that reprocessing is not provided for in national policy, direct disposal of spent nuclear reactor fuel, including of MOX fuel in suitable casks has proven to be a viable option to ensure the long-term safety of future generations for thousands to millions of years. Ceramic matrices for immobilization of separated PuO_2 are developed as well.

Current state of radioactive waste management programmes

Radioactive waste management programmes are large-scale industrial projects that start with pre-disposal and, depending on national waste management strategies and waste acceptance criteria, involve the separation of reusable materials from final wastes, the proper conditioning and categorisation of these waste types until storage and disposal either in surface facilities, awaiting sufficient decay and requiring institutional control for several hundred years (not provided for in all countries), or in near-surface facilities or deep geological repositories if the radioactivity and half-life are too high to leave the waste on the surface and protect it. The aim of these repositories is to ensure the long-term safety of future citizens through passive barriers that reduce the risk of contact with the waste or contamination of the surface environment before sufficient decay has occurred. The time span from conception to realisation, filling and closure of deep repositories covers time spans of 100 years or more and usually extends over storage areas of several km² at a depth of several hundred metres. At such a depth, the migration time of radionuclides into the human-accessible environment is several 10,000 to millions of years, depending on their mobility, the host rock and the location. Even though human intrusion through drilling into the repository can never be completely ruled out by future generations, the probability of such future (unintentional) intrusion is certainly much lower than for surface disposal sites. Therefore, geological disposal is a measure to minimise the occurrence of intrusions based on high requirements such as the absence of exceptional or special resources at the site.

Successful waste management and disposal programmes must be both socially and technically acceptable (Ewing, 2015). The disregard of this basic principle has led to the failure of several repository projects. As a result, programmes for the construction of repositories for high-level waste have recently been strongly promoted, with several advanced countries (Finland, Sweden, France, etc.) having granted or being close to granting licences for the deep geological disposal of high-level waste or spent fuel. Switzerland has also identified and proposed a site for a repository for high-level waste, which will also store low- and intermediate-level waste. The waste management organisations in these countries have demonstrated, with tens of thousands of pages of scientific evidence compiled over a period of 30 or more years, that the current state of fundamental understanding of the science and technology of long-term repository development is sufficiently developed to realise geological disposal projects. It is clear that the realisation of this progress was only possible through the combination of fundamental understanding of rock and waste properties with extensive site-specific investigations, including the development of a tailored engineered barrier system adapted to the properties of the waste and compatible with the sitespecific natural (geological) barrier. In these countries, a great deal of information was collected on the selected geological repository formations, including host rock extent, geometry, thermomechanics, mineralogical heterogeneity, groundwater flow mapping and diffusion and retention processes, etc. Detailed repository architectures have been designed, including a network of tunnels, ventilation and an engineered barrier system to be built around each waste product emplaced. Provisions for closure and sometimes reversibility are being considered.

Although repository projects in earlier-stage countries are often less developed, they will benefit from these advanced programmes by constraining the overall research agenda to focus less on generic basic research and more on local siteand project-specific and technical issues. Stakeholders in all countries must now recognise that the construction of repositories for high-level radioactive waste has become the current state of the art.

The role of research in radioactive waste management

While even in the most advanced repository programmes, general basic research continues until repository closure to keep pace with the evolution of science aimed at optimisation and providing a better understanding of the safety and expected longterm evolution of a repository, this research no longer has the character of a unique selling point for the realisation of national HLW repository projects, but recognises that each national repository project is unique in some way and that the required site-specific investigations and repository designs may take decades and require tailored solutions.

We can define nuclear waste management and disposal research as the systematic scientific investigation and exploration of features, events and processes that affect and interact with materials, fluids and territories to establish or disprove facts and lead to new conclusions regarding waste treatment and characterisation, optimal disposal pathways, and the siting, design, construction and closure of repositories, including engineered barrier systems, evaluating evolution over time and long-term performance to ensure safety during operation as well as over many generations. State-of-the-art scientific and technological approaches are often applied, sometimes using specialised underground or 'hot' laboratories.

The design of the architecture of the network of underground transport galleries and the emplacement sites of a repository requires a multitude of closely interlinked research and development activities. This requires the development of a suitable excavation technology, its quality assurance, with an adapted waste emplacement technology. Well-designed plugs and seals should permanently close once-open cavities and restore the properties of the primary rock as well as possible over many tens of kilometres.

Researchers need to acquire the necessary contextual background knowledge through interaction with the different categories of actors. Research addresses the different knowledge needs of the different categories of actors: waste producers initiate research on the characterisation of waste and its radionuclide inventories, on the separation of clearly identifiable waste streams, on optimal disposal routes, and on the production and characterisation of waste products. Waste management organisations (WMOs) conduct research to address outstanding issues in the design of repository projects, their safe implementation, their integration into the site, their operation, their closure and their long-term safety. Technical support organisations for regulators (TSOs) are engaged in the research required to verify the safety claims of WMOs and to assure public oversight bodies as a basis for decision-making (regulators, parliamentary offices, etc.) that no harm is to be expected from a particular disposal route and repository project.

While research managers tend to distinguish between basic research and project-oriented applied research, two pitfalls have become apparent in the planning of radioactive waste management research: 1) Research is limited to very specific day-to-day technological issues that arise in the development of the disposal project. The issues addressed may lack a general vision, and the issues may change rather erratically over time as the disposal programme develops, so that knowledge acquired in the past can easily be lost. 2) Basic research has been and is also being conducted, for example, in the fields of informatics, radionuclide chemistry, environmental geochemistry, hydrogeology, microbiology, and nuclear physics, studying both individual and coupled processes from the repository scale to the molecular level. Often these studies provide excellent scientific insights, are generic in nature and provide background information, but are not directly linked to a specific repository project, but contribute to support design and safety assessment. The link to geological disposal is often cited as a motivation, but the better understanding of the problems addressed that this provides does not necessarily lead to a better understanding of repository safety and implementation requirements. The scientists involved in these studies, with some important exceptions, often do not have a sufficient understanding of the technological or societal dimension of the repository projects.

Because of the unprecedentedly long time spans to be considered, repository safety assessments themselves sometimes lack credibility among the general public in some countries, as well as among some researchers. However, no safety analysis has been published in 30 years that shows that the repository is not safe. There is also a misunderstanding: safety analyses are not predictions for the future. The repository must be safe for every expected and "unlikely but not excluded" future development, still recognising that very unlikely scenarios can occur that are not taken into account due to the extremely low probability of occurrence (a meteor falling into a repository shaft). In developing the safety case, multiple and often redundant lines of evidence are considered, all potentially ensuring long-term safety.

While many scientists are specialists in one or another process relevant to repository safety, the overall picture of repository safety is often created by others, such as engineers from waste management organisations or technical support organisations for regulatory authorities. Since the safety analysis is the platform for integrating all the information into a coherent picture with the information provided by the scientists, the safety analyst moderates this process and is also the "operator" of some of the integration tools used in the performance assessment and dose calculations. Therefore, scientists need to be more involved in the whole context of producing evidence and analysis for repository safety.

However, contextual knowledge is sometimes lacking in repository safety research. For example, research managers often argue that better knowledge of the behaviour of actinides in the geological setting of a repository is needed because actinides are the main carrier of radiotoxicity of the waste in the long term. This ignores the fact that in the current European repository concepts, although actinides are the main carrier of radiotoxicity of the waste, they are not the dominant exposure risks from the repository, as actinides are extremely immobile in the reducing geochemical environments of the current repository concepts (Np, Am and Pu are expected to move only a few metres before decaying). The potential release of uranium from the wastes is also limited by the reducing conditions, and although only uranium from the wastes is considered in the safety analyses, as required by the regulations, its release from the wastes is much less than that of natural uranium, which is already present quite immobile in the repository rocks. On the other hand, more knowledge about Pu behaviour might be needed to address criticality issues. It is crucial to broaden the context of repository safety research to focus public spending on the real issues at stake in radioactive waste management research.

Due to the complexity, the pure project dimension and the involvement of multiple stakeholders in national radioactive waste management programmes across generations, no single person can have an overview of all the issues at stake. Therefore, it is essential to create a context by developing a cross-generational and cross-stakeholder knowledge management programme, including a roadmap for research as recently proposed in the EURAD programme or by NEA and IAEA.

Roadmap for research in the field of waste management, including final disposal, in the face of divergent thinking and controversies

A detailed roadmap for the safe implementation and related research and development and intergenerational knowledge management of radioactive waste management programmes has been developed as part of the recent European Joint Programme on Radioactive Waste Management (European Joint Programme on Radioactive Waste management, 2021), involving 23 countries and 51 stakeholders mandated by the various European governments with this project. In this roadmap, the key issues for research and for the development of national waste management programmes are divided into a series of objectives along seven themes starting from programme management, pre-disposal activities, the engineered barrier system that includes the waste to be disposed of, the required geoscientific knowledge, repository design and its optimisation, siting and licensing, and safety case. This requires a close interlocking of basic and applied research, industrialisation and knowledge management in a transparent manner.

Pathways are different for surface and geological disposal, for low- and high-level waste streams and for different geological formations such as crystalline rock, clay, salt, and tuff. Some countries with small inventories of radioactive waste are analysing the option of disposal in deep boreholes without using engineered deep repositories. Further changes in repository programmes result from societal requirements for transparency, retrievability and reversibility. Research needs are also different to some extent in countries with small and with large waste inventories, as the diversity of scientific and technological issues varies despite similar fundamental questions. Some countries have extensive experience in operating repositories for certain waste streams, while they are still developing solutions for others.

Transmutation of radioactive waste is sometimes cited in research as an alternative to waste disposal. With huge investments in nuclear reactors and the nuclear fuel cycle and centuries of reactor operation, transmutation can reduce the long-lived actinide inventory of the waste, but not its fission product inventory and the long-term risk of the disposed transmutated waste, which depends on the mobility of some fission products. The heat generation of the waste and the required disposal space are also only slightly reduced. Transmutation requires reprocessing. Reprocessing increases the number of different waste streams, at least in countries that opt for direct disposal, such as Germany, the United States, Canada, Sweden, Finland, etc. Even more, studies have shown that the total actinide inventory, which includes both accumulated waste and inventories in the nuclear reactor fleet, decreases only very slowly, even over transmutation periods of up to 200 years. Extensive research has shown that transmutation neither reduces the need for disposal in deep geological formations nor improves the long-term safety of the waste in final disposal, since the riskdetermining nuclide inventories, such as that of the rather mobile I129, are not reduced by transmutation. The term "rather mobile" should be used with caution, as it may be more than 100000 years before the first traces of I129 can escape from the repository into the environment accessible to humans.

Current research gaps

Open research gaps are identified by IGDTP, SITEX, and EURADSCIENCE and in EURAD through a joint Strategic Research Agenda SRA (EURAD EURD SRA, 2022), which has already been published and will be updated soon. A detailed description of the respective research topics can be found there, which also includes the properties of wastes and waste forms. Only a few typical examples are discussed here:

Spent fuel and vitrified high-level waste are well characterised and the disposal properties are largely known. Remaining uncertainties include inventories of some long-lived beta-emitting activation products such as Cl36, Cl4, etc., to better quantify the limits according to prudent hypotheses considered in the safety assessment. The EURAD SFC work package has reduced the uncertainties in the inventories of heat-generating radionuclides in order to optimise the occupancy of the repository space. A good understanding of the mobility of radionuclides from these waste products is available, including consideration of the effects of containers and the near field environments of the disposal places, allowing the assessment of the release characteristics (source term) for a large number of radionuclides.

Special attention is given to investigations at the interfaces between barriers and materials in the repository such as copper/ clay, steel/clay, steel/concrete or concrete/clay in the presence of fluids such as water or gas. The investigations range from the molecular level to the size of metres. Methods for validating such coupled models deserve more attention.

Full-scale coupled codes are being developed to mathematically simulate the multiple interacting thermal, geomechanical, hydrological and geochemical processes involved in waste emplacement, not ignoring possible radiation effects. Digital representations can be developed to represent specific parts of the repository concept or the entire repository.

In some repository programmes there is interest in developing container concepts with long (>10000–1000000 years) quality assured target lifetimes, which requires a good scientific understanding of loads, corrosion resistance, fracture mechanics, weld performance and highly reliable quality assurance. Despite the vast knowledge accumulated for the fundamental understanding and development of container materials up to industrialisation, it often seems that the weld and its quality assurance need the most attention. However, SKB's development of friction stir welding

demonstrates that very high quality welds that are fully inspectable can be produced and are accepted by regulators.

While being well and safely managed in current repository concepts, iron-based container concepts are often expected to generate large amounts of hydrogen gas, which is produced by anoxic corrosion in the groundwater. After repository closure, large pressures can build up and fluid transport (gas, water) in the repository can be affected. These problems are avoided by covering the container with copper, as planned e.g., in Finland, Sweden, and Canada, or by allowing easy passage of gas, as planned e.g., in France and Switzerland. Inert containers (ceramics) with appropriate closure concepts can offer new possibilities.

For long-lived low and intermediate level waste, there is a need in some countries for cost-effective rapid characterisation of gamma and fissile nuclide content and physical properties using available non-destructive technologies, and for advanced separation and conditioning techniques to be developed for some problematic waste types.

There is also a certain interest in developing better understanding of the source term of various waste forms and how this translates into waste acceptance criteria for a repository, as well as their use in safety assessment.

Longer than originally planned and approved interim storage leads to a certain need for research with regard to the validation of transportability, repackaging and disposability of the waste packages after longer storage.

The behaviour of backfill materials such as bentonite and concrete is well known for a wide range of disposal conditions and densities. The erosion resistance of bentonites has also been widely studied for granite. The effects on water and gas transport, mineralogical transformation and radionuclide migration are largely known. There is naturally somewhat less knowledge about newer materials such as low pH cements and geopolymers. Behaviour under transient thermohydromechanical properties is still under investigation, including conditions where pore spaces are only partially saturated with groundwater. There is also the question of how long it takes for voids to fill with groundwater, taking into account gas pressure and gas solubility in nanoscale clay pores. Important questions are also the sealing behaviour of large-scale plugs and seals produced under these conditions. Effects such as those of microorganisms or organic substances on some of these properties are being investigated as part of the ongoing EURAD programme.

The transport and retention properties of host rocks for radionuclides and chemotoxic waste components, as well as of gases and groundwater, have been studied in detail for many years, taking into account chemical, redox and salinity gradients, as well as the impact of repository construction on these properties. Some studies go as far as understanding at the molecular level. While this level of detail is generally not required to design repository projects and constrain the safety case, it provides useful background information that demonstrates the soundness and scientific excellence of the knowledge gained.

Considering that many individual and some coupled processes are being studied under a variety of relevant conditions, it must be acknowledged that any repository concept is a very complex, multicoupled system on a large scale. The goal of research is certainly not to predict the actual and detailed evolution of this system on a large time scale. Such predictions are impossible and can never be addressed by science. The aim is merely to cover a range of possible developments and to ensure that, for any reasonably expected or less likely but not excluded development, the safe isolation of the waste is guaranteed for the required isolation times of hundreds of thousands of years. However, it should be noted that the high level requirements and the design of the repository aim to limit the complexity of the phenomenological processes to promote a simple understanding of the evolution of the repository, such as limiting the temperature or placing the waste types in separate zones and/or emplacement cells to limit the physicochemical interactions. The safety case required for public decision-making draws on a broad base of site-specific and general knowledge gained through decades of scientific research and addresses every reasonably conceivable situation and scenario, that could occur from waste emplacement to the development of the containment system in the distant future, taking into account the scientific and technological uncertainties that always remain, covering potential failures in the barrier system as well as geological events and developments, and addresses "what-if" scenarios to test the robustness of the planned isolation system. The focus of course remains on the more likely reference situations/scenarios.

Work is currently underway to develop coupled hydrothermo-mechanical-chemical multiscale computer models that quantify the expected mobilisation, transport and immobilisation of radionuclides, taking into account gas production and microbial effects, to assess the barrier system from repository construction through waste emplacement to closure and long-term post-closure performance.

The overall impact on safety is considered in the development of the safety case, which involves various stakeholders. Scientific data and models and their linkages help in the preparation of the safety case. By coupling known relevant processes, current safety analyses, for example for the geological disposal of high-level radioactive waste and spent fuel in clay rock in France or Switzerland, have shown that no impacts of the repository on the safety of the population are to be expected in the next 10000 years and that regardless of the expected evolution scenario of the repository system, even for longer periods of time, the maximum dose to be expected is more than 1000 times lower than the present and future ambient background dose of the population. Similar conclusions can be drawn from the safety assessments prepared in Finland and Sweden for the final disposal of spent fuel in crystalline rock. As the complexity of such assessments is usually quite high, it is certainly important to increase the credibility and transparency of these assessments through more research and, if at all possible, to challange such assessments and/or increase their

redundancy and provide multiple lines of evidence in the safety demonstrations, but it is certainly not reasonable to steer research and development in a direction that reduces the already negligible calculated dose impacts to even lower values.

On the other hand, technological or conceptual research is still needed to develop, optimise and/or ensure the quality of various concepts for the final disposal of low and high-level radioactive waste, e.g. the choice of more cost-effective barrier materials with similar insulation properties, the optimisation of waste package concepts, the engineered barrier design for high level waste and of the construction, operation and production of its components, e.g., by robotics, the management of reactive interfaces at voids between materials by voidfilling materials, the management of zones disturbed by excavation, the development, optimisation and study of the long-term performance of full-scale shaft and gallery seals, the development of monitoring concepts with long-term sensors and secure power supply, etc., Research will also play a role in ensuring that technical solutions to reduce costs do not come at the expense of safety.

Conclusion

While current research provides important background information that can be used in current or planned repository projects, and improves the understanding or predictability and digital representation (digital twin) of the features and processes involved, as well as confidence in the safety case, it should be noted that no research results in recent years have challenged the continuation of ongoing repository projects. Research has shown that safe repositories can be built in many geological environments worldwide, and current research goals are more in the area of complex system-coupled 3D and THMC modelling and optimisation of repository concepts.

Author contributions

The author confirms being the sole contributor of this work and has approved it for publication.

Acknowledgments

The careful review of the draft of this paper by Piet Zuidema, Frédéric Plas, Astrid Göbel and Johan Anderson is gratefully acknowledged.

Conflict of interest

The author declares that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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