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Sustainability assessment of nanoscale zero-valent iron production processes for contaminated site remediation through life cycle analysis tools

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ABSTRACT

Nanoscale zero-valent iron (nZVI) is the main nanomaterial used in environmental remediation processes. This article aims to evaluate the sustainability of nZVI production methods used in environmental remediation. Three nZVI production methods were selected: milling, chemical reduction with sodium borohydride, and chemical reduction with hydrogen gas. Sustainability assessment was made through three life cycle analyses: life cycle assessment (LCA) for environmental impacts, life cycle cost (LCC) for economic impacts, and social life cycle assessment (S-LCA) for social impacts. All analyses were performed based on the functional unit of 10.00 kg of nZVI produced. The LCA and LCC were performed in SimaPro. The S-LCA was conducted via stakeholder participation, through a social questionnaire, weighting factors were determined and used in the Social Index calculation. The milling method resulted in the lowest environmental, economic, and social impacts; while the hydrogen gas reduction method had the greatest economic and social impacts and the reduction with sodium borohydride method had the greatest environmental impacts. Overall, this study significantly contributed to the state-of-the-art application of sustainability assessment using life cycle tools in nanomaterial studies.

Keywords: Life cycle sustainability assessment; life cycle cost; social life cycle assessment; sustainable remediation; milling; chemical reduction.

1 INTRODUCTION

The process of contaminated site remediation has undergone countless changes over the years (Braun et al. 2019). The concern with the impacts generated by the remediation processes at the environmental, economic, and social levels has resulted in the inclusion of sustainability in this context. In this sense, the term “sustainable remediation” illustrates a broader approach to remediation, which considers the complete process involving all the impacts that can be generated throughout the remediation process life cycle (Rizzo et al. 2016; Alvarenga 2022).

There are numerous techniques that can be applied in the remediation of contaminated sites. One of the techniques that is growing in popularity in both studies and application is nanoremediation (i.e., using nanomaterials in remediation) (Hussain et al. 2022). In this sense, nanoscale zero-valent iron (nZVI) stands out as one of the most studied nanomaterials used in environmental remediation (Zhao et al. 2016). For example, nZVI corresponds to 47% of the nanomaterial applications in soil remediation processes in the United States (EPA 2016). Its extensive use is attributed to its removal efficiency and due to its low toxicity and production cost (Visentin et al. 2019). In practice, nZVI can be applied in both soil and groundwater remediation through in situ injection. The importance of this nanomaterial in environmental remediation demonstrates the need for increasing knowledge of its production and application.

Many of the uncertainties associated with nanomaterials involve their production and the impacts of the production processes (Thomé et al. 2015). In addition, the impacts of material production used in remediation should also be considered. Thus, numerous methods have been developed to assess the sustainability of remediation, such as life cycle assessment (LCA). LCA is an important tool for scientific research in different areas, as it is one of the main methods of sustainability analysis (Li et al. 2018; Visentin et al. 2019). However, in essence, LCA does not cover sustainability as a whole, only the environmental dimension. In this sense, for a sustainability assessment, it is necessary to consider the three pillars of sustainability: environmental, economic, and social (Visentin et al. 2020). Thus, life cycle cost (LCC) and social life cycle assessment (S-LCA) are also considered (Costa, Quinteiro and Dias, 2019) (Masilela and Pradhan 2022).

As a decision support tool, sustainability assessment provides an overview of the sustainability performance of production systems, highlighting areas of significant negative impact, where improvements or positive impacts can be made, and where opportunities can be explored (Gbededo et al. 2018; Kabayo et al. 2019, Wu et al. 2022). Therefore, knowledge of the sustainability of remediation techniques and the materials used in remediation becomes an important ally of decision makers in choosing the best remediation alternatives, while considering the combined environmental, economic, and social aspects of the overall process. Thus, this work aims to analyse the sustainability of nZVI production methods using life cycle analysis tools.

2 METHODOLOGY

2.1 nZVI production methods

Although the production of nanomaterials can be performed via numerous methods, in both laboratory and industrial settings, there are two main production technologies: top-down and bottom-up. Top-down technologies comprise processes that begin with particles of a larger size, such as bulk material, and through the application of physical mechanisms are reduced until they reach a nanometric size (10-9 meters) (Crane and Scott, 2012). On the other hand, bottom-up technologies, which begin with materials of a size smaller than a nanometer, such as atoms and molecules, produce nanomaterials through chemical reactions (Zhao et al. 2016).

Three nZVI production methods were selected for life cycle analysis in the present study: milling, chemical reduction with sodium borohydride, and chemical reduction with hydrogen gas. These methods were selected because they involve both top-down and bottom-up technologies and can be applied in both laboratory and industrial settings (Crane and Scott, 2012). According to Visentin et al. (2019), the chemical sodium borohydride reduction method is the most commonly used method in laboratory studies, while the milling method is used by a United States company and the chemical hydrogen gas reduction method is used by a Japanese company.

The milling method is a physical, top-down process. In this method, particles of bulk iron are ground in a rotating chamber until they reach the desired nanometric size, which is approximately 20 nm (Jung et al. 2015). The sodium borohydride reduction method is mainly used in laboratory-scale production. This method is a chemical, bottom-up process. For the nZVI production, sodium borohydride and ferric chloride reagents are combined (Barreto-Rodrigues et al. 2017). Sodium borohydride acts as a reducing agent for ferric chloride in the reduction reaction used to produce nZVI. Finally, the hydrogen gas reduction method is a chemical, bottom-up process applied in the industrial production of nZVI by a Japanese company. In this method, goethite and hematite particles are reduced with hydrogen gas at high temperatures to produce nZVI (Uegami et al. 2009).

2.2 Methodological procedures for life cycle analyses (LCA, LCC and S-LCA)

Three life cycle analyses (environmental, economic, and social) were performed for the selected nZVI production methods (milling, reduction with sodium borohydride, and reduction with hydrogen gas), considering the ISO 14040 (2006) stages: goal and scope definition, inventory analysis, impact assessment, and interpretation and sensitivity analysis.

2.2.1 Goal and scope definition

The aim of the life cycle analyses (LCA, LCC, and S-LCA) is to evaluate the environmental, economic, and social impact of nZVI production methods applied in soil and groundwater remediation in order to identify the method with the lowest environmental impacts and costs, and greatest social benefits. The results will potentially increase awareness of the environmental, economic, and social implications related to nZVI production and identify possibilities for improvements. The target audience includes the decision makers for sustainable remediation, designers seeking remediation techniques, and managers and workers of companies directly affected by the production of nZVI.

This study is based on a cradle-to-gate approach, that is, the limits of the system involve the extraction of raw materials up until the production of nZVI. The S-LCA extends across both the city and country of the studied manufacturers. The functional unit employed was 10.00 kg of nZVI produced. For this study, the performance of nZVI remediation in the methods was not considered.

2.2.2 Inventory

All inventory data used were secondary, or in other words, were obtained through external sources from companies using nZVI production methods. Primary data was not used, as it should be obtained directly from the companies, which was not possible due to their privacy policies. Secondary data were obtained from peer-reviewed scientific publications, patents, similar processes, and available databases; raw material supply, industrial waste waterway treatment companies, and hazardous waste incineration companies; and government reports on energy balance. In addition, another data source used was the SimaPro software's ecoinvent database. Table 1 presents the environmental and economic inventory of the nZVI production methods, and table 2 shows the impact categories and indicators of the S-LCA

Table 1. *Environmental and economic inventory of the nZVI production methods.*

Stage synthesis process	Inputs/Outputs	Amount	Unit cost
Milling method			
Milling	Iron	10.00 kg	$87.50
	Energy	180 kWh	$0.15
	Steel spheres	18 kg	
	Particulate material	2.00×10^{-3} g	
Internal costs	Labor Costs	8 h	$7.25
Reduction with sodium borohydride method			
Mixture of reactants	$FeCl_3$	28.30 kg	$41.60
	$NaBH_4$	25.60 kg	$303.00
Stirring	Energy	1.35×10^{-1} kWh	$0.15
Filtration	Energy	2.1×10^{-2} kWh	$6.58
	Filter papers	23.70 kg	$32.75
	Solid waste	23.70 kg	$1.20
	Wastewater	7.02×10^{-1} m³	$0.15
Washing	Ethanol	38.40 kg	$0.12
	Deionized water	2.10 m³	$0.02
	Wastewater	2.43 m³	$0.15
Internal Costs	Labor Costs	1 h	$7.25
Reduction with hydrogen method			
Production of goethite particles			
Step 1	$NaCO_3$	38.30 kg	$39.08
	$FeSO_4$	26.50 kg	$166.00
	N_2 (gas)	6.23 kg	$0.04
	Energy	33.6 kWh	$0.14
	Wastewater	1.98×10^{-2} m³	
Step 2	$FeCO_3$	28.40 kg	$9.98
	$Al(OH_3)$	0.954 kg	$90.50
	N_2 (gas)	21.50 kg	$0.04
	Energy	126 kWh	$0.14
	Wastewater	12.45×10^{-2} m	$0.15
	Energy	0.4 kWh	$0.14
	Wastewater	8.67×10^{-2} m³	$0.15
Shaping	Energy	76 kWh	$0.14
nZVI production			

Reduction	Energy	207 kWh	\$0.14
	H_2 (gas)	$6.2x10^{-1}$ kg	\$0.04
	N_2 (gás)	$2.92x10^{-2}$ kg	\$0.04
	Water (vapor)	$6.23x10^{-1}$ kg	
Oxidation	Water	$6.10x10^{-2}$ m^3	\$0.04
Drying	Energy	2.40 kWh	\$0.14
	Wastewater	$3.89x10^{-2}$ m^3	\$0.15
Internal Costs	Labor Costs	18 \$	\$7.63

Table 2. *Impact categories and indicators of S-LCA of the nZVI production methods.*

Stakeholders categories	**Impacts categories**	**Indicators**
Workers	Freedom of negotiation and collective association	Cooperation in work-employer relations
		Hiring and dismissal practices
		Collective bargaining coverage
		Union
	Child labor	Child labor
		Number of children outside school
	Fair wage	Minimum wage (U\$/hour)
		Flexibility of wage determination
		Remuneration and productivity
	Working hours	Average working hours per week
	Equal opportunity/discrimination	Participation of women in labor force
		Equal pay for similar work
	Health and safety	Occurrence of occupational lethal accidents per year
		Occurrence of occupational non-lethal accidents per year
		Exposure of workers to chemical products
		Health risks during the production process
Local Community	Safe and healthy living conditions	Carbon intensity (kg per PPP \$ of GDP)
	Access to material resources	Population with improved access to clean water
		Population with improved sanitation access
		Electricity supply quality
Society	Market and work	Country unemployment rate
		Efficiency of the labor market
	Contribution to economic development	Extension of commercialization
		Sophistication of the production process
		Collaboration between university and industry
	Governance	Efficiency of government expenditure
		Transparency in the elaboration of governmental policies
		Total tax rate and% of profits
Value chain	Fair competition	Intensity of local competition
		Ethical behavior of companies
		Strength of audit and reporting standards

2.2.3 Impact assessment

The impact assessment of the LCA and LCC were performed in SimaPro according to Visentin et al. (2019) methodology. In the LCA, the impact analysis methodology used was Impact 2002+. The categories of endpoint impact were considered to be human health, ecosystem quality, climate change, and resources.

In the LCC, the impacts were analysed in SimaPro through the elaboration of an impact analysis methodology. The internal and external costs of nZVI production were analysed. Internal costs are costs directly associated with production, such as those that involve raw materials, energy, labor, waste disposal, and other effluent costs. External costs are the environmental costs resulting from the environmental impacts generated from production, such as global warming, acidification, eutrophication, and toxicity). These costs are obtained by evaluating the LCA results.

The S-LCA was performed according to the methodology of Visentin et al. (2022). The categories of endpoint impact considered were: human resources management; community development; society development and corporate social responsibility.

3 RESULTS AND DISCUSSION

3.1 Life cycle assessment

Figure 1 presents the LCA normalized results for nZVI production methods in relation to total environmental impacts and impact categories, considering the functional unit of 10.00 kg of nZVI produced. Furthermore, the data represent the production method scenarios in terms of location, being the United States for the milling and sodium borohydride reduction methods and Japan for the hydrogen gas reduction method. The impact results are expressed in millionths of points (mPt). The magnitude of this numerical value expresses the size of the global environmental impact: the higher the value of the indicator, the greater the environmental impact of the production method (Visentin et al. 2019).

For the treatment of waste generated in the production methods, the incineration of solid waste and industrial treatment of wastewater were considered. Thus, considering the inventory data, the sodium borohydride reduction method includes the incineration of solid waste and wastewater treatment, and the hydrogen gas reduction method only includes the treatment of wastewater. While in the milling method, no solid waste or wastewater are generated.

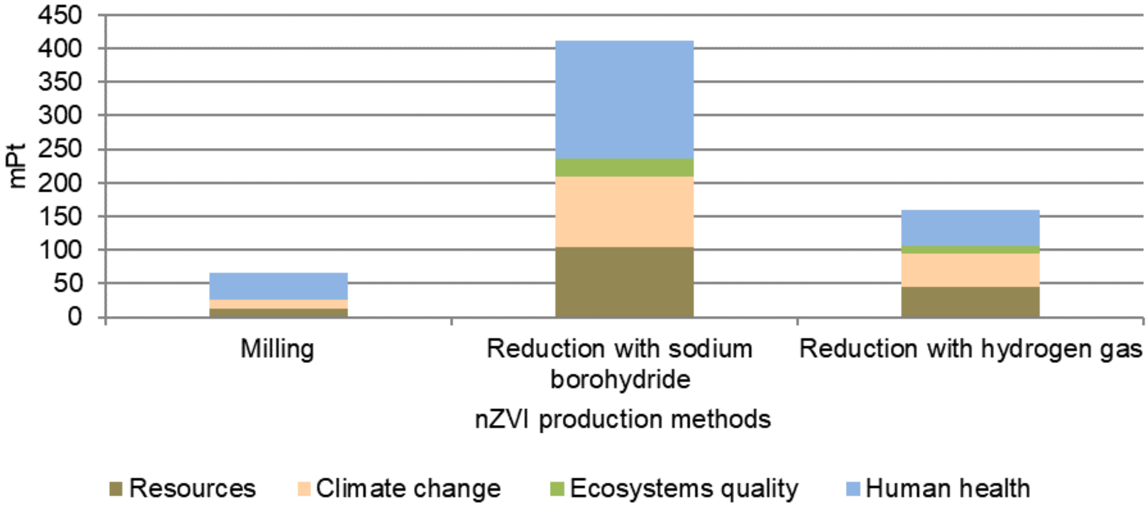

Figure 1. *LCA environmental impacts of nZVI production methods for the impact categories.*

The milling method resulted in the lowest total environmental impacts, followed by the hydrogen gas reduction method, while the sodium borohydride reduction method resulted in the greatest environmental impacts.

The milling method is a simple method that requires the use of rotating equipment such as a grinding chamber. In addition, in this method, the inputs used are simpler than those used in the other methods – in this case, it is necessary to use only large or micro-size iron particles and also energy for operation. In the sodium borohydride reduction method, reagents, such as sodium borohydride and iron chloride, are used, and these reagents contribute to greater environmental impacts. However, the energy consumption in the sodium borohydride reduction method is the lowest of the methods analysed, as it is a simpler and faster process (Visentin et al. 2019). The hydrogen gas reduction method is the most complex method of the three analysed, with more steps in the production of nZVI. In this method, reagents, such as calcium carbonate, iron sulfate (II), aluminium hydroxide, and nitrogen, are consumed, and these reagents are associated with the environmental impacts of their production, thus, contributing to the overall environmental impacts of this method. Another factor that also contributes to the environmental impacts of hydrogen gas reduction method is its high energy consumption, which is the largest of the methods analysed.

The impacts of the sodium borohydride reduction method are mainly related to the sodium borohydride reagent, corresponding to more than 90% of the method impacts. The impacts of the hydrogen gas reduction method depend mainly on the goethite particle production and energy consumption. The environmental impacts of this method can be reduced by changes in the production process, especially in relation to the goethite particle production stage (Visentin et al. 2019). Using pre-synthesized goethite particles can result in a 42% decrease in environmental impacts in human health, climate change and resources categories. However, the quality of the goethite particles may not be guaranteed in the same way as those produced by the company, and may influence the quality of the nZVI produced.

Two factors are associated with the environmental impact methods: the reagents used in the processes and the energy consumption. According to Visentin et al. (2019), sodium borohydride production is associated with a greater portion of the sodium borohydride reduction method's environmental impacts, while the greatest contribution to the environmental impacts of the milling and hydrogen gas reduction methods is related to energy consumption.

In the milling method, it is perceived that the greatest impacts are in the human health category, as well as the climate change and resources categories. In the sodium borohydride reduction method, impacts are perceived in all categories analysed, but are higher in the human health category. In this method, the impacts in the ecosystem quality category are the greatest of all the methods. In the hydrogen gas reduction method, the greatest impacts are determined to be in the human health category, followed by the climate change and resources categories.

The main impact to human health comes from respiratory inorganic pollutants emission, with 59% in the milling method, 10% in the sodium borohydride reduction method, and 26% in the hydrogen gas reduction method. In addition, the impacts in this category also contribute to carcinogenic and non-carcinogenic pollutants emission: 1% in the milling method, 3% in the sodium borohydride reduction method, and 7% in the hydrogen gas reduction method. In this category, the values of environmental impacts for the sodium borohydride reduction method are 4.4 times higher than the milling method values, and 3.2 times higher than the hydrogen gas reduction methods.

Regarding ecosystem quality, the main impacts come from terrestrial ecotoxicity and land toxicity in the sodium borohydride reduction method, which correspond to 2% and 23% of the impacts, respectively. The milling and hydrogen gas reduction methods do not significantly contribute to the impacts in this category, adding only 1% and 7% to the impacts, respectively. In this category, the values of environmental impacts for the sodium borohydride reduction method are 26.5 times higher than the milling method values and 2.4 times higher than the hydrogen gas reduction method values.

In terms of climate change, the main impact comes from global warming, with 58% in the sodium borohydride reduction method, 18% in the milling method, and 31% in the hydrogen gas reduction method. In this category, the values of environmental impacts for the sodium borohydride reduction method are 8.4 times higher than the milling method values and 2.1 times higher than the hydrogen gas reduction method values.

In relation to resources, the main impact comes from the use of non-renewable energies, which corresponds to 4% in the sodium borohydride reduction method, 20% in the milling method, and 28% in the hydrogen gas reduction method. In this category, the values of environmental impacts for the sodium borohydride reduction method are 7.8 times higher than the milling method values and 2.3 times higher than the hydrogen gas reduction method values.

3.2 Life cycle costs

Figure 2 presents the LCC results of the nZVI production methods, detailing the costs in the categories of materials, energy, waste and wastewater treatment, labor, and external environmental costs. The results are expressed in USD/10.00kg of nZVI. These costs were obtained after the Monte Carlo analysis was performed on the initial costs of the LCC in order to minimize the uncertainty of the data.

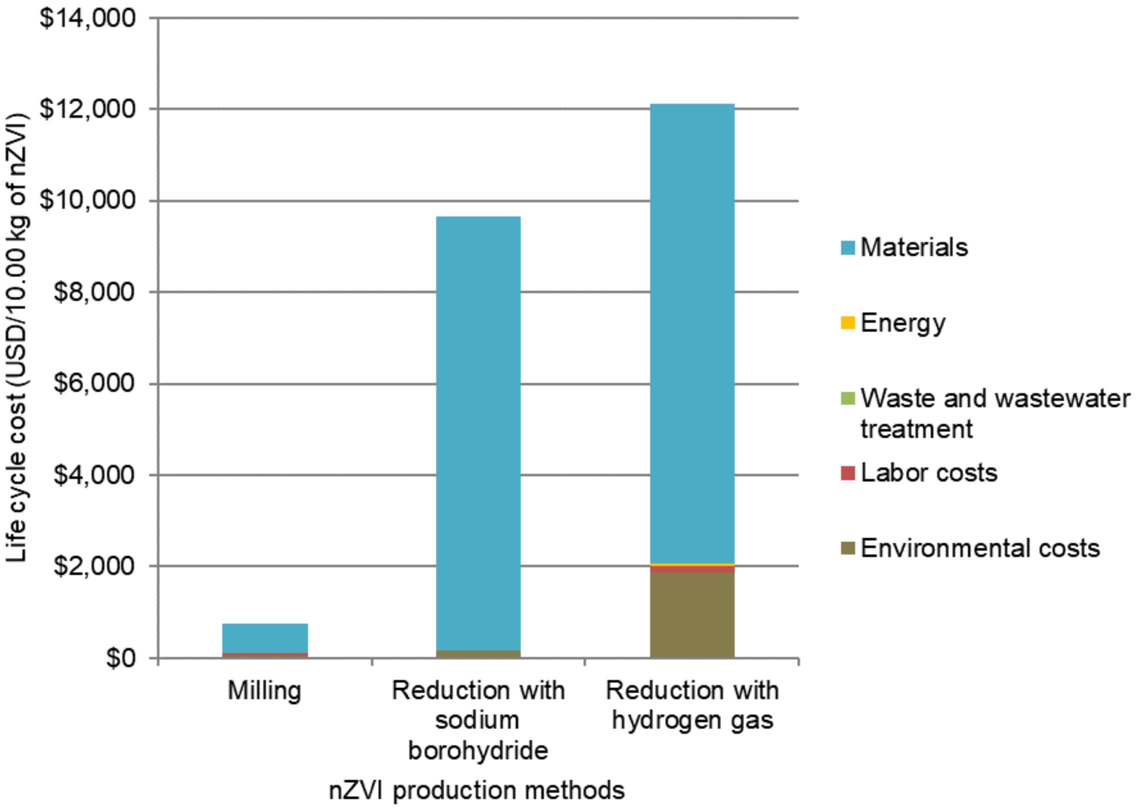

Figure 2. *LCC of the nZVI production methods for cost categories.*

The hydrogen gas reduction method resulted in higher LCCs, followed by the sodium borohydride reduction method. The milling method resulted in the lowest cost. Compared to other methods, the simplicity of the milling method, and the use of only one material in the process makes this method the cheapest.

In a cost detailing of the methods, it is perceived that the costs are mainly associated with the reagent inputs and the environmental costs. Material costs correspond to 85% in the milling method, 98% in the sodium borohydride reduction method, and 84% in the hydrogen gas reduction method. Environmental costs correspond to 6% in the milling method, 2% in the sodium borohydride reduction method, and 8% in the hydrogen gas reduction method.

The high association of materials costs (98%) in the sodium borohydride reduction method is primarily related to the high cost of sodium borohydride, which corresponds to 86% of the material costs. According to Visentin et al. (2019), the high cost of sodium borohydride is associated with its production, which involves numerous complex steps, with the reagents of sodium hydride (NaH) and trimethyl borate (B(OCH3)3). Thus, it is perceived that the costs obtained in this analysis corroborate what many authors affirm in relation to the sodium borohydride reduction method: That the high cost of sodium borohydride makes the feasibility of the use of this method on an industrial scale difficult. Moreover, a favourable economic change in this method would be the decrease in the cost of the sodium borohydride. However, in order to reduce these costs, the sodium borohydride synthesis process should be modified, or processes and reagents with lower costs should be considered. Or, alternatively, another chemical reducer that may be able to act as a substitute for this reagent should be employed.

The industrial energy cost applied within a country is relatively lower than the cost of domestic energy, so it is perceived that these costs do not end up contributing significantly to the total costs of the methods. Also, it is perceived that labor costs are directly related to the production time of each method. In the milling method, it takes 8 hours for the production of nZVI; in the hydrogen gas reduction method, it takes 18 hours; and in the sodium borohydride reduction method, it only takes 35 minutes. Thus, the labor costs correspond to 8% of the milling method costs, 0.1% in the sodium borohydride reduction method costs, and 1% in the hydrogen gas reduction method costs. Finally, wastewater treatment and waste incineration processes account for 0.1% of the costs in all methods.

In the hydrogen gas reduction method, as analysed in the LCA, a change in the production process through the use of pre-synthesized goethite particles could result in a 39% decrease in method costs.

3.3 Social life cycle assessment

Figure 3 shows the results of the S-LCA as one-dimensional data regarding the production methods of nZVI. The breakdown of social impact within the endpoint impact categories of each method is detailed. The social impact is expressed through scores from 0.00 to 1.00, with 1.00 representing a method with better social indices.

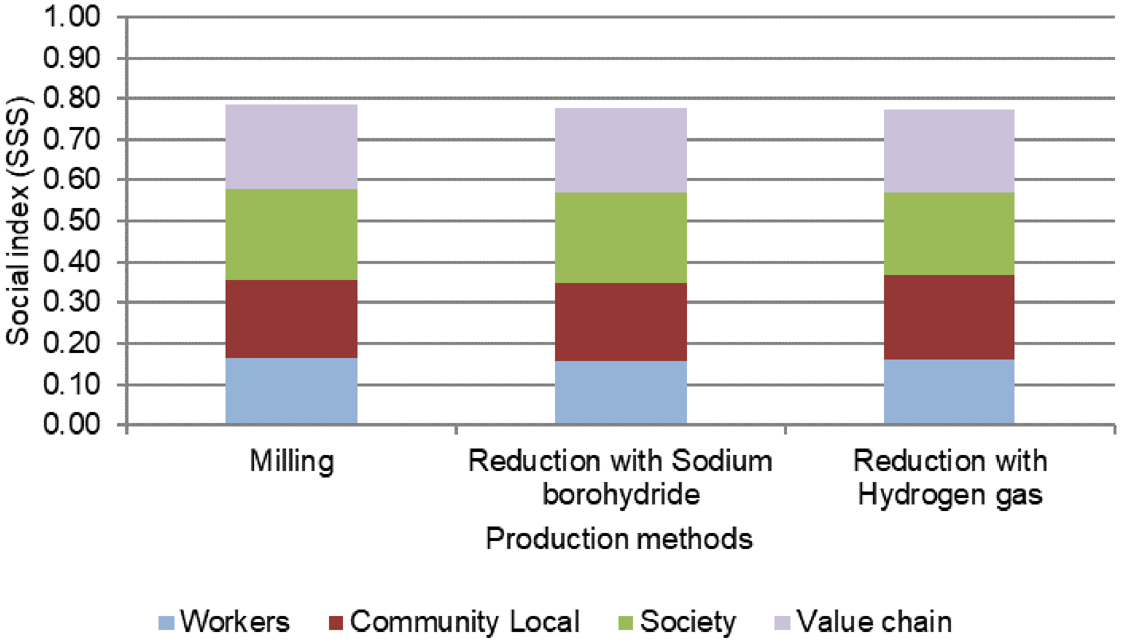

Figure 3. *S-LCA results of nZVI production methods for social impact categories.*

The three production methods of nZVI have the same global Social Index value. In a more detailed analysis, considering the decimal differences, one can say that the hydrogen gas reduction method resulted in the lowest social indices (SSS = 0.7726), followed by the sodium borohydride reduction method (SSS = 0.7806). The milling method displayed the best overall social indices (SSS = 0.7887).

The small difference between the inventory data of the social factors of the methods' localization countries (United States and Japan) resulted in small differences in the social index (0.016). The United States and Japan have many similarities regarding social issues, including, for example, the sophistication of the production process, flexibility in determining salaries, hiring and dismissal practices, etc. (see Table 4). However, some of the indicators observed differences that resulted in a lower score for Japan in the subcategories, such as equal opportunities and discrimination, working hours, child labor, etc.

The milling and sodium borohydride reduction methods have similar values in all the impact categories, except in human resources management. The indexes of both methods were determined from the United States inventory data, so data equality is expected. The difference between the methods is given in the category of human resource management due to the specific data of the methods in relation to the exposure of workers to chemicals, which is more significant in the sodium borohydride reduction method than in the milling method.

In the hydrogen gas reduction method, the lowest indices were verified in the human resources management category. In this method, the data was from Japan, and, regarding the many indicators of this category, the scores were lower than in other methods. In addition, the specific indicators of the method (i.e., exposure of workers to chemical products and health risks during the production process) also contributed to the lowest score in the category.

The community development category contributed positively to the hydrogen gas reduction method score, showing the highest score among the three methods. In this category, the carbon intensity indicator was the main contributor to the score. This indicator represents the carbon emission rate in relation to the intensity of a specific activity or to an industrial production process, which includes, for example, grams of carbon dioxide released by a megajoule of energy produced, or the ratio of greenhouse gas emissions produced and gross domestic product (GDP) (Brizga et al. 2017). Emission intensities were used to obtain estimates of atmospheric pollutants or greenhouse gas emissions based on the amount of burnt fuel present (Brizga et al. 2017). The values of this indicator highlighted the differences between the United States and Japan, respectively 46.4 and 31.9 kg per PPP $ of GDP.
In the development of the society category, the method of reduction via hydrogen gas displayed the lowest score. The indicators that contributed to this effect were the efficiency of the labor market, the extension of commercialization, the collaboration between a university and industry, and the efficiency of government expenditures. Regarding these indicators, Japan's score was lower than that of the United States, resulting in lower overall impact scores.

In the category of social and corporate responsibility, there were minimal observed differences between the methods due to both country's indicator score similarities. The difference depends on the intensity of the local competition indicator, which results in the lowest score for the reduction via hydrogen gas method.

4 CONCLUSIONS

Three life cycle analyses (LCA: environmental, LCC: economic, and S-LCA: social) were performed for three selected nZVI production methods: milling, chemical reduction with sodium borohydride, and chemical reduction with hydrogen gas. These methods were selected because they involve top-down and bottom-up production technologies, and because they are used in the industrial and laboratory production of nZVI.

The LCA results show that the milling method resulted in the lowest environmental impacts, followed by the hydrogen gas reduction method, while the sodium borohydride reduction method has the greatest impacts. Environmental impacts are mainly associated with reagents used in production and energy consumption. In relation to energy consumption, it is perceived that the environmental impacts are directly related to the energy matrix of the country considered. Thus, sensitivity analysis showed that the greatest impacts were observed in the India scenario, and the lowest in the Brazil and Switzerland scenarios.

The LCC results show that the milling method has the lowest life cycle costs, followed by the sodium borohydride reduction method. The hydrogen gas reduction method has the highest LCCs. The costs are mainly associated with the acquisition of the reagents used in the production processes, and the external environmental costs resulting from the environmental impacts generated. Sensitivity analysis has shown that the results are not sensitive to changing industrial energy costs in different countries.
The results of the S-LCA show that all the nZVI production methods have significantly equal Social Index values. In a detailed analysis, considering the decimal differences, the hydrogen gas reduction method resulted in the lowest social index, followed by the sodium borohydride reduction method. In contrast, the milling method exhibited the highest social index. All methods were classified as socially sustainable according to the implemented methodology. The lowest social scores were observed in the category of human resources management and worker stakeholders.

Finally, this work is essential since nanomaterials are increasingly present in daily life (e.g., pharmaceuticals, electronics, and food) and environmental remediation processes. Thus, the results of this research were able to address a gap in the scientific knowledge on the life cycle analysis application that compares the environmental, economic, and social impacts of nanomaterial production in soil and groundwater remediation. Future research should evaluate the impacts of using nZVI on the remediation of contaminated areas.

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