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Toward environmentally favorable nano-sensing by production of reusable gold nanoparticles from gold nano-waste: life cycle and nanocircular economy implications†

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The environmental impacts of gold nanoparticle (AuNP)-based sensing were investigated using a cradle-to-grave life cycle assessment (LCA). The LCA model considered AuNP synthesis, surface functionalization, an AuNP-based detection assay, and disposal. Additionally, the model incorporated two potential Au nano-waste reuse strategies reliant upon α-cyclodextrin (α-CD) or Triton X-114. The results show that, across ten midpoint categories that >80.4% of the environmental impacts arise from AuNP synthesis thus demonstrating the benefit of reuse of Au nano-wastes. Importantly, the two different reuse strategies enhance the environmental sustainability of the sensing application. Gold recovery contributed to a significant reduction in the amount of pristine Au^{3+} initially required despite the additional chemical and electrical demands of the reuse processes. Sensitivity analysis focused on two variables (*i.e.*, recovery efficiency and the number of reuse cycles) indicated that the environmental favorability of the sensing application is dominated by recovery efficiency. Finally, the reuse of Au nano-waste reduces the energy demand of nano-sensing and the total cost of AuNP-based industries, thus illustrating energy and circular economy implications.

Environmental significance

The manufacture and application of gold nanoparticles (AuNPs) have consistently expanded across many fields. However, the finite resource of gold ore, or lack thereof, continues to raise questions about the environmental sustainability of AuNP applications. There is limited knowledge of the environmental impacts of AuNP applications that must be addressed if practitioners want to support nanosustainability. Life cycle assessment (LCA) showed reduction of environmental burden of AuNP-based sensing through the reuse of Au nano-waste and illuminated greener alternatives to current AuNP production and use practices, thus illustrating energy and circular economy implications.

Introduction

The production of nanomaterials has been increasing in accordance with the surge in the development of nanotechnology-based techniques. Nonetheless, exploration of the environmental impacts of these techniques is relatively limited. It is imperative to rigorously assess the environmental sustainability of nanomaterial production to prevent unintended negative consequences.[1] Additionally, it is important to evaluate the environmental impacts of the nanomaterial use stage. Life cycle assessment (LCA) is a rigorous tool that allows practitioners to investigate the characteristics of a particular technique prior to field application.[2–4] To date, LCA has been extensively used to evaluate the environmental impacts of the manufacture of a range of nanomaterials (*e.g.*, Au, Ag, TiO_2, FeO_x).[5–10]

Gold nanoparticles (AuNPs) hold promise in many applications covering medical diagnostics and imaging for

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† Electronic supplementary information (ESI) available: (1) Experimental details; (2) detection of ssDNA using aptamer-functionalized AuNPs; (3) simulation details; (4) Tables S1–S5: inputs of synthesis, functionalization, detection, recovery, and recycling processes; (5) Table S6: 1 mg of customized chemicals from available databases; (6) the stoichiometry of the products and reactants for customization; (7) uncertainty analysis of environmental impacts (ozone depletion, smog, acidification, eutrophication, respiratory effect; (8) sensitive analysis of environmental impacts (global warming, carcinogenic, non-carcinogenic, ecotoxicity, fossil fuel depletion); (9) sensitive analysis of cumulative energy demand (CED). See DOI: https://doi.org/10.1039/d3en00505d

health care and dentistry, the electronics industry of inks in photovoltaics, and sensors.[11] The U.S. market value of AuNPs has reached ≈1180 million USD.[12] Free electrons on the AuNP surface oscillate when excited by incident light and generate unique localized surface plasmon resonance (LSPR).[13] AuNP-based sensing has been widely applied owing to their unique plasmonic properties and their high surface-to-volume ratio.[14,15]

While prior studies have evaluated the environmental impacts of AuNP synthesis methods,[5,6,10] the environmental impacts of specific AuNP-based applications have not been explicitly explored. The acceleration in widespread synthetic production of AuNPs has led to increasing amounts of Au nano-waste. We previously reported that the disposal of Au nano-waste results in negative environmental impacts.[16] Further, the declining amount and quality of natural gold ore grades remains a substantial challenge if Au-based nanotechnologies are to be sustainable.[17] The nanotechnology field has recently been inspired by the 'circular economy' concept, a sustainable design framework to return materials at the end of their service life into a circular product ecosystem.[18] This aspirational 'nanocircular economy' endeavors to build a closed-loop system involving the generation and reuse of nanomaterials that differs from the conventional 'make, use, dispose' linear model.[19] Under these circumstances, the reuse of Au nano-waste has received attention as a promising strategy to reduce environmental burdens and promote sustainability.

In this study, we evaluated the environmental impacts of AuNP-based optical sensing. Furthermore, we investigated the environmental sustainability of the reuse of Au nano-waste as a means to advance the nanocircular economy. We used LCA as a quantitative framework to evaluate the environmental impacts of AuNP-based sensing across the stages of synthesis, functionalization, detection assay, disposal, and reuse (as applicable). As an exemplary optical sensing approach, we examined an aptamer-functionalized AuNP-based colorimetric detection method. In the presence of target ssDNA, the color of an aptamer-functionalized AuNP suspension changes due to hybridization-induced aggregation, resulting in an absorbance peak shift to longer wavelength (*i.e.*, red-shift). The application of aptamer-functionalized AuNPs enables rapid and sensitive colorimetric detection with the potential for low-cost and simple operation.[20,21]

Materials and method

LCA framework

Herein, the environmental impacts of aptamer-functionalized AuNP-based sensing were assessed using LCA. As suggested by the International Organization for Standardization (ISO) this LCA included the following four steps: 1) goal and scope, 2) life cycle inventory (LCI) analysis, 3) life cycle impact assessment (LCIA), and 4) interpretation.[2]

Goal and scope. The goals of this study were 1) to assess the environmental impacts of a colorimetric detection method that uses aptamer-functionalized AuNPs; and 2) to investigate the environmental favorability of two Au nano-waste reuse strategies. The system boundaries included cradle-to-grave processes with the closed-loop of Au nano-waste reuse (Fig. 1). The system consists of AuNP synthesis, functionalization, detection assay, reuse (if applicable), and disposal. The functional unit employed was 100 femtomoles of functionalized AuNPs – this amount is equivalent to that used in a typical detection assay.[22] In this study, aptamer-functionalized AuNPs were used for the detection of ssDNA (ESI†). The class 1 integron-integrase gene (*int*I1) was benchmarked as a model ssDNA target. It has been recognized as a genetic marker for anthropogenic pollution and is commonly linked to genes conferring resistance to antibiotics, heavy metals, and disinfectants.[23]

Scenario 1 included all processes except a reuse step, representing the conventional 'make, use, dispose' linear model. Scenarios 2 and 3 examined two separate Au nano-waste reuse processes, representing a closed-loop circular model with reuse of Au nano-waste. At the grave stage, we simulated two recovery methods followed by strong acid-mediated recycling of Au nano-waste. Prior studies have reported several successful approaches to recovery from diverse nanomaterial wastes.[10,24–31] We selected two recovery pathways for Au nano-waste that are either Au-specific or solution-based recovery methods since the approach considered here was aqueous sensing. It is noted that the choice of recovery methods would impact the environmental burden of the process due to different chemical and electricity demands and recovery rates. First, we simulated the use of α-cyclodextrin (α-CD) which interacts with the square planar complex $AuBr_4^-$, after acid dissolution of Au nano-waste.[10] Second, we simulated the cloud point extraction (CPE) method that involves the thermo-reversible liquid–liquid phase transition of Au nano-waste using a nonionic surfactant (Triton X-114).[25,26] In the literature, the α-CD and Triton X-114 based recovery methods have reported ~60–80% (ref. 10) and >90% (ref. 32) mass recovery efficiencies, respectively. The efficiency presumably varies due to uncertainties in the experimental conditions.

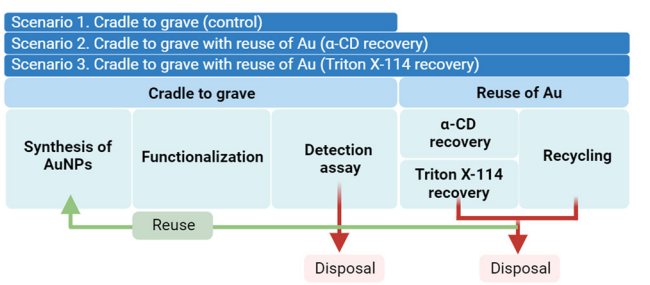

Fig. 1 System boundary of AuNP-based sensing with three different scenarios: control (without reuse) and two Au nano-waste reuse strategies.

Following one of these simulated recovery steps, Au was recycled *via* dissolution in strong acid (*aqua regia*; a mixture of HCl and HNO_3 at a volume ratio of 3 : 1) followed by a boil-off step.

Life cycle inventory (LCI) analysis. LCI inputs for experimental (synthesis, functionalization, and detection assay) and simulated (recovery and recycling) processes were obtained from the EcoInvent database (v3.5) in SimaPro (v9.0). Details on the experimental and simulated processes of AuNP-based sensing are provided in the ESI† and AuNP-based sensor development was successfully validated (Fig. S1 and S2†). The inputs for each process used to model the system are provided in ESI† Tables S1 to S5. Materials not found in the EcoInvent database were customized using reactions of other defined chemicals unless indicated otherwise (these are labeled as "custom defined"). For example, chloroauric acid ($HAuCl_4$) inventory inputs can be estimated using Au, chlorine gas (Cl_2), and hydrochloric acid (HCl) inputs. We designed such customizations based on the stoichiometry of the reactants and products assuming favorable chemical reactions and 100% yields. This was done because energy inputs from different chemical synthesis pathways can vary, and some synthesis methods have unknown pathways. The lists of customized chemicals are provided in Table S6.† The energy use for electricity (stirring, heating, and centrifugation) was determined based on the average electricity mix for the U.S. Northeast Power Coordinating Council from the EcoInvent database.[6]

The total amount of Au^{3+} needed ($[Au^{3+}]_{total}$) to make 100 femtomoles of AuNPs (functional unit) is ~0.089 mg based on the seed-mediated growth method (Table S1†). With the addition of a reuse step, the amount of reused Au^{3+} ($[Au^{3+}]_{reused}$) reduced the initially required pristine Au^{3+} ($[Au^{3+}]_{pristine}$). By varying the recovery efficiency and the number of reuse cycles, $[Au^{3+}]_{pristine}$ and $[Au^{3+}]_{reused}$ were determined as follows,

$$\left[Au^{3+}\right]_{pristine} = \left(\frac{1}{1 + r + r^2 + \cdots + r^n}\right) \times \left[Au^{3+}\right]_{total} \quad (1)$$

$$[Au^{3+}]_{reused} = (1 + r + r^2 + \cdots + r^{n-1}) \times [Au^{3+}]_{pristine} \quad (2)$$

where r is the recovery efficiency (0 to 1.0) and n is the number of reuse cycles (1 to 7).

The reduction of $[Au^{3+}]_{pristine}$ through the reuse of Au nano-waste can be favorable for the overall environmental impact of sensor development. Concomitantly, the reuse step requires use of additional chemicals and electrical energy burdens that are assumed to be proportional to $[Au^{3+}]_{reused}$. By summing up these positive and negative impacts, the environmental favorability of the reuse processes to the overall system was investigated. No matter how many reuse cycles were conducted, the sum of the inputs for functionalization and detection assay processes was assumed equal for all scenarios regarding a constant functional unit of aptamer-functionalized AuNPs. For disposal, the unrecovered Au and other used chemicals were assumed to be incinerated as hazardous waste.

Life cycle impact assessment (LCIA). To assess the environmental impact of the system, characterization factors must be defined. In this study, the Tool for Reduction and Assessment of Chemicals and Other Environmental Impacts 2.1 (TRACI 2.1, v1.05) with the US 2008 model for normalization/weighting sets were applied. The TRACI tool provides ten midpoint-oriented characterization factors that quantify the potential impact of inputs with common equivalence units: ozone depletion (OD), global warming (GW), smog (SG), acidification (AC), eutrophication (EP), carcinogenic (CG), non-carcinogenic (non-CG), respiratory effect (RP), ecotoxicity (ET), and fossil fuel depletion (FF).

Interpretation. ISO recommends ensuring two conditions to interpret the results: 1) the results are accurate and 2) they meet the objectives of the study.[2] To meet these conditions, this study identified the elements that significantly contributed to the impact categories based on the results, evaluated the sensitivity of the significant contributors, and drew conclusions. For sensitivity analysis, the number of reuse cycles and recovery efficiency were set as variables. Further, an uncertainty analysis was also conducted. In total 1000 iterated data sets, which followed a normal distribution for inventory data, were collectively analyzed using the Monte-Carlo method. Mean values with the confidence interval for each scenario and inventory uncertainty were calculated based on the datasets.

Results and discussion

Impact assessment of the primary processes in AuNP-based sensing

There are six primary processes involved in AuNP-based sensing: AuNP synthesis, functionalization, detection assay, recovery, recycling, and disposal. We simulated the recovery and recycling scenario to make a circular system from a linear one that ends with detection assay followed by disposal. The recovery and recycling steps were only simulated when evaluating the two reuse scenarios. Fig. 2 shows the ten overall midpoint environmental impacts of AuNP-based sensing for two different recovery method-mediated reuse scenarios (scenario 2: α-CD; scenario 3: Triton X-114). The environmental impacts were normalized by the no-reuse scenario (scenario 1). For scenario 1 (represented as "a" for each category), we found that >90.7% of the environmental impacts arise from AuNP synthesis. The functionalization and detection assays had much smaller impacts. This result highlights, for the first time, that the environmental burden of AuNPs-based sensing was relatively small compared to AuNP synthesis. We note, however, that the environmental burdens of aptamer production require additional delineation. There are several methods for AuNP functionalization with aptamers[33–36] and different hybridization buffer recipes.[37–39] Nonetheless, based on our

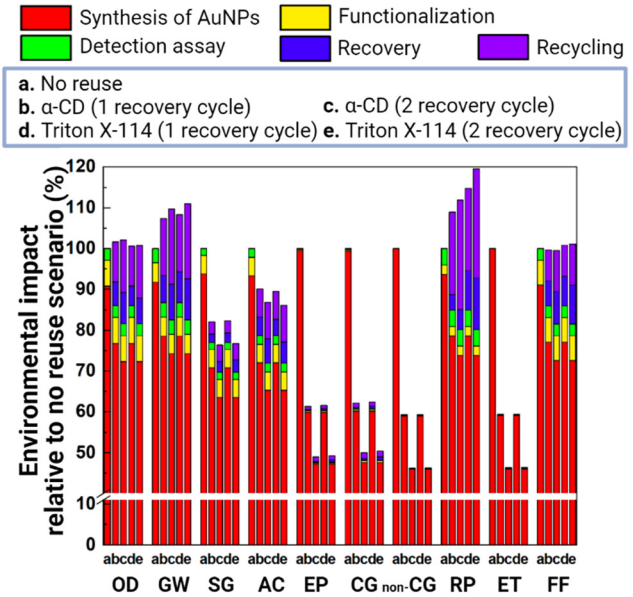

Fig. 2 Ten categorized relative environmental impacts of the sum of all processes (synthesis, functionalization, detection assay, recovery, and recycle) in the system boundary of the AuNP-based sensing application. No-reuse scenario (a) and two recovery method-mediated reuse scenarios (b and c: α-CD and d and e: Triton X-114) assuming a recovery efficiency of 0.7 and either 1 or 2 reuse cycles. For comparative purposes, all scenarios were normalized to the no-reuse scenario. Impact categories: ozone depletion (OD), global warming (GW), smog (SG), acidification (AC), eutrophication (EP), carcinogenic (CG), non-carcinogenic (non-CG), respiratory effect (RP), ecotoxicity (ET), fossil fuel depletion (FF).

LCA results, we expect that methodological variations in functionalization and the detection assay would not significantly affect the overall environmental impacts.

The relative environmental favorability of scenarios incorporating reuse was investigated (indicated in Fig. 2 as "b–e") and were compared to the no-reuse scenario. Higher recovery efficiency and multiple reuse cycles were expected to minimize environmental impacts owing to increased reuse of Au nano-waste. Based on the reported recovery rates of both recovery methods,[10,32] a recovery efficiency was conservatively assumed to be 0.7 with either one or two reuse cycles for simulation. We found that the relative environmental impacts were primarily favorable except for the global warming (GW) and respiratory effect (RP) impact categories. In particular, the environmental impacts of the two reuse-incorporating scenarios decreased by ~40 and ~54% for eutrophication (EP), carcinogenic (CG), non-carcinogenic (non-CG), and ecotoxicity (ET) for either one or two reuse cycles. Two reuse cycles showed more favorable impacts than one, implying the potential utility of multiple reuse cycles. Both recovery methods predict negative impacts for GW and RP, showing impacts of >106%. This means there was a greater environmental impact burden when there was a reuse scenario than the no-reuse one. For each of these categories, two reuse cycles had greater environmental burdens than one reuse cycle. This result can be attributed to the significant burden on GW and RP of the chemical and electricity demands required for recycling. The uncertainty analysis provides the statistical significance of the differences from the scenarios (Fig. S3†).

The results imply the environmentally favorable benefits through a closed-loop circular model of AuNP-based sensing application compared to a conventional linear one without reuse scenario. It was found that the environmental burden of AuNPs-based sensing applications can be more than halved for some categories which depend on the recovery rate and reuse cycle. It is worth noting that shifting the nanomaterial application paradigm from linear to circular models could significantly reduce environmental impacts. For better support to nanosustainability, the environmental favorability of other nanomaterial applications in the circular model should be investigated as well.

Impacts of extended reuse

To illustrate the relative contribution of synthesis and recovery/recycling processes with more extended reuse cycles, two representative categories that each had favorable (SG) and non-favorable (RP) environmental impact of reuse were further investigated. Fig. 3 indicates that smog (SG) and respiratory effect (RP) of synthesis, recovery, and recycling processes as a function of the number of reuse cycles assuming a recovery efficiency of 0.6. A lower recovery efficiency of 0.6 was used in this case to simulate different scenarios with underperformed recovery. As expected, with an increase in the number of reuse cycles, the synthesis process had decreased impacts in SG and RP owing to the reduction in $[Au^{3+}]_{pristine}$, while there were increased impacts for the recovery and recycling processes due to their additional chemical and electrical demands. Both the increasing/decreasing trade-offs plateaued with a higher number of reuse cycles because $[Au^{3+}]_{reuse}$ decreased. Such trends were the same across all categories. However, the overall environmental impacts were more mixed because the contribution of each reuse cycle to the categories was different. In the case of SG, the scale of the impact of the synthesis process was ~10× larger than that for the combined

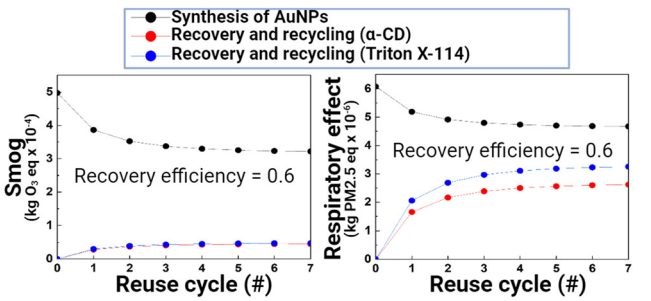

Fig. 3 Smog (SG) and respiratory effect (RP) of synthesis of AuNPs and two recovery/recycling processes with varying number of reuse cycles (#). The recovery efficiency was set to 0.6.

recovery and recycling processes. Accordingly, the overall system had a decreased impact relative to the no-reuse scenario regardless of the number of reuse cycles, thus demonstrating the environmental favorability of the reuse scenario towards this category. On the other hand, for RP, the increased impacts from the recovery and recycling processes were larger than the reductions arising from the synthesis process. The reuse strategies were not environmentally favorable with respect to RP. In the comparison of the two recovery methods, their environmental impacts were comparable in SG while the Triton X-114-assisted method had a higher impact than the α-CD-assisted one for RP. This may be attributed to the greater electricity consumption required for heating in the Triton X-114-assisted method.

Breakdown of process contribution

Fig. 4 summarizes the chemical and electricity contributions to the environmental impacts of each process. For AuNP synthesis, electricity and $HAuCl_4$ (*i.e.*, $[Au^{3+}]_{pristine}$) dominated the environmental impacts. In particular, $HAuCl_4$ constituted >95% of the impact for EP, CG, non-CG, and ET. These categories exhibited the most favorable impacts when either reuse strategy was applied. This demonstrates that the categories dominated by the environmental impacts of $HAuCl_4$ exhibited higher environmental favorability following Au reuse. Conversely, the categories that had smaller (*i.e.*, <40%) $HAuCl_4$ contributions (OD, GW, RP, FF) showed minimal impact of reuse. Across all categories, the next most highly contributing factors to the environmental footprint were electricity for boiling and stirring and nano-pure water production. Alternative synthesis methods that require less electricity or water (*e.g.*, room temperature mediated AuNP synthesis[5]), or that used electricity from renewable sources, would be more environmentally sustainable.

For the functionalization and detection assay, the use of a phosphate-buffered solution with 0.1% Tween-20 (PBS-T20) and electricity were dominant contributors, respectively. PBS-T20 was required to wash the nanoprobes after functionalization. The minimal impacts of the functionalization process support our prior assessment that alternative functionalization methods should not dramatically alter the overall environmental impacts. Similarly, the electricity required for the detection assay is an essential step required to denature the nanoprobes before hybridization occurred. Thus, any other hybridization buffer

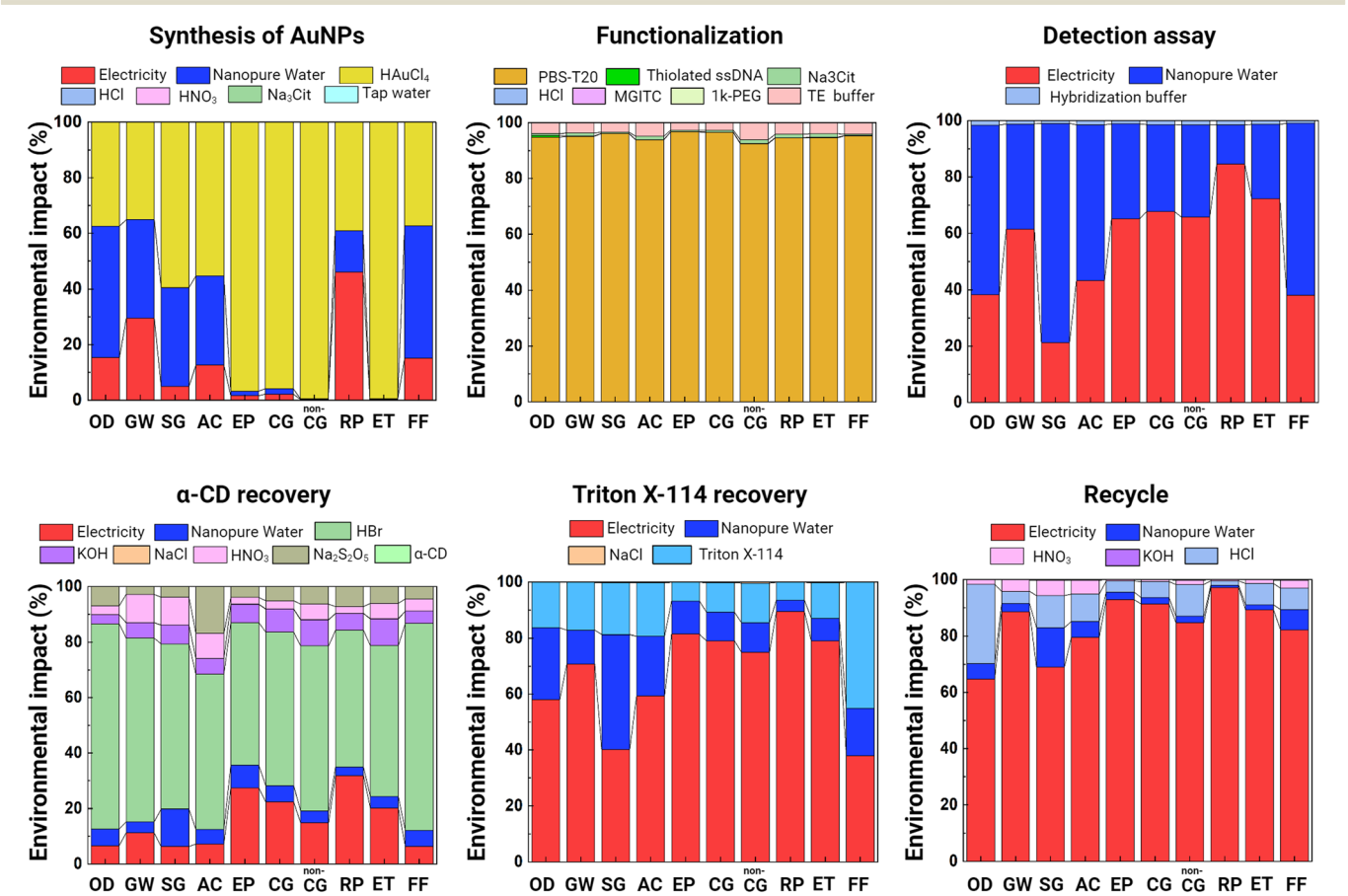

Fig. 4 Chemical and electricity contributions to environmental impacts of synthesis, functionalization, detection assay, recovery, and recycling processes. Ten categories: ozone depletion (OD), global warming (GW), smog (SG), acidification (AC), eutrophication (EP), carcinogenic (CG), non-carcinogenic (non-CG), respiratory effect (RP), ecotoxicity (ET), fossil fuel depletion (FF).

recipes would not increase the environmental impacts significantly.

For α-CD and Triton X-114 based recovery methods, the major environmental impact contributors were HBr (50–75%) and electricity (40–90%) across all categories, respectively. Bromine is a corrosive and highly toxic chemical that is expected to have high environmental impacts across all categories.[40] The main difference between the two methods was the amount of electricity required. The α-CD-based method requires electricity only for sonication (0.24 kJ), while the Triton X-114-based CPE protocol needs electrical energy for heating (1.67 kJ) to recover 100 femtomoles of AuNPs (Table S4†).

For recycling, electricity dominated the impact due to the required HNO_3 boil-off step. To recycle 100 femtomoles of AuNPs, 3.88 kJ of energy were required to remove residual HNO_3. This value is comparably high since the synthesis process required 4.44 kJ for heating (Tables S1 and S5†). Future research should focus on improving the efficiency of HNO_3 removal.

Sensitivity analysis of environmental impacts

The sensitivity of the environmental impacts of AuNP-based sensing was evaluated. Two variables (recovery efficiency and reuse cycle) were simulated and their concomitant environmental impacts were calculated. Recovery efficiency was the key factor determining the reduction in the amount of pristine $HAuCl_4$ required to make 100 femtomoles of functionalized AuNPs. Higher recovery efficiencies have a decreased environmental impact and reflect the promise of reuse strategies. Multiple reuse cycles increase the amounts of chemicals used and the electricity required for recovery and recycling, but also reduce the amount of pristine $HAuCl_4$ required. Fig. 5 shows the environmental impacts of the reuse scenarios with α-CD (top row) and Triton X-114 (bottom row) mediated recovery methods in five selected categories with distinct patterns (OD, SG, AC, EP, RP). In this figure, all environmental impacts were normalized relative to the no reuse scenarios. We simulated recovery efficiency from 0 to 1.0 over seven cycles of reuse. The other five categories (GW, CG, non-CG, ET, FF), which show similar patterns, are illustrated in Fig. S4.† The required amounts of pristine $HAuCl_4$ and the additional chemicals and electricity for recovery and recycling were calculated using eqn (1) and (2). Simply, if recovery efficiency and reuse cycles lie on the contour line of <100%, reuse strategies indicated greater environmental favorability than the no reuse case and *vice versa*.

The overall sensitivity patterns for both reuse strategies were quite similar. OD (and FF) showed the least sensitivity to the two variables, while EP (and CG, non-CG, ET) showed the most. In the range of the two variables in this study, the relative impacts were 90–125% for OD and 17–93% for EP. Overall, the many horizontal contour lines imply that the reuse cycles were less sensitive to environmental impacts than recovery efficiency for most cases. For the OD category, both reuse scenarios turned out to be more environmentally

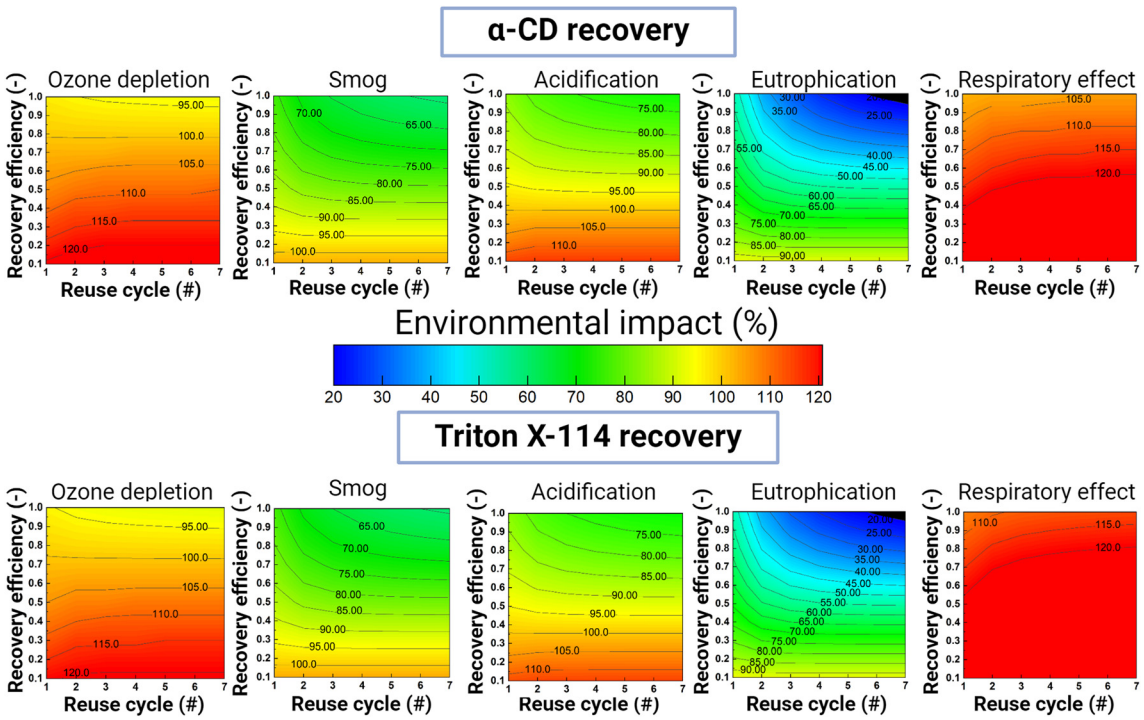

Fig. 5 Sensitive analysis of environmental impacts (ozone depletion, smog, acidification, eutrophication, respiratory effect) with varying recovery efficiency (0.0 to 1.0) and the number of reuse cycles (#). The environmental impacts were normalized by the no-reuse scenario.

favorable than the scenario with no reuse when recovery efficiency was higher than ~0.75 for any reuse cycle. OD was more sensitive to recovery efficiency than the number of reuse cycles. For the categories of SG and AC, if recovery efficiencies were >0.15 and >0.35, both relative impacts were <100% regardless of the number of reuse cycles. With an increase in the number of reuse cycles to 5–6, their impacts declined. For the EP category, at any recovery rate and number of reuse cycles, the impact was much less than the no-reuse scenario. Lastly, for the RP category, at any recovery efficiency and number of reuse cycles, the relative impact was higher than the control. Further, more reuse cycles showed a higher environmental impact. The higher recovery efficiency and the least number of reuse cycles would minimize RP. In short, except for RP, reuse strategies showed favorable environmental impacts compared to the no-reuse case if the recovery efficiency was higher than the contour line of 100%. Overall, the recovery efficiency is the dominant factor affecting the environmental favorability of AuNP-based applications and not the number of reuse cycles. Based upon this observation, the experimental recovery efficiency should be evaluated to determine if the sustainability of reuse strategies can ultimately be achieved. Also, the effect of the reuse cycles can be considered when designing reuse systems to minimize environmental impacts.

Energy and economic implications

The single point cumulative energy demand (CED, v. 1.05) for each reuse scenario was calculated in units of MJ. Sensitivity analysis of the relative CEDs implies that both reuse scenarios exhibit better environmental favorability than the no-reuse case when recovery efficiency was higher than 0.4–0.45 for any reuse cycle (Fig. S5†). Likewise, the effect of multiple reuse cycles was less than that for recovery efficiency.

We also investigated the direct implication of Au nano-waste reuse on the nanocircular economy. AuNPs had an estimated U.S. market value of ≈1180 million USD in 2022 that is expected to increase to ≈1511 million USD by 2025.[12] Regarding the mass production required by the growing demand for AuNPs and the decreasing amount of gold ore grades in nature, the sustainability of AuNP-based industry must be examined. We simulated the total costs of AuNPs by year in the U.S. for the reuse scenarios in comparison to those without reuse (Fig. 6). The market price of AuNPs is ≈80 USD per gram. Under the assumption that the market price is constant over the years, the costs of AuNPs can be proportionally reduced by the amount of reused AuNPs. For example, the market price of AuNPs in 2014 was 314.4 million USD which is equivalent to 3930 kg. With 30% recovery efficiency, only 3023 kg would have been required for production. Then, the market price would have dropped to 241.8 million USD. As a result, significantly reduced costs of AuNPs can be achieved in the years to come. The total cost of AuNPs decreased to ≈1162 and 889 million USD in 2025

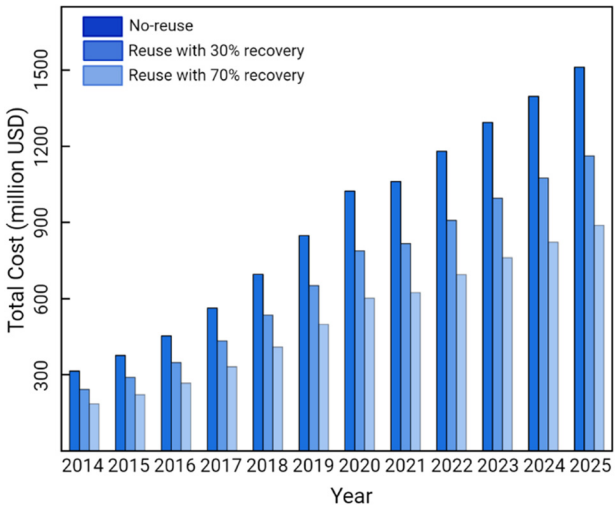

Fig. 6 Total cost of AuNPs in U.S. market by year with no-reuse scenario and reuse scenarios with 30 and 70% recovery efficiencies.

through AuNP reuse assuming either 30 or 70% recovery efficiency. There were 23.1 and 41.2% decreases in the costs from ≈1511 million USD with no-reuse scenario, respectively. The result implies the significant potential benefit of the nanocircular economy of AuNP-based industries.

Conclusions

We applied LCA to investigate the environmental impact of an AuNP-based sensing application. The system boundary consisted of AuNP synthesis, surface functionalization, detection assay, reuse of Au nano-waste (if applicable), and disposal. Two reuse strategies using α-CD and Triton X-114 were simulated and compared to the no-reuse scenario. The results showed that the environmental impacts of the AuNP-based sensing application were significantly reduced when Au nano-wastes were reused across a majority of the 10-midpoint categories. This finding was attributed to a significant reduction in the required amount of pristine Au^{3+} for AuNP synthesis despite the additional chemical and electrical demands required for the reuse of Au nano-waste. Furthermore, it was found that significant reductions in energy demand and AuNP total market costs can be achieved through Au nano-waste reuse. The results imply the potential of the reuse of Au nano-waste for the development of environmentally favorable AuNP-based sensing applications and help to quantitatively demonstrate the environmental benefits obtained from the reuse of Au nano-waste. Overall, we have shown the direct implications of the reuse of Au nano-waste for successful nanocircular economy development.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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