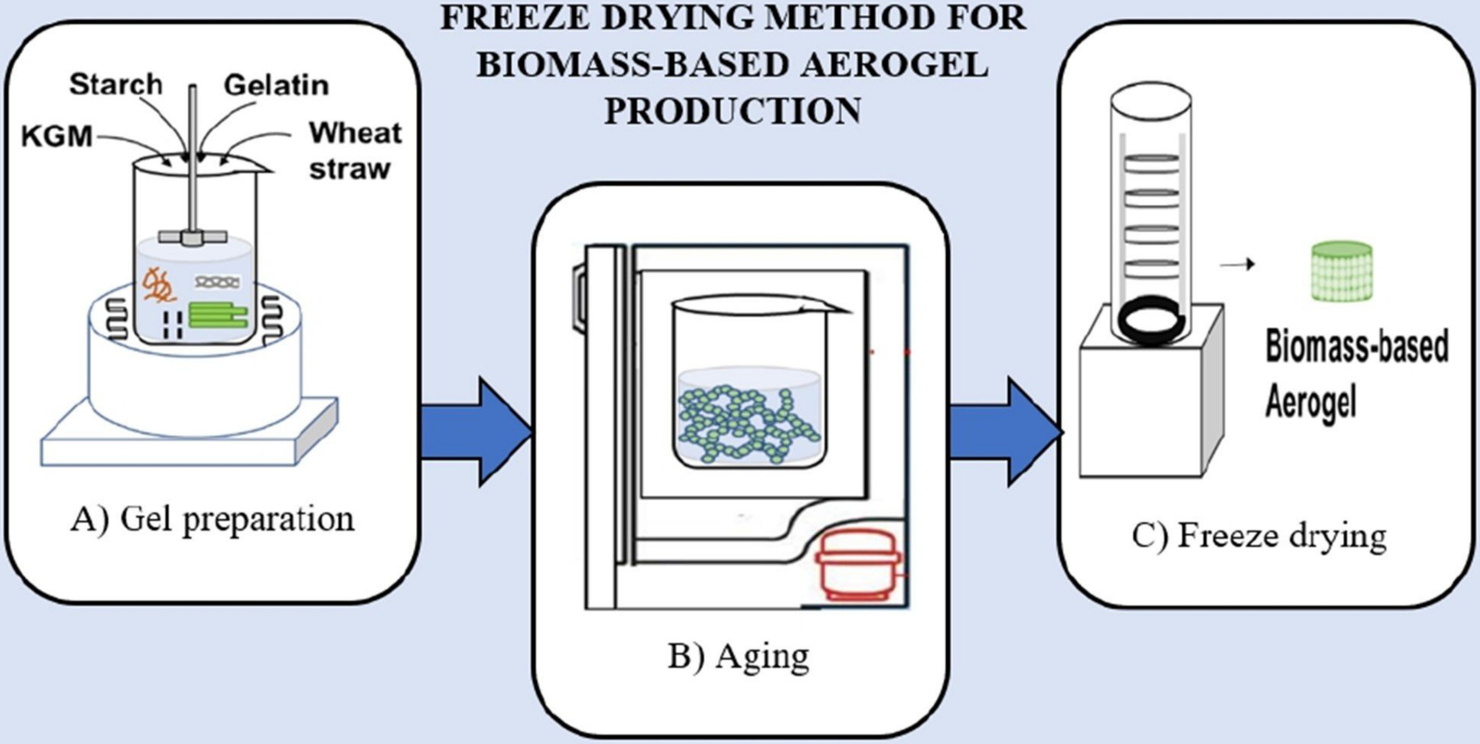
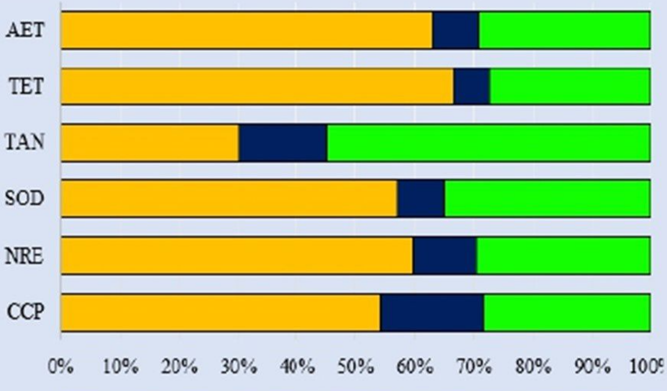
Graphical Abstract

FREEZE DRYING METHOD FOR BIOMASS-BASED AEROGEL PRODUCTION



ENVIRONMENTAL IMPACTS OF BIOMASS-BASED AEROGELS

Functional Unit: 1 m^3 of biomass-based aerogel



	CCP	NRE	SOD	TAN	TET	AET
🟧 Freeze Drying	3.67E+02	9.85E+03	2.40E-04	2.59E+00	1.38E+03	6.21E+03
🟦 Aging	1.18E+02	1.73E+03	3.38E-05	1.29E+00	1.29E+02	7.89E+02
🟩 Gel Preparation	1.91E+02	4.89E+03	1.47E-04	4.69E+00	5.65E+02	2.87E+03

🟧 Freeze Drying 🟦 Aging 🟩 Gel Preparation

6.76E+02 kg CO$_2$ eq. → Climate change potential

1.65E+01 MJ primary → Non-renewable energy potential

4.21E-04 kg CFC-11 eq. → Stratospheric ozone depletion

8.57E+00 kg SO$_2$ eq. → Terrestrial acidification potential

2.07E+03 kg TEG soil → Terrestrial ecotoxicity

9.87E+03 kg TEG water → Aquatic ecotoxicity

Life cycle assessment of a novel biomass-based aerogel material for building insulation

***Yixin Wang* [1], *Rizwan Rasheed* [2] *, *Fatang Jiang* [1, 3], *Asfra Rizwan* [2], *Hajra Javed* [2], *Yuehong Su* [1], *Saffa Riffat* [1]**

[1] Department of Architecture and Built Environment, University of Nottingham, NG7 4RD, UK
[2]Sustainable Development Study Centre, Government College University Lahore, 54000, Pakistan
[3]School of Bioengineering and Food Science, Hubei University of Technology, Wuhan, 430068, China

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Correspondence

*Rizwan Rasheed; email: *rizwanrasheed@gcu.edu.pk and riz_mian@hotmail.com*

Sustainable Development Study Centre, Government College University, Katchary Road, Lower Mall, Lahore,54000, Pakistan

ORCID: https://orcid.org/0000-0002-5441-7376

Abstract

There is a growing interest in accounting for the environmental externalities and the greenhouse gas (GHG) emissions associated with the building industry. This study examines the life cycle environmental impacts of a novel biomass-based aerogel building material manufactured via freeze drying method comprising of three process stages, *i.e.,* gel preparation, aging and freeze drying. The main focus of this study is to evaluate the contribution of each stage to the environmental load using life cycle assessment tool, figure out the main stage that has the greatest impact on the environment and propose some potential improvements by critical analysis of the production process. Life cycle impact scores are quantified as per functional unit of 1 m^3 biomass-based aerogel for six midpoint impact categories (climate change potential, non-renewable energy potential, stratospheric ozone depletion, terrestrial acidification potential, terrestrial ecotoxicity and aquatic ecotoxicity). The respective LCA scores for these categories are depicted as 6.76E+02 kg CO_2 eq., 1.65E+04 MJ, 4.21E-04 kg CFC-11 eq., 8.57E+00 kg SO_2 eq., 2.07E+03 kg TEG soil and 9.87E+03 kg TEG. While comparing individual process substages, the freeze drying stage of the manufacturing process presents the highest overall impact contribution. Comparative environmental scoring with other aerogel types further reveals that the biomass-based aerogels are environmentally promising alternatives. Since the production is done at a laboratory scale, these results can be regarded as a conservative estimate, however they can act as steppingstones for process optimization for commercial scale manufacturing.

Keywords: Eco-footprints, LCA, sustainable production, eco-friendly insulation, freeze drying

1. Introduction

In recent years, environmental concerns such as global warming, acid precipitation, ozone depletion and the destruction of ecological diversity are being noticed by the world. Therefore, protection of the environment has become a major global concern. With the industrial and other sectors paying greater attention to environmental protection and management, the eco-environment coordination of new materials has laid a foundation for controlling pollution at the source. After the invention of inorganic aerogels, many different raw material type have been employed to prepare aerogels. Owing to their excellent properties, biomass-based aerogels represent a promising class of novel materials which have gained wide interest of researchers. Globally environment-friendly materials are being promoted not only in terms of their chemical and physical properties but also in terms of their environmental efficiency indicators which has proven to be another important feature. Different combinations of raw materials, energy sources and manufacturing techniques have been widely researched for developing aerogels with minimum negative environmental externalities. For assessing the environmental viability and performance efficiency of these novel developed materials, various methodologies and modelling tools are being used [1]. One effective tool is the life cycle assessment (LCA) method, extensively used for the evaluation of the environmental impact of materials during their life cycle providing valuable insights into improvement of materials and their processing technology thereby, promoting the harmony of the material with its environment [2] [3].

1.1. Narrative of aerogels

Aerogel is a synthetic three-dimensional porous material produced by specific drying methods to replace the liquid part in gel with air [4]. Aerogels have a unique structure possessing low density, high porosity and large interior surface area, which contributes to specific functional properties. Silicon aerogels were first studied by Kistler in 1931, and their special functional properties gained great attention in academia [5]. Since their inception, different types of aerogels have been researched, developed and applied in various fields [6] [7]. Among these fields, thermal insulation is one of the most promising high-performance application fields because aerogels can avoid excessive heat dissipation with extremely low thermal conductivity [8] [9]. Aerogels have been widely applied in commercial buildings, with specialized applications in cavity insulation, glazing units [10] [11] and cladding systems owing to constant development in production process and their economic viability [12]. European Union (EU) presented the directive of optimizing and improving the design of construction products to minimize their environmental impacts [13]. Recently biomass-based aerogels have also been widely studied as they possess excellent thermal insulation properties. Using alternative, biomass-based construction materials is one of the most prominent trends nowadays due to its advantage in achieving sustainability [13]. Rudaz et al. (2014) prepared biomass-based aerogels with pectin through sol-gel and supercritical CO_2 drying method, which have high porosity (90%), low density (0.05-0.2 g/cm^3) and low thermal conductivity (0.016 ~ 0.020 W/m·K). It is essential to drive the development of aerogels while addressing the energy performance of the production process and environmental protection [14].

The literature on aerogels in the 20th century is primarily focused on silica aerogels [7] and metal oxide aerogels [15]. Recently, plant biomass is being used as raw material to produce environment-friendly and sustainable aerogels [16]. They are ideal raw materials for the preparation of aerogels in modern industrial applications [17]. At present, different biomass sources have been reported for aerogels' production such as cellulose [18] [19] [20], marine polysaccharides [21], starch [22] [23] [24], pectin [14], gelatin, whey [25] and casein [26].

There are two typical drying methods for preparing aerogels, supercritical drying method and freeze-drying method. In supercritical drying method, alcogel is produced by immersing in ethanol. After this step, the supercritical extraction of ethanol assisted by supercritical fluids (such as CO_2, CH_4) is carried out and then aerogels can be collected. Aerogels obtained by freeze drying method, undergo two important process stages including gel freezing and sublimation at ultra-low pressure [17]. A majority of studies have been reported using supercritical drying method for the preparation of aerogels [27]. However, this method has certain disadvantages such as high cost and significant CO_2 burden. On the other hand, freeze drying method is a novel technique that can be applied to the aerogel production. This method is comparatively more cost-effective, up scalable and requires relatively less continuous electricity supply. A major advantage of the freeze-drying method is that this process only uses a slight amount of energy (0.85 kW) and thus has lesser CO_2 burden [28]. Therefore, freeze drying is regarded as comparatively simple, economically and environmentally feasible, and can be efficiently replicated at industrial scale. This technology is now gaining wide attention and is being employed at various scales to prepare functional aerogels [29].

1.2 LCA progress of aerogels

The environmental impacts associated with the production and use of different types of aerogels have been studied in recent years, however the LCA study of biomass-based aerogels have rarely been done even the biomass-based aerogels have recently become increasingly popular. At present, LCA has been used to assess the environmental impacts of aerogel production focused on chemical materials, such as silica aerogel-based panel. In a study conducted by Dowson et al. [12], the energy consumption for production and CO_2 burden of aerogel with high and low temperature supercritical drying were investigated [12]. In another environmental assessment study of aerogel-based panel linked to the energy efficiency was researched, applied in 5 European climate zones to evaluate the regional and weather influence on the performance [30]. Global warming potential, Non-renewable primary energy use, Ozone depletion potential, Acidification potential have been given.

Moreover, the manufacturing techniques of biomass-based aerogels with freeze drying also need to be studied to devise improvements from environmental and economical points of view. In 2016, De Marco et al. [31] reported a lifecycle assessment of starch aerogels using supercritical drying methods in lab scale [31]. In 2018, further research was carried out which indicated environmental impacts made by different production scale plants: lab and pilot plant. However, similar studies about biomass-based aerogel with freeze drying methods have not been performed.

It has been established in view of previously conducted research work, that aerogels derived from inorganic sources such as silica aerogels and metallic aerogels negatively impact the environment in diverse ways. Moreover, the manufacturing techniques of biomass-based aerogels with freeze drying also need to be studied to devise improvements from environmental and economical points of view. For the environmental impact analysis, life cycle assessment (LCA) approach is regarded as an efficient tool which enables characterization and quantification of impact values [32][33]. This helps in evaluating the environmental footprint of any product to develop and select the most viable option in terms of long-term environmental efficiency. In the construction industry, the LCA methodology has been adopted to analyse the environmental impact of construction materials [34].

The current study is aimed at the analysis of life cycle environmental impacts of manufacturing a novel biomass-based aerogel insulation material derived from konjac glucomannan, wheat straw, starch and gelatin, manufactured by freeze drying technique. A measurable environmental burden in terms of climate change potential, non-renewable energy potential, stratospheric ozone depletion, terrestrial acidification potential, terrestrial ecotoxicity and aquatic ecotoxicity has been calculated and assessed using life cycle assessment (LCA) tool. The objective is to evaluate the environmental sustainability of the novel biomass aerogel based on the type of raw materials utilized and manufacturing process employed to highlight its positive environmental externalities.

2. Methodology

2.1. Streamlined life cycle assessment (LCA)

Life cycle assessment (LCA) is regarded as an efficient tool for the evaluation of the environmental footprint of any product for its entire existence. This study uses the LCA approach to identify and characterize the environmental impacts associated with biomass-based aerogel production. The software package utilized for the calculation of impacts is SimaPro (version 8.1) and the methodology is applied according to the guidelines encapsulated in the ISO 14044 standard.

For the impact analysis, background LCI data has been retrieved from the "Ecoinvent (Version-3)" database and literature focusing on the LCA of similar aerogels. Logical assumptions have been drawn where needed based on the regional conditions due to lack of process optimization and pilot scale conditions. The background unit process inventory has been presented in Table S1.

2.1.1 Goal and scope definition

The goal and scope of the current study are to evaluate the environmental implications of biomass-based aerogels by highlighting and quantifying the various ecological impacts associated with their manufacturing.

2.1.2 System boundary and functional unit

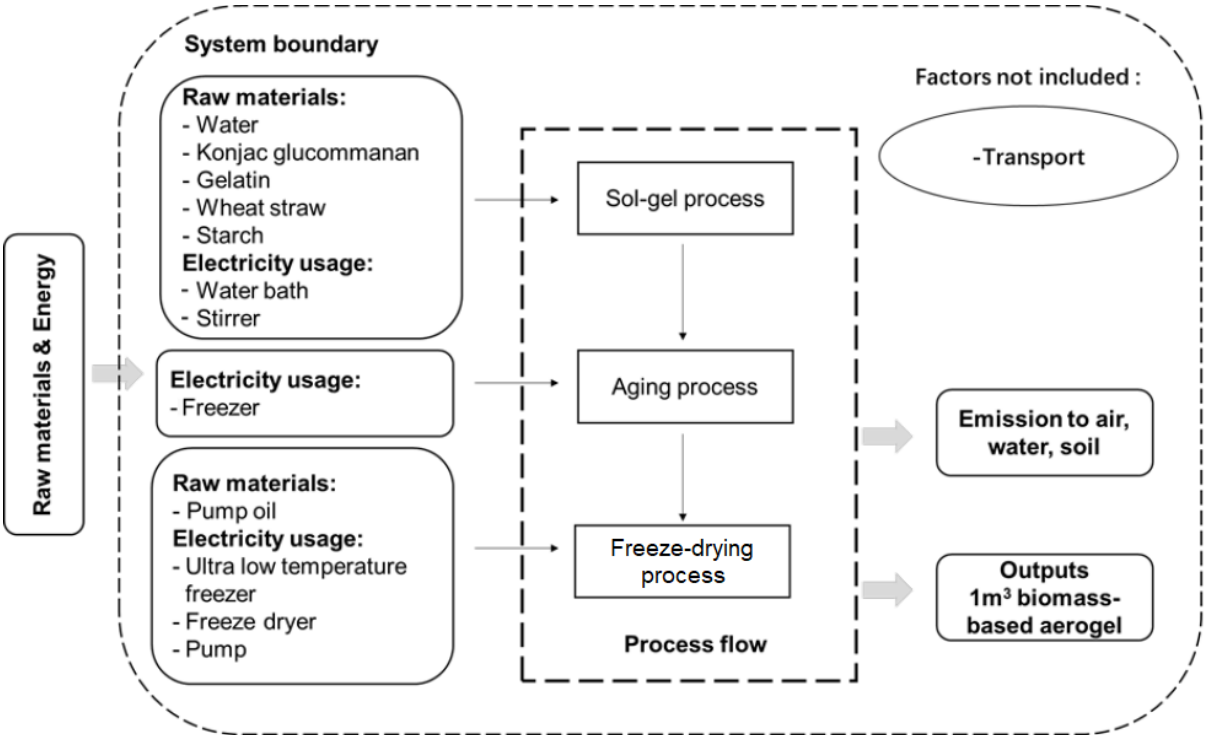


Figure 1. LCA system boundary of biomass-based aerogel production process (gate to gate)

2.2. Data collection

Defining a relevant system boundary and a representative functional unit that can be replicated at various scales globally is crucial to an LCA study. The system boundary drawn for this study focuses on the manufacturing stage of biomass-based aerogel i.e., ‘gate to gate’ consisting of three major substages: gel preparation, aging and freeze drying. As presented in the Figure 1, the inputs (raw materials and electricity usage) and outputs (biomass-based aerogel and emissions) have been listed. The transportation process has not been included currently. The functional unit (FU) selected for the environmental footprint analysis of novel biomass aerogel is 1 m^3 of biomass-based aerogel. The environmental impacts scores of the processes involved in the manufacturing of 1 m^3 of biomass-based aerogels have been calculated and the results are discussed in the subsequent sections.

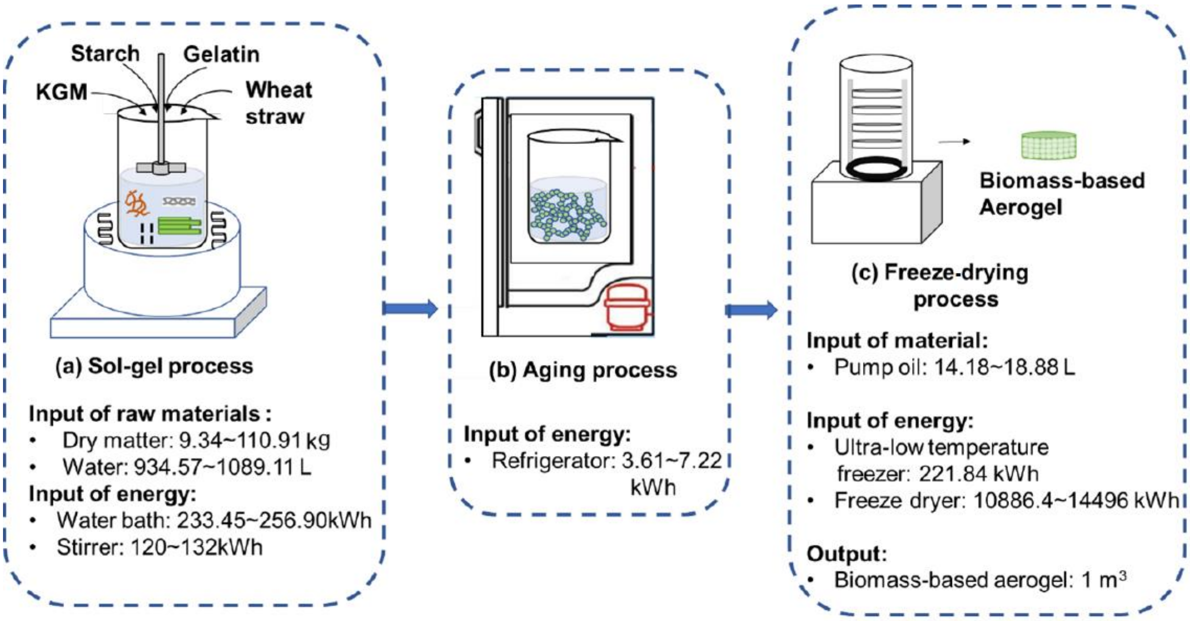


Figure 2. Process flow chart of biomass-based aerogels production by freeze-drying method in accordance with UK standards

The biomass-based aerogels’ manufacturing process consists of three basic stages *i*.e., gel preparation, aging and freeze drying. The respective material and energy inputs of these stages have been inventoried in this section while the summarized production flowchart is presented in Figure 2. In the lab-scale production process, there are negligible amount of waste of the raw materials and solution due to controlled conditions. In addition, when calculating the running times of the instrument the figure has been rounded off to the nearest value, in case of decimal values. The electricity consumption by the refrigerator and ultra-low temperature freezer is estimated based on their annual energy consumption.

2.2.1. Gel preparation

As the first stage of biomass-based aerogels production, gel is prepared by following some basic steps. Initially, raw materials are dispersed in the solvent, and gel is formed after the sol-gel process. For preparing 1 m^3 of biomass-based aerogel, 934.57~1089.11 L of water and 9.34~110.91 kg of raw materials including konjac glucomannan, starch, gelatine and wheat straw are required. Raw ingredients were continuously added in the water during the mechanical stirring at 600 rpm for 1 hour to obtain hydrogel. The sol was then injected into a cylindrical mould. The mass of all raw ingredients was measured using digital scales and the total energy use during this process was calculated. Table 1 presents the respective life cycle data inventory for the raw materials in biomass-based aerogel preparation. In the gel preparation stage, the energy has been calculated. The laboratory water bath can stir 1.5 litre of glue in one round. Therefore, it is necessary to run the water bath 666.67~733.33 times and the stirrer needs to be operated 2000.00~2200.00 times together. This will result in overall energy consumption of 233.45~256.90 and 120~132 kWh, by water bath and stirrer respectively.

It should be noted that the water baths, refrigerators, etc. are not working continuously on full power, but working intermittently. For the water bath, the total operation time and rated power consumption of one cycle are 1.0 hours and 1.4 kW, which is distinctly higher than actual. To calculate the authentic energy consumption, the actual working situation will be analysed. Based on the experiment, the water in the water bath is heated from room temperature to 90 °C in 15 minutes with the power draw 1.4 kW. When the water bath is in a stable operation state, the energy consumption is low and could be ignored. With the power draw of 1.4 kW, the actual energy consumption of one cycle is 1.4 kW × 0.25 h.

2.2.2. Aging and freezing

After the gel preparation, samples with mold were placed in refrigerator aging at 4 °C for 0.5~1 hour. The refrigerator has a capacity to hold 22 molds at a time, so the freezer needs to complete 515 freezing cycles to produce 1 m^3 biomass-based aerogels. According to the technical data of the refrigerator and ultra-low temperature freezer, the electricity power of two freezers is 0.014 kW (122 kWh / annual) and 0.094 kW (2.256 kWh / 24h), respectively. Energy consumption for one cycle of the refrigerator and ultra-low temperature freezer are 0.014 kW × 0.5 to 1 h (0.007 ~ 0.014 kWh) and 0.094 kW × 10 h (0.94 kWh), respectively.

2.2.3. Freeze drying

This final step is crucial to the manufacturing of aerogels. The freeze-drying system comprises of two major parts, freeze dryer and vacuum pump (as shown in Figure 3). The temperature and pressure are controlled by cold trap and vacuum pump. Under high vacuum, the ice in cryogenically frozen samples changes from solid-state to a gaseous state through sublimation process. To prepare 1 m^3 of biomass-based aerogels, 14.18~18.88 liters of vacuum pump oil is required.

Figure 3. Image of freeze dryer and vacuum pump

In the beginning, the chamber temperature of the freeze dryer needs to be cooled down, which usually requires approximately 0.5 hours. After this step, the samples in the plastic mold were dried in a freeze dryer at temperature of -60 °C and under the pressure of 20 Pa for 24 hours. It was assessed that 1324.65~1766.2 cm^3 of aerogels could be produced during one-time operation of freeze dryer, thus producing 1 m^3 of biomass-based aerogels, freeze dryer needs to run 567~755 times. During the freeze-drying process, the energy use of each component has been calculated and presented in Table 1.

The manufacturing of novel biomass aerogels has been conducted on trial basis i.e., at a small lab-scale. As there is a limited knowledge regarding the optimized process conditions and most suitable combination of raw materials' along with other parameters, so a precise value has not been quoted and instead a narrow range of values is presented in the LCI. Therefore, to calculate the environmental impact, average LCI values have been assumed for this production process. The unit process LCI data used for the modelling of environmental impacts of the novel biomass-based aerogel is presented in supplementary material (Table S1).

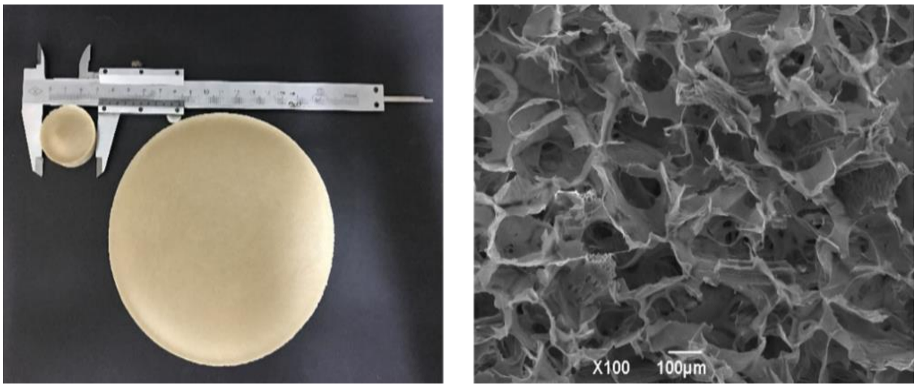
2.2.4. The aerogel

The macrograph of the prepared biomass-based aerogels is displayed in Figure 4 (a). The figure shows a greenish-brown structure with a flat, smooth surface. To better demonstrate the internal porous structure of biomass-based aerogels, the micrograph is presented in the form of Scanning Electron Microscope (SEM) image (Figure 4(b)), which were tested by a JEOL LV6060 model scanning electron microscope (Tokyo, Japan) as shown in Figure 5. As presented in previously published work [22][35], the thermal conductivity of the biomass-based aerogel reaches 0.046 ~ 0.052 W/m·K, which was measured by a thermal conductivity analyzer (Hot Disk TPS 2500, Uppsala, Sweden). The density of the biomass-based aerogel is 0.020 ~ 0.052 g/cm^3. The corresponding required insulation thickness is 46 ~ 52 mm for 1 m^{2}K/W thermal resistance as shown in Table 2.

Table 1. Life cycle inventory for the raw materials and the energy usage during freeze-drying in biomass-based aerogel preparation (1 m^3)

Raw materials	Volume (L)	Mass (kg)	Material supplier
Water	934.57~1089.11	-	-
Konjac glucomannan	-	9.34~10.89	Licheng Biological Technology Co., Ltd.
Gelatin	-	0~42.3	Sinopharm Chemical Reagent Co., Ltd.
Wheat straw	-	0~15.42	Farm
Starch	-	0~42.3	Wuhan Lin He Ji Food Co., Ltd
Pump oil	14.18~18.88	-	Edwards

Electrical equipment	Time of one cycle (h)	Electricity consumption (kWh)	Equipment supplier
Water bath	0.25	233.45~256.90	Grant Instrument
Stirrer	1.0	120~132	Scilogex
Refrigerator	0.5~1	3.61~7.22	Elecrtrolux
Ultra-low temperature freezer	10	221.84	Fisher
Freeze dryer	24	10886.4~14496	Boyikang



a) **b)**

Figure 4. Macrograph (a) and SEM image (b) of biomass-based aerogel

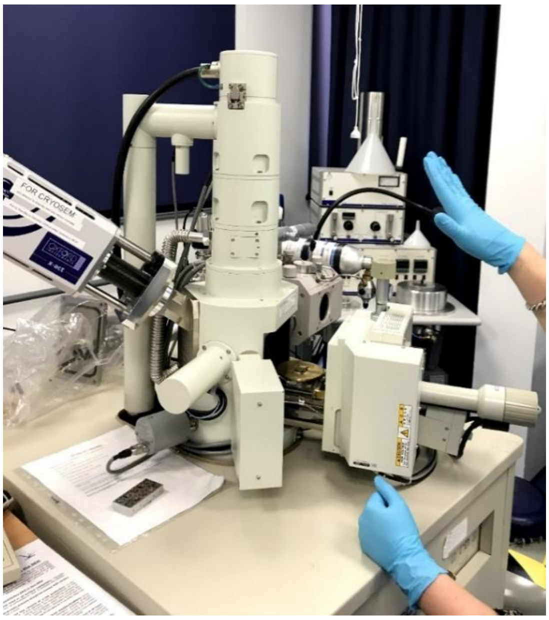


Figure 5. Image of scanning electron microscope

Table 2. The characteristic of biomass-based aerogel and the thickness required for 1 m^2K/W thermal resistance

Biomass-based aerogel properties	Unit	Value
Thermal conductivity (λ)	W/m·K	0.046 ~ 0.052
Corresponding insulation thickness	mm	46~52

3. Results and Discussions

3.1. Life cycle impact assessment (LCIA)

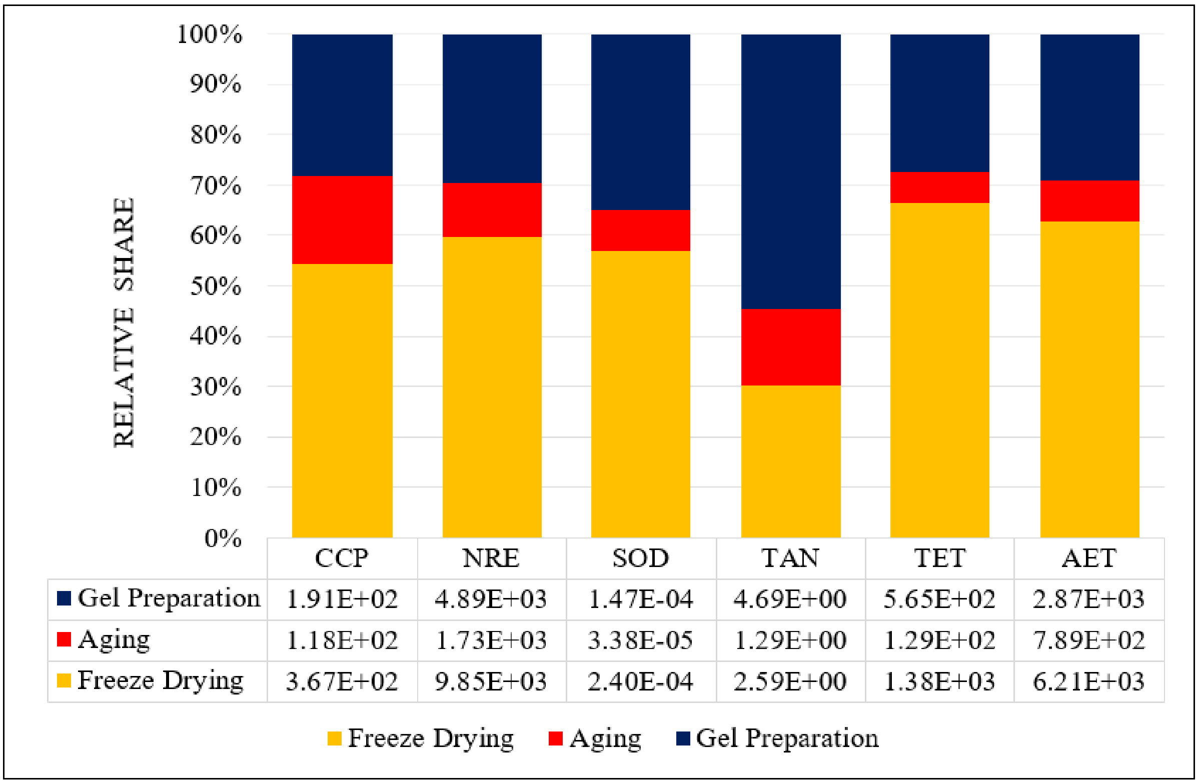
In this study the classification and characterization of the environmental impacts are carried out by deploying the

IMPACT 2002+ midpoint method, owing to its simplicity and wide recognition. It has the ability to translate complex life cycle inventory data into variable impact values without compromising on the accuracy and comprehensiveness of the study. Impact evaluation at the midpoint level provides opportunities to better comprehend the effects of a particular raw material or any specific air emission on different environmentally important issues such as climate change and resource depletion. Moreover, the analysis can be conducted at a scale that ensures higher degrees of certainty, accuracy and wide global acceptability.

For this study, six impact categories have been selected: (a) climate change potential; (b) non-renewable energy potential; (c) stratospheric ozone depletion; (d) terrestrial acidification potential;(e) terrestrial ecotoxicity and (f) aquatic ecotoxicity. They have been chosen by accounting various factors predominately the ease of quantification and their potential relevance to scientific researchers and concerned product stakeholders. Table 3 presents the overall impact values obtained for each of the selected impact categories. Figure 6 shows the relative contribution (in percentage) of these three substages to all the six impact categories. While the impact values in each category for the three processes involved in the manufacturing of biomass-based aerogels are depicted graphically in Figure 7-12.

Table 3. Impact values obtained for selected impact categories as per the selected functional unit (1 m^3).

				Impact share of different raw materials and energy			
	Impact category	Unit	Impact value	Electricity	Pump oil	Wheat straw and starch	Other raw materials (Konjac glucomannan, gelatin, etc.)
a)	Climate change potential	kg CO_2eq.	6.76E+02	3.54E+02	1.18E+02	1.05E+02	9.98E+01
b)	Non-renewable energy potential	MJ primary	1.65E+04	1.06E+04	2.11E+03	2.03E+03	1.72E+03
c)	Stratospheric ozone depletion	kgCFC-11eq.	4.21E-04	2.43E-04	7.14E-05	5.22E-05	5.47E-05
d)	Terrestrial acidification potential	kgSO_2eq.	8.57E+00	3.91E+00	2.07E+00	7.03E-01	1.89E+00
e)	Terrestrial ecotoxicity	kgTEG soil	2.07E+03	8.90E+02	5.18E+02	3.73E+02	2.90E+02
f)	Aquatic ecotoxicity	kgTEG water	9.87E+03	3.86E+03	2.77E+03	1.25E+03	1.99E+03



	CCP	NRE	SOD	TAN	TET	AET
■ Gel Preparation	1.91E+02	4.89E+03	1.47E-04	4.69E+00	5.65E+02	2.87E+03
■ Aging	1.18E+02	1.73E+03	3.38E-05	1.29E+00	1.29E+02	7.89E+02
■ Freeze Drying	3.67E+02	9.85E+03	2.40E-04	2.59E+00	1.38E+03	6.21E+03

Figure 6. Relative contributions of the main stages in the biomass-based aerogel production in the selected midpoint categories (CCP: Climate change potential, NRE: Non-renewable energy potential, SOD: Stratospheric ozone depletion, TAN: Terrestrial Acidification potential, TET: Terrestrial ecotoxicity, AET: Aquatic ecotoxicity)

3.1.1. Climate change potential

According to the results of LCIA, the manufacturing of 1 m^3 of biomass-based aerogels by freeze-drying method result in CO_2 burden of 6.76E+02 kg. The major share in this impact is of the drying stage of the manufacturing process accounting for a contribution of 3.67E+02 kg CO_2 eq. (Figures 6 and 7). The respective impact contribution of energy consumption and raw materials' processing in each process stage, depicted in Figure 7 reveals a relatively higher impact share of energy in this impact category. This can be attributed to the fossil fuels required to fulfill the energy requirements of the equipment employed during the process and the longer operation time. However, the overall impact value is less when compared to other types of aerogels owing to the use of less energy intensive freeze-drying method along with biomass-based raw materials in comparison to non-renewable inputs. In a study conducted by Dowson et al. [12], tetramethoxysilane aerogels were prepared using low and high temperature supercritical drying methods. The LCA results revealed that the manufacturing of 1 m^3 of tetramethoxysilane aerogels accounted for a CO_2 burden of 2.84E+03 kg with high temperature and 1.62E+04 kg with low temperature supercritical drying method while the supercritical drying process stage being the major impact contributor in terms of both material and energy [12]. It can be observed that the impact share of biomass-derived aerogels in this impact category is considerably less in comparison to silica aerogels, owing to the deployment of a novel freeze-drying method .

Another similar study by De Marco et al. [36] aimed at comparing the environmental impacts associated with the manufacturing of corn starch aerogels with supercritical drying method at bench scale and pilot scale. According to the results of the study, an impact contribution of 3.84E + 00 and 1.06E + 00 kg CO_2 eq. have been evaluated for 1 g corn starch aerogels manufactured at lab scale and industrial scale, respectively [36]. If we equate the functional unit of this study (1 g) with the one in our study (1 m^3) for comparing the impact values, then it can be comprehended that the manufacturing 1 g of biomass-based aerogels by freeze-drying method will account for a CO_2 burden of 1.69E-02 kg (density of manufactured biomass-based aerogels is 0.04 g/cm^3). The comparison between corn starch aerogel and biomass-based aerogels with different drying methods confirms that freeze-drying method for manufacturing aerogels accounts for substantially less environmental impacts and has a significant share is reducing the CO_2 burden associated with the process [36].

Aerogels typically have a surface area between 200-600 m^2/g, where biomass-based aerogels have surface area of approximately 220 m^2/g. When comparing the climate change potential of biomass-based aerogels with other commonly employed thermal insulation materials in construction industry such as polyurethane and rock wool, biomass-based aerogels depict enhanced environmental performance. In one such study, Yilmaz et al. [37] analysed the environmental impacts of 1 m^2 of 50 mm thick polyurethane and rock wool composite panels. The results depicted an impact contribution of 6.10E+01 kg CO_2 eq. and 5.88E+01 kg CO_2 eq. by polyurethane and rock wool board [37]. Upon equating the impact value of 1m^2 of biomass-based aerogels with this study, the results depict that biomass-based aerogels have the least contribution *i.e.* 8.45E-01 kg.

If we compare the environmental performance of our novel product with other biomass-based insulation boards, the significantly better environmental performance of the novel biomass aerogel can be assessed. In a study by Gomez-Campos et al. [38], the life cycle inventory of the flax fiber has been assessed based on the processes of flax cultivation, scutching, combing, spinning and weaving. The results showed that the climate change potential of 1m^2 technical textile is 7.79E+00 kg CO_2 eq. [38]. In another study by Ben-Alon et al. [39], LCA of the natural cob earthen material has been conducted which presented an environmental impact of 1.32E+01 kg CO_2 eq. for a FU of 1 m^2 cob wall [39]. While equating the impact value in-terms of FU of present study i.e., 1 m^2 of biomass-based aerogels, the results depict that biomass-based aerogels have lower climate change potential i.e., 8.45E-01 kg CO_2 eq.

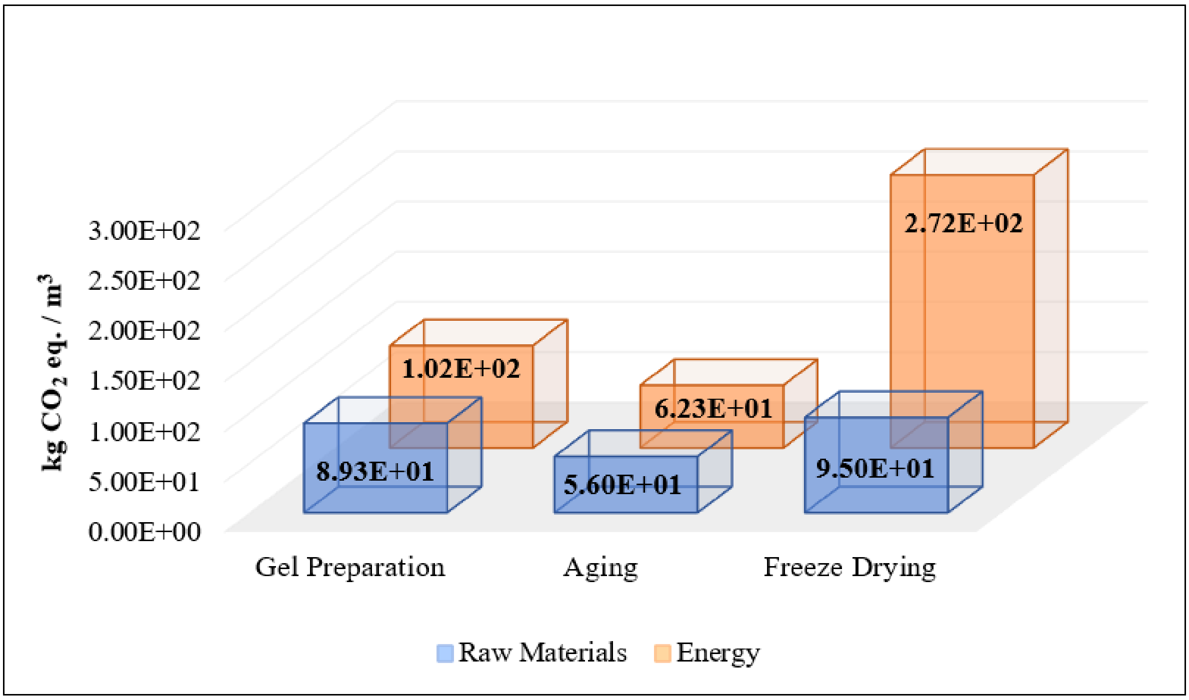


Figure 7. Overall impact of biomass-based aerogels manufacturing process stages on climate change impact category and the respective contribution of raw materials and energy

3.1.2. Non-renewable energy potential

It can be comprehended from Table 3 and Figure 6 that the highest impact value is achieved in this category *i.e.* 1.65E+04 MJ/m^3. The graphical depiction of the impact contribution of individual process stages and the relative share of raw materials' processing and energy consumption is presented in Figure 8. It can be observed that the freeze-drying stage of the manufacturing process has an impact share of about 59.6 % followed by the gel preparation stage i.e., 29.6 % (Figures 6 and 8). From the inventory data it can be analyzed that the electricity burden is maximum in the freeze-drying process *i.e.* 11108.24~14717.84 kWh, owing to the use of ultra-low temperature freezers and freeze dryers with a running time of 10 hours and 24 hours respectively. According to the results of the study by Dowson et al. [12], the total production energy involved in manufacturing 1m^3 of tetramethoxysilane aerogels was evaluated to be 2.28E+05 MJ with high temperature and 3.41E+05 MJ with low temperature supercritical drying method [12]. From the non-renewable energy potential data of tetramethoxysilane aerogels, it could be found that biomass-derived raw materials and freeze-drying method require less continuous electricity supply.

The results of another similar study by De Marco et al. [40], that compared the life cycle impacts of manufacturing 1 kg of maize starch aerogels by supercritical drying method at various production scales, depicted an impact value of 5.42E+04 MJ at bench scale and 1.25E+04 MJ at pilot scale for the same impact category [40]. On equating these results with our study, it can be analysed that the production energy involved with the manufacturing of 1 kg of biomass-based aerogels by freeze-drying method is 4.12E+02 MJ, which is far less in comparison to the results of the study by De Marco et al. [40]. This can be attributed to the use of sustainable freeze-drying process in terms of energy input and economic viability. The freeze-drying process requires a relatively less continuous electricity supply in comparison to supercritical drying method, as the temperature can be maintained within the ultra low temperature freezer and freeze dryer for longer periods.

Similarly, in a study by Pinto et al. [2], the environmental impacts of silica aerogels containing tetraethyl orthosilicate (TEOS) and 2-propanol (i-PrOH) manufactured by subcritical drying method were evaluated by deploying LCA tool. Subcritical drying method was employed as an environmentally sound alternative of supercritical drying process. The results of the study revealed an impact contribution of 7.42E+03 MJ per kg of finished aerogel product in non-renewable energy impact category which is still higher than 1 kg of biomass-based aerogels manufactured by freeze-drying method *i.e.* 4.12E+02 MJ [2].

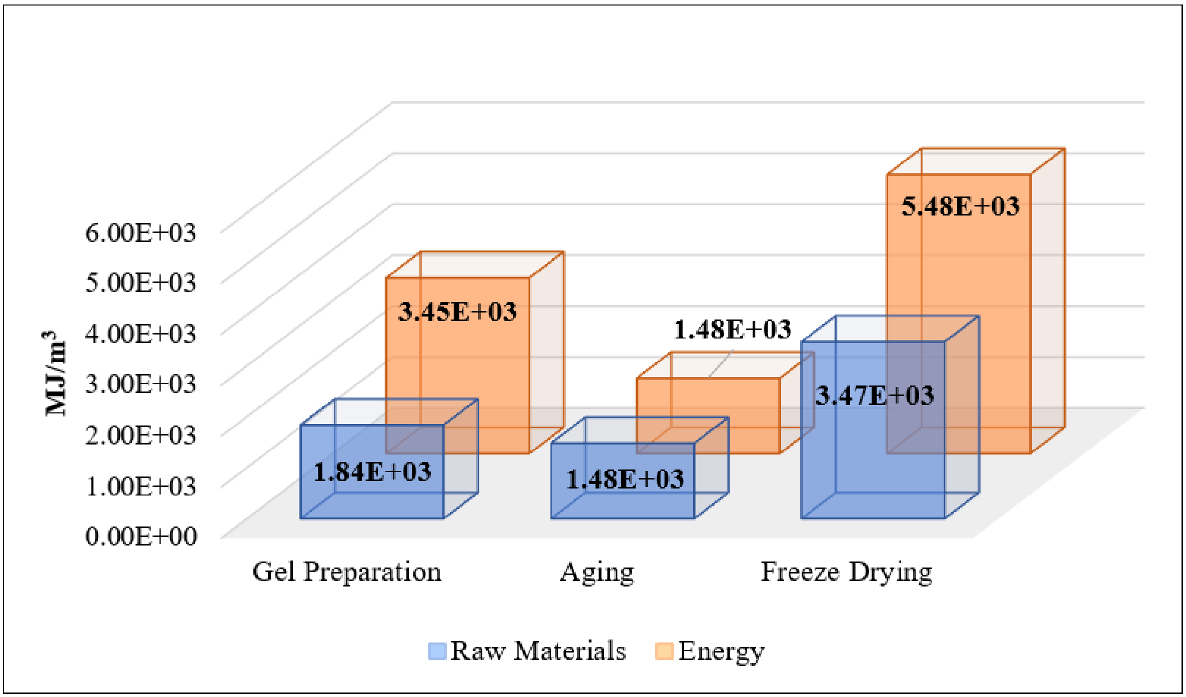


Figure 8. Non-renewable energy potential of biomass-based aerogels manufacturing process stages and the respective contribution of raw materials and energy.

3.1.3. Stratospheric ozone depletion

The impact contribution in this category is relatively less in comparison to other environmental parameters.

According to the results, an impact value of 4.21E-04 kg CFC-11 eq./m^3 is achieved in stratospheric ozone depletion category (Table 3). The freeze-drying stage accounted for an impact contribution of 2.40E-04 kg CFC-11 eq. presenting the highest share of approximately 57 % (Figures 6 and 9). This can be explained in the light of the findings by Pan et al. [41], which focused on the production of hybrid (silica aerogel based on MTMS/Water-glass co-precursor) aerogels by freeze-drying method and confirmed that the low temperature freezing process accounts for higher CFCs release in comparison to other process stages [41]. According to the results of a study conducted by Karatum et al. [42] which employed the LCA approach to analyze the environmental impacts of silica aerogels prepared at small and large scale, it can be concluded that manufacturing of 1 ml of monolithic silica aerogels by CO_2 supercritical extraction method resulted in an impact value of 1.80E-08 kg CFC-11 eq. and of 5.00E-09 kg CFC-11 eq. k by alcohol supercritical extraction method [42]. If we compare it with the results of our study, it can be evaluated that 1 ml of biomass-based aerogels manufactured by freeze-drying process account for a CFC eq. release of 4.21E-10 kg (1m^3=1.00E+06 ml). In another study by De Marco et al. [31], the stratospheric ozone depletion potential of 1 g starch aerogels manufactured by supercritical drying method was evaluated to be 6.32E-07 kg CFC-11 eq [31]. Equating the functional unit of this study with our functional unit reveals that 1 g of biomass-based aerogels prepared by freeze drying method will result in significantly less impact contribution of 1.05E-08 kg CFC-11 eq.

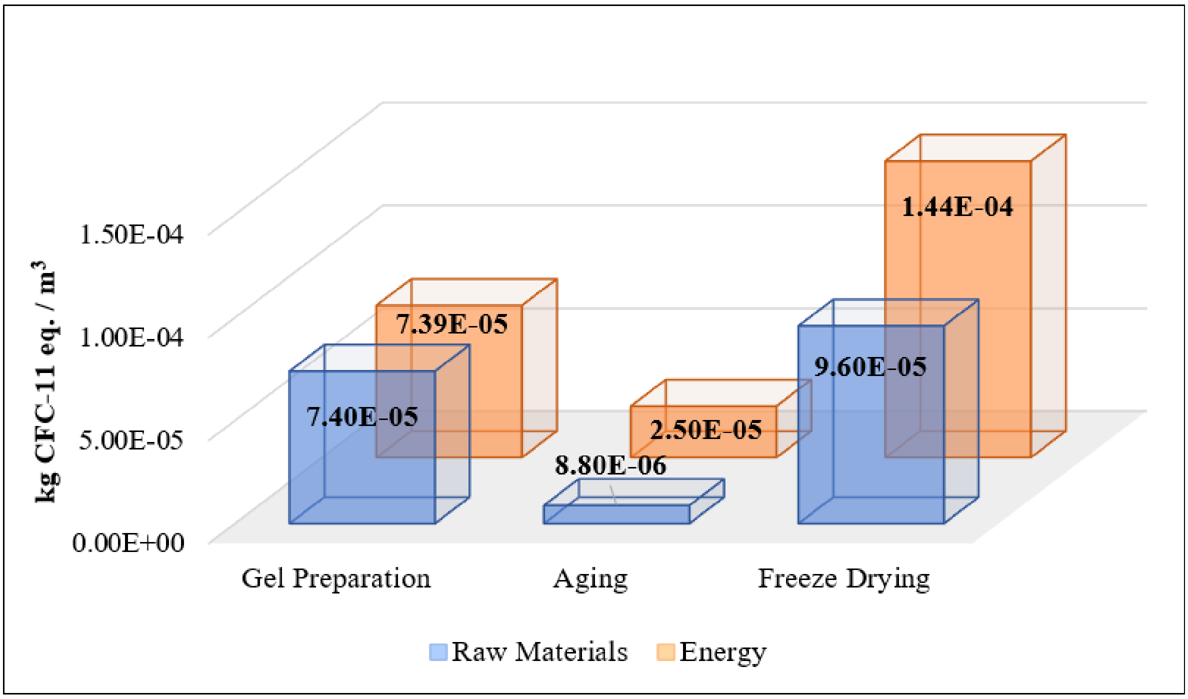


Figure 9. Impact of biomass-based aerogels manufacturing process stages on stratospheric ozone depletion and the respective contribution of raw materials and energy

The comparative assessment of environmental performance of different thermal insulation materials with biomass-based aerogels also reveals similar results. In a study by Yilmaz et al. [37], LCA-based environmental performance analysis of insulated composite façade panels was conducted. The results depicted a stratospheric ozone potential of 2.79E-06 and 3.17E-06 kg CFC-11 eq. for 1 m^2 of 50 mm thick polyurethane and rock wool composite panels respectively [37]. Whereas, upon equating biomass-based aerogels as per surface area of $1m^2$, the results reveal that biomass-based aerogels have the least contribution to this category i.e., 5.26E-07 kg CFC-11 eq. proving their environmental efficiency over conventional insulation materials.

3.1.4. Terrestrial acidification potential

The life cycle modelling revealed that the manufacturing of 1 m^3 of biomass-based aerogels results in a contribution of 8.57E+00 SO_2 eq. (Table 3). The gel preparation stage has the highest share of 54.7% i.e., 4.69E+00 kg SO_2 in the total impact followed by freeze-drying stage *i.e.* 2.59E+00 kg SO_2 (30.2%) (Figure 6 and 9). The use of biomass-based raw materials has resulted in a relatively lower impact in this category in comparison to other types particularly methanol based and silica aerogels. The results of the study by Pinto et al. [2], based on the environmental analysis of subcritical production of silica aerogels confirmed an impact share of 1.43E+00 kg SO_2 eq. per kg of aerogels prepared [2]. When compared with the acidification potential of 1 kg of biomass-based aerogels prepared by freeze-drying method i.e., 3.42 E-01 kg SO_2 eq., it can be concluded that the use of renewable organic raw materials results in significantly less environmental impacts.

Similarly, the environmental impact analysis of starch aerogels conducted in a study by De Marco et al. [30], demonstrated an impact contribution of 9.30E-02 kg SO_2 eq. per gram of aerogel manufactured by supercritical method [31]. In another study by De Marco et al. [36], the environmental impacts of 1 g of starch aerogels manufactured at bench scale and pilot scale were compared. The results showed an impact contribution of 5.09E-02 and 1.44E-02 kg SO_2 eq. respectively, in this midpoint impact category [36]. The comparative analysis of the results achieved for biomass-based aerogels with starch aerogels by equating the functional unit, demonstrated that manufacturing of 1 g of biomass-based aerogels by freeze-drying process results in relatively low terrestrial acidification potential of 2.14E-04 kg SO_2 eq., presenting them as an environmentally sound alternative.

In another study by Yilmaz et al. [37], the environmental impacts of 1 m^2 of 50 mm thick polyurethane and rock wool composite panels were quantified. It was evaluated that polyurethane panels accounted for a comparatively less acidification potential of 1.24E+00 kg SO_2 eq. than rock wool composite panels with an impact value of 1.30E+00 kg SO_2 eq. [37]. As the functional unit of this study is different from the one selected for our study (1 m^3), therefore, we have equated our functional unit by considering 1 m^2 of 50 mm thick biomass-based aerogels for effective comparison. Results highlight that biomass-based aerogels have the lowest impact value of 1.07E-02 SO_2 eq.

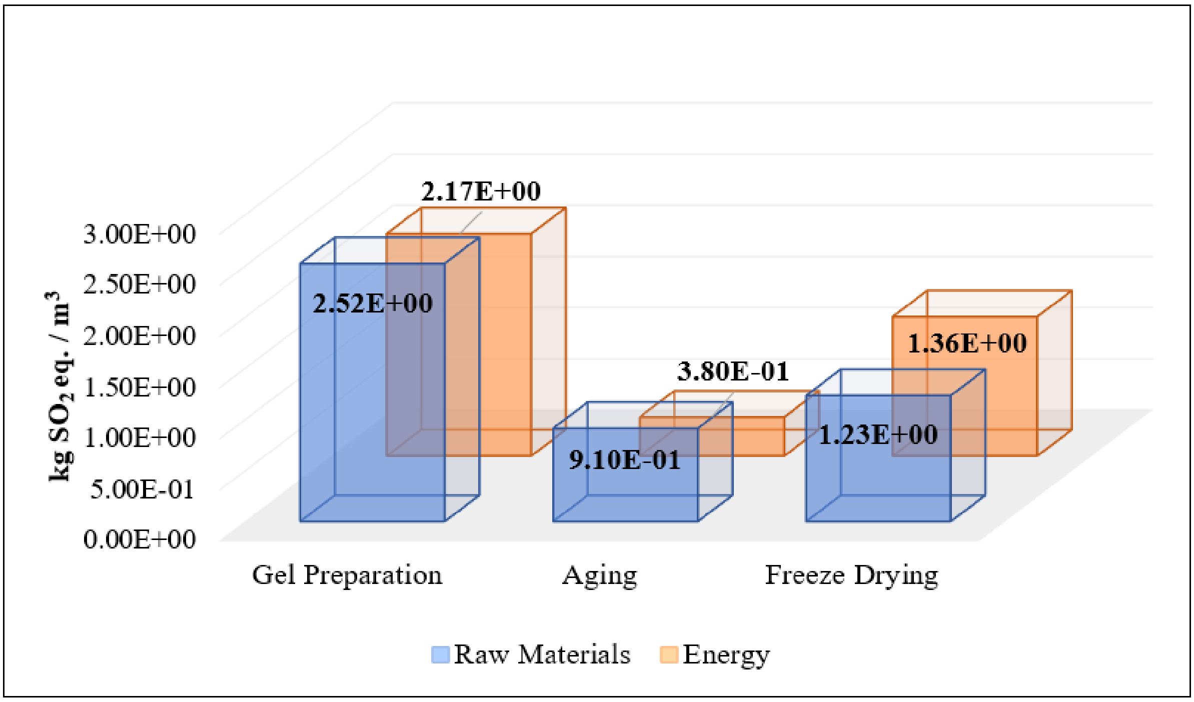


Figure 10. Impact of biomass-based aerogels manufacturing process stages on terrestrial acidification and the respective contribution of raw materials and energy

3.1.5. Terrestrial ecotoxicity

Table 3 highlights an impact contribution of 2.07E+03 TEG soil in this category per m^3 of biomass-based aerogel produced. The relative contributions of each of the process substages are 5.65E+02, 1.29E+02 and 1.38E+03 kg TEG soil in gel preparation, aging and freeze-drying, respectively (Figure 6 and 11). The environmental study on starch aerogels used in drug delivery and other medical applications conducted by De Marco et al. [36] demonstrated an impact score of 5.67E+01 and 1.58E+01 kg TEG soil per gram of aerogel produced at laboratory scale and industrial scale respectively [36]. In another similar study by De Marco et al., an impact contribution of 1.03E+02 kg TEG soil per 1 g of starch aerogels manufactured, was evaluated [31]. When compared with the LCA results achieved for biomass-based aerogels, it can be concluded that 1 g of biomass-based aerogels manufacturing account for a terrestrial ecotoxicity potential of 5.18E-02 kg TEG soil. The obtained impact value is significantly low in comparison with the results for starch aerogels evaluated in similar studies as discussed.

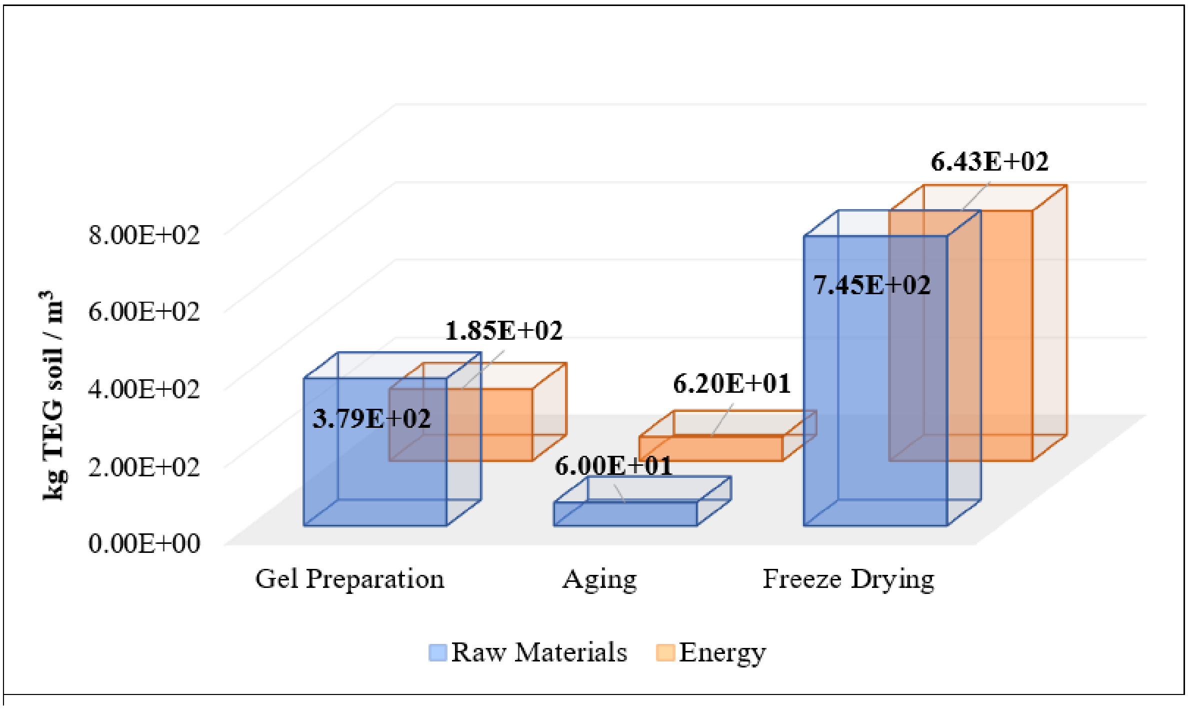


Figure 11. Impact of biomass-based aerogels manufacturing process stages on terrestrial ecotoxicity and the respective contribution of raw materials and energy

3.1.6. Aquatic ecotoxicity

The total impact contribution to the category of aquatic toxicity is reported to be 9.87E+03 kg TEG water as per 1 m^3 of biomass-based aerogels produced (Table 3). The individual contribution of the manufacturing substages highlights the greatest share is of freeze-drying stage *i.e.* 6.21E+03 kg TEG water, with the aging and gel preparation stage contributing 7.89E+02 and 2.87E+03 kg TEG water, respectively (Figure 6 and 12). In a similar study by De Marco et al. [40], the environmental impacts of starch aerogels produced by the supercritical drying method were evaluated, the results showed an impact value of 2.17E+02 kg TEG water for bench scale and 6.53E+01 kg TEG water for pilot scale manufacturing of 1 g of aerogel [40]. In another study, De Marco et al. [31] evaluated an aquatic ecotoxicity potential of 4.04E+02 kg TEG water associated with the pilot scale manufacturing of maize starch aerogels by supercritical drying method [31]. On equating the functional units, it can be comprehended that the aquatic ecotoxicity potential of 1 g of biomass-based aerogels manufactured by freeze-drying method is 2.46E-01 kg TEG water. With the comparison between these biomass-based aerogels manufacturing technology, there has no alcohol used in the freeze-drying process attributing to the lower impact value in the aquatic ecotoxicity. This highlights the potential positive impact of the biomass-based aerogels produced by the freeze-drying method making it comparatively more environment friendly.

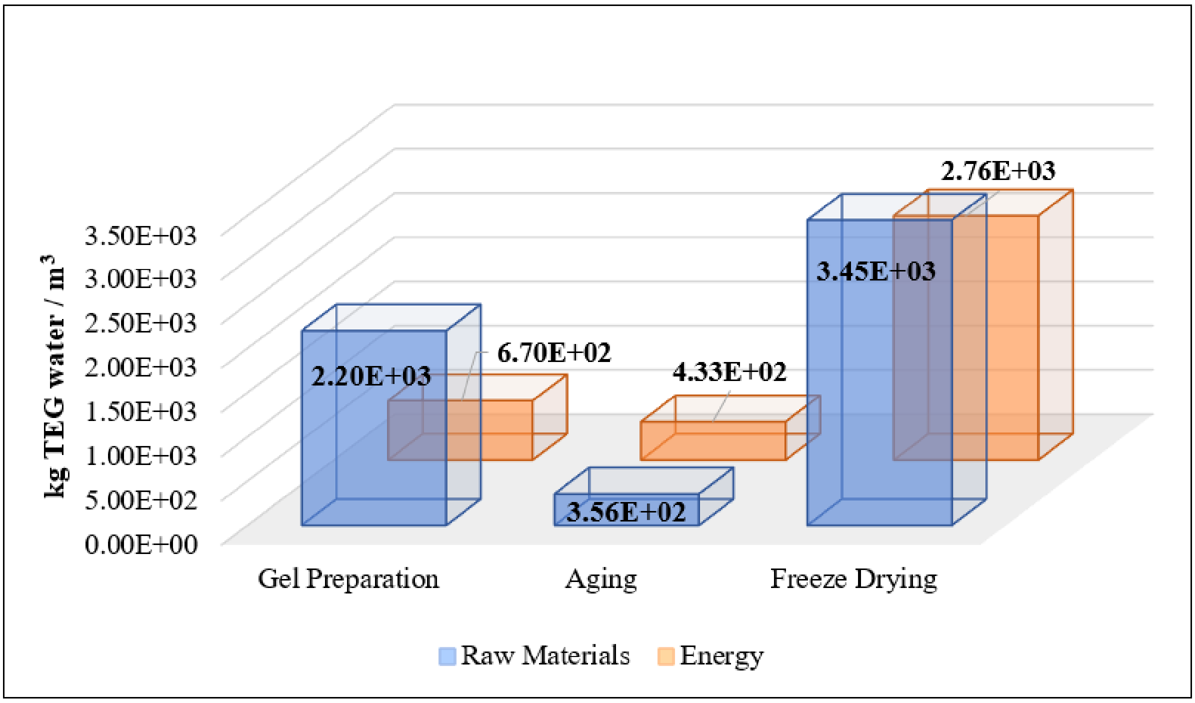


Figure 12. Impact of biomass-based aerogels manufacturing process stages on aquatic acidification and the respective contribution of raw materials and energy

3.2. Practical implications of the study

The results obtained from streamlined LCA, reveal that the use of biomass-based raw materials and deployment of freeze-drying method for manufacturing, make the finished product, an environment friendly and efficient alternative to conventionally used aerogels. When compared with other similar materials, the relative environmental impact scores of biomass-based aerogels have been evaluated to be considerably less. In the case of starch aerogels derived from organic sources like corn and maize, manufactured by subcritical or supercritical drying method, the relatively less impact value of biomass-based aerogels can be attributed to the deployment of freeze-drying method for manufacturing. This novel method is considered less electricity intensive as the low temperature once achieved within the freeze dryers can be maintained for longer periods. While in the case of conventional production methods, large volumes of CO_2 are often used to maintain the supercritical flow, which further adds to the adverse environmental impacts. Similarly, when compared with inorganic silica aerogels, the novel biomass-based aerogels proved to be more sustainable with lower impact scores in all the selected impact categories. This can be justified in terms of the natural raw materials used. Table 4 presents a comparative analysis of the results achieved by our study, with other similar previous studies.

In addition to environmental benefits, these biomass-based aerogels possess a wide range of applications including thermal insulation, biomedical and environmental fields with improved efficiency and promising mechanical performance [22]. In the construction industry, the use of silica aerogels as thermal insulators is highly encouraged due to their exceptionally low thermal conductivity i.e., 0.012 - 0.015 W/m·K, which is even lower than air (0.025 W/m·K). However, there are certain other limitations like brittleness [43], high production cost and large environmental footprint [9]. The biomass-based aerogels produced by freeze-drying method have no such disadvantage of brittleness as verified by previous study in which its elasticity was well observed [22]. Table 5 presents a comparison of the major physical performance parameters of these novel biomass-based aerogels with conventionally used aerogel types such as silica and starch. The thermal conductivity of these novel aerogels is comparable with other similar materials while the density and tensile strength is better. The method of production of biomass-based aerogel contributes to the low production cost and low impact on the environment compared with other aerogels produced by supercritical methods. Meanwhile, it also possesses relatively good thermal insulation properties. Based on these characteristics, the biomass-based aerogels are expected to act as the core material of the lightweight structural panels in temporary buildings, which commonly appear in the rapid housing reconstruction program after the natural disaster or emergencies such as earthquakes, floods or COVID-19 pandemic [44].

These panels are a form of sandwich panel structure, which is composed of two-layer thin metal panels and lightweight biomass-based aerogels as the middle layer. This fully considers the structure, strength and water resistance requirements. After the usage, these panels could be reused if needed or disassembled to take out the biomass-based aerogel for landfill treatment, in which they will be degraded rapidly._For green production, resorcinol-formaldehyde derived aerogels have certain disadvantages like toxicity and cost intensiveness [45]. Similarly, for tissue engineering purposes, the use of silica aerogels is limited as it is non- biodegradable and cannot undergo decomposition in human body [36]. These novel biomass-based aerogels can overcome all these limitations. The green chemistry of biomass-based aerogels presents the opportunity to extend its applications to other diverse fields.

Table 4. Comparative assessment of LCA results obtained from similar previous studies

Impact category	Biomass-based aerogels manufactured by freeze-drying method				Results from previous studies			
	1 m^3	1 m^2	1 kg	1 g	Method of production	Functional unit	Impact value	Reference
Climate change potential ($kg\ CO_2$ eq.)	6.76E+02	8.45E-01	1.69E+01	1.69E-02	High temperature supercritical drying	1 m^3 of silica aerogels	2.84E+03	[12]
					Low temperature supercritical drying	1 m^3 of silica aerogels	1.62E+04	[12]
					Bench scale supercritical drying	1 g corn starch aerogels	3.84E+00	[36]
					Pilot scale supercritical drying	1 g corn starch aerogels	1.06E+00	[36]
					Belt lamination	1 m^2 polyurethane panel	6.10E+01	[37]
					Belt lamination	1 m^2 rock wool board	5.88E+01	[37]

					Scutching, spinning and weaving	1 m^2 flax technical textile	7.79E+00	[38]
					-	1 m^2 cob wall	1.32E+01	[39]
Non-renewable energy potential (MJ primary)	1.65E+04	8.25E+02	4.12E+02	4.12E-01	High temperature supercritical drying	1 m^3 of silica aerogels	2.28E+05	[12]
					Low temperature supercritical drying	1 m^3 of silica aerogels	3.41E+05	[12]
					Subcritical drying	1 kg of silica aerogels	7.42E+03	[2]
					Bench scale supercritical drying	1 kg of maize starch aerogels	5.42E+04	[40]
					Pilot scale supercritical drying	1 kg of maize starch aerogels	1.25E+04	[40]
Stratospheric ozone depletion (kg CFC-11 eq.)	4.21E-04	5.26E-07	1.05E-05	1.05E-08	CO_2 supercritical extraction	1 ml of monolithic silica aerogels ($1m^3=1.00E+06$ ml)	1.80E-08	[42]
					Alcohol supercritical extraction	1 ml of monolithic silica aerogels	5.00E-09	[42]
					Supercritical drying	1 g starch aerogels	6.32E-07	[31]

					Belt lamination	1 m^2 polyurethane composite panels	2.79E-06	[37]
					Belt lamination	1 m^2 rock wall composite panels	3.17E-06	[37]
					Scutching, spinning and weaving	1 m^2 flax fiber textile	1.55E-07	[38]
Terrestrial acidification potential (kg SO_2 eq.)	8.57E+00	1.07E-02	2.14E-01	2.14E-04	Subcritical drying	1 kg of silica aerogels	1.43E+00	[2]
					Bench scale supercritical drying	1 g starch aerogels	5.09E-02	[36]
					Pilot scale supercritical drying	1 g starch aerogels	1.44E-02	[36]
					Supercritical drying	1 g starch aerogels	9.30E-02	[31]
					Belt lamination	1 m^2 polyurethane composite panels	1.24E+00	[37]
					Belt lamination	1 m^2 rock wall composite panels	1.30E+00	[37]

Terrestrial ecotoxicity (kg TEG soil)	2.07E+03	1.04E+02	5.18E+01	5.18E-02	Bench scale supercritical drying	1 g starch aerogels	5.67E+01	[36]
					Pilot scale supercritical drying	1 g starch aerogels	1.58E+01	[36]
					Supercritical drying	1 g starch aerogels	1.03E+02	[31]
Aquatic ecotoxicity (kg TEG water)	9.87E+03	4.94E+02	2.46E+02	2.46E-01	Bench scale supercritical drying	1 g starch aerogels	2.17E+02	[36]
					Pilot scale supercritical drying	1 g starch aerogels	6.53E+01	[36]

Table 5: Comparative analysis of performance parameters of biomass-based aerogels

Parameters	Starch aerogel	Silica aerogel	Biomass-based aerogel	Source
Thermal conductivity W/m·K	0.024	0.012-0.015	0.046 ~ 0.052	[22] [46] [47]
Density g/cm^3	< 0.2	0.29	0.020 ~ 0.2	[22] [46] [47]
Tensile strength (MPa)	0.065-0.069	0.081	0.0675	[22] [46]

3.3. Impact score and sensitivity analysis

Sensitivity analysis based on the LCI values has been conducted to present a clearer perspective of the environmental externalities associated with novel biomass-based aerogels. For process optimization and to select the best combination of raw materials along with other parameters, an exact precise value has not been quoted and instead a narrow range of values is presented in the LCI. While this sensitivity analysis has also considered any uncertainties involved in the calculation of impact scores due to probable deviations in raw material and energy use during biomass aerogel manufacturing. In Figure 13, the vertical bars depict the actual impact scores calculated based on the average LCI values while the red capped line presents the possible variations in weighted impact scores highlighting the minimum and maximum score for each selected impact category. The scores have been calculated based on the highest and lowest value for raw material used and energy consumed during various process stages (Table 1). As the production of novel biomass aerogels was done at bench scale, some disparity in the modelled environmental impacts can be anticipated with industrial scale manufacturing in larger batches. Therefore, sensitivity analysis for presenting the highest and lowest possible impact scores has been included to better understand the process dynamics and environmental footprint at different scales of production.

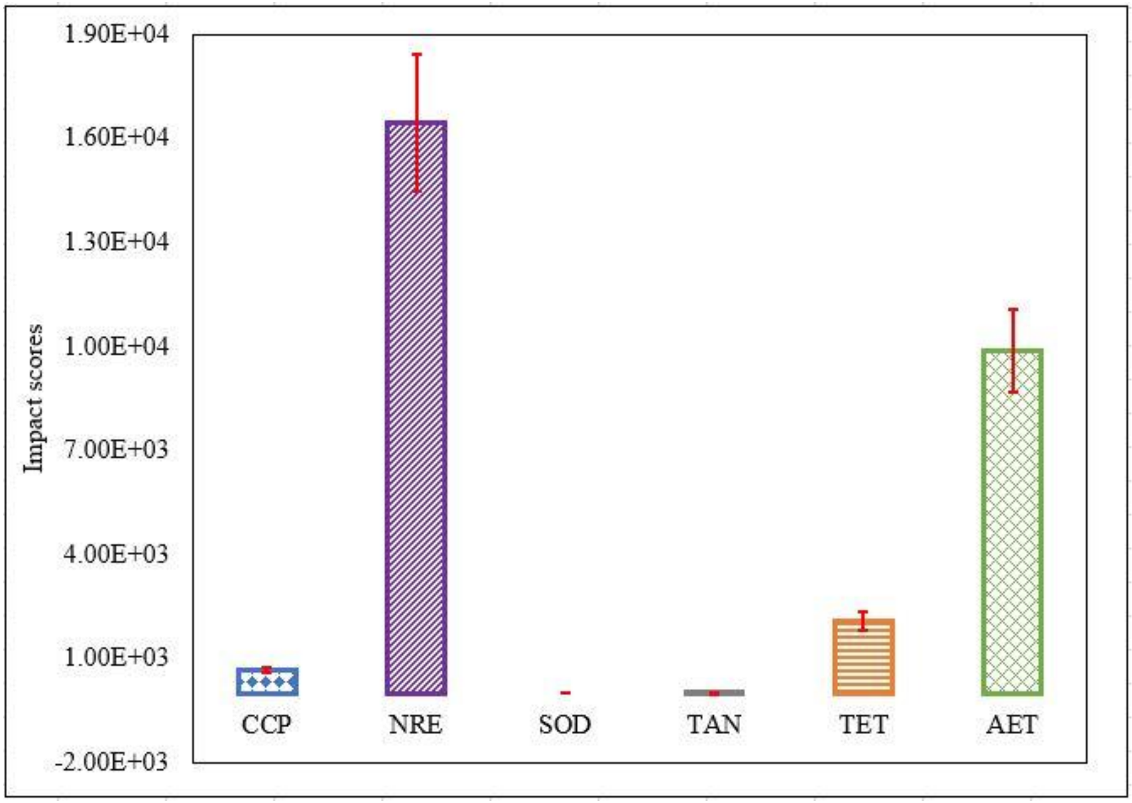


Figure 13. Sensitivity analysis of the life cycle impact scores based on LCI values

4. Conclusion

The study has analyzed the environmental footprint of manufacturing of a promising biomass-based insulation material manufactured by freeze-drying method, comprising of three major process stages: 1) gel preparation; 2) aging; 3) freeze-drying. The results revealed an overall impact value of 6.76E+02 kg CO_2 eq., 1.65E+04 MJ, 4.21E-04 kg CFC-11 eq., 8.57E+00 kg SO_2 eq., 2.07E+03 kg TEG soil and 9.87E+03 kg TEG water in climate change potential, non-renewable energy potential, stratospheric ozone depletion, terrestrial acidification potential, terrestrial ecotoxicity and aquatic ecotoxicity impact categories, respectively. On comparing the impact values quantified in this study with previous studies focused on conventional aerogel types including silica aerogels, it has been depicted that aerogel manufactured by biomass-based raw materials using the freeze-drying method is an environmentally sound and efficient alternative. To be more specific, compared with the silica aerogel, the biomass-based aerogel with freeze drying method can bring decrease of the climate change potential, non-renewable energy potential and terrestrial acidification potential impacts by 76.20%, 92.76%, 85.03%, respectively. Besides, only the impact of production stage is involved in the comparison between silica aerogel and biomass-based aerogel. If the impact of material disposal is considered, the residual impact of silica aerogel will be added while that of biomass aerogel can be ignored as it is 100% biodegradable.

This study has provided valuable insights into a sustainable construction material based on both raw material and process modifications. As discussed, the performance parameters of the proposed material are comparable with other conventional types with significantly less environmental impacts. Although the novel freeze-drying method do pose some negative environmental externalities too due to a large demand of energy for processing which can be highlighted as a major limitation of this study, the potential guidance and improvements can also be derived based on this study. The energy usage can be reduced by incorporating the use of equipment with higher energy efficiency, using renewable energy sources for meeting the electricity need for freeze drying and reducing the processing time. Once the production and maufactring process will be largerly commercialized the greater product batches, or a continous flow process will further improve the environmental performance of the product and process due to the concept of economies of scale.

Author statement

Yixin Wang: Data Collection, Methodology, Formal analysis, Investigation, Writing, Review and Editing. **Rizwan Rasheed:** Data Administration, LCA Design, Methodology, Modelling, Formal analysis, Investigation, Writing, Review and Editing. **Fatang Jiang:** Resources, Conceptualization. **Asfra Rizwan:** LCA Calculations and Interpretations, Review and Editing. **Hajra Javed:** LCA Interpretations, Writeup, Review and Editing. **Yuehong Su:** Conceptualization, Supervision, Administration, and Review. **Saffa Riffat:** Supervision.

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