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# Implementing a sustainable process for the recovery of palladium from spent catalysts at industrial scale: A LCA approach

Annachiara Ceraso [a], Grazia Policastro [b], Marica Muscetta [c,*], Laura Clarizia [c,**], Alessandra Cesaro [a]

[a] Department of Civil, Architectural and Environmental Engineering, University of Naples Federico II, via Claudio 21, 80125, Naples, Italy
[b] Department of Engineering, Telematic University Pegaso, Centro Direzionale Isola F2, 80132, Naples, Italy
[c] Department of Chemical, Materials and Industrial Production Engineering, University of Naples Federico II, P.le V. Tecchio 80, 80125, Naples, Italy

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ABSTRACT

Due to its unique physicochemical properties, palladium is widely used in several industry applications (e.g., vehicle emission control). In view of the circular economy, it is essential to explore secondary sources of palladium, such as urban mines. Current technologies for effective palladium recovery involve high energy consumption and severe environmental impact. More recently, a novel green method for recovering palladium from spent catalysts through a combination of mild acidic leaching and photodeposition on ZnO nanoparticles was proposed on a laboratory scale. In the present study, the environmental impacts of this recovery method, properly upscaled and modelled, was assessed by employing the LCA approach. Specifically, a comparative LCA was carried out for the process with as well as without recycling key components, such as Cu (II) and NaCl for the leaching solution and ZnO. The outcomes identified critical areas and drove the investigation of alternative process configurations to reduce its environmental footprint, such as the use of carbon dioxide in the photo-deposition process with the aim of decreasing the resulting terrestrial ecotoxicity. This study marks a significant step forward in advancing research toward industrial–scale implementation of palladium recovery. It provides valuable insights for researchers in the field of green physicochemical processes for metal recovery, thus offering guidance for future decision–making towards more sustainable practices.

## 1. Introduction

The Earth's Crust is known to contain a limited supply of valuable elements such as silver, gold, palladium, platinum, etc., known as precious metals (PMs). Their scarcity, combined with the numerous applications in which they can be used, justifies the high cost of these materials. Used as a means of payment since ancient times, PMs are increasing in demand and consumption year by year, being extensively used in catalysis, chemical engineering, petroleum, electricity, and electronics (Chen et al., 2021; Vancea et al., 2020). The reason for their widespread application is related to their unique physical and chemical properties: palladium, for instance, due to its low melting point, high resistance and unique catalytic properties represents the preferred metal in the automotive industry to purify exhaust emissions (Xun et al., 2020). As reported in the Annual report of MMC Norilsk Nickel (Potanin et al., 2019), a higher palladium consumption was registered in 2019, with 294 t used in the automotive industry, due to the stricter emission regulations on a global scale. In China, the adaptation of the automotive industry to international emission regulations, indeed, has increased the metal consumption, obtaining about 70% of the manufactured cars meeting the new standards.

Less than 1 billion tons of palladium are present worldwide (Yaneva, 2022): the main sources are located in Russia and South Africa, which together provide around 75% of the metal mined. In this context, the recovery of this metal from secondary sources can be a key point to reduce the depletion of natural sources while providing the proper treatment of Pd–rich waste, thus lowering the growing price of this metal (Ciopec et al., 2021). Moreover, metal–containing waste can be defined as “urban mines”, which are a very appealing alternative to traditional mining for those countries lacking natural mineral ores. Currently, pyrometallurgy and hydrometallurgy are the main technologies employed for metal recovery from secondary sources. Nonetheless,

* Corresponding author.
** Corresponding author.
*E-mail addresses:* marica.muscetta@unina.it (M. Muscetta), laura.clarizia2@unina.it (L. Clarizia).

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off−gas treatment, the use of different process units, the need to operate at high temperature and pressure as well as to keep the pH within specific range have been identified among the drawbacks associated with conventional recovery processes. In the case of hydrometallurgical processes, the use of strong mineral acid to leach the target metals from the solid waste stands as an additional disadvantage, so that the utilization of mildly acidic conditions has been proposed as alternative in recent years (Astuti et al., 2018; Lei et al., 2018). In this context, the effective recovery of palladium through a combination of mildly acidic leaching and a photodeposition on ZnO has been proposed by Muscetta et al. (2022) on a laboratory scale. The process was applied to different metals, thus demonstrating the possibility of applying this methodology to a wide range of matrices (Muscetta et al., 2023a, 2023b). Despite the number of advantages of this kind of system (i.e., high selectivity, easy applicability, low temperatures and ambient pressure, moderate acidic conditions, and non−toxic chemicals), the evaluation of its possible environmental burdens is of paramount importance to drive its sustainable scale−up.

The environmental impact of industrial metallurgic processes can be quantitatively assessed with the support of Life Cycle Assessment (LCA) studies (De Meester et al., 2019). Recently, the increasing interest towards the recovery of resources from waste has led to the widespread of LCA of novel processes for the recovery of precious elements, including palladium, from waste. Researchers have focused on both Waste Electrical and Electronic Equipment (WEEE) and spent catalysts, with a significant gap related to the latter (Gu et al., 2016; Hagelüken and Goldmann, 2022; Le and Lee, 2021; Padamata et al., 2020; Souza et al., 2017).

Thompson et al. (2018) conducted a LCA study comparing a traditional nitric acid leaching process with a bioleaching process, to extract rare−earth elements (REE) from fluidized catalytic cracking (FCC) catalysts. The results showed that the chemical leaching caused higher impacts due to a greater electricity consumption, while the glucose utilized in the bioleaching process led to slightly higher impacts in the non−carcinogenic and eutrophication categories. Another study compared two different hydrometallurgical alternatives for the recovery of platinum and cobalt from catalysts. The first was based on ion exchange resin and the latter on solvent extraction. The solvent alternative appeared to be more impacting than the resin process in nearly all considered categories, except for the ozone depletion potential (Duclos et al., 2020).

Recently, Ruiu et al. (2023) performed a comparative LCA of a novel sustainable process for palladium recovery from spent catalysts. The process was based on the use of supercritical $CO_2$, a green solvent, operated in mild conditions of pressure and temperature and was compared with a standard recovery process. The innovative process resulted to be less sustainable than the other, as the data for both technologies were gathered from the laboratory−scale, limiting the results representativity.

These outcomes suggest the importance of a targeted evaluation of the environmental sustainability of newly developed recovery approaches. Therefore, aim of this work was in evaluating the potential impacts of a novel process for palladium recovery from exhausted catalysts, developed on a lab scale in a previous work (Muscetta et al., 2022), with or without the recirculation of the main species involved in the process (i.e., Cu (II) and NaCl for the leaching solution and ZnO). To the best of author's knowledge, this is the first work proposing the use of LCA to optimize the sustainable development of a Pd−recovery process since its lab scale setting up.

By analysing the results obtained through this methodology, critical points were identified and used to modify the process, in order to enhance its environmental sustainability for the possible scale up.

## 2. Materials and methods

### 2.1. Experimental

Copper chloride ($CuCl_2$, 99%), sodium chloride (NaCl, >99%), and zinc nitrate hexahydrate ($Zn(NO_3)_2 \cdot 6H_2O$, reagent grade, 98%) were obtained from Aldrich Chemistry. Sodium hydroxide pellets (NaOH) and nitric acid ($HNO_3$) were purchased from AnalaR BDH. Ethanol (EtOH, 99.8%) was obtained from Fluka. A Cary 100 UV–vis spectrophotometer from Agilent Technologies was used for the colorimetric determination of palladium (II), cupric and cuprous ions in the solution, according to previous methods (Gahler, 1954; Muscetta et al., 2020, 2021c). Palladium dissolution and photocatalytic palladium reduction processes were executed in accordance with the methodologies delineated in earlier publications (Muscetta et al., 2021a, 2021b, 2022). In each experiment, a fixed quantity of a spent catalyst (comprising palladium supported on titanium dioxide), prepared through the photodeposition technique, was employed. Particularly, 2.1%wt. of Pd was deposited on the photocatalyst surface. Before using the material in the present investigation, the metal−doped photocatalyst was used several times to assess the photocatalytic hydrogen generation (the data are not shown, being out of the scope of the present study). After these photocatalytic experiments, the spent material was washed three times with double−distilled water and then dried at 80 °C for 24 h to obtain the powder. A schematic representation of the process is reported in Fig. 1.

As reported previously (Muscetta et al., 2022), a leaching solution (comprising of $NaCl/CuCl_2$) was employed to convert Pd(0) into Pd(II), following the reaction (R.1), then Pd(II) can be subsequently subjected to a photocatalytic treatment in the presence of ZnO nanoparticles within a photocatalytic reactor, based on the reactions R.2 – R5, where $P_1$ and $P_2$ are generic by−products obtained from ethanol oxidation.

$$\frac{1}{2}Pd^0 + Cu^{2+} + 4Cl^- \rightleftarrows \frac{1}{2}PdCl_4^{2-} + CuCl_2^- \left(\begin{matrix}\text{Leaching}\\ \text{step, R.1}\end{matrix}\right)$$

$$ZnO \xrightarrow{h\nu} h^+ + e^- \xrightarrow{k_r} \text{heat} + \text{light} \left(\begin{matrix}\text{Photodeposition}\\ \text{step, R.2}\end{matrix}\right)$$

$$EtOH_{ads} + h^+ \xrightarrow{k_{h^+}} EtOH^* + e^- + h^+ \xrightarrow{fast} P_1 \left(\begin{matrix}\text{Photodeposition}\\ \text{step, R.3}\end{matrix}\right)$$

$$Pd^{2+} + \left(Pd^0\right) + e^- \xrightarrow{k_1} Pd^+ + e^- \xrightarrow{fast} Pd^0 \left(\begin{matrix}\text{Photodeposition}\\ \text{step, R.4}\end{matrix}\right)$$

$$Pd^{2+}_{ads} + EtOH_{ads} \xrightarrow{k_2} Pd^0 + P_2 \left(\begin{matrix}\text{Photodeposition}\\ \text{step, R.5}\end{matrix}\right)$$

The resulting composite material of Pd/ZnO was separated from the solution, thus obtaining the effective palladium recovery by promoting the dissolution of zinc oxide nanoparticles via the adjustment of the suspension pH to below 6.30. With the aim to reduce the produced waste, Muscetta et al. (2022) proposed (1) the reuse of the solution coming from the photodeposition unit (mainly composed of ethanol, NaCl and $CuCl_2$) in a new leaching stage, and (2) the use of the zinc ions and nitrates coming from the ZnO dissolution unit to synthesize fresh zinc oxide particles for recycling and use as a photocatalyst in the photodeposition stage.

The detailed description of the experimental setup for each stage is reported elsewhere (Muscetta et al., 2022). However, as concerns the photodeposition unit, an inertization of the suspension was necessary to avoid the reaction between photogenerated electrons and oxygen. Specifically, to assess the environmental impact of the whole process and based on the results of the LCA, three different inertization procedures were followed: 1) a nitrogen stream bubbling starting 40 min before the photocatalytic run, at a flow rate of 0.3 L $min^{-1}$, then adjusted at a flow rate at 0.2 L $min^{-1}$ during the photocatalytic experiment; 2) a nitrogen stream bubbling starting 30 min before the photocatalytic run, then

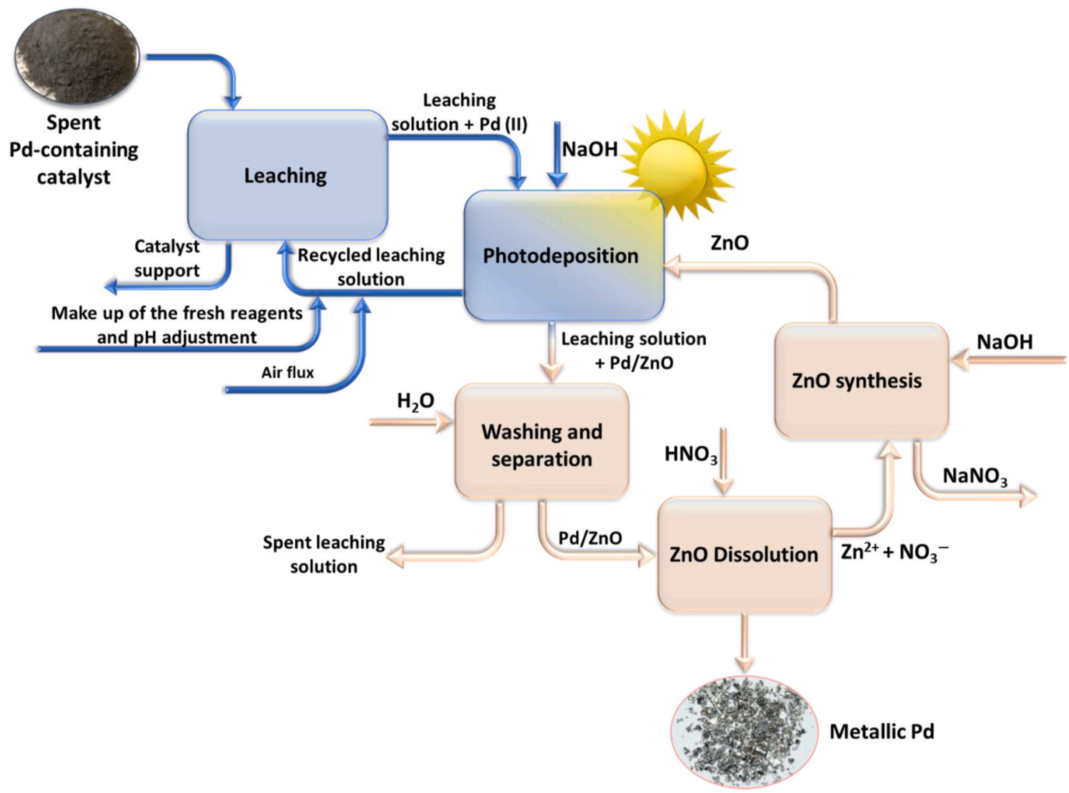


**Fig. 1.** Schematic representation of the proposed process for the recovery of palladium from spent catalysts.

close the system during the photocatalytic experiment; 3) the use of condition (2) in the presence of carbon dioxide as the inert gas in the recovery process.

### 2.2. Life Cycle Assessment

The Life Cycle Assessment (LCA) was developed in accordance with the international standards ISO 14040 and ISO 14044, in which the several phases of an LCA are described: goal and scope definition, inventory analysis, impact assessment, and results interpretation.

### 2.3. Goal and scope definition

The goal of this study was to assess the potential impacts of a novel process for palladium recovery from exhausted catalysts proposed by Muscetta et al. (2022). As already mentioned, the analysis was meant to identify hot spots in the process as well as to suggest possible ways to reduce its overall impact.

To this end, the analysis focused on the alternative method for palladium recovery presented in Fig. 1, as well as on the method without the recirculation of the leaching solution and ZnO nanoparticles. Therefore, the foreground system applied a “gate−to−gate” approach, as the boundaries of the life cycle included the activities finalised to the recovery of palladium from spent catalysts but excluded both the pretreatment and the final disposal of the catalysts themselves.

The adopted functional unit (FU) corresponded to the final product of the process, namely 21 g of palladium recovered from one spent catalyst (1 Kg).

### 2.4. Life cycle inventory

The energy and mass flows of the recovery processes, both with and without the recirculation of ZnO nanoparticles, are described in Muscetta et al. (2022) and were upscaled for the purposes of the LCA, using the framework provided by Piccinno et al. (2016). Successively, the inputs and outputs were scaled in accordance with the FU.

Afterwards, the recovery methods were modelled with the software *SimaPro v9.4.0.2 PhD.*, using the processes from the *Ecoinvent 3.8* database, which provides six different libraries according to the allocation method and the type of processes (unit or system). For this study, the database version “*allocation cut−off by classification − unit*” was used.

The Life Cycle Inventory of the recovery systems of palladium from one spent catalyst is given in Table 1 (Muscetta et al., 2023c). The data about the environmental burdens of the feedstock chemicals and energy

**Table 1**
LCI for recovery of palladium from 1 kg spent catalyst.

| Compounds | Process without recirculating | Process with recirculating | Unit | Description |
|---|---|---|---|---|
| | Amount | Amount | | |
| Sodium chloride | 70 | 10.5 | kg | Market, GLO |
| Copper (II) chloride | 0.067 | 0.01008 | kg | Market, GLO |
| Sodium hydroxide | 0.89 | 0.1 | kg | Market, GLO |
| Zinc oxide | 0.10 | 0.02 | kg | Market, GLO |
| Ethanol | 20 | 3 | L | Market, RoW |
| Perchloric acid | – | 0.89 | L | Market, GLO |
| Nitric acid | 0.10 | 0.10 | L | Market, RoW |
| Water | 1280 | 1110 | L | Market, RoW |
| Nitrogen | 36,000 | 36,000 | L | Market, RoW |
| Air | – | 36,000 | L | Natur |
| Electricity, low voltage | 81.14 | 102.30 | kWh | Market, GLO |

for the recovery processes were obtained directly from the database (Xue et al., 2015). The electricity used by the equipment necessary for the processes was assumed to have a low voltage (Karal et al., 2021) and it was supposed to be produced by an average global combination of multiple energy sources, both renewable and non−renewable.

Similarly, the measures about emissions strictly connected to the chemicals used during the process were provided by Muscetta et al. (2022). Even though most of them were supposed to either end in a water matrix (Table 2) or constitute liquid waste to be handled, the treatment for this stream was not part of the defined foreground system and it was, therefore, excluded from the analysis. Instead, the inert gases (air and nitrogen) were modelled as emissions in the air, after being used in the recovery systems.

### 2.5. Life cycle impact assessment and interpretation

For the impact assessment (LCIA), CML−IA baseline method was adopted, being one of the most frequently used for environmental assessment (Ramos and Rouboa, 2020; Villares et al., 2016; Xue et al., 2015). It is a method developed by the Institute of Environmental Sciences of Leiden and it includes the following impact categories: abiotic depletion, abiotic depletion (fossil fuels), global warming, ozone layer depletion, human toxicity, freshwater aquatic ecotoxicity, marine aquatic ecotoxicity, terrestrial ecotoxicity, photochemical oxidation, acidification, and eutrophication.

The LCIA results were calculated for the recovery process with and without recirculation.

## 3. Results and discussion

The results of the comparative LCA (See Fig. 2) show that, as expected, the recirculation of the leaching solution in the recovery process proposed by Muscetta et al. (2022) reduced the environmental burden in most impact categories in comparison to the process without recirculation, with high benefits especially for the abiotic depletion (ADP) category (−56.9%), due to the minor consumption of several chemical compounds, including sodium chloride, copper chloride, sodium hydroxide, zinc oxide, and ethanol, which differently contribute to the investigated impact categories.

Nonetheless, the recirculation was found to cause a higher impact on the category of terrestrial ecotoxicity, due to the energy demand of this additional phase, which determines a general increase in the electricity needed for the entire process (Xu et al., 2015).

For all remaining impact categories, it is possible to notice a general improvement in the environmental performance of the process, which can be partly attributed to a minor consumption of chemicals, which, in turn, reduces the background impact associated to their production and management (Villares et al., 2016), as well as to their reduced availability for future use (Rosenbaum et al., 2018). At the same time, the reduced emissions associated to the process with recirculation was proved to be particularly beneficial for the impact categories ODP, FWAEP, MAEP, and POCP, which are influenced by the release of substances into the environment and from their interaction with air, water, and soil (Rosenbaum et al., 2018).

Our findings are consistent with prior research on LCA regarding innovative metal recovery techniques. Karal et al. (2021) conducted an LCA study on neodymium (Nd) recovery from neodymium iron boron (Nd−Fe–B) magnets using a contemporary hydrometallurgical approach. The researchers reported that the extensive usage of sulfuric acid and other chemicals in the hydrometallurgical recycling of Nd resulted in a substantial depletion of abiotic resources. In addition, Li et al. (2019) investigated the environmental impacts of a novel electrochemical recovery process to recover base metals, precious metals, and rare earth elements (REEs) from e−waste, comparing it with the already existing pyrometallurgical and hydrometallurgical processes. The study proved that the limited use of chemical products, in contrast to the other processes, determined a lower impact on the acidification category. Similarly, Rezaee et al. (2023) compared the environmental impacts of hydrometallurgical processes using different chemicals and methods for leaching and recovery from waste printed circuit boards of both base and precious metals. The comparison showed how highly efficient processes with minimal consumption of chemicals have a low environmental burden, and that rejuvenated leaching liquor can be implemented in multiple leaching cycles, further reducing the need for chemical substances and water.

### 3.1. Assessment of alternatives to reduce the terrestrial ecotoxicity impact

The LCA results showed the potential environmental benefits of the novel recovery system with recirculation proposed by Muscetta et al. (2023c) for all the impact categories, except the terrestrial ecotoxicity. More specifically, according to the LCA analysis results, the energy demand was the most influential input to the environmental burden on this category, followed by the consumption of chemicals, with the greatest contribution provided by the input of nitrogen for the process atmosphere inertization (Fig. 3).

These outcomes align with an earlier study conducted by Akahori et al. (2014). The authors performed an LCA of a novel method for extracting rare earth metals from used magnets and the results indicated that the increased energy demand had a significant environmental impact.

Similarly, Li et al. (2019) led a study on a novel electrochemical recovery process, establishing that electricity consumption gives the greatest contribution to the environmental burden of the process, especially for the impact categories of global warming and fossil fuel depletion. Besides, the researchers established that the impact on global warming was mostly induced by the consumption of natural gas, while the use of alternative renewable energy could increase the sustainability of the process.

To reduce the terrestrial ecotoxicity impact of the process with recirculation, it would be necessary to either reduce the amount of used energy or substitute the electricity mix with a more sustainable one. While the first option is inhibited by the energy demand of the equipment necessary for running the process properly, the second option is a valid alternative, as renewable energy technologies have been proven to have several environmental benefits on all impact categories, in comparison to traditional power plants (Asdrubali et al., 2015). Nevertheless, the availability of energy from renewable sources depends on

**Table 2**
Emissions for recovery of palladium from 1 kg spent catalyst.

| Emissions | Process without recirculating | Process with recirculating | Unit | Description |
|---|---|---|---|---|
| | Amount | | | |
| Sodium chloride | 70 | 10.5 | kg | Emission in Water |
| Copper (II) chloride | 0.067 | 0.01008 | kg | Emission in Water |
| Sodium nitrate | – | 0.207 | kg | Emission in Water |
| Zinc oxide | – | 0.02 | kg | Emission in Water |
| Zinc | 1.22 | – | mol | Emission in Water |
| Nitrate | 2.44 | – | mol | Emission in Water |
| Ethanol | 20 | 3 | L | Emission in Water |
| Water | 1280 | 1110 | L | Emission in Water |
| Nitrogen | 36,000 | 36,000 | L | Emission in Air |
| Air | – | 36,000 | L | Emission in Air |

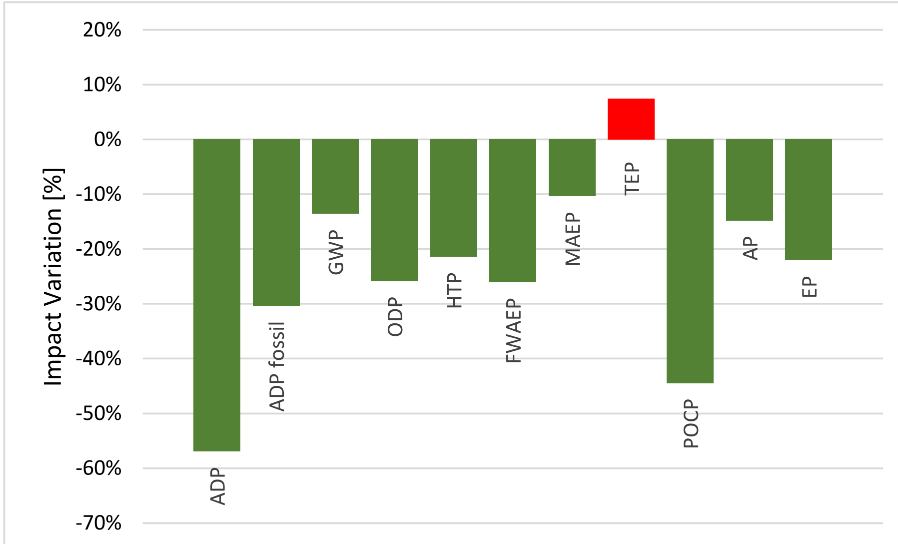


**Fig. 2.** LCIA comparison of the palladium recovery process with and without recirculation.

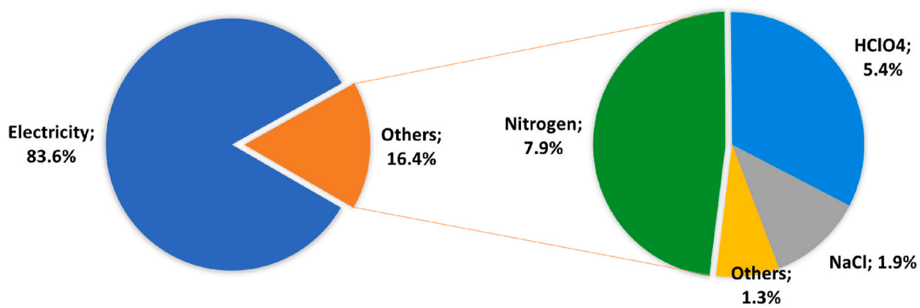


**Fig. 3.** Influence on the terrestrial ecotoxicity.

factors external to the recovery process itself, such as the geographical area where the method is being implemented.

Consequently, to reduce the impact of the recovery system with recirculation, it was decided to conduct additional research on the burden caused by the consumption of chemicals to the process with recirculation. In particular, it was analysed the influence of the inert gas on the impact assessment, as it provides the highest contribution to the impact on the terrestrial ecotoxicity category after the energy consumption, and to evaluate the possibility to either minimise the amount of used nitrogen or to substitute it with a different gas. In this regard, three different inertisation options were tested and compared: i) the use of a volume of 36,000 L of nitrogen, as in the original scenario described by Muscetta et al. (2022); ii) the application of a smaller amount of nitrogen (3000 L), iii) the usage of 3000 L of carbon dioxide as the inert

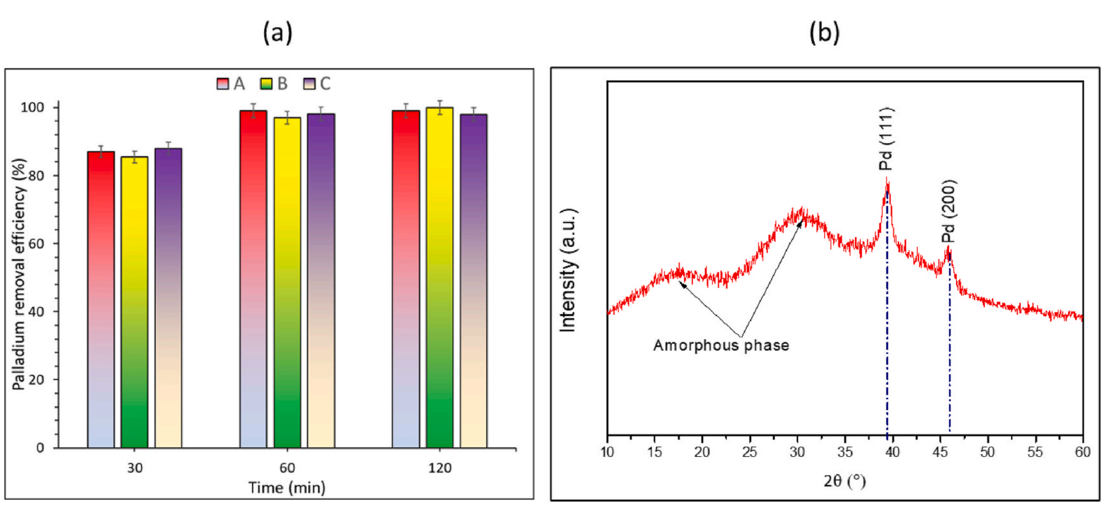


**Fig. 4.** (a) Efficiency of palladium recovery (A) with standard inertisation (36,000 L of nitrogen), (B) reducing the amount of inert used (3000 L of nitrogen) and (C) in the presence of a $CO_2$ instead of nitrogen (3000 L). (b) XRD patterns of the recovered metallic palladium.

gas in the recovery process. These experimental tests proved that the efficiency of the recovery process with recirculating was unvaried, even when a limited amount of nitrogen or carbon dioxide was being used for the inertisation, during the photodeposition phase of the palladium recovery process, as plotted in Fig. 4 (a). The characterization tests performed on the recovered materials were comparable in all cases, and in accordance with previous results (Muscetta et al., 2021a, 2022). Specifically, Fig. 4 (b) shows the XRD patterns of the recovered palladium when carbon dioxide as the inert gas is used in the recovery process; the peaks are in agreement with the reported literature diffraction peak values for metallic Pd (JCPDS no. 05−0681).

Once proved the process efficiency under different inertisation conditions, an additional impact assessment was conducted to verify whether the process modification could have led to improved potential environmental effects.

The impact assessment results of these scenarios were normalised regarding the environmental burden of the process without recirculation and then compared to each other (Table 3).

The use of a smaller amount of inert gas, either nitrogen or carbon dioxide, was found to reduce the impact of the process with recirculation on all categories, including terrestrial ecotoxicity. More specifically, a reduction up to 58.6% was found depending on the considered inert gas as well as on the impact category. In addition, the use of a minimal quantity of gas for the inertisation, further improved the environmental sustainability of the process with recirculation, as it allowed to reduce the impact on abiotic depletion (fossil fuels), global warming, marine aquatic toxicity, acidification, and eutrophication by over 10% (Table 4). Besides, the results showed how the use of $CO_2$ determined an even better environmental performance than the use of nitrogen in all impact categories, except for the global warming. In fact, the use of carbon dioxide strongly influences this category, as it is a greenhouse gas (Tran et al., 2022).

### 3.2. *Future decision−making support*

Starting from the LCA results of the novel palladium recovery process, it was possible to identify several options meant to improve its sustainability.

The process with recirculation, generally more environmentally friendly, was found to be characterised by a higher impact on the terrestrial ecotoxicity category than the one without recirculation, mainly due to the greater energy consumption. Hence, employing a sustainable energy mix from renewable sources would help to reduce the environmental burden of the recovery technology.

A second strategy to improve the sustainability of the process with recirculation is the reduction of the used inertisation gas, which would determine lower values for all impact categories. Besides, replacing nitrogen with $CO_2$ could further improve the LCIA results, except for the global warming category (Table 4). Nonetheless, reusing the carbon dioxide stream multiple times and/or using the $CO_2$ from exhausted gaseous emissions can further minimise the impact of using it in the process (Alsarhan et al., 2021; Garcia et al., 2021).

**Table 3**
CML−IA impact categories.

| Impact categories | Abbreviations | Units |
|---|---|---|
| Abiotic depletion | ADP | kg Sb eq |
| Abiotic depletion (fossil fuels) | ADP fossil | MJ |
| Global warming | GWP | kg $CO_2$ eq |
| Ozone layer depletion | ODP | kg CFC−11 eq |
| Human toxicity | HTTP | kg 1,4−DB eq |
| Freshwater aquatic ecotoxicity | FWAEP | kg 1,4−DB eq |
| Marine aquatic ecotoxicity | MAEP | kg 1,4−DB eq |
| Terrestrial ecotoxicity | TEP | kg 1,4−DB eq |
| Photochemical oxidation | POCP | kg $C_2H_4$ eq |
| Acidification | AP | kg $SO_2$ eq |
| Eutrophication | EP | kg $PO_4^{3-}$ eq |

**Table 4**
Inert gases impact assessment comparison.

| Impact categories | Abbreviations | Units | $N_2$ = 36000L | $N_2$ = 3000L | $CO_2$ = 3000L |
|---|---|---|---|---|---|
| Abiotic depletion | ADP | kg Sb eq | −56.9% | −58.4% | −58.6% |
| Abiotic depletion (fossil fuels) | ADP fossil | MJ | −30.3% | −40.8% | −41.8% |
| Global warming (GWP100a) | GWP | kg $CO_2$ eq | −13.6% | −27.4% | −24.0% |
| Ozone layer depletion | ODP | kg CFC−11 eq | −25.8% | −35.3% | −36.2% |
| Human toxicity | HTTP | kg 1,4−DB eq | −21.4% | −28.2% | −28.8% |
| Freshwater aquatic ecotoxicity. | FWAEP | kg 1,4−DB eq | −26.0% | −33.1% | −33.8% |
| Marine aquatic ecotoxicity | MAEP | kg 1,4−DB eq | −10.3% | −23.4% | −24.6% |
| Terrestrial ecotoxicity | TEP | kg 1,4−DB eq | +7.5% | −0.3% | −1.0% |
| Photochemical oxidation | POCP | kg $C_2H_4$ eq | −44.5% | −51.4% | −52.1% |
| Acidification | AP | kg $SO_2$ eq | −14.8% | −27.2% | −28.3% |
| Eutrophication | EP | kg $PO_4^{3-}$ eq | −22.1% | −33.2% | −34.2% |

To integrate both strategies (i.e., renewable energy and carbon dioxide utilization) in the novel palladium recovery process, future research should focus on the integration of renewable energy plants that produce carbon dioxide as a by−product. For example, the implementation of incineration plants would close the loop of the recovery method, in a circular economy perspective. Ideally, after the recovery of palladium the leftover spent catalysts would be disposed through incineration, thus producing energy and $CO_2$ emissions, both of which could be reused to properly run the process (Christensen and Bisinella, 2021).

## 4. Conclusions

In this study, the sustainability of a new green physicochemical method for recovering palladium from spent catalysts through a combination of mild acidic leaching and photodeposition on ZnO nanoparticles was assessed by employing the LCA approach.

The comparison of the LCA results obtained by modelling the proposed process with and without the recirculation of the leaching solution and ZnO photocatalyst revealed an improvement in environmental sustainability in the first case.

Among the crucial operating parameters to be considered in view of decreasing the environmental impact, the choice of a suited inert gas was identified as one of the most important. Indeed, either minimizing the amount of nitrogen employed or replacing it with carbon dioxide may turn to be possible solutions to reduce the potential terrestrial ecotoxicity of the process.

Overall, the study provided valuable insights into the environmental impact of the palladium recovery process from spent catalysts and offered recommendations for future decision−making, emphasizing the importance of monitoring the environmental sustainability in resource recovery processes since the very early stage of their development.

## CRediT authorship contribution statement

**Annachiara Ceraso:** Data curation, Methodology, Software, Writing

– original draft. **Grazia Policastro:** Data curation, Investigation, Methodology, Visualization, Writing – original draft. **Marica Muscetta:** Data curation, Writing – original draft, Conceptualization. **Laura Clarizia:** Conceptualization, Data curation, Supervision, Writing – review & editing. **Alessandra Cesaro:** Conceptualization, Software, Supervision, Validation, 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

Data will be made available on request.

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