Heliyon 11 (2025) e41527

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Research article

Assessment of greenhouse gas emissions and environmental impacts in the manufacturing process of thermoelectric coolers: A life-cycle impact perspective

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ARTICLE INFO

Keywords:
Life cycle inventory assessment
Thermoelectric coolers
Carbon emission
Ecotoxicity
Scarcity
Integrated pollution prevention and control

ABSTRACT

A path to carbon neutrality requires the development of refrigeration units that use no refrigerant or emit less greenhouse gas (GHG), such as Thermoelectric coolers (TECs). Using the life cycle inventory assessment (LCIA), the environmental impacts of the manufacturing process of TECs were analyzed, including greenhouse gas emissions, human carcinogenic toxicity (HCT), terrestrial ecotoxicity (TE), freshwater ecotoxicity (FE), mineral resource scarcity (MRS), and fossil resource scarcity (FRS). The alumina plate manufacturing process produces the most GHG emissions because it uses a lot of electricity in the sintering process. The type of energy source significantly affects GHG emissions, HCT, FE, and FRS but has only a limited impact on TE and MRS. Also, TE, FE, and MRS are affected by the mineral resources used to manufacture the legs.

Also, GHG reductions in the manufacturing process have been predicted based on Korea's electricity supply and demand plan for 2030. According to the plan, fossil energy is expected to decrease in 2030 compared to 2021, while renewables and nuclear power are expected to increase. For every 1 MWh of cooling amount, GHG emissions are predicted to decrease from 2.9 kg CO_2-eq in 2021 to 1.95 kg CO_2-eq in 2030 with a greener energy mix.

In addition, generating 2.1 % with green hydrogen would reduce total GHG emissions by 1.7%p more than grey hydrogen generation. Increased use of nuclear and hydrogen energy and decreased use of coal energy are likely to be the biggest drivers of reductions. This study suggests that alternatives to alumina plates that are more environmentally friendly should continue to be explored along with process improvements such as fast heating rate or sintering aids.

1. Introduction

The use of refrigerants in the operation of refrigeration units results in increased greenhouse gas (GHG) emissions and contributes to climate change [1–3]. The main F-gases emitted include chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) which have an atmospheric lifetime of up to 50,000 years, resulting in long-term environmental impact [4]. To mitigate environmental concerns, alternative cooling methods that do not use refrigerants or that emit less greenhouse gases are essential.

In a warming world, civilizations rely more on access to cooling, and the increased demand for cooling contributes significantly to climate change [5]. The development of refrigerant substitutes in refrigeration is a promising way to reduce the environmental and

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https://doi.org/10.1016/j.heliyon.2024.e41527
Received 18 June 2024; Received in revised form 23 December 2024; Accepted 26 December 2024
Available online 26 December 2024
2405-8440/

climate system impacts of the global increase in cooling demand on climate change [6]. Thermoelectric coolers (TECs) are refrigeration units that do not use refrigerants and have the advantage of not posing a threat to the environment and climate system.

Using the Peltier effect, TECs can convert electrical energy into temperature gradients. TEC systems consist of three key elements: thermoelectric legs (p-type and n-type semiconductors), heat sinks, and heat exchangers. Electrons move from the lower energy region inside the p-type to the higher energy region inside the n-type through the cold junction, transferring the absorbed heat through the cold side to the hot junction, causing the heat to be dissipated in the heat sink [7]. Additionally, TEC devices have advantages such as light weight, no moving parts, no or little noise, and no vibration. As a sustainable energy technology, TECs are utilized in compact refrigerators, wearable medical devices [8], automotive air-conditioners [9], and cooling devices for electronics [10].

Life cycle assessment (LCA) is used to assess the potential environmental impacts from raw material acquisition, via the use phases, to waste management [11]. LCA has been conducted in various fields, including construction, the chemical industry, agriculture, metals, waste management, transport, etc. LCA quantitatively and systematically evaluates environmental impacts such as global warming, scarcity, toxicity, etc. In addition, the results of LCA analysis can be used for marketing, strategic planning, and more [11]. LCA can even lead to new insights during the development of new technologies and can support policymakers in their work [13].

To date, there have been several LCA analyses to analyze the environmental performance of thermoelectric materials, with a focus on analyzing the environmental impact of different types of modules, one of the components used in TECs. Iyer and Pilla [14] evaluated the environmental impact of seven types of commercial or non-commercial TE modules (applied to thermoelectric generators, instead of coolers) over their life cycle. They found that, with some exceptions, TE modules provide significant positive environmental benefits over their lifecycle, regardless of the type of module used. They also found that the benefits of TE are comparable to those of renewable energy, including solar and wind energy. Soleimani et al. [15] divided thermoelectric modules into three categories (inorganic, organic, and hybrid), and an LCA was performed to consider the environmental performance of thermoelectric materials based on their efficiency. The results showed that the life cycle impact of inorganic thermoelectric materials was significantly higher than that of organic and hybrid because the manufacturing process of inorganic materials is very energy intensive.

Research has been conducted to combine thermoelectric coolers with PV systems or phase change materials to reduce energy consumption during the use process. Irshad et al. [16] investigated the carbon emissions of a PV-connected TE air-cooling system, a grid-connected TE air-cooling system, and a split air-conditioner. Based on a capacity of 0.75 tons, the electricity consumption of a split air-conditioner is 2.65 MWh/year, while a grid-connected TE system can save 1.09 MWh/year of electrical energy compared to the split air-conditioner. CO_2 emissions were calculated using a CO_2 intensity of 1.21 kg/kWh, and the grid-connected TE system reduced CO_2 emissions by 35.49 tons/life compared to a split-air conditioner.

Bozorgi et al. [17] analyzed the global warming potential of a phase change material (PCM)-based thermoelectric (TE) refrigerator. The CO_2 emissions of the system were found to be 1190 kg over the 15-year lifetime. In terms of hotspots in the life cycle, they concluded that the use stage emits nearly 97 % of the total CO_2, with most emissions coming from the use stage in the system. The PCM-based TE refrigeration system can be used without electricity for 29.4 % of a week, thereby reducing power consumption and the resulting emission during use process.

For comparison, the study by Solano-Olivares et al. [18] is analyzed, which experimentally evaluated the life cycle of a solar absorption and commercial air conditioning system. The functional unit was defined as "114,400 kW [sic] of cooling amount during lifetime 10 years". The solar absorption air-conditioning system has an emission of 0.139 kg CO_2-eq per kW and the commercial air-conditioning system has an emission of 0.697 CO_2-eq per kW.

A general review of the existing literature shows that environmental impact analyses have been conducted mainly based on leg type, which is the main material related to the efficiency of TECs or analyzed the use stage rather than the manufacturing process. Accordingly, it is important to analyze the impact of the input energy, which is expected to be as influential as the input material, since only the input material of the module has been mostly analyzed in previous studies.

Since electricity is used in all the production processes of the materials that make up TECs, changes in the electricity grid mix directly affect GHG emissions and environmental impact. Therefore, it is necessary to analyze the impact of the electricity grid mix. However, existing studies on the life-cycle environmental impact of TECs have been very limited in their analysis of changes in the electricity grid mix.

The Ministry of Trade, Industry, and Energy of South Korea develops a basic electricity supply and demand plan every two years to forecast mid-to long-term electricity demand for the next 15 years and to expand electricity facilities. The 9th Plan [19], released in 2020, and the 10th Plan [20], released in 2022, have somewhat different projections (targets) for the share of electricity generation by 2030. The main direction is to reduce the amount of coal-fired power generation and increase nuclear power and renewable sources. The 9th Plan projected coal at 30.3 % of 2030 generation, but the 10th plan projects an even larger decline to 20.1 %. Compared to 34.3 % in 2021, this is a significant decrease. In addition, nuclear power generation is expected to increase its target by 7.4%p from 25.0 % in the 9th to 32.4 % in the 10th, while renewable energy is expected to increase its target by about 3.9%p from 20.0 % in the 9th to 23.9 % in the 10th. With a renewable energy share of 6.2 % in 2021, the plan is to more than triple that by 2030. In the 10th Plan, the share of hydrogen and ammonia co-firing power generation was newly established at 2.1 %. The 9th and 10th Plans also established a new category for power generation using fuel cells as of 2030, which was 2.8 % in the 9th Plan and 2.6 % in the 10th Plan.

Therefore, the first goal of this study is to analyze the GHG emissions and environmental impacts of ecotoxicity and scarcity in the production of TECs by region. It is assumed that the manufacturing process is the same, but all the materials used as inputs to the process are produced in the country or available in the global market, and the GHG emissions and environmental impacts are accordingly calculated. The results of this study are expected to serve as a reference for exploring hotspots by showing the difference in emissions due to differences in raw materials and energy-producing countries with the same process using the same materials. The second objective of this study was to analyze the changes in GHG emissions from the production of TECs in South Korea in response to changes in the projected energy mix of Korea's electricity generation in 2030. In addition, we wanted to identify the effectiveness of policy changes by checking the GHG reduction effect of the energy mix and applying the LCA methodology as a tool to support policymaking.

2. Methods

2.1. Modules to analyze

A TEC is made of three main components: p-type and n-type legs, ceramic plates, and copper tabs. TECs are composed of a module in which a pair of p-type and n-type semiconductor legs is coupled to form a row. The copper tab is used to combine semiconductor legs and ceramic plates to allow electricity to flow. In addition, a ceramic plate that absorbs or releases heat is attached to the legs.

In this study, a bismuth-telluride module (Kelk Ltd.) [21] was selected and analyzed. According to Soleimani et al. [15], Bi_2Te_3 modules generate the least negative environmental impact among inorganic thermoelectric materials. Bi_2Te_3 modules have reached commercially viable levels of performance and are one of the most widely used thermoelectric materials available today.

The legs of the module are composed of a p-type leg ($Bi_2Sb_{1.6}Te_3$) and an n-type leg ($Bi_2Te_{2.4}Se_{0.6}$) . The ceramic plate is made of aluminum oxide (Al_2O_3) and copper tabs are thin lines made of copper (Cu). Detailed information on the weight of each material is provided in Table S1 in the Supplementary Information. The module created by assembling the main materials is assumed to be mounted in a TEC refrigerator that may store food and pharmaceuticals at low temperatures.

2.2. Life cycle assessment

LCA was conducted according to the guidelines of ISO 14040 and 14044 [11,12]. ISO 14040 provides the principles and framework of the LCA, and ISO 14044 is the requirements and guidelines of the LCA. Referring to ISO 14040, there are four phases in an LCA: Goal and Scope definition, Life Cycle Inventory (LCI), Life Cycle Impact Analysis (LCIA), and Interpretation. Therefore, following the guidelines of ISO 14040, the four phases of this LCA study are described below.

1) Goal and Scope definition: The LCA goal of this study was to evaluate the environmental impacts, including greenhouse gas emissions of thermoelectric coolers over their manufacturing process, and find ways to reduce them. For this goal, the environmental impacts of the electricity grid mix change used in all processes were analyzed in detail. Therefore, p-type and n-type legs, ceramic (alumina) plates, and copper tabs consisting of thermoelectric coolers were analyzed. The regions analyzed were France, the United States, Australia, China, and South Korea. The regions were selected because they have a special grid mix with a high proportion of generation from nuclear or renewable or fossil fuels.

In addition to greenhouse gas emissions (GHG), the analysis focused on resource scarcity and ecotoxicity. Resource scarcity of fossils (FRS) and minerals (MRS) and ecotoxicity of freshwater (FE), terrestrial (TE), and human carcinogenic toxicity (HCT) were analyzed.

Also, the goal and scope definitions include LCA standards and functional units. A functional unit is a quantitative measure of the functionality provided by the product [22]. The functional unit is 1 MWh of cooling amount from 47.89g module set during lifetime. A module with a similar specification, KSM-09071C [23], exhibits a $Q_{c,max}$ value of about 50 W. If we assume that the COP of the thermoelectric module is slightly less than 0.3 [24], the input power required can be estimated to be about 170 W. The lifetime is calculated as 100,000 h [25]. The calculation method and results for cooling amount and input power are described in Table S2 in the Supplementary Information (see Fig. 1).

2) Life cycle inventory (LCI): LCI is an inventory of input and output data of a product system [12]. LCI data were collected using Ecoinvent v3.8 database [26] and modeled using the openLCA v2.0.3 program [27]. In this study, we refer to the input/output data of the bismuth-telluride module manufacturing process in the paper by Iyer and Pilla [14]. The system boundary in Fig. 2 shows the

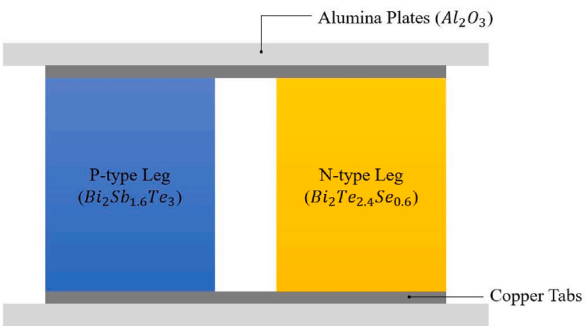

Fig. 1. Structure and composition of a TEC.

input/output data. The input data are energy, water, and chemicals that are required to form the final components through the process using the powder as shown in Fig. 2. Output data includes output materials and chemicals released into water, soil, and air.

3) Life cycle impact assessment (LCIA): The purpose of an LCIA is to analyze the environmental significance of a product system by providing additional information to evaluate the results of an inventory of its inputs and outputs [12]. In this study, LCIA was conducted with the ReCiPe2016 Midpoint method. ReCiPe2016 is a state-of-the-art method for converting life cycle inventories into life cycle impact scores at the midpoint and endpoint levels [28]. The midpoint analysis of the ReCiPe2016 method combines the output inventory that affects each item and analyzes human toxicity, mineral depletion, tec. The endpoint analysis combines the midpoint items and is calculated into the groups of human health, ecosystem health, and resource availability. The reason for the midpoint in this study is to specifically analyze the environmental impact of the manufacturing process of TECs.
4) Interpretation: Interpretation is a systematic technique for quantifying and evaluating the results of the previous step, LCI or LCIA. Interpretation provides further confirmation of the goal and scope parameters and can be used to assess whether the system boundaries are appropriate. In this study, the results were interpreted within the system boundary considering the input/output inventory and TEC manufacturing process.

2.3. System boundary

Fig. 2 illustrates the system boundary of the manufacturing process. In this study, the environmental impact was calculated by assuming that the module was later applied to a refrigeration unit.

The manufacturing stage includes the process of each material from raw material to finished material. For this stage, we refer to the module's process in the paper by Iyer and Pilla [14], and the inventory details are given in Table S3~S6 of Supplementary Information.

For the p-type and n-type legs, the manufacturing process starts with the raw materials of the elemental powders that make up each leg. Each leg is formed after powder mixing, hot pressing, ingot polishing, and leg dicing. Alumina plates are produced by ball milling, sintering, and polishing alumina powder available in the market. Copper tabs are produced by purchasing copper sheets available on the market and cutting them according to the specifications. The materials are then assembled and connected in pairs to form a module of 55 mm × 51.5 mm × 4.4 mm.

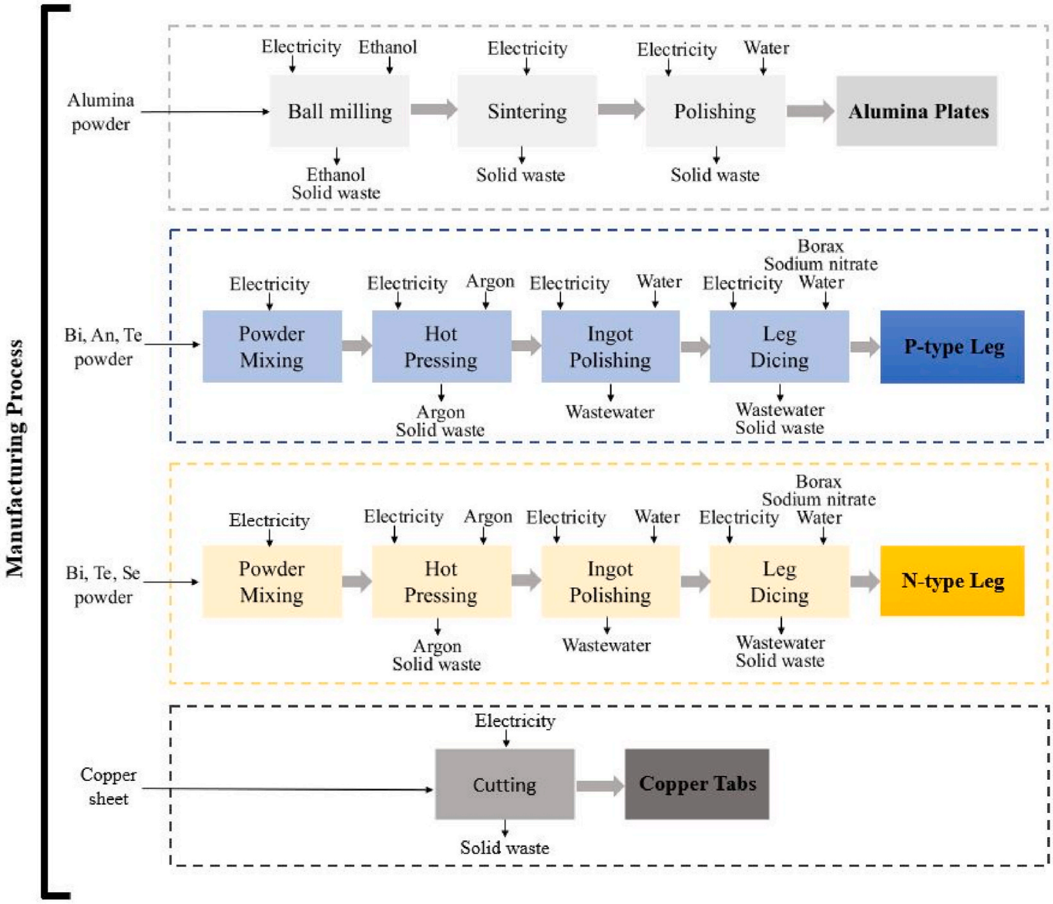

Fig. 2. The system boundary of the TEC manufacturing stage.

2.4. Electricity data

The environmental impact of electricity generation plays a crucial role in LCA, as almost all production processes use grid electricity [29]. Various types of energy sources generate electricity, and the environmental impact value and GHG emissions of electricity generated from different energy sources vary. Generating electricity from renewable sources is expected to make a better environmental contribution than generating electricity from fossil fuels.

We analyzed the differences in GHG and environmental impacts by electricity generation share using the 2021 electricity generation energy share data of five countries (France, United States, Australia, China, and South Korea) provided by the IEA (International Energy Agency) [30]. The Ecoinvent database of energy generation types in each country was used for analysis. Among the five countries analyzed, United State and China have regional data in the Ecoinvent database, while France, Australia, and South Korea have representative national values only. Therefore, we analyzed the Western Electricity Coordinating Council (WECC) in the U.S. and the Guangdong (GD) area in China as representative values.

Additionally, in South Korea, MOTIE (Ministry of Trade, Industry and Energy) prepares the basic electricity supply and demand plan every two years. It is a long-term plan for the next 15 years and is designed to forecast mid-to long-term electricity demand and expand electricity facilities accordingly. Comparing the 9th Plan (covering 2020–2034) and the 10th Plan (covering 2022–2036), the 10th Plan shows a decrease in fossil fuels and an increase in nuclear and renewable energy (as of 2030) compared to the 9th Plan, as in Table 1. In contrast to the baseline 2021 grid mix, the 9th Plan adds fuel cells as a new generation source, and the 10th Plan adds hydrogen and ammonia co-firing on top of that. The calculation of future GHG reductions expected by 2030 is based on the basic supply and demand plan in Table 1.

2.5. GHG emission calculation methods

The GHG emissions of electricity generation using fossil fuels (coal, natural gas, oil), renewables (solar PV, solar thermal, wind, biogas), nuclear, and hydro used in this study were obtained from the Ecoinvent v3.8 database. GHG emissions from electricity production using waste included biogas. Emissions from fuel cells and hydrogen and ammonia co-firing generation, which are currently being developed to keep up with the plans to increase the proportion of green energy in the future, are calculated directly using government sources, industry reports, and other sources. In 2.5.1, fuel cell generation and, in 2.5.2, hydrogen and ammonia co-firing generation are discussed in detail, including the references used and the calculations.

2.5.1. Fuel cell generation

A fuel cell is a system that produces electricity and heat without combustion. In this study, it is assumed that natural gas steam reforming (NGSR) [31] is used to produce hydrogen for a molten carbonate fuel cell (MCFC) system. The 47 % efficiency of the molten carbonate fuel cell, the model DFC 3000 produced by FuelCell Energy Corporation, was used for the calculation. The Gyeonggi green energy park, one of the world's largest, built by Posco Energy in South Korea, a private energy producer, consists of a 2.8 MW FuelCell Energy DFC3000 power plant to provide continuous baseload power to the country's power-starved grid [32]. The density and heating value of the municipal natural gas used in the fuel cell are based on the lower heating value data (9800 kcal/Nm^3) provided by the Korea gas corporation [33]. The formula for calculating GHG emissions from combustion of natural gas is given in (1). Since the unburned fraction is typically small [34], it is assumed that the carbon in the fuel is completely oxidized.

$$\text{Combustion GHG emission} = \text{Energy use} \times \text{Emission factors} \tag{1}$$

Emission factors for stationary combustion in the energy industry are based on the IPCC 2006 guideline. The emission factors for natural gas are 56100 g CO_2/GJ, 1.0 g CH_4/GJ, and 0.1 g N_2O/GJ. Three representative greenhouse gases, CO_2, CH_4, and N_2O, were used to calculate the combined GHG emission value. The calculation was based on the global warming potential, which is equal to 1 (CO_2) , 28 (CH_4), and 265 (N_2O), respectively [35]. The upstream process of natural gas used in South Korea was calculated based on Choi and Song [36]. The GHG emissions from the recovery, processing, and liquefaction in natural gas-producing countries, as well as the import, regasification, and distribution process in South Korea, were obtained [36].

Table 1
Energy generation forecast details (as of 2030).

Energy source	9th Plan	10th Plan
Fossil fuels	53.6 %	43.0 %
Coal	30.3 %	20.1 %
Natural gas	23.3 %	22.9 %
Renewables	20.0 %	23.9 %
Solar PV	7.8 %	9.5 %
Wind	6.9 %	6.3 %
Biogas (including Waste)	2.6 %	3.4 %
Hydrogen and ammonia co-firing	–	2.1 %
Fuel cell	2.8 %	2.6 %
Nuclear	25.0 %	32.4 %
Hydro	1.4 %	0.7 %

2.5.2. Hydrogen and ammonia co-firing generation

A new addition to the 10th basic electricity supply and demand plan of South Korea, hydrogen and ammonia co-firing power generation is a method of generating hydrogen and ammonia, which are carbon-free fuels, using existing coal and LNG generators.

There are three types of hydrogen: green, blue, and grey hydrogen. Green hydrogen is produced by electrolyzing water using solar or wind energy, and grey hydrogen is produced by reforming fossil fuels. And blue hydrogen is clean hydrogen that captures and removes carbon dioxide from fossil fuel reforming. Currently, South Korea plans to import green hydrogen produced in Australia and other countries with abundant renewable energy resources using ammonia carriers. However, of the hydrogen planned to be produced domestically in 2030, 48.4 % of grey hydrogen and 38.7 % of blue hydrogen are produced by reforming fossil fuels, compared to 12.9 % of green hydrogen [37].

Table 2 summarizes the greenhouse gas emissions according to the green and grey hydrogen energy systems in the references. In terms of greenhouse gas emissions, green hydrogen ranges from 0.35 to 4.38 kg CO_2-eq per kg H_2 and grey hydrogen ranges from 8.50 to 11.24 kg CO_2-eq kg of H_2, with the minimum value of grey hydrogen being about twice as large as the maximum value of green hydrogen. Therefore, it is necessary to analyze green hydrogen and grey hydrogen separately, and the emissions of green hydrogen and grey hydrogen per kg H_2 are calculated as 2.37, and 9.87 as the median of the minimum and maximum ranges, referring to the reference in Table 2.

To calculate the total GHG emissions, the lower heating value of hydrogen of 120.2 MJ/kg [38] and the hydrogen power generation efficiency of 44 % were used [39].

$$\text{Efficiency } \eta = \frac{P_{out}}{P_{in}} \tag{2}$$

$$P_{in} = mass_{hydrogen} \times \text{LHV} \tag{3}$$

The value of P_{in} is calculated using formula (2), and the mass of hydrogen is calculated using formula (3).

$$\text{Total GHG emissions} = mass_{hydrogen} \times \text{GHG emissions per kg } H_2 \tag{4}$$

Finally, total greenhouse gas emissions are calculated using formula (4) as the mass of hydrogen and the GHG emissions per kilogram of green or grey hydrogen.

3. Results

3.1. Life cycle impact analysis of the TEC manufacturing process

The results of five countries, France, the United States, Australia, China, and South Korea, were analyzed to identify differences in environmental impact depending on the provider of input materials and the composition of input energy in the manufacturing process of TECs. There are five types of environmental impacts analyzed along with GHG emissions, and the meanings of the abbreviations are as follows. HCT stands for human carcinogenic toxicity, TE for terrestrial ecotoxicity, FE for freshwater ecotoxicity, MRS for mineral resource scarcity, and FRS for fossil resource scarcity. Fig. 3 shows the percentages of electricity generation from energy sources in the five countries in 2021.

France has a noticeably higher share of nuclear energy (68.3 %) than the other countries, followed by renewables at 11.7 % and hydroelectric at 11.5 %. France uses significantly less fossil fuel than other countries, comprising only 8.4 % of its electricity. All the other countries considered in this study use fossil fuels as the primary energy source for electricity generation. The United States generates its highest share of energy from fossil fuels at 60.9 %, followed by nuclear at 18.6 % and renewable at 14.2 %. Australia has the highest use of fossil fuels (73.3 %) among the five countries analyzed. In particular, as shown in Fig. 3, the share of coal is very high at 52.8 % of fossil fuels, followed by those of renewables, including solar PV (10.4 %) and wind (9.2 %), and does not use nuclear power.

The largest proportion of China's electricity is obtained from fossil fuels (66.3 %), followed by hydroelectric (15.6 %) and then renewables (13.4 %). China generates a significant amount of its electricity from coal, 63 % of the total. China also has the highest percentage of hydroelectricity generation among the five countries. Finally, South Korea has the highest proportion of nuclear (26.1 %)

Table 2
GHG emissions per kg H_2 by hydrogen type and energy system.

Hydrogen	Energy System	GHG emission [kg CO_2-eq/kg H_2]	Location	Reference
Green	Solar	1.58–2.95	USA	[40]
	Wind	0.35–4.38		
	Solar	3.1	USA	[41]
	Wind	0.86		
	Solar	1.06–1.57	Chile	[42]
	Solar	1.7–4.4	EU	[43]
Grey	Natural gas	11.24	USA	[41]
	Natural gas	8.43	Spain	[44]
	Natural gas	8.50	–	[45]

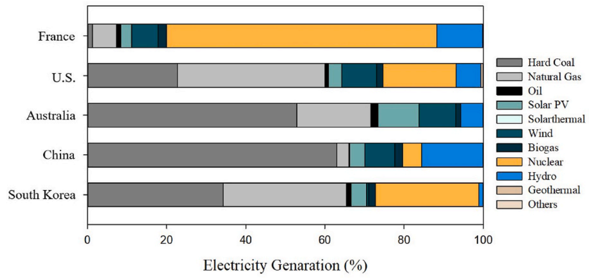

Fig. 3. Generation of electricity in France, the U.S., Australia, China, and South Korea.

and renewable (6.2 %) energy after fossil fuels (66.5 %). In South Korea, coal and natural gas account for 34.1 % and 31.1 % of electricity generation, respectively, and nuclear power is the second highest among the five countries after France.

The results of GHG emissions and environmental impact analysis of the TEC manufacturing process can be seen in Fig. 4. All materials and manufacturing processes are the same in the analysis. However, the results are compared considering that the only differences are the place of production of each material, the place of production of electricity, and the generation mixes of the electricity grid.

3.1.1. GHG emissions

In terms of GHG emissions, the largest amount of GHG emissions come from alumina plate manufacturing, followed by n-type, p-type, and copper tabs in all countries. Alumina plate manufacturing uses significantly more electricity than other processes because the sintering process consumes a large amount of electricity to heat the powder after ball milling to ensure tight adhesion. Therefore, the amount of GHG emissions is most influenced by energy sources. France, which has a significantly lower proportion of electricity generation from fossil fuels, has significantly lower GHG emissions than other countries. The GHG amounts of the largest emitter, China (3.99 kg CO_2.eq), and the smallest emitter, France (0.49 kg CO_2-eq), differ by a factor of 8.

The attribution analysis shows that except in France, the largest amounts of GHG are emitted by coal generation. France uses little coal for electricity generation, emitting most of its GHG from natural gas processing. Although South Korea and China use the same proportion of fossil fuels, as well as those of other materials (average values are considered due to lack of country-specific data), they

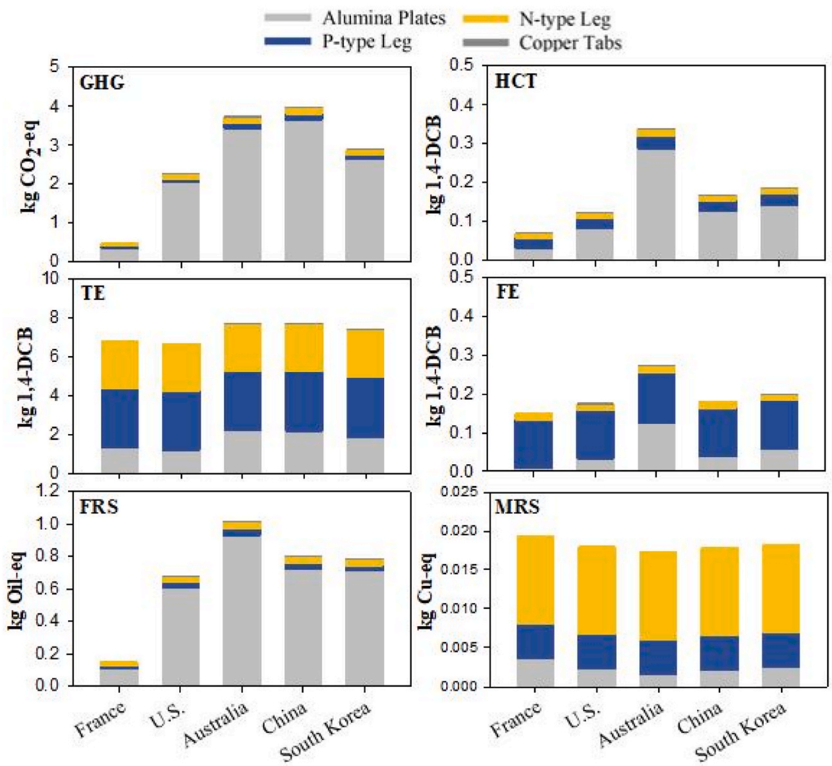

Fig. 4. GHG emissions and environmental impact by country according to TEC component materials.

show a difference in GHG emissions. The reason for the difference is that China uses 63.0 % coal, while South Korea uses 34.1 % coal and 31.1 % natural gas. The GHG emissions from coal are greater than those of oil and natural gas [46]. The present contribution analysis also shows that coal generation has a greater impact on GHG emissions. China and Australia, which rank first and second in terms of GHG emissions, use a high proportion of coal among fossil fuels.

3.1.2. Human carcinogenic toxicity

HCT does not differ significantly between countries in the production process of p-type and n-type materials, but the difference in impact between countries in the production of alumina plates is significant. In particular, Australia has a particularly large HCT impact compared to other countries (twice that of China and about 5 times that of France). According to the contribution analysis of HCT, France's electricity generation using nuclear power is the largest factor in the production of alumina plates, while electricity generation using hard coal is the most influential factor in the USA, Australia, China, and South Korea. Coal-fired power plants emit large numbers of particles into the air as aerosols. The continuous inhalation of fine coal particles and byproducts can adversely affect human health such as the immune system, heart, DNA, brain, and reproductive system [47].

The contribution analysis for Australia, which has a high HCT, shows that coal has the largest impact on human health, followed by aluminum oxide and solar PV. Australia is dominated by solar PV among renewables, with related studies showing that a higher share of solar PV in power generation results in greater HCT impact [48,49]. As in Fig. 3, Australia's share of solar PV in 2021 was 10.4 %, the highest among the tested nations. Rashedi and Khanam [50] reported that mono-silicon panels in solar PV emit NH_3, NO_x, and SO_2 into the air, affecting particulate matter formation. The contribution analysis shows that the main contribution from p-type legs is due to the production of antimony. According to Fu et al. [51], symptoms of chronic antimony poisoning have been observed in residents living in mining areas in China, and various toxicological experiments have proven that antimony primarily causes cell damage, tissue degeneration, and protein denaturation, potentially leading to cancer. In this study, the global average value of antimony was used, producing equal impacts among countries, but the value is subject to uncertainty based on the scarcity of the resource.

3.1.3. Terrestrial ecotoxicity

For TE, only alumina plates show differences between countries, while p- and n-type legs show minimal differences between countries. Analysis of the p-type leg materials shows that tellurium production overwhelmingly affects TE. In the n-type leg process, the treatment of wastewater generated in the ingot polishing and leg dicing stages greatly affects TE. The contribution analysis shows that the impact of wind generation is significant in all countries. The wastewater and oil generated at the construction site of the wind energy farm can affect terrestrial ecotoxicity as they can seep into the underground soil and cause serious environmental problems [52].

In France, the contribution of nuclear generation to TE is large, with high concentrations of copper, vanadium, silver, zinc, and chromium in the emissions. Among them, copper is coated around the containers used to store nuclear fuel waste, and it corrodes over time [53]. The relatively high salinity of groundwater can promote the general dissolution of copper [54]. Since tellurium production data are not available in Ecoinvent v3.8 by country (only as a global average), the TE values due to n- and p-type legs are the same for all countries analyzed. As such, they can be subject to uncertainty. The production of tellurium and antimony has a significant impact on the leg production process. Industrial and mining activities have contributed large amounts of antimony (Sb) to nearby soils, adversely affecting soil quality and contaminating associated ecosystems [55].

3.1.4. Freshwater ecotoxicity

FE has a large impact due to the production of p-type legs. There is no difference between the country-specific impacts from the production of p-type and n-type legs because the global average is used due to the lack of country-specific data. However, in practice, the reserves of antimony resources show significant regional variations, so the results are subject to high uncertainty. In the case of p-type legs, the main impact is due to the production of antimony, the material that makes up the module. Mining and industrial activities result in the continuous migration of antimony into the aquatic environment, affecting the Earth's biogeochemical cycles [51]. China, one of the world's leading exporters of antimony, is experiencing water pollution from intensive antimony mining. There are country differences due to the manufacture of alumina plates. The trend is the same as for HCT. Solar cells have adverse impacts on FE as well as HCT, and LCA analyses of solar cells have shown that the main impacts are freshwater ecotoxicity and human toxicity [48, 49].

3.1.5. Fossil resource scarcity

FRS increases as the percentage of fossil fuels used increases. That is, a higher proportion of energy generated using fossil fuels yields a greater likelihood of fossil resource depletion. The contribution analysis shows that the largest impacts are from the use of hard coal and natural gas in all countries.

3.1.6. Mineral resource scarcity

MRS is affected by n- and p-type legs and alumina plates, with no significant differences between countries. N-type has the largest impact on MRS because bismuth production, treatment of wastewater, and tellurium production are affected. The P-type legs affect bismuth production, followed by tellurium and antimony production.

3.1.7. GHG reduction methods of hotspots

According to Section 3.1.1, the hotspot of GHG emissions is alumina plates. As an alternative, materials based on thermally conductive and electrically insulating polymers [56,57] can be considered. Recently, polymers have been explored for their potential to balance high thermal conductivity with high electrical insulating properties [58]. Therefore, the consideration of substitutes is limited, and the efficiency of the only input, electricity, is considered.

As shown in Table S3 in Appendix A. Supplementary Information, the process of manufacturing alumina plates consists of three steps. Based on South Korea, 2.4 % of the total emissions of alumina plates are from ball milling, 87.3 % from the sintering process, and 10.3 % from polishing. The contribution of the sintering process is significant. The only input material for the sintering process in Table S3 is electricity, except for the powder after ball milling in the previous step. Therefore, reducing electricity consumption to improve the sintering process is the most effective way to reduce greenhouse gas emissions.

To reduce electricity consumption in the sintering process, it is important to control the parameters to ensure complete combustion and stable operation to minimize energy consumption [59]. Zhu et al. [60] found that high temperature exhaust gas recirculation sintering dramatically improved sintering performance, which contributed to reducing CO_2 emissions. Heidary et al. [61] found that in various ceramic sintering processes, fast heating rate results in much less heat dissipation, which can reduce energy consumption by 20 times, and adding sintering aids can be the simplest way to reduce energy consumption, which can reduce energy consumption by 1.4 times.

Fast heating rate, sintering aids, energy recovery system, etc. can be expected to reduce energy consumption and reduce GHG emissions through process improvement of the sintering process.

3.2. GHG emissions in the TEC manufacturing process according to electricity grid mix change

The carbon footprint of a manufacturing process is affected by many variables, including material substitution, process improvements, and productivity gains due to energy efficiency improvements. The results obtained in Section 3.1 above show that the highest GHG emissions are generated during alumina plate manufacturing. Therefore, we focused our attention on electricity, which is used in all processes and sensitively affects GHG emissions.

In Fig. 5, the 2021 grid mix is the same as the value obtained from the country-specific GHG emissions in Section 3.1, calculated using the grid mix in 2021. Plan A is the projected grid mix included in South Korea's 9th national basic electricity supply and demand plan, while Plan B is the 10th national basic electricity supply and demand plan. In addition, the value of GHG generated to produce TECs was analyzed assuming use of Plan A.

According to Fig. 5, Plan B will result in a 23.5%p decrease in fossil fuels, a 6.3%p increase in nuclear, and a 17.7%p increase in renewables in 2030 compared to the 2021 generation. Plan B also creates a new share of 2.1 % for hydrogen and ammonia co-firing generation. In Plan A, the fuel cell share was 2.75 %, but in Plan B, the share of the fuel cell (2.57 %) and hydrogen and ammonia co-firing (2.1 %) increases to 4.67 %.

Calculations were made using electricity generation data analyzable with Ecoinvent v3.8 data and the GHG analysis methods described in 2.5.1 and 2.5.2. As shown in Fig. 6, compared to producing TECs using the 2021 grid mix, the GHG emissions for Plan A were reduced by 11.5%p, while those for Plan B were reduced by 32.6%p for green hydrogen and 30.9%p for grey hydrogen. Although hydrogen accounts for only 2.1 % of the power generation, using green hydrogen can reduce overall GHG emissions by an additional 1.7 % compared to using grey hydrogen. And even with the larger emissions of grey hydrogen, Plan B still has lower emissions compared to Plan A.

Comparing the 2021 grid mix to Plan A, renewables increase significantly by 13.8%p and fossil fuels decrease by 12.9%p. Comparing Plan A to Plan B, renewables increase by 3.9%p and fossil fuels decrease by 10.6%p. The change from the 2021 grid mix to Plan A results in a larger decrease in the share of fossil fuels and an increase in the share of renewables than the change from Plan A to Plan B. However, there is a larger reduction in GHG emissions when changing from Plan A to Plan B. The reason for this can be determined by comparing the 9th and 10th electricity supply and demand plans.

In Table 1, coal and nuclear are the most important categories. In Section 3.1, we showed that France has significantly lower GHG emissions with a high share of nuclear energy use. The large increase in the value of nuclear from Plan A to Plan B had a significant impact on the overall reduction in GHG emissions. The decrease in coal also contributed to the significant decrease in GHG emissions.

Fig. 7 shows the results of the attribution analysis. The major contributors in 2021, Plan A, and Plan B are shown, with the minor contributors as others. In 2021, GHG emissions were produced by electricity generation using hard coal (66.4 %), natural gas (22.8 %), and oil (2.1 %). After fossil fuels, tellurium production contributed 0.9 % and biogas 0.8 %. The contribution of fossil fuels to electricity generation decreases with Plan B, and the contribution of renewables increases. In Plan B with green hydrogen, the order of contribution is hard coal (57.8 %), natural gas (25.0 %), fuel cell (3.2 %), biogas (2.2 %), tellurium production (1.3 %), and solar PV (1.1 %).

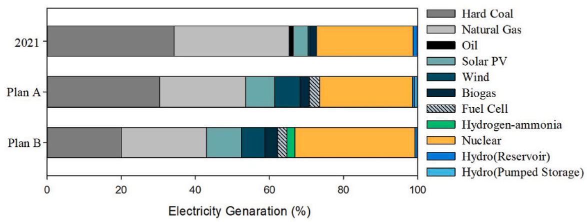

Fig. 5. Percentage of electricity generation by energy type in South Korea's 2021 and 9th and 10th basic electricity supply and demand plans.

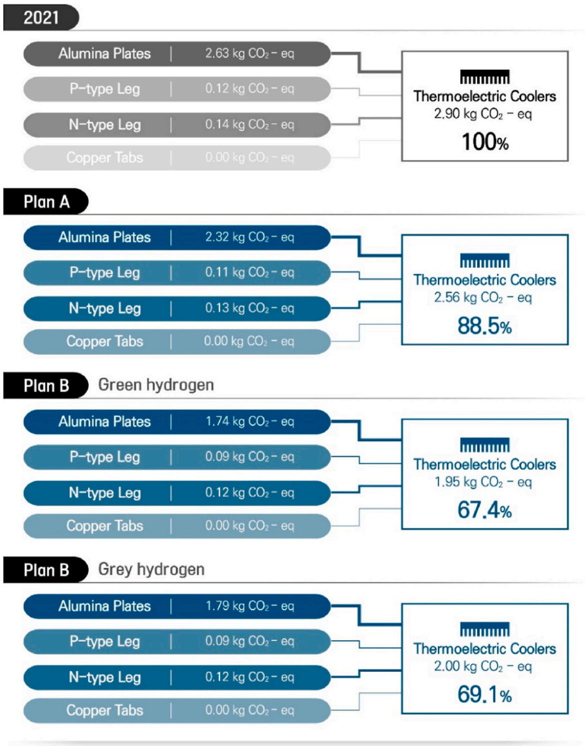

Fig. 6. GHG emissions and potential reductions in the TEC manufacturing process.

The share of GHG emissions from coal generation are expected to decrease, while the share of emissions from natural gas generation are projected to increase. The share of emissions from renewables is also expected to increase slightly by energy type. The large contribution from fuel cells, which are categorized as renewable energy, is significant because in South Korea, fuel cells are powered by natural gas.

Fig. 8. Shows the sources of increase and decrease that contribute to the decrease in GHG emissions from 2021 to 2030. (a) is the result of applying Plan A, and (b) is the result of applying Plan B with green hydrogen. And (c) is the result of applying grey hydrogen in Plan B. Plan B shows a significant decrease in emissions from hard coal compared to 2021, and a significant decrease compared to Plan A. Emissions from fuel cell and hydrogen power generation in (b) and (c) have increased, but this is because they are new additions to the 2030 plan that do not exist in 2021.

4. Discussion

In this study, the calculations assume that each material is produced in the countries where the TECs are assembled or purchased from the market, but the data scarcity of some materials in the Ecoinvent database can be a limiting factor. For example, aluminum oxide production data used in the manufacture of alumina plates are available in the EU27, China, and South America. Also, ethanol production data are only categorized into the European region (RER) and the rest of the world (RoW). Therefore, in regions where

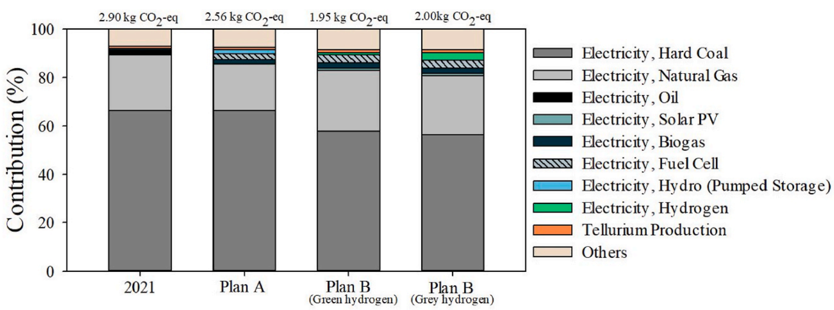

Fig. 7. Attribution analysis of greenhouse gas emissions.

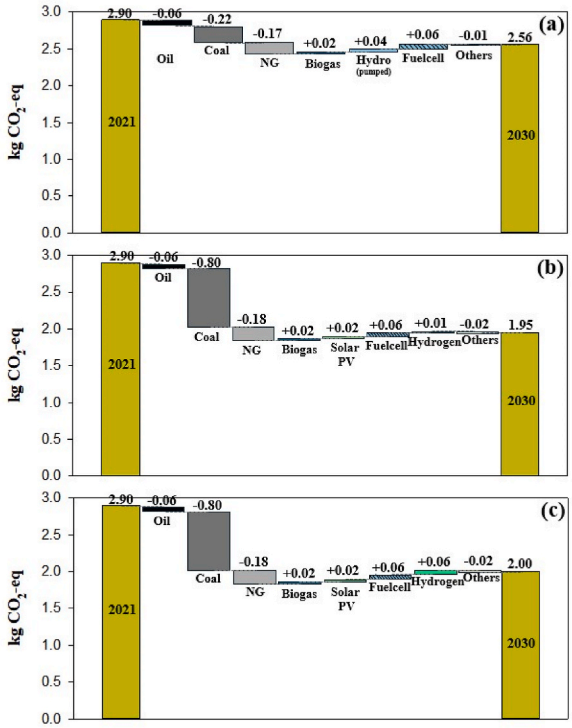

Fig. 8. (a) is the change of GHG emissions under Plan A compared to 2021, (b) is the change of GHG emissions with green hydrogen under Plan B compared to 2021, (c) is the change of GHG emissions with grey hydrogen under Plan B compared to 2021.

there are gaps in data for a particular material and only average values exist, the results of some analyses may be closer to the global average rather than the specific region.

Additionally, some analyses use secondary data rather than primary data, so results are subject to uncertainty due to the lack of precisely measured data and lack of complete information by region. The life-cycle impact of nuclear power use cannot be fully assessed without considering the final disposal of high-level nuclear wastes, which, at present, have never been realized commercially. The results related to nuclear power in this study must rather be considered preliminary.

This study shows that additional carbon reductions are possible depending on the type of eco-friendly power generation in production activities for carbon neutrality. As a heat sink and heat source, alumina plates are a widely used material due to their low cost, light weight, and high thermal conductivity. However, this study found that alumina plates had the greatest impact on greenhouse gas emissions, so further analysis of other low-emission materials was considered. However, unlike thermoelectric generators, the ideal heat sink material for thermoelectric coolers must have high thermal conductivity as well as high electrical insulation. This limited the consideration of alternatives, and the development of alternatives that could be environmental benefits is essential.

In addition, process improvements such as reducing energy consumption in the manufacturing process require improving the efficiency of the sintering process, which is the most energy-consuming and GHG gas emitting process. In the sintering process, only electricity is required to adhere to the ball-milled powder, so improving the efficiency of the sintering process can lead to a reduction in energy consumption. During the sintering process, methods such as fast heating rate, sintering aids, energy recovery system, etc. can be applied.

5. Conclusion

The main objective of this study was to analyze the GHG emissions and environmental impacts of TEC manufacturing by country. The results of the study are expected to serve as a reference for exploring hotspots of GHG based on differences in raw materials and electricity generation of countries, even when TECs, which are eco-friendly cooling devices, are manufactured by the same process using the same materials. The ultimate goal is to analyze ways to help reduce GHG emissions. To achieve the objective, France, the United States, Australia, China, and South Korea were selected, and the impact of GHG emissions on human carcinogenic toxicity (HCT), terrestrial ecotoxicity (TE), freshwater ecotoxicity (FE), mineral resource scarcity (MRS), and fossil resource scarcity (FRS) were calculated through LCIA assuming that TECs are manufactured in each country.

The manufacture of TECs requires electricity, and GHG emissions depend more on the source of electricity than on the origin of the materials and the substances used in the process. In particular, the largest source of GHG emissions was the alumina plate manufacturing process because the sintering of alumina plates consumes a large amount of electricity. France, which uses a large share of nuclear power, emits the smallest amount of GHG, while China and Australia, which use a large share of coal, emit the most. For HCT, the alumina plate manufacturing process is a hot spot, and the impact is especially dominant in Australia due to the high share of solar PV in electricity generation.

The production of the antimony that makes up the p-type leg affects the HCT. We found that tellurium production in the p-type leg process has a significant impact on terrestrial ecotoxicity, while treatment of wastewater from ingot polishing and leg dicing of the n-type leg process has a significant impact on TE. Among renewables, wind generation also has a large impact on TE. Freshwater ecotoxicity, like HCT, is also affected by solar PV and varies by country when manufacturing alumina plates, and the impact is high in Australia, where the share of solar PV is high. However, the reason for the high impact of p-type is due to antimony production. Mineral resource scarcity does not show a significant difference between counties, but the impact of n-type is higher due to the use of telluride, which is a rare element. In addition, the larger is the proportion of fossil fuels used, the higher is the Fossil Resource Scarcity. When fewer fossil fuels are used (i.e., greener electricity), there is a higher impact on GHG, HCT, FE, and FRS. Also, TE, FE, and MRS are affected by the mineral resources used to manufacture the legs.

The environmental impact of the use and transportation process is further analyzed in the Supplementary Information, and the environmental impact of the manufacturing process is negligible compared to the use process. TECs use a significant amount of electricity during their life cycle to convert electrical energy into heat energy, so the more eco-friendly electricity grid mix, the more effective the use of TECs can be. Also, compared to production, transportation emissions are very small. If it is assumed that a country with low manufacturing emissions exports to a country with high emissions, then all combinations do not exceed the emissions of the country with high emissions. From a life-cycle perspective, emissions during use are overwhelming, so it is the best environmental interest to use TECs with eco-friendly power sources.

Additionally, we analyzed the change in GHG emissions when TECs are manufactured using the grid mix electricity considering the South Korean electricity supply plan for 2030. The original plan (Plan A) was to reduce fossil fuels by 12.9%p from 2021 to 2030, reduce nuclear by 1.1%p, and increase renewables by 13.8%p. However, the updated plan (Plan B) is to reduce fossil fuels by 23.5%p from 2021 to 2030, increase nuclear by 6.3%p, and increase renewables by 17.7%p. The national electricity plan was revised to significantly reduce fossil fuels and increase the use of nuclear and renewable energy compared to existing plans.

Manufacturing TECs using electricity in the Plan A would total 2.56 kg CO_2-eq of GHG, an 11.5%p reduction in emissions compared to 2.90 kg CO_2-eq in 2021. In the Plan B, the reduction differs between green and grey hydrogen, with a 32.6%p reduction to 1.95 kg CO_2-eq using green hydrogen and a 30.9%p reduction to 2.00 kg CO_2-eq using grey hydrogen. Despite the small amount of hydrogen energy used (2.1 %), the difference in emissions between green and grey hydrogen is 1.7%p. By using green hydrogen, 1.7%p more GHGs (0.05 kg CO_2-eq) can be reduced than if grey hydrogen is used.

On the other hand, significant uncertainty exists on how hydrogen energy will be integrated into the energy sector. For instance, hydrogen compression for storage and transportation can be very energy intensive. It is suggested that hydrogen storage pressures typically range between 20 and 25 MPa [62]. The energy required for hydrogen compression for the suggested pressure range can roughly be anywhere between 5 % and 15 % of the HHV of hydrogen itself [63]. Accordingly, the environmental impact from the use of hydrogen will also be proportionally increased. Numbers can vary as the situation in the future unfolds, and unaccounted effects may introduce sizeable variations.

The results of this study are projections based on the basic energy supply and demand planning and assume that the grid mix projections for 2030 are realized. The Korean government revises its electricity supply and demand plan every two years for the next 15 years, and if carbon-free energy is further expanded, greater GHG reductions can be expected from the production of TECs.

CRediT authorship contribution statement

Hyo Young Kim: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Jeong Eun Kim:** Visualization, Validation, Investigation. **Daehyun Wee:** Writing – review & editing, Supervision, Project administration, Funding acquisition.

Data availability statement

The data associated with the study has not been deposited into a publicly available repository, but the data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was primarily supported by a National Research Foundation of Korea (NRF) grant funded by the Korean government [NRF-2022R1F1A1060204] and by the Korea Ministry of Environment (MOE) Graduate School as part of the Integrated Pollution Prevention and Control Project. This work was also supported by the National Research Foundation of Korea's Brain Korea 21 FOUR Program "REFINANCE: Research and Education team For an INtegrated Approach on Newly emerging Contaminants in multiple

Environmental media" at the Department of Environmental Science & Engineering, Ewha Womans University.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e41527.

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