Pathways for Sustainable Adoption of Chitosan-Based Beads in Water Treatment

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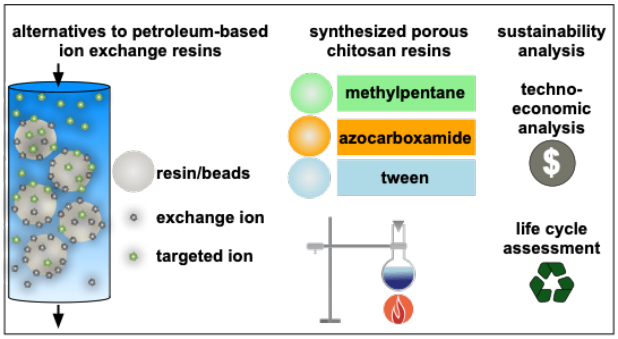
Graphical Abstract:



Keywords: circular economy, life cycle assessment (LCA), techno-economic analysis (TEA), water treatment, biopolymers, chitosan, ion exchange

Abstract

The increasing environmental concerns associated with petroleum-based ion exchange resins have spurred interest in sustainable alternatives, such as biopolymeric beads derived from chitosan. This study evaluated the sustainability of three porous chitosan particles synthesized with low-toxicity solvents (methylpentane porous chitosan particles, azocarboxamide porous chitosan particles, and tween porous chitosan particles) using techno-economic analysis and life cycle assessment. The results, normalized to both mass of particles produced and percent removal of methylene blue, revealed that azocarboxamide porous chitosan particles were the most cost-effective variant, despite the methylpentane ones exhibiting the highest removal efficiency. Environmental impacts were consistent across most categories, with azocarboxamide porous chitosan particles showing higher impacts for human toxicity (carcinogenic) and ozone depletion potential. Sensitivity analysis identified precursor costs, synthesis yield, chitosan and NaOH amounts, and electrical energy consumption as key drivers of sustainability. The findings emphasize the importance of considering both synthesis yield and treatment efficacy when evaluating the sustainability of chitosan-based ion exchange resins. Process optimization and exploration of eco-friendly alternatives are recommended to enhance the sustainability of these materials. This study contributes to the development of sustainable water treatment methods and promotes the transition towards a circular economy in the ion exchange resin industry.

Synopsis

Chitosan-based ion exchange resins offer a sustainable alternative to petroleum-based resins for water treatment, promoting a circular economy.

1.0 Introduction

The global market size of ion exchange resins for water treatment is estimated to be USD$5.85 billion/yr by 2028.[1] Many ion exchange resins used for water treatment are petroleum-based, often synthesized from styrene and divinylbenzene derived from petrochemicals.[2] These resins are polymeric structures that can be functionalized to carry specific ions, enabling ion exchange.[3] After their effective lifetime, spent ion exchange resins undergo degradation and gradually lose their efficacy. In addition to the disadvantage of being generated from petrochemicals, these spent resins decompose, leading to the formation of microplastics and posing further challenges to the environment. The predominant concern in the production of petroleum-based polymers release of greenhouse gas emissions.[4] Also, disposal requires careful consideration due to potential hazardous content.[5] Driven by the urgent need to safeguard our water resources, water treatment technologies are evolving unprecedentedly with focus on both novel and sustainable solutions.[6–11] A variety of methodologies for alternatives to petroleum-based resins have been proposed and implemented; however, finding the right balance between treatment efficacy and sustainability is the focus of ongoing studies.[12] Among the promising solutions are biopolymeric beads, which offer a potentially more sustainable alternative to conventional petroleum-based resins.

The advancement of biopolymers provides a pathway towards attaining environmental sustainability through the reduction of dependence on non-renewable fossil fuels.[13] Biopolymers (e.g., cellulose, chitin, and their derivatives) are known for their abundance, biocompatibility, biodegradability, and non-toxicity.[14] Biopolymers and their synthesized composites can be developed for targeted removal of various pollutants such as organic dyes, pesticides/herbicides, pharmaceuticals, polycyclic aromatic hydrocarbons, heavy metals, radioactive substances, and per- and polyfluoroalkyl substances.[4] The ability of biopolymers to be modified for improved functionally allows for the optimization of performance and the mitigation of specific contaminants.[15] Chitin has been explored as a precursor for alternative to petroleum-based ion exchange resins in numerous studies.[4,16,17] Despite the widespread interest in developing and testing biopolymers for ion exchange, only a handful of studies have assessed the sustainability of these beads as an alternative to synthetic polymers in water treatment.[14,18] Thus, a need exists to characterize the costs and environmental impacts of chitosan derived biopolymeric beads to develop sustainable water treatment methods.

Here, we assessed the costs and environmental impacts of biopolymeric beads for water treatment through techno-economic analysis and life cycle assessment, respectively. Sensitivity analysis was conducted to understand drivers for sustainability. This urgent evaluation focuses

on chitosan derived beads for the removal of model organic pollutant with opportunities to expand this framework to other biopolymers and contaminants. This work aims to help inform conversations on whether it is time to consider a pivotal shift toward the early adoption of sustainable alternatives to petroleum-based resins.

2.0 Methodology

2.1 Synthesis process for the three different chitosan beads

The synthesis procedure for the porous chitosan particle synthesis consists of two main phases (Figure 1).[19] In the general procedure (gray boxes), 2 grams of chitosan polymer is dissolved in 80 ml of a 2% glacial acetic acid solution at 70°C. The resulting chitosan solution is then added dropwise into a 1 M NaOH aqueous solution, leading to the formation of beads due to chitosan's insolubility in alkaline conditions. The aged beads are rinsed with deionized water to a neutral pH and then cross-linked with a 5% glutaraldehyde solution overnight. The cross-linked beads are subsequently washed and dried at room temperature. To enhance the porosity of the microsphere particles, three modified synthesis methods were employed, resulting in 2-methylpentane porous chitosan particles (MPCP), azocarboxamide porous chitosan particles (APCP), and tween porous chitosan particles (TPCP) being produced. In the MPCP method, an organic surfactant mixture of 10 mL 2-methylpentane and 24 mL tween 20 was added to the polymer solution. The APCP method involved incorporating a porogen agent, 4 g of azocarboxamide, into the chitosan solution. Lastly, the TPCP method utilized 35 mL tween 20 as a porogen agent.

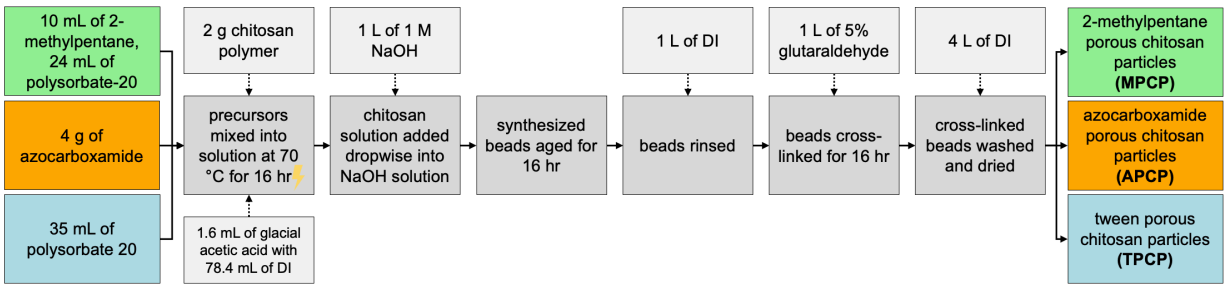


Figure 1. The color-coded boxes on the right illustrate the introduction of precursors into the general synthesis steps for chitosan particles, that are represented by a lighter color. The final chitosan particles are depicted with the same color as the precursors.

2.2 Sustainability framework with techno-economic analysis and life cycle assessment

The objective of this study was to conduct a thorough sustainability analysis on the synthesis processes of three porous chitosan particles. This analysis aims to evaluate the environmental and economic sustainability linked to these synthesis processes. The indicators employed to quantify these impacts were first normalized to per gram of beads produced from one synthesis cycle. The next level of analysis normalized indicators to percent removal of methylene blue, since water treatment was the primary motivation for this work. The methods for the methylene blue removal experiments are described in the Supplementary Information (SI) Section 1 with the results in Figure S1. The assessment of sustainability as well as the uncertainty and sensitivity analyses were completed in Python. The scripts and underlying assumptions for this work are openly available on GitHub for implementation.[20]

To estimate the costs of the synthesis processes for the three porous chitosan particles, TEA was used. This cost estimation was performed for materials and utilities, including chemicals, supplies, and electricity. The electricity cost was determined based on the energy consumed throughout the synthesis process. Although the overall cost of the general method used for general chitosan bead synthesis remained constant, variations emerged in the costs linked to procedures using different precursors. The environmental impacts for the different chemicals, inputs and energy were estimated by using LCA. Environmental impacts were estimated for acidification potential (kg SO2-Eq), global warming potential (kg CO2 eq), ecotoxicity: freshwater (CTUe), eutrophication potential (kg N-Eq), human toxicity: carcinogenic (CTUh), human toxicity: non-carcinogenic (CTUh), ozone depletion potential (kg CFC-11-Eq), particulate matter formation potential (kg PM2.5-Eq), and maximum incremental reactivity (kg O3-Eq). These impacts for the inputs used in the synthesis of different chitosan particles were taken from the ecoinvent v3.10 database.

2.3 Uncertainty and sensitivity analyses

To incorporate uncertainty into the analysis, a range of 25% uncertainty distribution was applied to all assumptions and data points for each parameter, depending on the level of confidence and data availability. This approach captures the potential fluctuations in materials cost and impacts. A total of 10,000 Monte Carlo simulations were conducted to quantify and address the uncertainty in the system. The results obtained for the uncertainty analysis includes the averages in the form of median, 5^{th} percentile, and 95^{th} percentile values. Additionally, a sensitivity analysis was performed using Spearman's rank correlation coefficients to identify the key drivers of changes in the system's cost and environmental impacts. The sensitivity of

individual parameters was analyzed for three different chitosan beads. In this study, we present the Spearman's rank correlation coefficients (absolute values > |0.05| with p-values < 0.05) for costs and environmental impacts.

3.0 Results and Discussion

3.1 Economic viability and environmental implications of chitosan bead production and treatment efficacy

In our initial level of analysis, the indicators for cost and environmental impacts were normalized based on the mass of porous chitosan particles produced. These results revealed that TPCP and MPCP were the highest cost to produce at 123.27 [93.28-161.80] USD/g (denoted as median [5th–95th percentile]) and 104.07 [80.76-135.07] USD/g, respectively (Figure S2). In contrast, APCP proved to be the most cost-effective method, with a price of 32.89 [26.13-41.00] UDS/g. The cost differences among chitosan particle variants stemmed from the precursors used during synthesis while maintaining a uniform general technique. The incorporation of azodicarbonamide as foaming agent in the preparation process a cost-effective approach as also demonstrated by several other studies.[21,22] It is suggested that future research endeavors focused on enhancing porosity to improve adsorption capacity via tuning the amount of azodicarbonamide.[23] Similar trends were observed in our next level of analysis that normalized costs to percent removal of the methylene blue (Figure 2). Specifically, the cost of TPCP was 3.78 [2.92-4.89] USD/% removed, MPCP was 1.96 [1.55-2.48] USD/% removed, and APCP was 0.74 [0.60-0.89] USD/% removed. This analysis provides a more meaningful comparison of the costs associated with each chitosan particle variant by considering their effectiveness in removing the target pollutant. The results highlight the importance of not only considering the production costs but also the performance of the particles in their intended application. In another study, azodicarbonamide utilized in the preparation of polymeric lignin composite and revealed a high removal efficiency of methylene blue.[24]

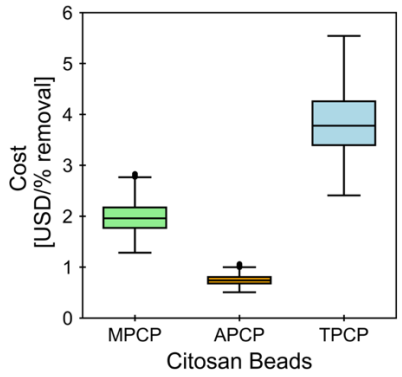


Figure 2. Estimated costs for the synthesis of 2-methylpentane porous chitosan particles, (MPCP), azocarboxamide porous chitosan particles (APCP), and tween porous chitosan particles (TPCP). The plot shows the costs on the ordinate normalized to percent removal of methylene blue and three different particles on the abscissa. The uncertainty analysis was conducted by 10,000 Monte Carlo simulations. Boxes and whiskers show the median values (centerline), 25th and 75th percentiles (bottom and top of the box), and 5th and 95th percentiles (lower and upper whiskers).

When results were normalized to mass of the porous chitosan particles produced, similar trends were observed in the estimates for acidification potential at 0.0047 [0.0041- 0.0053] kg SO2-Eq/g, global warming potential at 0.91 [0.81-1.03] kg CO2 eq/g, ecotoxicity: freshwater at 7.29 [6.49-8.24] CTUe/g, eutrophication potential at 0.0064 [0.0054-0.0075] kg N-Eq/g, human toxicity: non-carcinogenic at 1.79×10^{-7} [1.59×10^{-7} - 2.02×10^{-7}] CTUh/g, particulate matter formation potential at 0.0011 [0.010-0.013] kg PM2.5-Eq/g, and maximum incremental reactivity at 0.47 [0.42-0.53] kg O3-Eq/g (Figure S3). These results indicate that the general chitosan synthesis procedure is the main contributor to the environmental impacts when considering the mass of particles produced. Despite the advantages of conventional agents like azodicarbonamide, significant environmental issues remains, such as contribution to global warming and ozone depletion, as well as safety concerns regarding potential carcinogenicity and flammability.[25] Notably, this study found out APCP had drastically higher estimates for human toxicity: carcinogenic at 0.15 [0.12-0.18] CTUh/g and ozone depletion potential at 5.96×10^{-8} [14.95×10^{-8} - 7.13×10^{-8}] kg CFC-11-Eq/g. These findings suggest that the precursors used in the APCP synthesis may have a more significant impact on these specific environmental categories. Further investigation into the specific components and their potential toxicity would be necessary to identify opportunities for mitigating these impacts.

Normalizing the environmental impacts to percent removal of methylene blue revealed different trends compared to the mass-normalized results (Figure 3). MPCP had the lowest environmental impacts across all the categories, indicating that its higher removal efficiency compensates for the impacts associated with its production. TPCP had the highest impacts for all categories except human toxicity: carcinogenic, suggesting that its lower removal efficiency exacerbates the environmental burdens when considering its functional performance. The divergence in trends between the mass-normalized and percent removal-normalized results highlights the importance of considering the intended application and functionality of the chitosan particles when assessing their environmental sustainability. While the mass-normalized results provide insights into the inherent environmental impacts of the production process, the percent removal-normalized results offer a more comprehensive understanding of the particles' environmental performance in the context of their pollutant removal capabilities.

These findings reveal the complex tradeoffs that exist among the costs and environmental impacts of the different chitosan particle variants. The results emphasize the need for a holistic approach when evaluating the sustainability of these materials, considering both their production costs and their effectiveness in the intended application. By normalizing

the results to the functionality of the produced chitosan particles, decision-makers can make more informed choices that balance economic viability with environmental sustainability.

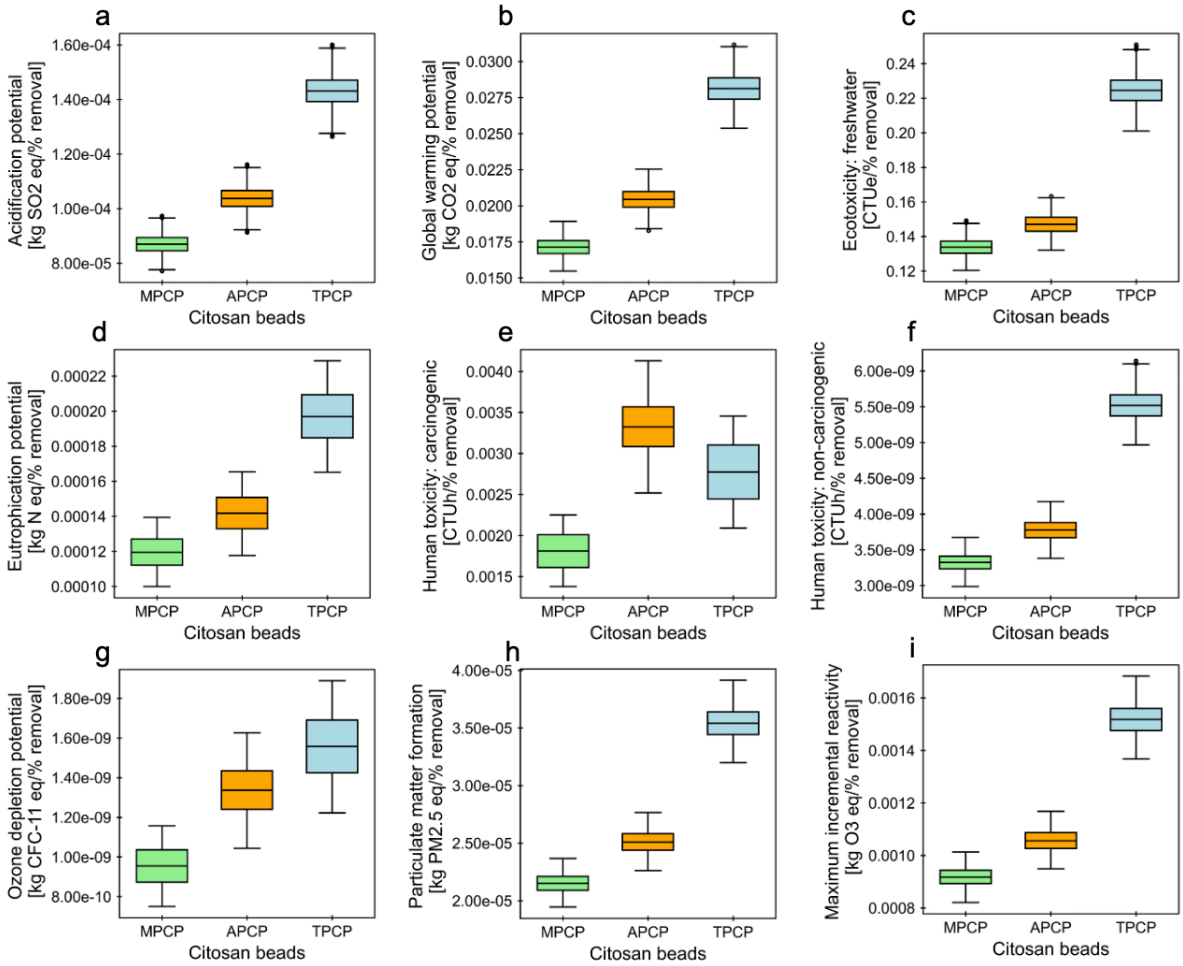


Figure 3. Estimated environmental impacts for the synthesis of 2-methylpentane porous chitosan particles, (MPCP), azocarboxamide porous chitosan particles (APCP), and tween porous chitosan particles (TPCP). The plots show the specific impact categories on the ordinate normalized to percent removal of methylene blue and three different particles on the abscissa. Environmental impact categories are global warming potential (a), acidification potential (b), ecotoxicity (c), eutrophication (d), human toxicity: non-carcinogenic (e), human toxicity: carcinogenic (f), ozone depletion (g), particulate matter formation potential (h), and maximum incremental reactivity (i). The uncertainty analysis was conducted by 10,000 Monte Carlo simulations. Boxes and whiskers show the median values (centerline), 25th and 75th percentiles (bottom and top of the box), and 5th and 95th percentiles (lower and upper whiskers).

3.2 Charting sustainability pathways

In our next level of analysis, we explore which assumptions are driving the results through sensitivity analysis. Overall, 14 different assumptions were found to be significant (Figure 4). The precursors associated with the production of chitosan particles are found to drivers for costs, including tween 20, polysorbate 20, and glutaraldehyde. Additionally, the synthesis yield affects both environmental impacts and cost. Thus, opportunities exists to

enhance the synthesis conditions, implement process efficiency enhancements, and explore opportunities to scale up. The amounts of chitosan and NaOH also impact the environmental sustainability, including eutrophication, human toxicity: carcinogenic, and ozone depletion potential. To address these impacts, further optimization in the synthesis process can be explored. This optimization will help reduce the environmental impact and promote a more sustainable approach to synthesis. Other research has noted that the viability of chitosan production facilities stems from the utilization of NaOH and HCl.[26,27] The consumption of electrical energy by hotplates contributes greatly to increased environmental stress, which is further influenced by the duration of heat application. A viable solution is to explore energy-efficient alternatives and optimize the duration of heat application.

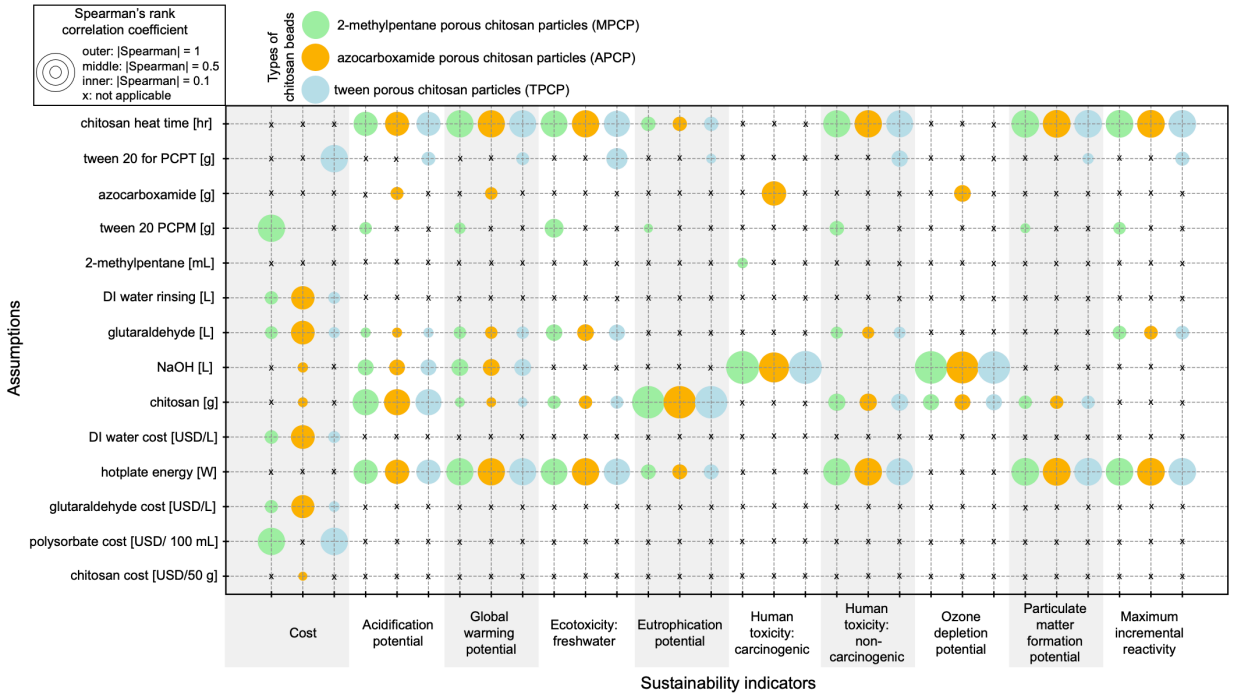


Figure 4. Spearman's rank correlation for costs and environmental impacts for the three different particles. The key drivers are on the ordinate corresponding with particle's cost and environmental impact on the abscissa.

3.3 Conclusions

In this study, we conducted a comprehensive sustainability assessment of three porous chitosan particles as potential alternatives to petroleum-based ion exchange resins for water treatment. When normalized to mass produced, APCP was the most cost-effective variant, while TPCP and MPCP were significantly more expensive. However, when normalized to percent removal of methylene blue, MPCP exhibited the lowest environmental impacts, while TPCP had

the highest impacts for most categories. Sensitivity analysis identified precursor costs, synthesis yield, chitosan and NaOH amounts, and electrical energy consumption as key drivers of sustainability. To enhance the sustainability of chitosan-based ion exchange resins, it is crucial to optimize synthesis conditions, implement process efficiency enhancements, optimize precursor quantities, and improve energy efficiency. This study provides valuable insights into the economic viability and environmental implications of chitosan-based ion exchange resins as alternatives to petroleum-based materials, emphasizing the importance of considering both production costs and treatment efficacy when evaluating their sustainability. Further research is recommended to optimize synthesis conditions, investigate the applicability of this framework to other biopolymers and contaminants, and explore the potential for scaling up the production of these sustainable alternatives.

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