APPLYING A LIFE CYCLE ENVIRONMENTAL PERSPECTIVE TO THE DEVELOPMENT OF RADIOACTIVE WASTE TREATMENT TECHNOLOGIES

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Abstract

Environmental life cycle assessment (LCA) is a standardised approach to evaluating and improving environmental sustainability in a holistic manner, and has been applied quite extensively to nuclear energy generation life cycles. However, there are very few examples of LCA being used to ensure more sustainable radioactive waste management. The paper outlines the prior use of LCA in this field and the potential benefits of its application, before focusing on work conducted as part of the EU/Euratom-funded PREDIS project which has applied LCA and the methodologically-related approach of life cycle costing (LCC) to a variety of novel treatment options for metallic, organic and concrete package waste streams. This has been conducted in close collaboration with partners across Europe to identify candidate waste forms and pre-treatment methods and to coordinate data collection processes, which will be outlined. Subsequent LCA has been conducted using Sphera LCA for Experts (GaBi) software with input from the Ecoinvent 3.9 database. The paper draws on this work to analyse cases of specific waste treatment techniques and to illustrate how LCA can be used to ensure that new waste treatment processes in the nuclear sector are designed with sustainability in mind from the outset.

1. INTRODUCTION

LCA is an environmental sustainability tool that applies the concept of life cycle thinking to assess the consequences of human activities. Broadly speaking, as specified in ISO 14040 and 14044 [1, 2], LCA involves:

(a) quantification of environmental burdens of a product, process or activity via assessment of the energy and materials used and wastes released to the environment;
(b) quantification of environmental impacts (i.e. translating the above burdens into potential impacts); and
(c) identification of opportunities for environmental improvements along the life cycle via the identification of ‘hot spots’.

Thus far, LCA in the nuclear sector has focused on electricity generation, deriving estimates of environmental impacts per kWh or MJ of electricity produced. Such studies have considered light water reactors, heavy water reactors, advanced gas cooled reactors, and fast reactors, operating in closed and open fuel cycles. The results have been reviewed in literature [e.g. 3, 4, 5], typically focusing on greenhouse gas emissions. Many individual LCAs of nuclear power have included a broader range of environmental impact categories — spanning issues such as carbon footprint, acidification, eutrophication, ecosystem toxicity, human toxicity, and resource depletion, among others — and have used the results to compare the impacts of nuclear systems with other power technologies [e.g. 6, 7, 8]. The insights provided by these and other LCAs have been instrumental in determining policy positions, including the inclusion of nuclear power in the EU taxonomy for sustainable activities (‘green
taxonomy”) [9]. LCA has also been used to investigate the impacts (and mitigation of impacts) arising from potential nuclear-based energy vectors such as hydrogen and ammonia [10, 11].

However, as revealed in a forthcoming review by the authors, only one existing LCA study [12] has an explicit focus on the back-end of the nuclear fuel cycle. It evaluates the entire decommissioning process for a UK Magnox reactor and, in doing so, highlights the paucity of detailed inventory data (such as mass and energy flows) for waste treatment and decommissioning processes. In the energy generation-focused studies mentioned above, the back-end is either greatly oversimplified, lacking specification, or missing entirely [3, 5]. One potential explanation for this knowledge gap is simply a lack of data, as suggested above. Another is the suggestion that the majority of environmental impacts in the nuclear power life cycle occur in the front end (e.g. mining, milling, enrichment), as typically found by prior LCAs [3]. However, given the lack of detailed quantification of back-end processes in all existing LCAs, this suggestion is somewhat unsubstantiated.

This general lack of environmental knowledge surrounding the back-end of the nuclear fuel cycle is particularly problematic due to the large volumes of waste needing treatment and disposal: the arisings of radioactive waste in the EU by 2030 are estimated at 5.2 million m$^3$ (VLLW, LLW, ILW, HLW and spent fuel) [13]. The impacts of treating and disposing of this waste are currently unknown and impossible to estimate accurately without further study, meaning data-driven minimisation of those impacts is also not possible.

In light of the evident benefits that could be achieved by considering cradle-to-grave environmental impacts in the nuclear back end, this paper presents a set of practical methods to conduct such analyses. Two case studies are then used to highlight the application of these methods, with selected results discussed in more depth to illustrate the outcomes and their potential implications.

2. METHODS

LCA is a well-established technique with a wealth of existing literature demonstrating its use. It is standardised via ISO 14040 and 14044 [1, 2], in which four key phases are identified: goal and scope definition, inventory analysis, impact assessment, and interpretation. This work aligns with these standards. The LCA modelling presented here has been conducted using Sphera LCA for Experts (GaBi) [14].

2.1. Goal and Scope Definition

To retain flexibility in view of the developing technologies under study, the functional unit (FU) and system boundaries are defined separately for each case study. The FUs are defined as follows:

(a) For the use of gel to decontaminate a radioactive metallic surface, the FU is the treatment of 10 m$^2$ of planar stainless steel surface. Stainless steel was chosen as it is regularly used in parts of a nuclear power plant that will become irradiated over time such as areas that will come in to contact with coolant, and in radioactive material management areas such as hot cells.

(b) For geopolymcer encapsulation of radioactive solid organic waste (RSOW), the FU is the pre-disposal processing of 1 kg of intermediate level RSOW surrogate. This was chosen as it represents the encapsulation of ash from the IRIS plant [15], received as a product of thermally treated ion exchange resins and contaminated personal protective equipment.

The system boundaries proposed in this approach begin at the receipt of waste to be treated (from waste generator organisations) and end at a predefined point appropriate for each treatment technology.

For the gel decontamination case study, the system boundary starts at the production of reagents and equipment used to decontaminate the metallic surface and ends at the removal of the treatment gel from the target stainless steel surface once there has been sufficient contact time. All upstream and downstream burdens associated with these processes are included.

For the study of geopolymer encapsulation of radioactive solid organic waste, the system boundary includes the receipt of waste at the treatment facility to the handoff of a conditioned waste drum to a final repository. All upstream and downstream burdens associated with these processes are included. This represents a typical route which would be utilised in the up-scaled operation of this process.
2.2. Inventory Analysis

The technical data required to specify the foreground systems were collected iteratively with PREDIS project partners. This was achieved using a data collection template and collated in Excel files. The template was relatively generic to maximise flexibility, and the data received from partners varied from lab-scale data with many missing data points to detailed inventory based on upscaled systems. Therefore, interpretation and adaptation by the authors was required. This was enabled by the collection of indicative values, ranges, and explanatory comments in the template, followed by iterative meetings online and in person.

For all LCA case studies in the PREDIS project, including the two presented here, a ‘base case’ was established representing the present-day default treatment/disposal process that each wasteform would undergo, allowing comparison against the treatment processes under development. In all cases, background data were sourced from the ecoinvent database v3.9 [16], as it is the most widely used and accepted life cycle inventory database, particularly for Europe-focused systems. We note that many other similar databases exist, such as the U.S. LCI Database managed by NREL [17], which may be more suitable in some cases.

The case study assessing the use of decontaminating gel is based on data provided by CEA [18] including masses and ratios for the gel ingredients, as well as information on the type of equipment used to apply and remove the gel from the target surface. Once all the mass and energy flow data were collated they were used to create a model illustrating the environmental impacts associated with decontaminating an exemplar 10 m² contaminated stainless steel surface.

The case study addressing the geopolymer encapsulation of radioactive solid organic waste (RSOW) surrogate is based on data provided by Politecnico di Milano [19]. The inventory information provided was based on a 20% waste loading of a bespoke zeolitic volcanic tuff based geopolymer. The waste, tuff and respective activation materials are mixed in-situ within a standard waste drum.

Due to data confidentiality, full detail of each inventory dataset is not presented here, but rather the approach of data collection and iteration with partners is emphasised in order to arrive at a mutually agreed representative dataset.

2.3. Impact Assessment

The third phase of LCA, Impact Assessment, uses environmental impact coefficients, often referred to as characterisation factors, to estimate the potential environmental impacts caused by the burdens identified during the Inventory Analysis phase. The exact list of indicators generated by an LCA depends on the impact assessment method adopted, and a variety of options exists. Much prior literature has used the CML method [20] but this is now generally considered outdated. Alternatives include IMPACT2002+ [21], TRACI [22, 23], ILCD [24], Environmental Footprint [25] and ReCiPe [26, 27]. Of these options, ReCiPe and Environmental Footprint are often seen as the state-of-the-art and there is some evidence to suggest that ReCiPe is the most widely used method, although a plurality is evident in the community [28].

We note that the standard life cycle impact assessment methods have a relatively simplistic approach to radiological impacts, leading to recent work which has developed and recommended new methods – known as CGM and UCrad – to be used in the context of nuclear power and waste management activities [29, 30]. These methods are not applied here, but are recommended for inclusion in future studies.

Consequently, this work adopts the ReCiPe impact assessment methodology, providing information on all included impact categories, with the UCrad method for radiological impact also considered.

2.4. Interpretation

The interpretation of results is being conducted iteratively with PREDIS project partners to evaluate key findings, validate models and identify strategic avenues for future impact reduction. These issues are addressed in the results section below.
2.5. **Life Cycle Costing**

While not included in this paper, the methods adopted by the PREDIS project do include economic life cycle costing (LCC). LCC is not standardised in the same way as LCA, although it shares a common aim of accounting for (economic) impacts across the entire life cycle. A general methodology exists for LCC [31] based on work by the Society of Environmental Toxicology and Chemistry (SETAC). This methodology mirrors that of the LCA cradle-to-grave approach in that LCC should include capital costs, fixed operating costs, variable operating costs, waste management and recycling costs, end of life costs and transport costs.

Various metrics are permissible within LCC, based on discounted or non-discounted costs presented per life cycle stage, net present value, annualised costs and value added [32].

The approach proposed here stresses alignment of LCC models with their corresponding LCA models in terms of system boundary, system specification and functional unit, to maintain internal consistency. Cost estimates should be attached to each flow into and out of the system, and the choice of metrics and discounting should then be based on discussion with partners.

3. **RESULTS AND DISCUSSION**

3.6. **Gel Decontamination**

For the treatment of 10 m² of radioactive stainless steel surface using a decontaminating gel, the model was separated into four sub-processes: treatment (including gel production), application (including sprayer production), removal (including vacuum cleaner production) and energy used by equipment.

FIG. 1 shows all 18 midpoint impact categories arising from the ReCiPe 2016 methodology. It is clear that the impacts are dominated by the production of the decontamination gel and vacuum cleaners that are used to remove the gel from the treated surface: these processes account for an average of 41% (range 12-90%) and 58% (range 10-88%), respectively, across the impact categories. For the gel this is mainly due to the production of the oxidising agent present in the mixture. For the vacuum cleaners this is due to their short lifespan and therefore the quantity of cleaners necessary: once full, the vacuum cleaner drum must be safely disposed of, which involves compaction and encapsulation. Therefore, the lifespan of the cleaner is restricted by the volume of the drum. Arising from these hotspots, avenues for improvement include identifying ways to reuse the vacuum cleaners or some of their components, and investigating alternative oxidant species in the gel formulation. These potential changes must of course be undertaken bearing in mind any resultant effects on waste acceptance criteria.

The main benefit of treating large metallic surfaces with this approach is that the treated metal can be recycled rather than being disposed of in a radioactive waste store. This recycled metal could then be made available to the wider market, displacing the need to produce virgin steel and therefore reducing the impacts of the steel manufacture sector.

FIG. 2 illustrates the huge difference in impacts of producing virgin steel compared to decontaminated and recycling the stainless steel being treated. This illustrates the base case approach in which the contaminated stainless steel would be sent to deep geological disposal and new steel would have to be produced.
This treatment option also frees up valuable space in deep geological disposal facilities. When built, GDFs will have a finite amount of space to store both current inventory and any future arisings, therefore any approach which can minimise the volume consigned to this disposal route is valuable.

Therefore, from these results it can be inferred that treatment of large planar metallic surfaces with prospects to reuse or recycle greatly reduces the overall carbon footprint, as well as the time and money required to send contaminated wastes to deep geological disposal.
3.7. Geopolymer Encapsulation

Results from the geopolymer encapsulation case study are shown in FIG. 3 which compares a base case treatment technique for radioactive solid organic waste — encapsulation in cement — with a geopolymer alternative. As geopolymer encapsulation is still under development, uncertainties remain around waste loading in different circumstances. A baseline assumption of 20% was used for the direct comparison to cement, as this matches the typical loading for cementitious matrices. However, preliminary data suggest that higher loadings may be possible with geopolymers, therefore additional models were developed to investigate increased waste loading (30% and 40%).

\[\text{FIG. 3. Life cycle environmental impacts of utilising varying methods of RSOW encapsulation}\]

A clear distinction can be made between the climate change potential of utilising novel geopolymer technologies in comparison to typical Portland cements at the same waste loading factor. A clear correlation can also be drawn between the increase of waste loading factor and resultant decrease in climate change potential. Similar trends were also identified for the other 17 impact categories in the ReCiPe 2016 LCIA methodology.

Hence, it can be inferred that a shift towards utilising geopolymer matrices is likely to reduce carbon footprint by at least ~63% for similar waste loading factors, validating the environmental driver for further research into geopolymer treatment processes. However, the cost of implementation at scale requires further investigation and is the subject of future work by the authors.

Conclusions can also be drawn surrounding the increase in waste loading. Higher loading increases the energy consumption during waste processing activities, but the results demonstrate that the resultant impacts are outweighed by the reduction of impacts associated with the use of materials for encapsulation. Therefore, this would suggest that research and development of these advanced matrices should be focused on assessing the feasibility of increased waste loading. In terms of implementation across a campaign of RSOW processing, this would also see a reduction in the final number of drums allocated to a final repository which would have consequential reductions in environmental impact and cost, but quantification of these issues is outside the scope of this study.

These recommendations could potentially have a significant impact on waste acceptance criteria (WAC) set out by WMO’s, which could be adjusted to include environmental conditions in parallel with a robust safety case.

4. CONCLUSIONS

The paper has outlined the approach taken in the PREDIS project to incorporate life cycle thinking into the development of radioactive waste treatment technologies, illustrated by two case studies addressing gel-based decontamination of metallic waste and geopolymer encapsulation of organic solids. The approach applies environmental life cycle assessment (LCA) and economic life cycle costing, following appropriate international standards where applicable. Key elements include the appraisal of multiple impact assessment categories – 18 in this case – to provide a broad understanding of the environmental profile of the developing technologies, as well
as the use of hotspot analysis to identify future focal points for R&D, alongside benchmarking against base case treatment techniques to establish potential benefits and/or drawbacks of new approaches.

The two case studies presented have identified the following:

(a) Gel manufacture and vacuum cleaner production have broad, multi-criteria dominance over the environmental impacts of metal decontamination, with energy consumption and application playing almost negligible roles. Therefore future efforts should identify alternative oxidants in gel formulation and explore opportunities to minimise or reuse vacuum cleaners or their components.

(b) The impact of gel decontamination of planar stainless steel surfaces has a carbon footprint three orders of magnitude lower than the production of virgin metal, suggesting substantial climate benefits to this approach over the base case of direct disposal.

(c) Geopolymer encapsulation of solid organic wastes outperforms conventional cementation at similar waste loadings, by >60%, while higher waste loadings increase this advantage despite the concomitant increase in energy consumption. The latter is true even without considering downstream benefits resulting from repository space savings, which would amplify these benefits.

(d) Costs and optimisation of waste loadings should be the focus of future geopolymer encapsulation research to maximise sustainability.

Given the large volume of radioactive waste in need of treatment/disposal (5.2 million m$^3$ across the EU by 2030), the insights gained from LCA during process development could enable far-reaching future benefits. This requires close collaboration with technical partners to ensure the validity of input data and suitability of improvement suggestions, as has been practised in the PREDIS project.

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