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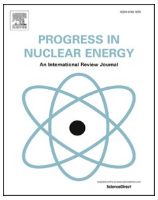

Using life cycle assessment to quantify the environmental benefits of circular economy strategies in the nuclear industry

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ABSTRACT

In the United Kingdom nuclear energy is expected to play a key role in decarbonising the power generation sector. The implementation of circular economy approaches in the nuclear industry can minimize the amount and the associated environmental impacts of nuclear wastes. In this article, we demonstrate how Life Cycle Assessment (LCA) can be used to investigate the environmental benefits of two circular economy strategies that aim at reducing the amount of intermediate level waste (ILW) destined for disposal in a geological disposal facility. The first case study focuses on a novel technology for recycling zirconium alloy cladding waste, whilst the second case study investigates the environmental benefits of using depleted (instead of natural) uranium to produce uranyl nitrate, a chemical used in the PUREX process. Our results show that both circular approaches outperform conventional ones across all environmental categories and particularly in terms of resources depletion, with reductions up to 25% and 94% for respectively zirconium alloy recycling and depleted uranium reuse. The environmental benefits of both approaches are due not only to a reduction in the amount of ILW to be disposed of, but also because they are assumed to induce a reduction in the demand for mining of primary zirconium or uranium respectively. When both approaches are combined, the environmental benefits range from 4% in the category freshwater and up to 94% in the category resource use, energy carriers.

1. Introduction

In 2019 the UK Government established a legally binding target to reduce the country's greenhouse gas (GHG) emissions to net-zero by 2050 (Committee on Climate Change, 2019). This is in line with the European Green Deal (European Commission, 2019) and the outcomes of the Conference of Parties (COP26) held in Glasgow, which reiterated the climate emergency status (UNFCCC, 2021) and the utmost importance of employing the best available science for effective climate action and policymaking. In the Glasgow Climate Pact (UNFCCC, 2021) the parties welcomed the latest recommendations of the Intergovernmental Panel on Climate Change (Masson-Delmotte et al., 2018), and stressed the urgency of applying the stricter target of the 2015 Paris Agreement that aims at keeping the increase in global average temperatures to below 1.5 °C above pre-industrial levels (UNFCCC, 2015). The net-zero target entails profound and far-reaching transitions in nearly all sectors of the economy, with the power sector being in the spotlight because it has the potential to decarbonise almost fully and more quickly than other sectors. Nuclear energy is expected to play a key role in the future UK energy mix as a low-carbon, baseload source of energy. The UK Government's 2020 Ten Point Plan for a Green Industrial Revolution (HM Government, 2020) envisages investments in large-scale nuclear plants to replace, and potentially expand, the country's ageing nuclear fleet as well as additional research and development investments in advanced nuclear fuel cycles, including small and advanced modular reactors.

The management and disposal of nuclear wastes represents one of the most controversial aspects of nuclear power generation. Nuclear wastes are classified according to radioactivity and heat generation level: as high level waste (HLW), intermediate level waste (ILW), low level waste (LLW) and very low level waste (VLLW) (Wilson, 1996). Whilst VLLW, LLW and some ILW can be disposed of in near-surface repositories, higher activity wastes (which include HLW and some ILW) require disposal in deep repositories - known as Geological

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Disposal Facilities (GDFs) or as Deep Geological Repositories (DGRs) - which are to be built several hundred meters underground in geographically stable environments. To date, there aren't many operational GDFs in the world (Conca, 2021; Nuclear Waste Partnership, 2021). In the UK, construction of the GDF is not expected to start for at least 25 years; with construction and operation of the facility projected to last for approximately 100 years (Paulillo et al., 2020d).

The circular economy approach may help mitigate some challenges of nuclear waste disposal (Taylor et al., 2022b). The approach represents a new economic paradigm that aspires to replace the "take-make-dispose" of the traditional linear economy (Morseletto, 2020). Circular economy aims to minimize wastes and retaining materials' value in the long-term, thus promoting efficient use of natural resources (Haupt and Zschokke, 2017; Morseletto, 2020). The approach enables decoupling economic growth from the constraints of natural resources (Haupt and Zschokke, 2017; Niero and Olsen, 2016), and thus from the negative environmental consequences that are linked to the extraction of resources, including environmental pollution and biodiversity losses (Peña et al., 2021). However, the environmental benefits of circular economy strategies should not be presumed, rather they must be questioned and assessed quantitatively; an appropriate tool to do so is Life Cycle Assessment (LCA) (Cordella et al., 2020; Peña et al., 2021). LCA is an ISO standardised (ISO 14040:2006; ISO 14044, 2006) and widely adopted methodology to assess the environmental impacts of products, including goods and services such as the generation of electricity or the management of wastes. The methodology adopts a life-cycle perspective and covers a wide range of environmental issues, including climate change, resources depletion, eutrophication, acidification, ecotoxicity. This holistic perspective enables the identification of trade-offs, thus making LCA a robust tool for supporting decisions.

The majority of LCA studies in the nuclear industry available in the scientific literature are aimed at assessing the environmental performance of electricity generation from nuclear energy from an attributional perspective. The earliest LCA studies were published in the early 2000s (Lee and Koh, 2002; Lee et al., 2000); these studies demonstrated that a twice-through nuclear cycle based on the PUREX (Plutonium Uranium Redox Extraction) process outperforms the once-through cycle as well as the direct use of PWR spent fuels in CANDU reactors. The advantage of a twice-through cycle is that it avoids or reduces the need for several front-end activities, including mining and enrichment of uranium, that represent notable sources of environmental impact. The environmental benefits of the twice-through cycle were confirmed by Poinssot and colleagues (2016, 2014) for the French context and by Paulillo et al. (2021) for the UK. Additional studies focused on less conventional fuel cycles are provided by Ashley et al. (2015), Carless et al. (2016) and Serp et al. (2017). The environmental impacts of decommissioning nuclear plants are currently uncertain; they have rarely been assessed, largely because few sites have been decommissioned. The only LCA study available in the scientific literature was authored by Wallbridge et al. (2013), who quantified the environmental impacts of decommissioning the Magnox power plant at Trawsfynydd, in Wales UK.

A significant portion of literature focused on a single environmental issue: climate change. This is because the primary objective of these studies is to compare energy sources based on their potential of curbing GHGs emissions, and perhaps because of the lack of data regarding other kinds of emissions. Lenzen (2008), Sovacool (2008) and Warner and Heath (2012) collected and reviewed estimates of life-cycle GHG emissions from a significant number of literature studies. They demonstrated that the variability of literature estimations depends on three factors: the uranium ore grade, the enrichment process and its source of electricity. Considering this, Warner and Heath attempted to harmonize the values reported in the scientific literature; their analysis yielded a median carbon footprint of ~12g CO_2-eq./kWh and a range (from minimum to maximum) of ~110 g CO_2-eq./kWh. Norgate et al. (2014) investigated in detail the effect of uranium ore grade on life cycle GHG emissions; their results show that when the ore grade declines from 0.15% to 0.001% (e.g. for uranium contained in some granites) the GHG emissions ramp up from 34 kg CO_2 eq./MW_e to 594 kg CO_2 eq./MW_e. Parker et al. (2016) performed a similar study for mining and milling of uranium in Canada; their analysis yielded an average carbon emission intensity of 42 kg CO_2 eq./kg U_3O_8 with an average ore grade of 3.81%.

The lack of an appropriate and standardised methodology in LCA for quantifying the environmental impacts of radioactive emissions, including those arising from the disposal of nuclear wastes, has hindered the application of LCA to the nuclear industry, especially in comparison to other power generation technologies (Paulillo et al., 2018). Paulillo and colleagues developed a novel methodology – UCrad – for quantifying radiological impacts from routine radioactive discharges and from nuclear waste disposal, that is based on the traditional approach for predicting human toxicity impacts in LCA (Paulillo et al., 2020a, 2020c); the methodology was then further developed to estimate human health impacts in terms of Disability Adjusted Life Years, DALY (Paulillo et al., 2023). UCrad was deployed in Paulillo et al. (2020d) to evaluate the environmental impacts of recycling Used Nuclear Fuel (UNF) in the Thermal Oxide Reprocessing Plant (THORP) at the Sellafield site in the UK. The results revealed that a great proportion of the environmental impacts is linked to two specific causes: indirect environmental impacts associated with the production of uranyl nitrate, which was used to separate plutonium from uranium in THORP, and of copper, proposed in one scenario to be used as the outer layer of the disposal canister for HLW in a future GDF. Paulillo et al. (2020d) also identified the fuel cladding as a significant contributor to the radiological impacts of the GDF,

In this article we aim to demonstrate how LCA can be deployed to evaluate circular economy strategies and support decisions in the nuclear industry, by identifying environmentally advantageous options that could be part of a future nuclear fuel cycle in the UK. Our work addresses two of the environmental hot-spots identified by Paulillo et al. (2020d): uranyl nitrate and cladding. The first case study focuses on a novel technology for recycling zirconium alloy cladding from used nuclear fuels (Taylor et al., 2022b), whilst the second investigates an alternative approach to manufacture uranyl nitrate – a chemical used in the THORP generation of the PUREX process. This work significantly advances the application of LCA in the nuclear industry, for several reasons: first, our case studies address considerable environmental issues and have not been investigated previously; second, the LCA methodology is yet to be used to assess circular strategies in the industry; third, unlike most studies in the literature, our analysis is not limited to carbon emissions, rather we assess a wide range of environmental impacts; fourth, we adopt a consequential approach for quantifying the environmental benefits.

2. Methods

In this Section we start by briefly introducing the LCA methodology (Section 2.1) and we continue (in Section 0 and 0) with a detailed description of the LCA case studies. Finally, in Section 2.4 we report the impact assessment methods use for calculating the results.

2.1. Life cycle assessment methodology

The Life Cycle Assessment (LCA) is a standardised methodology by the International Organisation for Standardisation (ISO) (ISO 14040:2006; ISO 14044, 2006). The ISO defines LCA as "the compilation and evaluation of the inputs and outputs and the potential impacts of a product system throughout its life cycle". It is based on a quantified functional unit (FU): the function of the product system considered, expressed as a service or good(s) delivered. The methodology quantifies potential impacts relative to the throughput needed to deliver the FU, and obtained from best estimate, location-generic environmental models. The ISO standardised framework, shown in Fig. 1, consists of

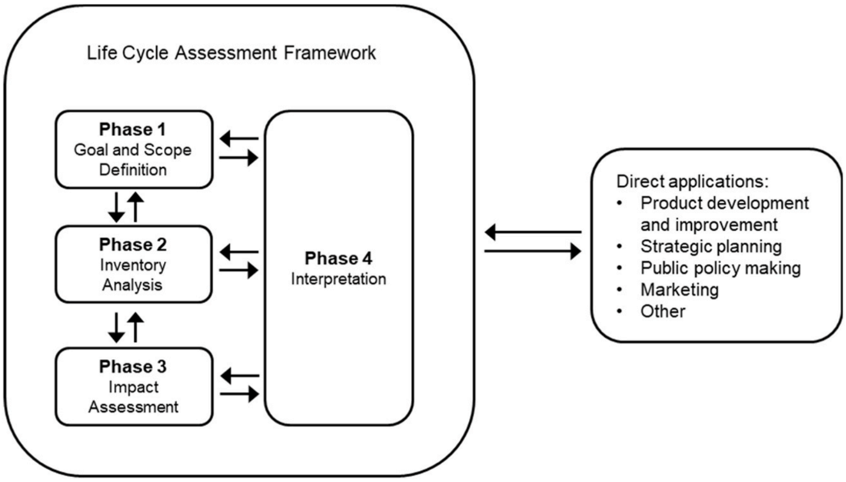

Fig. 1. Phases of the Life Cycle Assessment (LCA) methodology (ISO 14040:2006).

four phases, namely Goal and Scope Definition, Inventory Analysis, Impact Assessment and Interpretation. The LCA phases are iterative in the sense that during a study each phase may be revised considering the other phases.

The first phase, goal and scope definition, frames the study: the goal includes the reason for carrying out the study (why the study is done), its intended application (what the study wants to achieve), the intended audience and the commissioner of the study and other potential influential stakeholders to highlight conflicts of interest. The scope, on the other hand, defines the functional unit and establishes the focus of the study in terms of the processes to be included in the product system (system boundaries).

Following the first phase, inventory analysis collects information about the physical flows in terms of input of resources, materials, semi-products, products and by-products and the output in terms of emissions, waste and the final product. It must be noted that that the validity and accuracy of LCA results are strongly dependent on that of the underlying inventory; for this reason, having access to high-quality data, e.g. collected on site or extrapolated from design flowsheet, is of utmost importance. Partly to facilitate compilation of the inventory data, it is common practice to distinguish between the *foreground* system, i.e. those activities that are the focus of the study and may be affected by the results, and the *background,* i.e. activities in the rest of the economy linked to the foreground by exchange of materials and energy (Clift et al., 2000). The foreground activities are described by primary data while the background activities by industry-average data. Several commercial databases are available as sources of background data; notable examples are ecoinvent (Wernet et al., 2016) and Sphera (formerly Thinkstep) (Kupfer et al., 2020).

Taking the life cycle inventory as a starting point, impact assessment "translates" the physical flows of the product system into potential impacts on the environment and human populations using knowledge and models from environmental and medical science. Impacts are expressed as their contributions to a set of pre-defined impact categories, each addressing a specific issue; for instance, the climate change category includes all gases contributing to the greenhouse effect. Finally, in the Interpretation phase, the results of the study are checked for consistency and completeness, and conclusions and recommendations based on the results of earlier phases are developed.

2.2. *Case study n°1: zirconium alloy recycling*

The first LCA case study concerns the potential recycle of zirconium alloy, an alloy of zirconium and other metals (including niobium, tin, nickel and chromium) that is used as cladding of nuclear fuels due to its low neutron absorption cross section, its corrosion resistance and adequate high-temperature mechanical strength (Yagnik and Garde, 2019a). The recycling process of zirconium alloy has been investigated for years, mainly in the United States (Taylor et al., 2022b). It represents an attractive process because cladding significantly contributes to the potential radiological impacts arising from the GDF. (Paulillo et al., 2020d).

Historically, when used nuclear fuels were recycled at Sellafield site in the UK, zirconium alloy cladding was separated from the fuel, encapsulated in a cementitious matrix and sent to a temporary storage facility, with the ultimate objective being their permanent disposal in a future national GDF. This approach represents the "take-make-dispose" principle of the traditional linear economy. The recycling of zirconium alloy, on the other hand, embodies the principle of the circular economy, with two notable advantages. First, zirconium alloy cladding represents a substantial portion (~70% by volume (Collins et al., 2011; J. D. Vienna et al., 2016)) of ILW that arise from UNF recycling and need to be disposed of in a GDF (Paulillo, 2018); therefore, reducing the amount of zirconium alloy waste could entail a reduction of the GDF footprint (required for disposing of UNF recycling wastes) and, potentially, also in the radiological impacts that will arise from the GDF. Second, the recycling of zirconium alloy makes available zirconium that would otherwise be wasted, thus reducing the demand for extraction of primary zirconium - an activity with high environmental impacts; the recovered zirconium could be used, for example, in a closed loop to manufacture new zirconium alloy cladding. We also note that the recycling of zirconium alloy may also unlock the possibility of controlling the isotopic enrichment of the alloy, which among other things could reduce the radiological dose of the used cladding; this scenario is outside the scope of this work but could be part of future research efforts.

Zirconium is primarily used in the nuclear industry and in some applications in the chemical industries (including catalysis and gas purification), and has an annual production exceeding 7000 tones/year (Yagnik and Garde, 2019a). Zirconium alloy is commercially obtained from zirconium ore via the Kroll process; this produces a zirconium sponge that is alloyed with other elements (such as iron, nickel, tin and chromium) and then arc melted under vacuum. The produced zirconium alloy ingots then undergo a series of processes including forging, extrusion, pilgering and heat treatment that generate seamless cladding tubes (Yagnik and Garde, 2019a). Fig. 2 shows a schematic diagram of the life-cycle of zirconium alloy cladding, including both linear and circular approaches.

Lab-scale experiments have demonstrated that zirconium can successfully be extracted from zirconium alloy cladding through

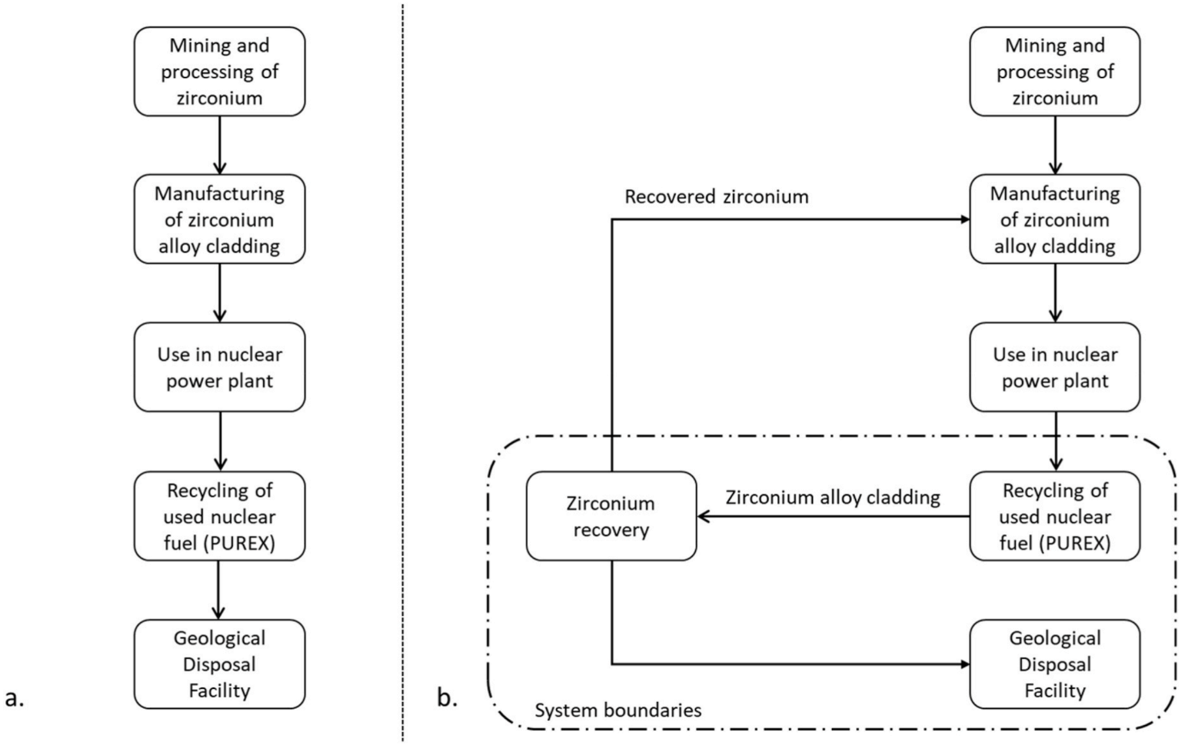

Fig. 2. Simplified schematic diagram of the life-cycle of zirconium alloy cladding for linear (a.) and circular (b) approaches. The system boundary considered in this study for the circular approach is represented by a grey dash-dotted line. The zirconium alloy cladding undergoes a re-processing in a recovery facility; the outputs are recovered zirconium that is re-circulated into the manufacturing of zirconium alloy cladding, and a waste stream going to the GDF.

chlorination or hydrochlorination (Collins et al., 2011, 2012). The reactions, which are carried out at high temperature (∼350 °C), generate a salt, zirconium tetrachloride ($ZrCl_4$), which represents the input to the Kroll process (Collins et al., 2012; Yagnik and Garde, 2019b). A subsequent purification step is required to remove alloying elements like niobium and iron. Collins et al. (2012) maintain that this step can be conducted in an unshielded facility because of low radiation doses (<5.6 nSv/s) of the salt product, whilst another study based on high burn-up fuel cladding (Spencer et al., 2017) suggests the opposite, due to high concentrations of high volatile species such as ^{94}Nb and ^{125}Sb in the $ZrCl_4$.

2.2.1. Goal and scope definition

The goal of this case study is to investigate the potential environmental benefits of recycling zirconium alloy that is used as cladding for nuclear fuels. To this end, we first quantify the environmental impacts associated with zirconium alloy recycling; we then compare the environmental impacts of the circular approach with the traditional, linear approach that envisages the disposal of zirconium alloy cladding waste in a GDF. The study adopts a consequential perspective, answering the question “what are the environmental consequences of recycling zirconium alloy cladding waste?” (Guinée et al., 2011; Hauschild et al., 2018). We only account for short-term effects, which include first order direct physical effects, but not second and third order effects, which are known as negative and positive feedback effects[1] (Sandén and Karlström, 2007). Fig. 2 reports the system boundaries of the LCA study, for both circular and linear approaches. For both approaches, the system boundaries start from the activity the generates zirconium alloy cladding waste through the recycling of used nuclear fuels. We include this activity in the system boundary even though it is in common to both approaches because we aim at quantifying the environmental benefits of moving from a linear to a circular approach for the entire back-end of the nuclear fuel cycle, that is the recycling of nuclear fuels and the disposal of radioactive wastes. We assume that zirconium alloy cladding waste arise from Light Water Reactor (LWR) used fuels that are recycled through the PUREX process, which was the UNF recycling process historically adopted at Sellafield’s THORP prior to its closure in 2018. A description of the PUREX process is provided by Wilson (1996), whilst the environmental impacts of the THORP plant have been investigated in detail by Paulillo et al. (2020d). Cladding waste includes chopped up sections of tubing also known as “hulls” that arise in the “Head End” step of the PUREX process, where sheared fuel assemblies are dissolved in nitric acid; this enables separating the cladding from the solution containing the nuclear fuel. The linear approach (with respect to Zr), historically implemented at Sellafield, envisages that the hulls are sent to the waste encapsulation plant (WEP), where they are immobilized in cement grout and encapsulated in 500 L stainless steel drums. The final waste product is classified as ILW and temporary stored onsite pending its permanent disposal in the GDF (Paulillo et al., 2020d).

In a future circular approach we assume that the hulls are instead sent to a two-step recycling process, which reduces the concentration of impurities enabling the re-use of zirconium. However, the application of recovered zirconium is limited to the nuclear industry because it contains ^{93}Zr, which has low radioactivity and weak beta emissions (Collins et al., 2011; Spencer et al., 2017). In the first step, zirconium alloy hulls are treated with chlorine (Cl_2) at high temperatures to generate $ZrCl_4$ (Spencer et al., 2017). In the second step, a sublimation-based purification process is carried out to remove niobium, iron and other alloying elements or impurities that were present in the cladding. The purification step consists in two chemical treatments: the first one removes

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[1] Note that first order are direct effects related to changes in the material/energy flows of production/consumption of products (e.g. replacing natural with recycled uranium yields lower environmental impacts). Second orders effects take into account negative feedback mechanisms related to economic aspects, like changes in price due to a constrained supply. Third order effects encompass positive feedback mechanisms related to e.g. improvements in technologies or knowledge generation.

niobium in the form of high volatility metal chloride ($NbCl_5$), through chlorination with thionyl chloride ($SiOCl_2$); the second treatment targets iron by reducing the volatile $FeCl_3$ into the non-volatile species $FeCl_2$ with hydrogen gas and collection of zirconium chloride via bulk sublimation. We make two important assumptions in modelling the circular approach: first, that the recycled zirconium retains the same properties of primary zirconium, and therefore that it can be replaced in a 1:1 ratio; and second, that unreacted cladding and other wastes arising from the recycling process are classified as ILW and treated similarly to cladding waste as in the linear approach. This conservative assumption stems from lack of data in the literature and the fact that treatments for other waste types (e.g. hazardous, industrial waste) typically have lower environmental impacts. The functional unit corresponds to the recycling of 1 tonne of uranium pre-irradiation at its end-of-life, which generates 323 kg of zirconium alloy cladding waste (Paulillo, 2018; Paulillo et al., 2020d).

2.2.2. *Allocation*

The circular approach to managing zirconium alloy cladding waste described above provides two functions: it manages a waste stream whilst producing a valuable output – zirconium – that can be used, for example, in a closed loop to manufacture additional zirconium alloy cladding (see Fig. 2). Multi-functional systems represent a methodological challenge in LCA because they require allocating the environmental impacts amongst multiple functions. Numerous allocation strategies are available in the literature, and each strategy can lead to significantly different results. In this study we implement a consequential perspective (with inclusion of first order direct effects; see section 2.2.1) by accounting for the avoided burdens associated with changes in demand or production. In this case, the recovery of zirconium is assumed to induce a reduction in demand for primary zirconium; the credits represent the avoided environmental impacts associated with the production of primary zirconium.

2.2.3. *Life cycle inventory*

Inventory data for the linear approach – which includes a PUREX plant, ancillary waste management plants and the geological disposal facility – is obtained from the work of Paulillo and colleagues (2018; 2020d) and is based on high-quality collected on site. We adapted their inventory to be applicable to the management of LWR fuels, whilst the original model was developed for Advanced Gas-cooled Reactors (AGR) ones. The adapted inventory features two key differences: first, the weight of hulls per ton of uranium pre-irradiation, which is 323 kg for LWR fuels; and second, the absence of graphite and stainless steel fuel assembly components that are removed from AGR fuels prior to recycling.

The recycling of zirconium alloy cladding is modelled using data from laboratory experiments from the literature. Inventory data for the first step, i.e. the chlorination of zirconium alloy cladding, was obtained from Spencer et al. (2017) and is summarised in Table 1. The laboratory-based process only requires chlorine as input, the amount has been estimated from the stoichiometric reaction. We estimate the consumption of thermal energy from the temperature at which the process is carried out (∼320–350 °C), using the approach proposed by Piccinno et al. (2016) and assuming an efficiency of the heating device of 72%. As noted in Section 0, we also assume that wastes generated from chlorination, which include unreacted cladding and ash residues, are classified as intermediate-level waste (ILW) and treated accordingly. Laboratory experiments showed that about 96% of cladding are chlorinated. 70% of the zirconium in chlorinated cladding reacts to generate $ZrCl_4$, whilst a small amount (8%) is found in the ash as zirconium oxide (ZrO_2). The remaining zirconium was retained in the test apparatus, implying that the $ZrCl_4$ salt product contains a greater amount of zirconium (than 70%) though the exact amount could not be determined (Spencer et al., 2017).

Table 1
Inventory data for the chlorination step.

INPUTS	QUANTITY & UNIT
Zirconium alloy **cladding**	323 kg
Chlorine, Cl_2	251.01 kg
Thermal energy	483.80 MJ
OUTPUTS	**QUANTITY**
Unreacted cladding	12.92 kg
Chlorinated cladding	310.08 kg
Of which	
Zr, as $ZrCl_4$ salt	217.06 kg
Zr, as ZrO_2 in ash residues	23.57 kg
Other residues	116.58 kg

Table 2 reports the inventory data for the purification step, which is based on (Barnes et al. n.d.). Like in the previous step, we estimated material inputs from stoichiometric reactions and the consumption of thermal energy using the process temperature, and we assumed that all wastes generated are treated as ILW. The purification step is modelled with a yield of 89%, in accordance with results from laboratory experiments.

Overall, the two-step recycling process features a yield of 60%, that is, 194 kg of purified $ZrCl_4$ are obtained from 323 kg of zirconium alloy claddings. Finally, the background system for both linear and circular approaches is modelled using average market data obtained from Ecoinvent (version 3.6) (Wernet et al., 2016) and Sphera (service package 40) databases (Kupfer et al., 2020; Sphera, 2020).

2.3. *Case study n°2: uranyl nitrate from depleted uranium*

The second LCA case study deals with uranyl nitrate – $UO_2(NO_3)_2$ – a water-soluble, yellow uranium salt that is prepared by reaction of uranium with nitric acid and that is used in the PUREX process. Paulillo et al. (2020d) found that a substantial portion of the historical environmental impacts of recycling used nuclear fuels in THORP at Sellafield originated from the usage of uranyl nitrate, and more specifically from the mining and further processing of uranium from which uranyl nitrate is obtained. Building upon these findings, we investigate the environmental performance of recycling used nuclear fuels when uranyl nitrate is obtained from depleted uranium (DepU) (US Nuclear Regulatory Commission (NRC), 2021), which for this analysis is assumed to arise from enrichment tails waste. This approach, which also embodies the principles of the circular economy, has two advantages: first, it avoids mining of uranium – a high-impact activity; and second it prevents disposal of DepU, reducing the amount of waste (though very small) to be accommodated in the national GDF.

2.3.1. *Goal and scope definition*

The goal of this high-level study is to assess the environmental benefits of recycling used nuclear fuels in a PUREX process using uranyl nitrate obtained from DepU. Uranyl nitrate was used as feedstock to make a reductant used in the THORP process to facilitate the separation of plutonium from uranium. The uranyl nitrate feedstock is mainly

Table 2
Inventory data for the purification step.

INPUTS	QUANTITY & UNIT
Zr, as $ZrCl_4$ salt	217.06 kg
Nitrogen, N_2	21.6 kg
$SOCl_2$	13.7 kg
Hydrogen, H_2	0.5 kg
Thermal energy	327 MJ
OUTPUTS	**QUANTITY**
Purified Zr, as $ZrCl_4$ salt	193.83 kg
Residues	23.23 kg

produced from natural uranium but using the depleted uranium (DepU) left from enrichment operations would avoid utilising freshly mined uranium. (Note that DepU could also be used to produced Mixed Oxide Fuel, a scenario that was investigated in Paulillo et al. (2021). We compare the environmental impacts of this approach with the baseline scenario that envisages using natural uranium to produce uranyl nitrate. Like the first case study (Section 0), this study adopts a consequential perspective with inclusion of first order direct physical effects.

Fig. 3 reports schematic diagrams of the system boundaries for the baseline and the circular economy DepU-based alternative, respectively. In both approaches, LWR used fuels are assumed to be recycled via the PUREX process. Uranium and plutonium that are obtained from recycling are outside the system boundaries and therefore not considered any further; this is in line with the previous study (Paulillo et al., 2020d). Fission products and other waste streams that arise from PUREX are treated by several ancillary plants, with the resulting solid radioactive waste being prepared for final disposal in the national GDF. The baseline and circular economy scenarios differ only with respect to uranyl nitrate, which is obtained from natural uranium in the baseline and from depleted uranium in the alternative scenario. Specifically, it is assumed that the circular economy scenario uses DepU that is generated by enrichment activities and that would otherwise have to be disposed of in a GDF. Following Paulillo et al. (2020d), the functional unit corresponds to the management of 1 tonne of uranium pre-irradiation at its end-of-life.

2.3.2. *Allocation*

The circular economy scenario, which assumes uranyl nitrate being obtained from depleted uranium, represents a multi-functional system that recycles UNF and, at the same time, that manages depleted uranium, a waste that would otherwise be disposed in a GDF. Similar to the previous case study (section 0), we assume that the use of depleted uranium in uranyl nitrate reduces the demand for disposal of DepU in a GDF; therefore, the associated credits represent the avoided environmental impacts of DepU disposal.

2.3.3. *Life cycle inventory*

As noted in Section 0, this case study builds upon the work of Paulillo (2018; 2020d). This provides the inventory data which is based on a combination of site-specific data from Sellafield Ltd. and literature data;

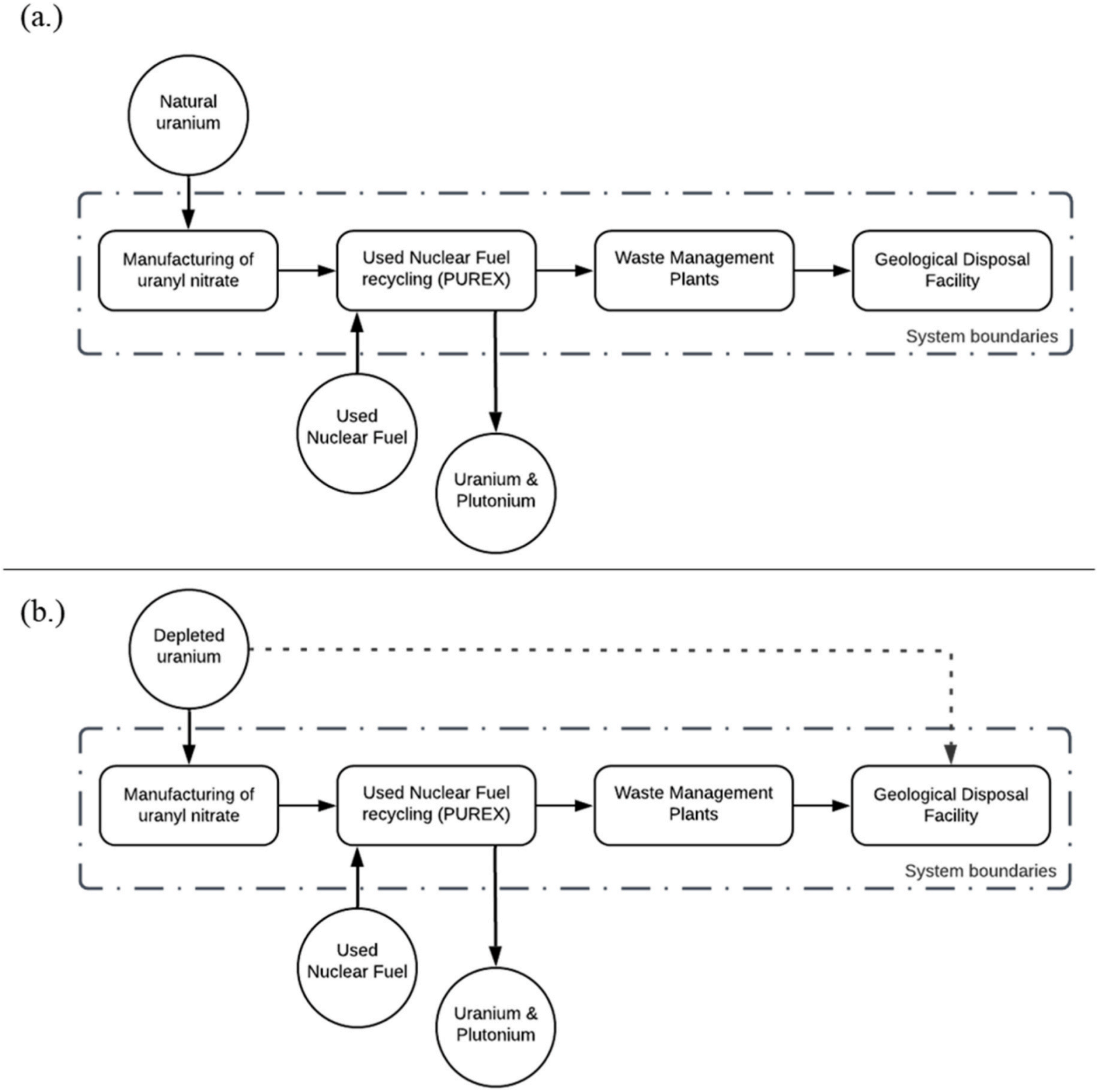

Fig. 3. Schematic diagrams of the system boundaries for the baseline scenario (a) and the alternative (b), where the depleted uranium substitute natural uranium. The dashed-line in figure (b) represent the avoided impacts due to such substitution.

the inventory for the baseline scenario is fully reported in (Paulillo, 2018). The background system is modelled with average market data obtained from ecoinvent database (v. 3.6) (Wernet et al., 2016). Uranyl nitrate, which is not covered in LCA databases, has been accounted only for the burdens of the reagents required for its production, according to stoichiometric ratios between yellowcake (i.e. uranium ore) and nitric acid (50% mol) in a ~3.3:1 M ratio (Paulillo et al., 2020d). The amount of uranyl nitrate and that of its reagents for the management of 1 ton of uranium pre-irradiation (i.e. the functional unit) are reported in Table 3.

2.4. Impact assessment

Environmental impacts are calculated using the Environmental Footprint 2.0 method of the Joint Research Centre (European Commission, 2013; Zampori and Pant, 2019) and the UCrad model for radiological impacts developed by Paulillo et al. (2020b, 2020c, 2020a) (see Section 0). The environmental categories investigated are reported in Table 4. The life-cycle modelling has been performed in GaBi software by Sphera (2020).

3. Results and discussion

3.1. Case study n°1: zirconium alloy recycling

The environmental impacts of both linear and circular approaches to managing zirconium alloy cladding waste are reported in numerical form in the Appendix (Table A1). Fig. 4 reports the relative changes between the circular approach and the traditional linear approach; these are obtained as the difference between the environmental impacts of the circular approach and the linear approach, relative to those associated with the linear approach, according to equation [1]:

$$\Delta\% = 100 \times \frac{circular\ app. - linear\ app.}{linear\ app.} \quad [1]$$

Fig. 4 shows that the circular approach achieves a reduction in the environmental impacts associated with UNF recycling across the full spectrum of environmental categories. The most significant environmental benefits are found in the categories “resource use, minerals and metals” (−25%), “water scarcity” (~-14%) and “ionising radiation, waste” (−5.5%). The reduction in the depletion of resources is primarily due to the avoided environmental impacts of mining primary zircon sand where zirconium is found as zirconium silicate (Gediga et al., 2019). On the other hand, the reduction in the amount of ILW that is sent to the GDF is responsible for the decline in radiological impacts from radioactive waste disposal and water use. A more modest reduction is found for the climate change category (~2%), whilst impact scores in the remaining categories range from 0.1% for “resource use, energy carriers” and up to 1% for “ozone depletion”.

The recycling of zirconium alloy cladding waste brings only modest improvements in the environmental performance of UNF recycling; this is because the management and disposal of cladding waste do not represent a large contribution to the overall environmental impacts (Paulillo et al., 2020d). The difference in environmental performance between the circular and linear approach is in fact significant when excluding all activities that are in common between the linear and circular approaches (i.e. PUREX and its ancillary plants, and the GDF). This comparison, which is reported in Figure A1 in the Appendix, demonstrates that at process-level the recycling of zirconium alloy attains a 2.8 times reduction in the category “resource use, minerals and metals”, and between 50% and 80% in the remaining categories.

Table 3
Amount of uranyl nitrate and reagents required for the functional unit.

	QUANTITY & UNIT
Uranyl nitrate	82.6 kg
Of which	
Natural uranium (yellowcake)	97.6 kg
Nitric acid	87.4 kg

Table 4
Environmental categories analysed.

IMPACT CATEGORY	UNIT
Acidification terrestrial and freshwater	Mole of H^+
Cancer human health effects	CTUh
Climate Change	kg CO_2
Ecotoxicity freshwater	CTUe
Eutrophication freshwater	kg P eq.
Eutrophication marine	kg N eq.
Eutrophication terrestrial	Mole N eq.
Land use	Pt
Non-cancer human health effects	CTUh
Ozone depletion	kg CFC-11 eq.
Photochemical ozone formation	kg NMVOC eq.
Resource use, energy carriers	MJ
Resource use, mineral and metals	kg Sb eq.
Respiratory inorganics	Disease incidents
Water scarcity	m^3 world eq.
Ionising radiation, waste (in GDF)	Bq U238 ILW- eq.
Ionising radiation	Bq U235 air- eq.

Fig. 5 reports a hot-spot analysis at process-level for the recycling of zirconium alloy cladding, which includes the two-step process for recovering zirconium, treatment and disposal of wastes and the credits for producing zirconium. The chart shows the percentage contribution of each process to the absolute impact value in each environmental category; note that we consider impacts in absolute terms to enable comparison between impacts and credits (i.e. impacts with negative sign). For example, the category climate change is dominated by the disposal of wastes (green bar); the two-step recycling process contributes to 20% (pink bar) of the absolute impacts whilst the credits for zirconium recycling have even lower contribution, equal to −6%. From the graph it can be seen that most of the environmental impacts originate from the treatment and disposal of the wastes generated by the recycling process, whilst the zirconium recovery process has only minor contributions. On the other hand, the credits associated with the displacement of primary zirconium are substantial, and even greater than the environmental impacts in the category “resource use, minerals and metals”; notably, this means that in this category the process brings a reduction in environmental impacts. This demonstrates the exceptional performance of the circular approach in this category when compared to the linear approach (see Fig. 4 shows and Figure A1 in the Appendix).

It must be noted that the results presented in this section present a high level of uncertainty due to the fact that inventory data on the recycling process comes from experiments at laboratory scale. On the other hand, the use of laboratory-based data makes the our results conservative because they overestimate the environmental impacts associated with the recycling of zirconium alloy cladding (see Section 0); this implies that the environmental benefits of the circular approach could be even more significant. The hot-spot analysis shows that the majority of the environmental impacts are due to treatment of ILW wastes; the process environmental performance would improve considerably if the wastes were not to be treated as ILW – e.g. they could be classified as low-level waste (LLW) and disposed in a near-surface repository, or as hazardous or industrial waste. The low technological readiness level (TRL) of the recycling process also entails that its environmental impacts are likely to diminish when moving to an optimised commercial scale; this however would affect the environmental performance to a lesser extent than the assumption on waste classification. Finally, in showing the credit contributions, the hot-spot analysis demonstrates the relative importance of the allocation strategy, i.e. the avoided burdens approach described in Section 2.2.2, on the LCA results. This implies that the LCA results may change if zirconium

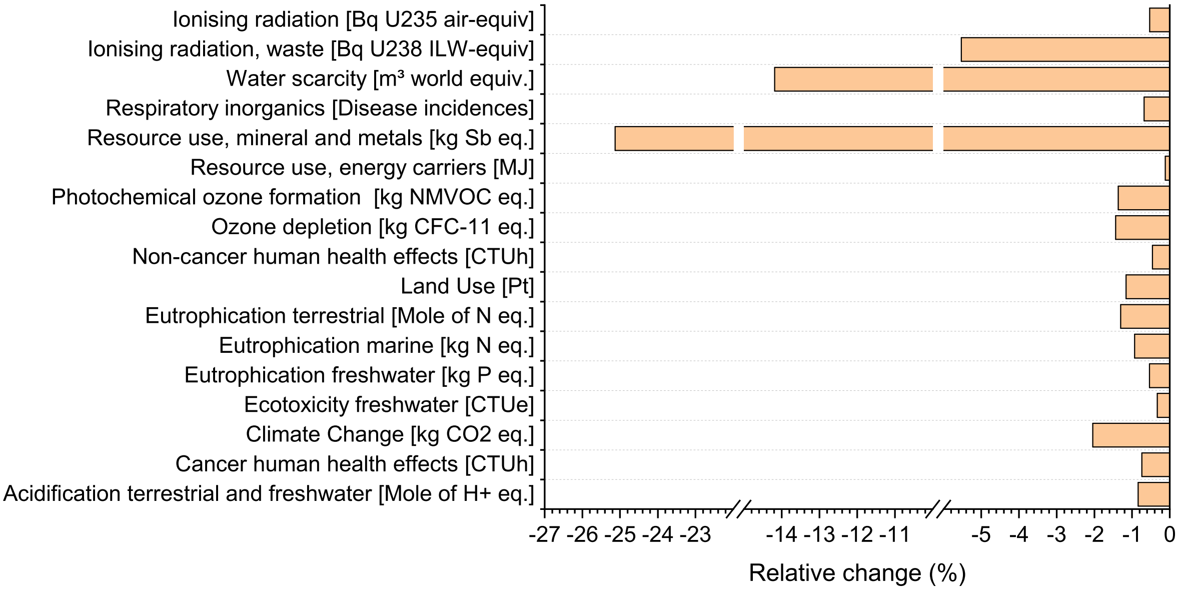

Fig. 4. Relative changes in environmental impacts of used nuclear fuels recycling when moving from a linear to circular approach for the management of zirconium alloy cladding waste.

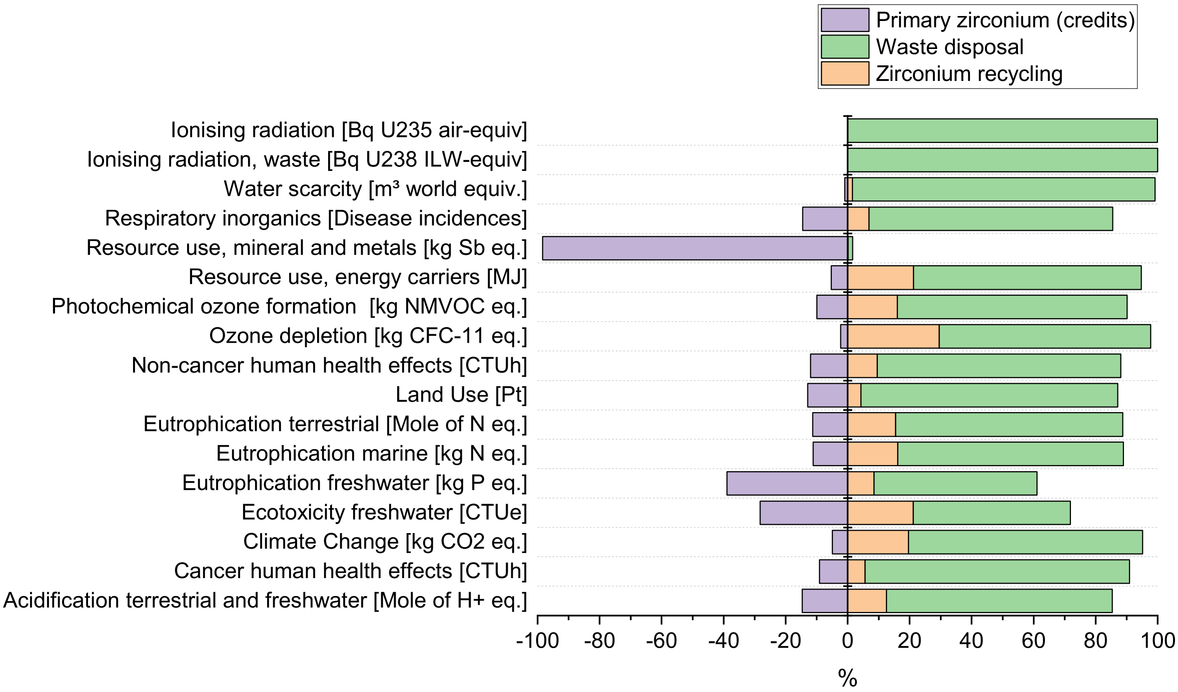

Fig. 5. Hot-spot analysis for zirconium alloy cladding recycling. “Primary zirconium (credits)” shows the negative contribution associated with the avoided activity of mining primary zirconium to the total environmental impacts.

recycling fails to induce a reduction in demand for primary zirconium.

3.2. Case study n°2: uranyl nitrate from depleted uranium

Fig. 6 reports the relative changes between the DepU-based alternative and the baseline, with the numerical values reported in the Appendix (Table A2). The relative changes are obtained as difference between the environmental impacts of the DepU-based alternative and the baseline, relative to those associated with the baseline, according to equation [2]:

$$\Delta\% = 100 \times \frac{DepU\ alternative - Baseline}{Baseline} \qquad [2]$$

The chart shows that producing uranyl nitrate from depleted, instead of natural, uranium yields substantial environmental benefits. The circular economy scenario decreases the environmental impacts of UNF recycling from 3% in the category “land use” and up to 94% in the category “resource use, energy carriers”, with GHG emissions being reduced by ~4%. Most of the environmental benefits are due to the avoided activities related to the extraction and post-processing of natural uranium, which are particularly significant in the categories dealing with radiological impacts (−18%), resource use-energy carriers (−94%),

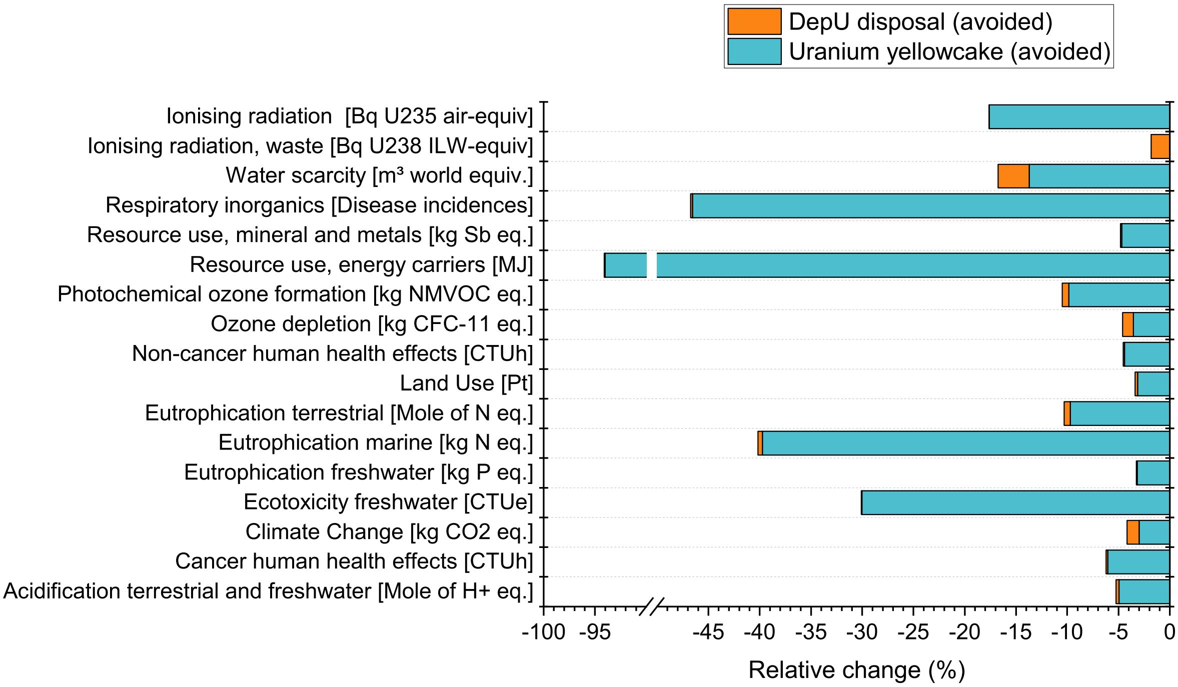

Fig. 6. Relative changes between DepU alternative and baseline.

respiratory inorganics (−47%), marine eutrophication (42%) and freshwater ecotoxicity (−30%). The credits associated with the avoided disposal of DepU in a GDF have non-negligible contributions only with respect to radiological impacts from nuclear waste disposal, water scarcity and climate change. Overall, these results confirm the findings of Paulillo et al. (2020d) in identifying uranyl nitrate (used as a reagent) as a key contributor to the environmental impacts of recycling used nuclear fuels. We note that, similar to Section 3.1, the LCA results presented in this Section are also uncertain, partly because the case study is based on extrapolation from historical operational data in the UK where DepU was not routinely utilised.

3.3. Integration of case study n°1 and n°2

In Fig. 7 we combine both case studies – i.e. zirconium alloy hulls recycling and uranyl nitrate from DepU – which we compare with a baseline recycling scenario that envisages no recycling of cladding and producing uranyl nitrate from natural uranium; the numerical values are reported in the Appendix (Table A3). The comparison is based on the same functional unit (see Section 2.2.1 and 0), which corresponds to the recycling of 1 tonne of uranium pre-irradiation at its end-of-life. The chart shows that the implementation of both circular approaches would reduce the environmental impacts of UNF recycling from ~4% in the category freshwater eutrophication and up to 94% in the depletion of resources-energy carriers. GHG emissions would be reduced by 6%, whilst radiological impacts of direct operational emissions and those

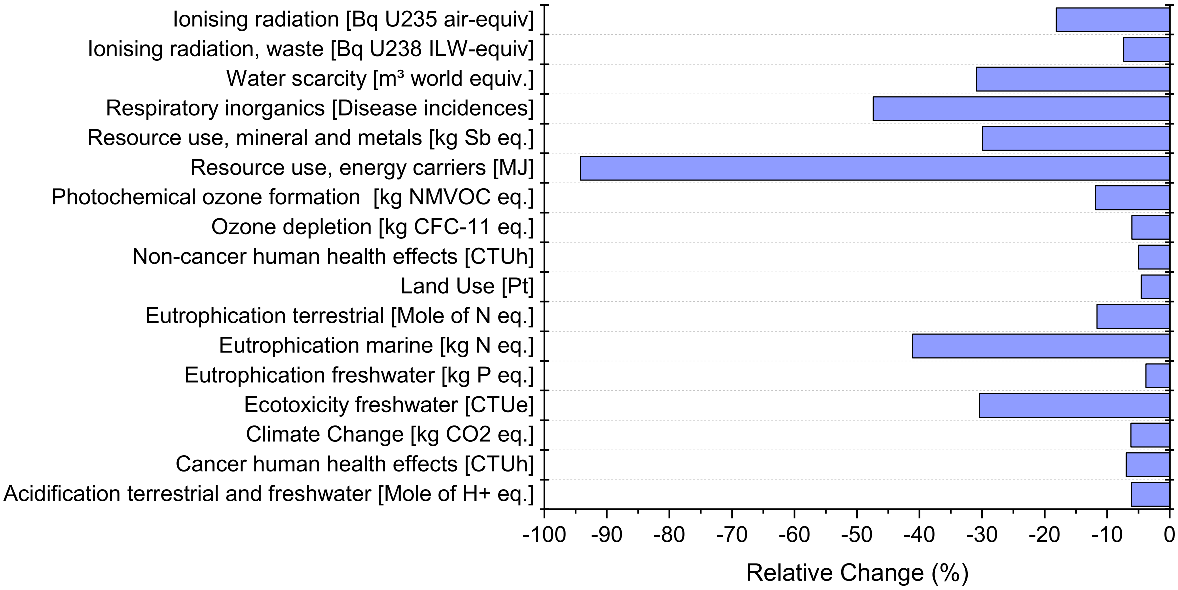

Fig. 7. Relative changes between the baseline scenario and the implementation of recycling zirconium alloy cladding and producing uranyl nitrate from DepU.

arising from nuclear waste disposal would decrease by 18% and 2%, respectively. Table A3 shows that the improvements in most of the environmental categories are driven by the replacement of natural with depleted uranium for the manufacture of uranyl nitrate; this is expected given the magnitude of the relative changes reported in Figs. 4 and 6. The recycling of hulls, on the other hand, is the primary cause for the reduction in the radiological impacts from wastes and use of mineral and metal resources; the two scenarios contribute similarly to the reduction in water consumption. As noted in the previous sections, the uncertainty of our results is high due to the nature of the underlying inventory data. No uncertainty analysis could be performed because we could not find any information on the data's potential variability; this aspect should be assessed in future works.

The interpretation of these LCA results – including those presented in Section 3.1 and 3.2 – is straightforward because one scenario is environmentally preferable across all environmental categories; this is because we compare traditional approaches with circular one that are assumed to induce a reduction in the demand for mining and other high-impact activities. The application of LCA not only confirms the environmental advantages of circular approaches, but it also provides a quantitative basis using a standardised methodology that ensures robust and replicable results. However, the majority of LCA studies require consideration of trade-offs between environmental categories. A notable example is the comparison between once- and twice-through cycles: the former may be environmentally advantageous with respect to radiological impacts from direct discharges, but the latter is preferable in the remaining categories (Paulillo et al., 2021). The comparison between fossil fuels and first generation biofuels represents another notable example, with the latter being preferable in terms of carbon emissions but disadvantageous with respect to land use and other categories that are affected by the use of fertilizers (Vedel Hjuler and Balle Hansen, 2018).

Finally, we note that the environmental impacts determined by LCA are only part of the sustainability of a system. The economic impacts of the changes to the technology and the societal benefits should also be considered in the overall sustainability assessment for a robust decision-making. For example, the economic advantages of the open fuel cycles compared to the closed counterpart (in terms of lower uranium prices as well as short-term capital investment) (Taylor et al., 2022b) are among other aspects likely to have led most countries to opt for an open fuel cycle (IAEA, 2022), despite the environmental benefits of recycling used nuclear fuels (Paulillo et al., 2021; Taylor et al., 2022a). Future work should therefore complement our LCA results with socio-economic indicators. Another key consideration concerns the technological maturity of the proposed changes to meet increased circularity of the recycling process, noting that other changes to processes may bring even greater benefits.

4. Conclusions

LCA represents an appropriate tool to quantitatively investigate the environmental benefits (or drawbacks) of circular economy strategies. This article demonstrates by example how LCA can be used to assess circular economy strategies and hence to support decisions in the nuclear industry. LCA can be used as a standalone tool for decision-support or as part of multi-criteria assessment considering various aspects including economic and social ones.

Building on previous analyses of direct disposal in an open cycle versus recycling in a closed fuel cycle, we presented two case studies related to the recycling of used nuclear fuels using the PUREX process as implemented in THORP. The first investigates the potential environmental benefits of recycling zirconium alloy cladding waste, compared to the traditional approach that envisages their encapsulation in a wasteform ready for disposal in a geological disposal facility (GDF). The LCA results indicate that the circular approach outperforms the traditional one across the full spectrum of environmental categories; and that switching from a linear to a circular approach would bring sizeable improvements in the environmental performance of recycling used nuclear fuels (UNF). The environmental performance of the circular approach is expected to be even higher when the process is developed at a commercial scale. The second case study investigates the environmental performance of producing uranyl nitrate using depleted uranium, instead of natural uranium. The LCA results show considerable environmental benefits across all environmental categories, which are primarily due to avoided activities that are related to the production of uranium.

The application of LCA to the case studies presented above enabled i) confirming the environmental superiority of circular economy strategies for recycling zirconium and reusing depleted uranium compared to a baseline scenario where these are directed to disposal and ii) quantifying the extent of the environmental benefits with respect to the back-end of the nuclear fuel cycle. The interpretation of the results was straightforward because the circular economy strategies investigated resulted in improvements across all environmental categories; however, it must be noted that the majority of LCA studies require consideration of trade-offs (like in the comparison between once- and twice-through fuel cycles). The LCA methodology enables considering these trade-offs during the decision-making process; notably, this should be complemented with analyses of the wider socio-economic aspects so that all aspects of sustainability are considered.

CRediT authorship contribution statement

Martina Pucciarelli: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Visualization, Writing - original draft. **Stephen J. Palethorpe:** Conceptualization, Writing - review & editing. **Julian Spencer:** Conceptualization, Project administration, Writing - review & editing. **Anthony Banford:** Resources, Writing - review & editing. **Paola Lettieri:** Supervision, Writing - review & editing. **Andrea Paulillo:** Conceptualization, Methodology, Project administration, Supervision, Writing - original draft, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Inventory data is provided in the article or in the Appendix.

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Appendix

Table A1
Environmental impacts of the management of zirconium alloy cladding waste in linear and circular approaches.

IMPACT CATEGORIES	Linear approach - Baseline	Circular approach		
	PUREX & WTP- TOTAL	Zircaloy™ cladding disposal – avoided impacts	Zircaloy™ recycling	TOTAL
Acidification terrestrial and freshwater [Mole of H+ eq.]	2.77E+03	−3.83E+01	1.50E+01	2.75E+03
Cancer human health effects [CTUh]	4.37E-04	−5.28E-06	2.05E-06	4.33E-04
Climate Change [kg CO2 eq.]	3.04E+05	−1.20E+04	5.79E+03	2.98E+05
Ecotoxicity freshwater [CTUe]	3.27E+07	−1.66E+05	5.76E+04	3.26E+07
Eutrophication freshwater [kg P eq.]	2.39E+02	−1.56E+00	2.63E-01	2.38E+02
Eutrophication marine [kg N eq.]	5.86E+02	−9.63E+00	4.16E+00	5.81E+02
Eutrophication terrestrial [Mole of N eq.]	4.78E+03	−1.09E+02	4.64E+01	4.72E+03
Land Use [Pt]	2.21E+06	−4.03E+04	1.46E+04	2.19E+06
Non-cancer human health effects [CTUh]	2.44E-02	−1.85E-04	7.24E-05	2.43E-02
Ozone depletion [kg CFC-11 eq.]	4.03E-02	−1.34E-03	7.60E-04	3.97E-02
Photochemical ozone formation - human health [kg NMVOC eq.]	1.29E+03	−3.13E+01	1.36E+01	1.27E+03
Resource use, energy carriers [MJ]	6.88E+07	−1.61E+05	7.92E+04	6.87E+07
Resource use, mineral and metals [kg Sb eq.]	1.39E+01	−1.29E-01	−3.36E+00	1.04E+01
Respiratory inorganics [Disease incidences]	2.48E-02	−2.66E-04	9.68E-05	2.47E-02
Water scarcity [m^3 world equiv.]	1.37E+05	−3.27E+04	1.33E+04	1.17E+05
Ionising radiation, waste [Bq U238 ILW-eq.]	6.30E+10	−5.84E+09	2.36E+09	5.95E+10
Ionising radiation [Bq U235 air-eq.]	2.28E+09	−2.04E+07	8.27E+06	2.27E+09

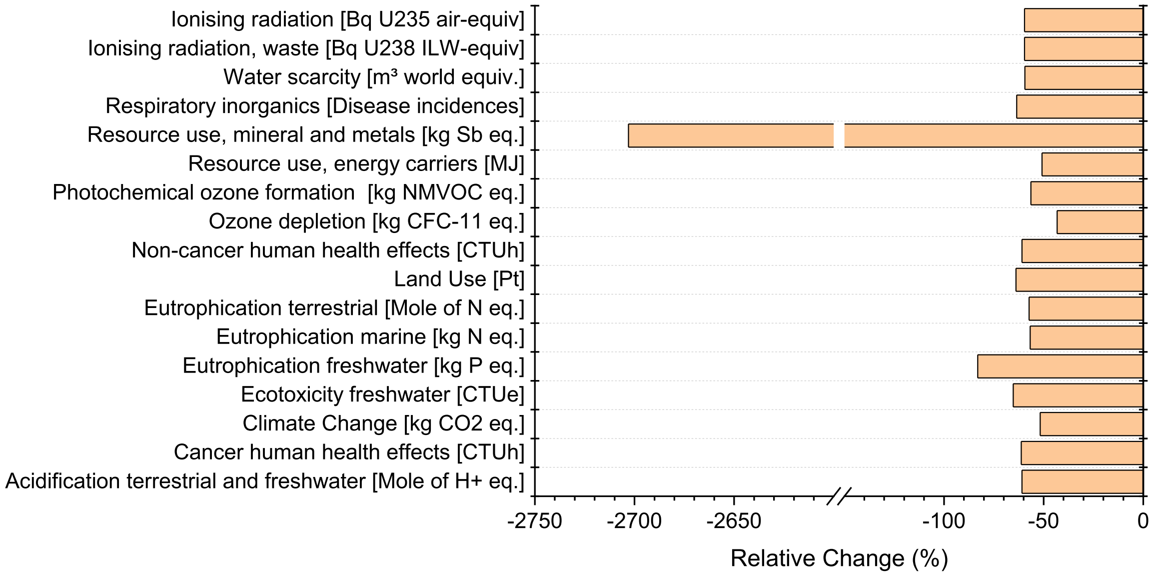

Fig. A2. Relative changes at process level between circular and linear approaches to management of zirconium alloy cladding waste.

Table A2
Environmental impacts of baseline and DepU-based alternative for uranyl nitrate scenarios.

IMPACT CATEGORIES	PUREX - baseline	PUREX – Uranyl nitrate from DepU	Relative change (%)
Acidification terrestrial and freshwater [Mole of H+ eq.]	2.77E+03	2.63E+03	−5%
Cancer human health effects [CTUh]	4.37E-04	4.09E-04	−6%
Climate Change [kg CO2 eq.]	3.04E+05	2.91E+05	−4%
Ecotoxicity freshwater [CTUe]	3.27E+07	2.29E+07	−30%
Eutrophication freshwater [kg P eq.]	2.39E+02	2.31E+02	−3%
Eutrophication marine [kg N eq.]	5.86E+02	3.51E+02	−40%
Eutrophication terrestrial [Mole of N eq.]	4.78E+03	4.29E+03	−10%
Land Use [Pt]	2.21E+06	2.14E+06	−3%
Non-cancer human health effects [CTUh]	2.44E-02	2.33E-02	−5%
Ozone depletion [kg CFC-11 eq.]	4.03E-02	3.84E-02	−5%
Photochemical ozone formation - human health [kg NMVOC eq.]	1.29E+03	1.16E+03	−10%
Resource use, energy carriers [MJ]	6.88E+07	4.06E+06	−94%
Resource use, mineral and metals [kg Sb eq.]	1.39E+01	1.32E+01	−5%

(*continued on next page*)

Table A2 (*continued*)

IMPACT CATEGORIES	PUREX - baseline	PUREX – Uranyl nitrate from DepU	Relative change (%)
Respiratory inorganics [Disease incidences]	2.48E-02	1.32E-02	−47%
Water scarcity [m^3 world equiv.]	1.37E+05	1.14E+05	−17%
Ionising radiation, waste [Bq U238 ILW-eq.]	6.30E+10	6.18E+10	−2%
Ionising radiation [Bq U235 air-eq.]	2.28E+09	1.88E+09	−18%

Table A3

Environmental impacts of PUREX-baseline system and the alternative system implementing both zirconium alloy recycling and DepU-based alternative for uranyl nitrate production.

IMPACT CATEGORIES	PUREX - baseline	PUREX – zirconium alloy recycling and uranyl nitrate from DepU	Relative change (%)	*Relative changes due to the zirconium alloy recycling scenario (%)*	*Relative changes due to the uranyl nitrate from DepU scenario (%)*
Acidification terrestrial and freshwater [Mole of H+ eq.]	2.77E+03	2.60E+03	−6.11%	*−5.3%*	*−0.9%*
Cancer human health effects [CTUh]	4.37E-04	4.06E-04	−6.97%	*−6.3%*	*−0.8%*
Climate Change [kg CO2 eq.]	3.04E+05	2.85E+05	−6.21%	*−4.3%*	*−2.1%*
Ecotoxicity freshwater [CTUe]	3.27E+07	2.28E+07	−30.43%	*−30.2%*	*−0.5%*
Eutrophication freshwater [kg P eq.]	2.39E+02	2.30E+02	−3.80%	*−3.3%*	*−0.6%*
Eutrophication marine [kg N eq.]	5.86E+02	3.45E+02	−41.12%	*−40.6%*	*−1.6%*
Eutrophication terrestrial [Mole of N eq.]	4.78E+03	4.23E+03	−11.62%	*−10.5%*	*−1.5%*
Land Use [Pt]	2.21E+06	2.11E+06	−4.55%	*−3.4%*	*−1.2%*
Non-cancer human health effects [CTUh]	2.44E-02	2.32E-02	−5.01%	*−4.6%*	*−0.5%*
Ozone depletion [kg CFC-11 eq.]	4.03E-02	3.78E-02	−6.05%	*−4.7%*	*−1.5%*
Photochemical ozone formation - human health [kg NMVOC eq.]	1.29E+03	1.14E+03	−11.86%	*−10.6%*	*−1.5%*
Resource use, energy carriers [MJ]	6.88E+07	3.98E+06	−94.22%	*−94.2%*	*−2.0%*
Resource use, mineral and metals [kg Sb eq.]	1.39E+01	9.72E+00	−29.94%	*−6.4%*	*−26.4%*
Respiratory inorganics [Disease incidences]	2.48E-02	1.31E-02	−47.41%	*−47.1%*	*−1.3%*
Water scarcity [m^3 world equiv.]	1.37E+05	9.44E+04	−30.93%	*−19.5%*	*−17.0%*
Ionising radiation, waste [Bq U238 ILW-eq.]	6.30E+10	5.83E+10	−7.35%	*−1.9%*	*−5.6%*
Ionising radiation [Bq U235 air-eq.]	2.28E+09	1.87E+09	−18.15%	*−17.7%*	*−0.6%*

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