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ORIGINAL ARTICLE

Comparative Life Cycle Assessment Of Edta-Modified and Amine Graphted Silica Xerogels

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ABSTRACT – NaI-131 removal from hospital wastewater using various silica xerogel adsorbents has been studied in order to meet the clearance level of radioactive waste. The contaminants emitted from the adsorbent manufacturing may affect the environment and human health. This study aimed to assess and minimize the environmental impacts of two adsorbents: EDTA-modified xerogel silica (EDTA Si-Xe) and Amine grafted silica xerogels (Amine Si-Xe), utilizing life cycle assessment (LCA) with the cradle to gate approach. OpenLCA 1.7 was used to estimate the impact, where background data were acquired from the European reference Life Cycle Database (ELCD) 3.2. The results show the dominant environmental impact resulting from adsorbent manufacturing is climate change. The impact of climate change on EDTA Si-Xe manufacturing and Amine Si-Xe manufacturing is about 0.258 CO_2-eq and 0.510 CO_2-eq, respectively. EDTA Si-Xe manufacturing performed the best (lower environmental impact) compare to Amine Si-Xe manufacturing. The process stage that contributes dominantly to environmental impact is calcination which gives high environmental impacts to climate change. The percent contribution of calcination to the environmental impact on EDTA Si-Xe manufacturing and Amine Si-Xe manufacturing is about 87% and 66%, respectively. The environmental impacts of the adsorbents manufacturing especially in climate change can be reduced by using Capture Carbon Storage (CCS) technology. Improvement analysis shows EDTA Si-Xe performed lower environmental impact compare to Amine Si-Xe with the value of impact categories lower than without applied CCS technology, especially value of climate change.

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INTRODUCTION

Nuclear medicine service activities that use open radioactive sources for diagnostic, therapeutic, and clinical medical research purposes have increased recently. In nuclear medicine the use of radiopharmaceuticals as internal radiation therapy is widely used. One of radiopharmaceutical that is widely used in nuclear medicine is NaI-131 which it is used in the therapy of thyroid cancer. The therapy of thyroid cancer is carried out by means of radiopharmaceuticals in the patient's body which will then follow the metabolic processes in the body [1]. NaI-131 is released into the environment through urine which is the result of kidney excretion [2]. Due to the radioactivity and toxicity, NaI-131 in hospital wastewater needed a technology that could be applied in handling it. A fairly effective, inexpensive, and simple method for handling it is adsorption method using adsorbent [3].

Adsorbent of silfica xerogel has been synthesized and investigated to remove of I-131 from wastewater to fulfill clearance level of radioactive waste [4]. The results showed that I-131 could be adsorbed, but the adsorption capacity is still low. To increase the adsorption capacity, it is necessary to modify the silica xerogel adsorbents. The silica xerogel adsorbents can be modified using EDTA [5] and Amine [6]. EDTA Si-Xe and Amine Si-Xe manufacturing involves a number of chemicals and process stages. The process consist of several stages that is treatment, calcination, screening, drying, washing, sol-gel, size reduction, and reflux. The involvement of chemicals and process stages in the synthesis process will actually have an impact on the surrounding environment. Thus, the LCA of the adsorbent is important for sustainable utilization to get a suitable adsorbent. Since currently unknown, the extent of those impacts have to be estimated scientifically.

According to The United Nations Conference on Environment and Development, the activity that is probably to have impacts to the environment should be assessed [7]. Conformation to that principle, this study focuses to provide LCA of adsorbents for NaI-131 removal that elucidates the adsorbent environmental impacts. LCA can determine which products and technologies give lower environmental impact in the process of comparing these products and technologies. This study assesses the environmental impacts of two adsorbents: EDTA Si-Xe and Amine Si-Xe. Hopefully, this research can provide information to support the progress of adsorbents for processing waste, especially radioactive wastewater.

In 2015, the comparative study of LCA of four adsorbent: activated alumina, alum, bone char, and wood char for fluoride removal in East Africa has been performed by [8]. The production of aerogel that used as carrier for drug delivery has been studied by [9]. In 2018, the comparative study of LCA of acid-base, acid, and thermally treated laterite for fluoride removal has been studied by [10]. Also there have been quite a lot of LCA studies on manufacturing of various adsorbents, such as manufacturing adsorbent in the form of activated carbon derived from coconut shells [11] and the

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manufacture of adsorbents in the form of activated carbon from wood waste [12]. The environmental impacts of manufacturing of EDTA Si-Xe and Amine Si-Xe adsorbents for NaI-131 removal have not been investigated to date.

The LCA was performed according to International Organization for Standardization (ISO) 14040, that consists goal definition, functional unit, and scope, inventory of life cycle, impact assessment and categories, and interpretation stages [13]. Not only to quantify and compare the environmental impacts, but hopefully this study was also to give improvement processes and technologies associated with the manufacturing EDTA Si-Xe and Amine Si-Xe to minimizing the environmental impacts. The results of this LCA study can be used as a basis for decision making for producers and consumers in selecting a product and manufacturing process that will be environmentally sustainable.

EXPERIMENTAL METHOD

This research was performed according to International Organization for Standardization (ISO) 14040 [13] that consists of goal definition, functional unit, and scope, inventory of life cycle, impact assessment and categories, and interpretation stages. This research was performed in Polytechnic Institute of Nuclear Technology, Yogyakarta, Indonesia.

Goal Definition, Functional Unit, and Scope

The goal of study is to compare the environmental impacts of two adsorbent for NaI-131 removal: EDTA-Si-Xe and Amine Si-Xe, to identify process stages associated with these adsorbents that gives lower environmental impact, and to minimize the environmental impacts caused by the manufacture of the adsorbents.

The functional unit of this LCA is about 1300 grams of EDTA Si-Xe and 1600 grams of Amine Si-Xe which interpreted as the necessary adsorbent to remove the same quantity of NaI-131 (radioactive substance) concentration (activity) about 97% (1100 MBq/day to 37 MBq/day). The functional unit in this study is based on the necessary for adsorbents for NaI-131 treatment in Indonesia using primary data generated from experimental data by [4]–[6], [14]. To meet the industrial level, the following assumptions are used to scaled up the primary data:

- The urine of therapy patients per day was 41.51 kg in one hospital
- Every 0.40 kg of urine in therapy patients contained 14.86 mL of NaI
- There are 60 hospitals in Indonesia that serve radiotherapy
- The adsorption capacity of Amine Si-Xe is 60 mg/g and EDTA Si-Xe is 74.074 mg/g

This study was performed using cradle to gate analysis through energy and mass balances of each process stage. Scope of the study is set from raw material acquisition, transportation to plant, manufacturing, and end at adsorbent attainment (Figure 1).

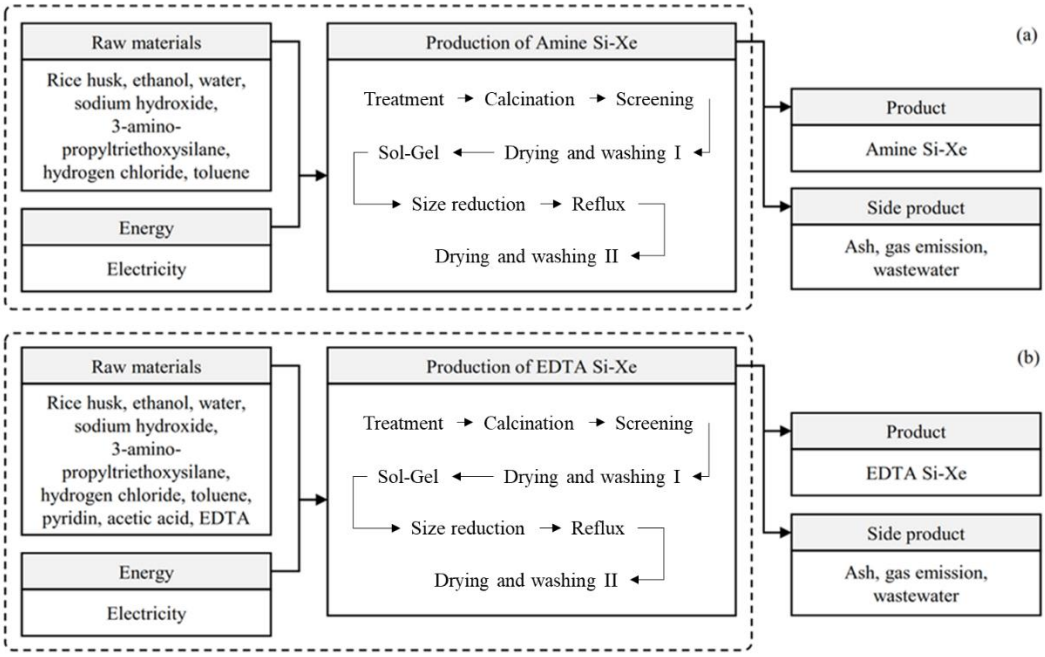

Figure 1. Scope of study for EDTA Si-Xe and Amine Si-Xe manufacturing

Life Cycle Inventory (LCI)

Set from raw material acquisition, transportation to plant, manufacturing, and end at adsorbent attainment, a LCI of inputs and outputs was acquired using a literature review [4]–[6], [14] and European reference Life Cycle Database (ELCD) 3.2 [15]. The data of system input were calculated using mass and energy balances method and tabulated in Table 1 and Table 2 for the same functional unit. Process details of adsorbent manufacturing are described in the flow process in Figure 2 and Figure 3. Each process stage that produces wastewater, COD and BOD specifications (Table 3) for wastewater are based on analytical laboratory wastewater data.

Table 1. Life cycle inventory of Amine Si-Xe manufacturing

Stage	Input	Unit
Electricity	339	kWh
Rice husk	42.6	Kg
Water	203.7	Kg
Sodium hydroxide	1.1	Kg
Hydrogen chloride	1.6	Kg
Ethanol	0.2	Kg
3-aminopropyltriethoxysilane (APTES)	0.44	Kg
Toluene	13.1	Kg

Table 2. Life cycle inventory of EDTA Si-Xe manufacturing

Stage	Input	Unit
Electricity	356.54	kWh
Rice husk	21.6	Kg
Water	127.91	Kg
Sodium hydroxide	1.23	Kg
Hydrogen chloride	1.24	Kg
Ethanol	3.48	Kg
APTES	0.202	Kg
Toluene	6.53	Kg
Pyridin	2.27	Kg
Acetic acid	3.45	Kg
EDTA	1.45	Kg

Table 3. Value of COD and BOD wastewater

Parameter	Value (mg/L)
BOD	20.16
COD	81.67

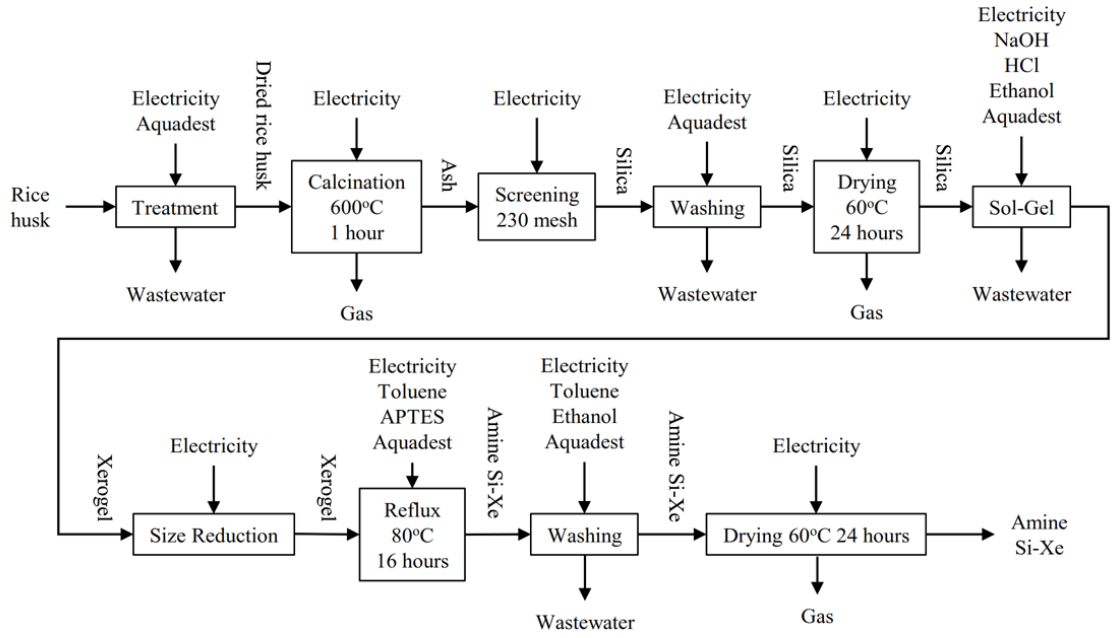

Figure 2. Flow process of Amine Si-Xe manufacturing

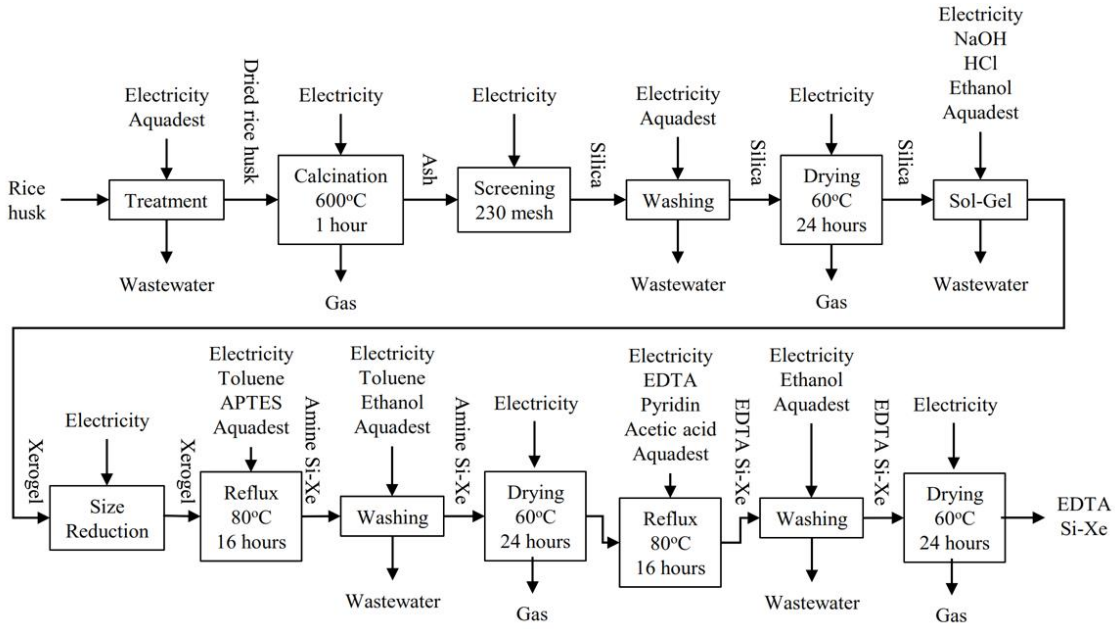

Figure 3. Flow process of EDTA Si-Xe manufacturing

The process of transporting raw materials such as rice husks and chemicals is transported using a van with a capacity of less than three tons. Selection of vans is based on availability from the ELCD database. The distance from the rice husk area (Trukan Village in Prambanan) to the storage area (Polytechnic Institute of Nuclear Technology, Yogyakarta) is about 27.2 km and the distance from chemical raw materials (Gondokusuman in Yogyakarta) to the production area is 10 km. The database on the number of emissions during the transportation of raw materials is based on the [15] (Table 4).

Table 4. Value of COD and BOD wastewater

Category for air pollutant load calculation	CO (g/km)	HC (g/km)	NOx (g/km)	PM_{10} (g/km)	CO_2 (g/kg BBM)	SO_2 (g/km)
Motorbike	14	5.9	0.29	0.24	3180	0.008
Car (gasoline)	40	4	2	0.01	3180	0.026
Car (diesel)	2.8	0.2	3.5	0.53	3172	0.44
Bus	11	1.3	11.9	1.4	3172	0.93
Truck	8.4	1.8	17.7	1.4	3172	0.82

Furnace capacities for calcination and reactor for reflux are 512 L and 60 L, respectively. In the drying process, an oven that has a capacity of up to 200 kg is used. The equipments are then calculated for their electricity needs based on their power specifications. The total electricity needs are tabulated in Table 1 and Table 2. Electricity intake is supplied from State Electricity Company.

Impact Assessment Methods and Impact Categories

Life cycle models was created by using OpenLCA 1.7. Infrastructure that are used repeatedly and common to all processes, such as manufacturing facilities, was assumed that it did not include in the assessment. To assess the environmental impact and to evaluate the contributions in each process stages, CML baseline 2000 method was used, similarly to the previous reference LCA study [16]. This method can be downloaded from the Leiden University website and is available over 1700 different flows. The impact categories studied were photochemical oxidation, acidification potential, climate change, eutrophication, and human toxicity.

RESULT AND DISCUSSION

Process Contribution and Dominant Impact Category Analyses

Acidification potential refers to the compounds that cause acidifying effects to the environment. These include anthropogenically derived sulphur (S) and nitrogen (N) such nitrogen dioxide (N_2O), nitrogen oxides (NO_x), sulfur dioxide (SO_2), nitrogen monoxide (NO), ammonia (NH_3), and other various substances. Acidification potential is usually calculated in $SO_{2\text{-eq}}$ [17]. EDTA Si-Xe and Amine Si-Xe manufacturing causes acidification potentials of 0.045 $SO_{2\text{-eq}}$ and 0.106 kg $SO_{2\text{-eq}}$, respectively (Figure 4). This shows Amine Si-Xe manufacturing produces higher SO_2 emissions than EDTA Si-Xe manufacturing. According to [18], SO_2 gas is formed as a result of the decomposition of rice husks to ash during the calcination process. Figure 4 show the calcination process is a process that has the highest contribution to the two manufactures compared to other process stages (sol-gel, drying, washing, reflux, size reduction, and transportation). However, the amount of SO_2 produced in Amine Si-Xe manufacturing is higher than in EDTA Si-Xe manufacturing. It

is because the input of rice husk in Amine Si-Xe manufacturing is more than that of EDTA Si-Xe manufacturing so that the resulting emissions are also comparable. In Amine Si-Xe manufacturing, other process stages that contribute besides calcination are transportation with 20% of 0.106 kg $SO_{2\text{-eq}}$. Transportation of raw materials contributes to producing SO_2, NO_x, and other. The SO_2 and NO_x gases emitted during manufacturing will later react with water vapor to form acid rain (sulfuric acid and nitric acid) in the atmosphere which will later fall along with rainwater. Rainwater that falls on land will cause an increase in soil acidity and surface water. Acid rain also has an impact on the soil, which can dissolve heavy metals in it. These heavy metals will later dissolve in groundwater and surface water. This will reduce the quality of water, especially for household water. One of the household water needs is consumption water. Consumption water that contains heavy metals will harm the health of humans, animals and plants [19]. In addition to causing acidification potential, the assessment results in Figure 4 also show that manufacturing Amine Si-Xe and EDTA Si-Xe are higher causing climate change.

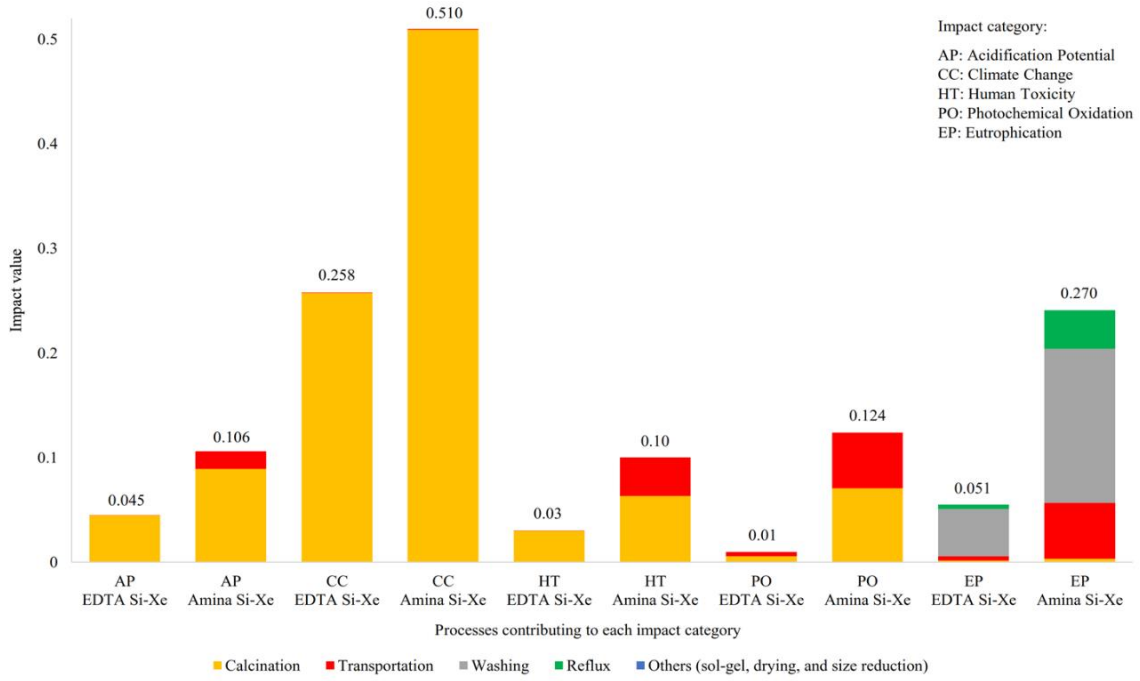

Figure 4. Processes contributing to each impact category

Climate change refers to global temperature changing. Not only increase global temperatures, but climate change also causes rising sea levels, climate disturbances, and the spread of disease [20]. Greenhouse gases such as carbon dioxide (CO_2), chlorofluorocarbons (CFCs), and methane (CH_4) contribute to climate change, especially the element carbon. Climate change is usually calculated in CO_2 equivalence ($CO_{2\text{-eq}}$) [17]. The results in Figure 4 show that EDTA Si-Xe manufactures have a lower impact on climate change than Amine Si-Xe. EDTA Si-Xe and Amine Si-Xe manufacturing causes climate change of 0.258 $CO_{2\text{-eq}}$ and 0.510 kg $CO_{2\text{-eq}}$, respectively. In both EDTA Si-Xe and Amine Si-Xe manufacturing, almost 100% (~ 99.8%) of the process stages that contribute to climate change are calcinations. Not only emitting SO_2, calcination produces exhaust gases from the decomposition of rice husks to rice husk ash, including CO_2 [18]. However, the amount of CO_2 produced in Amine Si-Xe manufacturing is higher than in EDTA Si-Xe manufacturing. It is because the input of rice husk in Amine Si-Xe manufacturing is more than that of EDTA Si-Xe manufacturing so that the resulting emissions are also comparable. Figure 4 also shows other impact categories during manufacturing that have a lower value than climate change, namely photochemical oxidation and human toxicity.

Human toxicity is mainly concerned with the effects of toxic substances on the human environment. It is expressed with the Uniform System for the Evaluation of Substances in 1,4-dichlorobenzene equivalents ($C_6H_4Cl_{2\text{-eq}}$) [21]. Meanwhile, photochemical oxidant formation (or photochemical smog) refers to a phenomenon that caused by the reaction of pollutant-forming emissions such as CO, CO_2, SO_2, NO, NO_2, NH_4, and volatile organic compounds (VOC) from combustion process with sunlight. This impact category causes eye irritation, breathing problems, crops, and damage to some materials (eg: rubber and plastic). The measure of kg $C_2H_{4\text{-eq}}$ (ethylene equivalents)/kg emission is used to calculate photochemical oxidation impacts [22]. The results in Figure 4 show that EDTA Si-Xe manufacturing provides a lower photochemical oxidation and human toxicity impact than Amine Si-Xe manufacturing. Because the input of rice husk in EDTA Si-Xe manufacturing is lower than Amine Si-Xe manufacturing (Table 1 and Table 2), pollutant-forming emissions produced from decomposition of rice husk to ash are also comparable.

In both EDTA Si-Xe manufacturing and Amine Si-Xe manufacturing, calcination contributes to the impact of human toxicity and photochemical oxidation which is more dominant than other process stages. In Amine Si-Xe manufacturing (Figure 4), calcination contributed about 63% of 0.1 kg $C_6H_4Cl_{2\text{-eq}}$ in the human toxicity impact category and about 57% of 0.124 kg $C_2H_{4\text{-eq}}$ in the photochemical oxidation impact category. In EDTA Si-Xe manufacturing (Figure 4), calcination

dominantly contributed about 99% of 0.03 kg $C_6H_4Cl_{2\text{-eq}}$ in the human toxicity impact category and about 58% of 0.01 kg $C_2H_{4\text{-eq}}$ in the photochemical oxidation impact category. The high contribution of calcination to photochemical oxidation is because the calcination process produces more pollutant-forming emissions than other process stages such as drying, washing, and others. These pollutant-forming emissions react with sunlight to form photochemical smog that cause photochemical oxidation potential.

In Amine Si-Xe manufacturing, transportation contributes 37% to human toxicity and 43% to photochemical oxidation. Besides calcination, transportation also produces pollutant-forming emissions from the combustion of fuel. Emission specification data for van (cars using gasoline) are tabulated in Table 4. These pollutant-forming emissions react with sunlight to form photochemical smog that cause photochemical oxidation potential. Another impact category that has a higher impact value than photochemical oxidation and human toxicity but has a lower impact on climate change is eutrophication.

Eutrophication is defined as an accumulation of chemical nutrient concentrations in an ecosystem that leads to abnormal productivity [23]. It can cause aquatic plants death and makes water turbidity increase. Eutrophication is calculated in phosphate equivalence ($PO_{4\text{-eq}}$) [17]. Phosphorus and nitrogen are the main control factors for algae propagation that caused water eutrophication [24]. Hence, the amount of nitrogen also contributes to the eutrophication value.

Nitrogen and phosphorus contained in rice husks will decompose during calcination, for example some nitrogen becomes pollutant gas (NO, N_2O, and NO_x), but nitrogen and phosphorus in certain amounts also still stick to the ash. The ash is further processed at the washing stage using water so that impurities such as nitrogen and phosphorus will be involved in the washing wastewater and produce silica products (Figure 2 and Figure 3). Because the rice husk used in EDTA Si-Xe manufacturing is lower than in Amine Si-Xe manufacturing, the resulting amount nitrogen and phosphorus is also lower. This is in line with the results in Figure 4 that show Amine Si-Xe manufacturing and EDTA Si-Xe manufacturing have an eutrophication effect of 0.270 kg $PO_{4\text{-eq}}$ and 0.051 kg $PO_{4\text{-eq}}$, respectively which EDTA Si-Xe manufacturing provides about lower eutrophication impact than Amine Si-Xe manufacturing.

In contrast to other impact categories which are predominantly due to calcination, the impact of eutrophication during adsorbent manufacturing is predominantly contributed by washing stage. The contribution of washing to eutrophication in Amine Si-Xe and EDTA Si-Xe manufacturing was 55% of 270 kg $PO_{4\text{-eq}}$ (about 0.147 kg $PO_{4\text{-eq}}$) and 89% of 0.051 kg $PO_{4\text{-eq}}$ (about 0.045 kg $PO_{4\text{-eq}}$), respectively. This shows that the contribution of washing in EDTA Si-Xe manufacturing is lower than in Amine Si-Xe manufacturing. It is because the amount of water used for washing in EDTA Si-Xe manufacturing is less than in Amine Si-Xe manufacturing. Apart from washing, transportation and reflux also contribute to eutrophication. Transportation produces NO_x pollutant emissions. NO_x contributes to the magnitude of the impact of eutrophication.

Interpretation and Improvement Analysis

Based on the impact category in the CML Baseline 2000 method, overall EDTA Si-Xe manufacturing contributes to a lower environmental impact than Amine Si-Xe manufacturing (Figure 4). Flow process of EDTA Si-Xe manufacturing (Figure 3) is longer than the Amine Si-Xe manufacturing (Figure 2). In EDTA Si-Xe manufacturing, the reflux, washing, and drying are performed thrice while in Amine Si-Xe manufacturing it is performed twice. Although flow process of EDTA Si-Xe manufacturing is longer, the resulting environmental impact is lower than the Amine Si-Xe manufacturing. It is because the amount of raw material (rice husk) fed in EDTA Si-Xe manufacturing is less than in Amine Si-XE manufacturing so that the resulting gas emissions are also lower, especially at the calcination.

Amine Si-Xe manufacturing and EDTA Si-Xe manufacturing contributed to a more dominant impact on climate change (kg $CO_{2\text{-eq}}$) (Figure 5). The impact of climate change on EDTA Si-Xe manufacturing is about 65% while Amine Si-Xe manufacturing is about 46%. The process stage which contributed more dominantly for both manufactures was

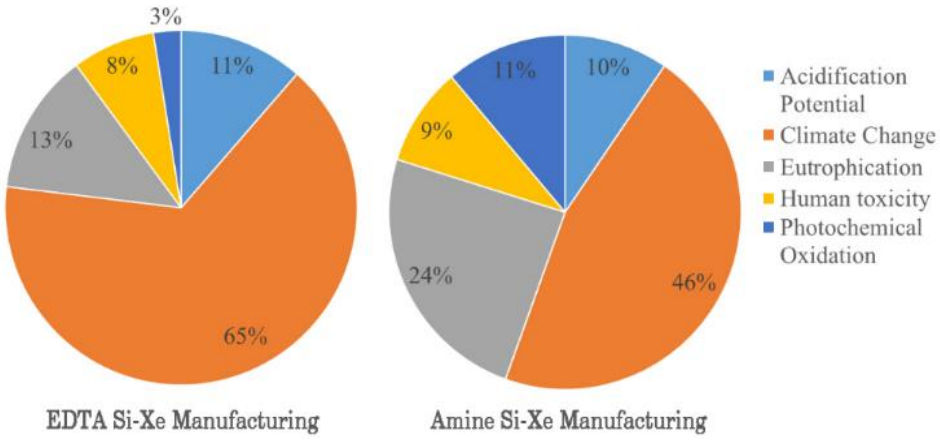

Figure 5. Comparative results of environmental impacts of EDTA Si-Xe and Amine Si-Xe manufacturing

calcination (Figure 6). About 87% of the environmental impact caused by EDTA Si-Xe manufacturing is due to the calcination process stage while Amine Si-Xe manufacturing is around 66%. The decomposition of rice husks to ash during the calcination process produces emissions of gases such as SO_2, NO_2, CO_2, N_2O, CH_4, CO, and $PM_{2.5}$, in particular dominated by CO_2 gas about 95% [18]. To reduce the respective impacts (especially on climate change) and the sustainable production of Si-Xe adsorbent, an analysis of improvements is performed. Improvement analysis is performed by reviewing the process and providing recommendations. In this study, we introduce to use Capture Carbon Storage (CCS) technology to treat the emissions gases, especially CO_2.

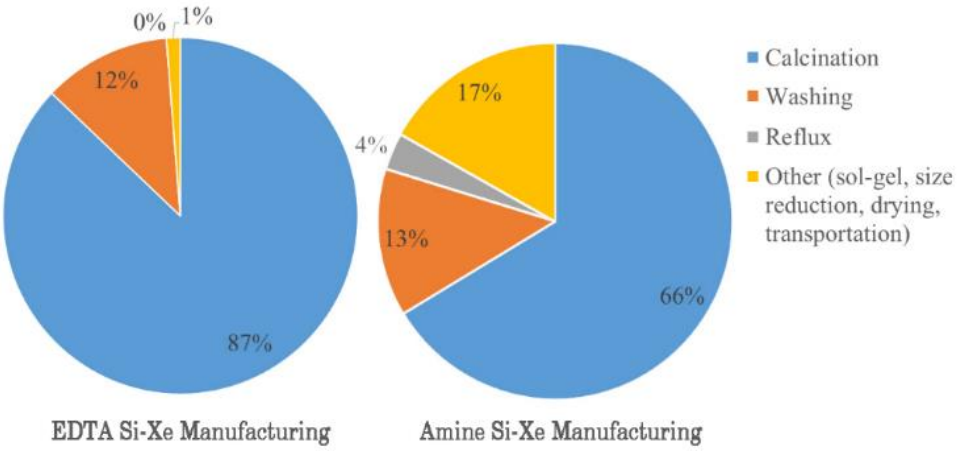

Figure 6. Comparative results of processes contributing of EDTA Si-Xe and Amine Si-Xe manufacturing

CCS prevents greenhouse gas emissions from fleeing into the atmosphere. This technology is usually applied to large-scale industries [25]. For both EDTA Si-Xe and Amine Si-Xe manufacturers, the results of reassessment by applying CCS in manufacturing show that there has been a decrease in the value of the impact in each impact category, especially in climate change (Figure 7 and 8). Climate change is the impact category with the highest percentage reduction after interpretation and analysis of improvements using CCS. The impact reduction of climate change in Amine Si-Xe manufacturing and EDTA Si-Xe manufacturing is about 90% each. However, other impact categories also decreased but not significantly.

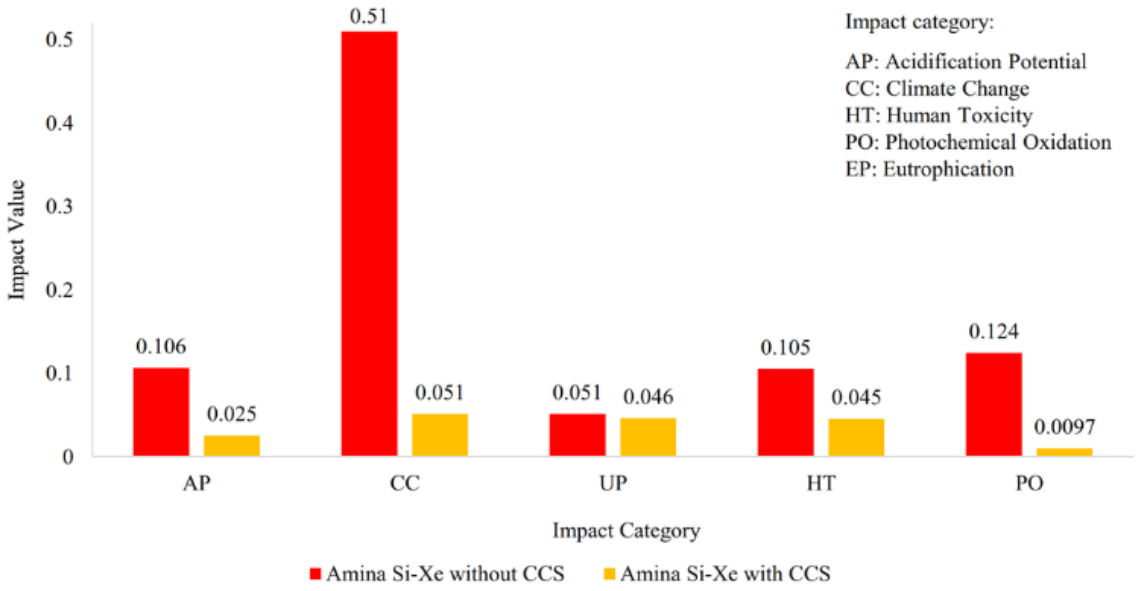

Figure 7. Comparative results of environmental impacts of Amine Si-Xe before and after improvement

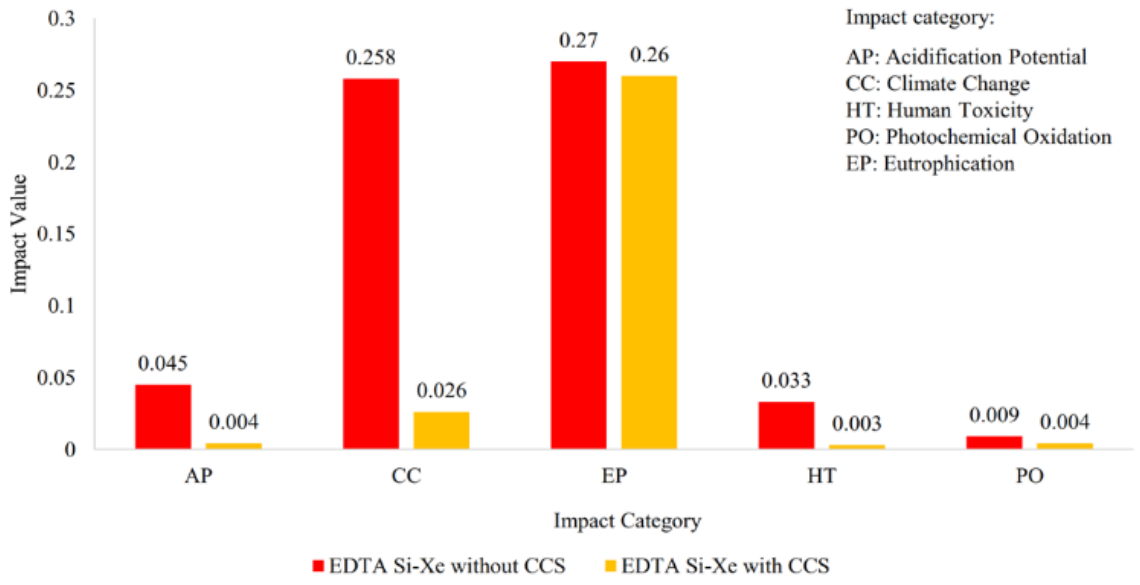

Figure 8. Comparative results of environmental impacts of EDTA Si-Xe before and after improvement

CONCLUSION

Climate change is the dominant environmental impacts resulting from EDTA Si-Xe manufacturing and Amine Si-Xe manufacturing which is about 0.258 $CO_{2\text{-eq}}$ and 0.510 $CO_{2\text{-eq}}$, respectively. EDTA Si-Xe manufacturing performed lower environmental impact compare to Amine Si-Xe manufacturing. The stages that contributes dominantly to environmental impact is calcination. Percent contribution of calcination to environmental impact on EDTA Si-Xe manufacturing and Amine Si-Xe manufacturing is about 87% and 66%, respectively. The environmental impacts of the adsorbents manufacturing especially in climate change can be reduced by using Capture Carbon Storage (CCS) technology. Improvement analysis shows EDTA Si-Xe performed lower environmental impact compare to Amine Si-Xe with value of impact categories lower than without applied CCS technology, especially value of climate change.

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