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Review

Sustainability and Circular Economy Perspectives of Materials for Thermoelectric Modules

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Abstract: The growing demand for energy and the environmental problems derived from this problem are arousing interest throughout the world in the development of clean and efficient alternative energy sources, which involve ecological processes and materials. The materials used in the processes associated with thermoelectric generation technology will provide solutions to this situation. Materials related to energy make it possible to generate energy from waste heat residues, which are derived from various industrial processes in which significant fractions of residual energy are deposited into the environment. However, despite the fact that thermoelectric technology represents some relative advantages in relation to other energy generation processes, it in turn faces some technical limitations such as its low efficiency with respect to the high costs that its implementation demands today, and this has been the subject of intense research in recent years. On the other hand, the sustainability of the processes when analyzed from a circular economy perspective must be taken into account for the implementation of this technology, particularly when considering its large-scale implementation. In this article, a systematic search focused on the sustainability of thermoelectric modules is carried out as a step towards a circular economy model. The review aims to examine recent developments and trends in the development of thermoelectric systems in order to promote initiatives in favor of the environment. The aim of this study is to present a current overview, including trends and limitations, in research related to thermoelectric materials. As a result of this analysis, it was found that aspects related to costs and initiatives related to circular economy models have been little explored, which represents not only an opportunity for the development of new approaches in the conception of thermoelectric systems, but also for the conception of optimized designs that address the current limitations of this technology.

Keywords: thermoelectric modules; cost-efficiency ratio; sustainability; life cycle; circular economy

1. Introduction

The progressive deterioration of the global ecosystem, as well as the decline of fossil fuel reserves, are some of the consequences of the current global energy problem. This is primarily due to the excessive use of conventional energy sources. In recent years, this has led to a great developments in the use of alternative energy sources that are more environmentally friendly and that in turn have renewable characteristics, which would guarantee that are exhausted in the short or medium term. Among these alternative sources of energy, the sources based on direct solar radiation stand out [1], as well as the sources of use of indirect solar energy, such as hydraulic [2], wind [3], and tidal sources [4]. Other sources are based on biomass [5] and on the use of energy from waves [6]. Likewise, there is potential for geothermal energy [7] and nuclear energy [8]. However, a significant energy source is based on the use of energy residues from industrial processes, and recently there has been a notable boom in the use of thermoelectric systems, which are based on the use of energy waste in the form of heat that is regularly discarded into the atmosphere. In recent years there have been various advances in technologies that have made it possible to make better use of this energy waste [9], since it is not enough to achieve high efficiency, but it is also important to consider the useful life of thermoelectric systems, especially for systems that operate in low temperature ranges. This enables both long-term reliability and the recyclability of the system's components, in order to certify as sustainable development and conserve natural resources for the benefit of present and future generations. However, based on a recent literature review, it has been found that recent developments in these areas are not paying enough attention to the use of sustainable processes and materials, which are fundamental aspects in the development of current technology. This is why the present study focuses on reviewing the main thermoelectric materials used today, and also analyzes the types of modules used and the main applications of these thermoelectric systems. Therefore, the main objective of this analysis is to establish which of the current technologies are designed in a way that favors recycling processes, and therefore, to determine the degree of sustainability of these technologies. In this sense, the present review seeks to characterize, using recent publications from the last decade, which technologies are being implemented, and to what degree they are contributing to a more sustainable production system.

These developments result from a growing demand from the community to produce cleaner manufacturing processes and materials in sectors such as manufacturing, construction and transport that represent around 75% of the energy consumed throughout the world [10].

Recently, considerable attention has been given to the use of thermoelectric (TE) materials as a complement to enhance the efficiency of renewable energy systems based on solar and biomass energy sources [1]. In addition, other applications in industrial sectors are making progress with these systems to become more efficient and reduce the impact on the environment, which translates to a significant recovery of wasted heat normally disposed into the environment. According to various reports, the energy consumption levels of 2019 will significantly increase by 2040 in sectors such as transport and construction, and the use of renewable energy and natural gas use will grow faster than the use of coal and oil [11].

These increases in the energy consumption of the different sectors and in renewable energies result in a promising outlook for TE materials. Studies have reported that about 60% of the energy consumed worldwide is dissipated as waste heat in the transport sector, with the manufacturing and construction sectors also making large contributions of residual heat [12–14]. The recovery of this residual heat, even partially, would represent a significant saving in world energy consumption, as well as significant reductions in greenhouse gas emissions [15], and would be a major contribution to a better environment and to a better circular economy model [16].

Residual heat can be classified into three categories according to the temperatures of the heat sources [11]. In this classification, low temperature ranges are temperatures below 100 °C; average temperatures when the temperatures are between 100 °C and 300 °C; and high temperatures when they are above 300 °C, such as in calcining, forming, heat treating, thermal oxidation and metal reheating processes [11]. Considering these ranges, the operating temperatures of commercial thermoelectric generation modules (TEG) show a great potential for the implementation of heat recovery systems using TEG systems in many different processes [17–20]. It is known that TE materials do not allow 100% heat loss recovery due to their current low efficiency. Furthermore, the high cost of the elements used to construct commercial TEG modules is another factor preventing their widespread implementation in many systems and economies [21–23]. Currently, one of the main goals is to improve the performance of TE materials so that they are competitive and efficient, which is can be obtained with a high Seebeck coefficient and electrical conductivity, while the thermal conductivity has to be low. Factors such as profitability and complexity of processing have limited this efficiency increase [24].

In the recent literature, there is a wide variety of studies that examine the development of sustainable alternatives and promote the development of alternative generation systems to reduce dependence on traditional fossil fuels and to reduce the current rates of emissions. Furthermore, numerous works aim to improve environmental conditions based on the development of clean generation systems. Clearly, the generation alternatives based on thermoelectric systems are viable and reliable; however, the present study goes beyond ensuring the implementation of cleaner generation alternatives to demonstrate that those same proposed solutions are in themselves sustainable. Therefore, in this analysis we intend to present some of the work that is being carried out today to contribute to the construction of a better future for future generations.

This work presents a systematic search on TE materials focused on the sustainability and profitability of TEG modules, and is divided in the following topics: materials, TEG modules, costs, efficiency, comparisons with other types of energy collection or cooling systems, and modeling of properties and construction. Finally, the recycling and end disposal of this type of TE material is reviewed (see Figure 1), with a particular focus on the circular economy for thermoelectric modules.

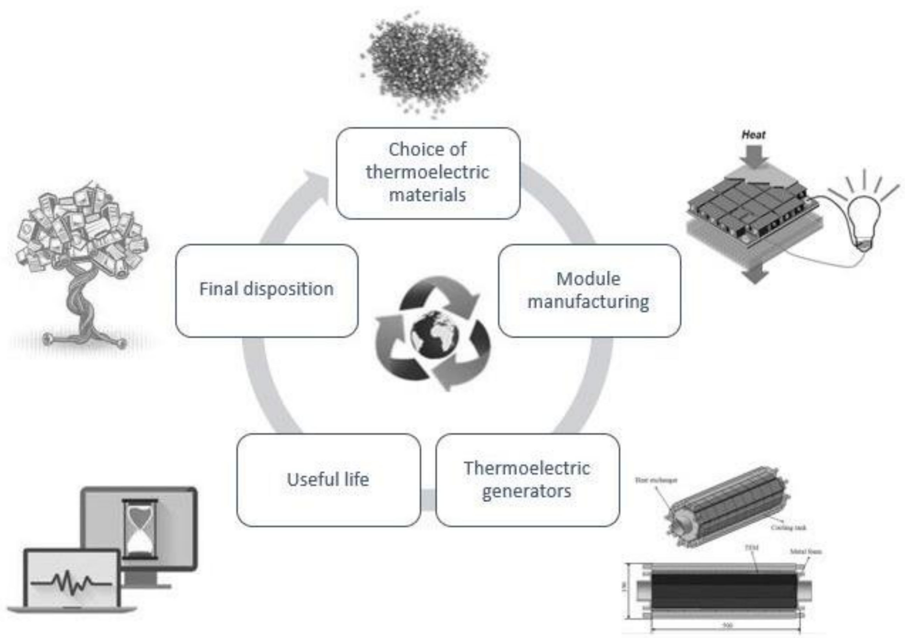

Figure 1. Themes of the article related to the circular economy of thermoelectric modules.

2. Methodology

The methodology proposed for this study was based on a comprehensive systematic review in which the most relevant publications related to the development of thermoelectric generation systems were taken into consideration. Based on this preliminary investigation, a relationship was established between the most relevant sources in order to establish comparisons between them in order to critically analyze the information collected in relation to the sustainability of these systems and to subsequently classify the documentation collected.

In this review, the most relevant publications were selected by the authors and summarized to identify their most notable aspects, and tables and graphs were developed, seeking to synthesize the collected evidence in the best way.

Based on the proposed methodology, a systematic search was initially made in the Scopus database using the keywords "thermoelectric AND modules" with the AND logical connector. In this initial search, a total of 4676 results were obtained. Then, two filters were used to refine the search. In the first filter, only selected review type documents were selected, and in the second filter, the publication date was restricted to the range from 2015 to 2020, resulting in a final total of 61 reviews, as shown in Table 1. These filters were based on reviews by Alam and Ramakrishna [25] and Zheng et al. [26], who provided an overview of research in the area of thermoelectric materials incorporating semiconductor-doped materials and highlighted efforts to date to increase thermoelectric efficiency from different perspectives and the potential for economic and environmental benefits through the improvement of thermoelectric systems.

Table 1. Summary of the initial systematic search for the subject of the review.

Database	Number of Articles Found		
	Unfiltered	Filter 1	Filter 2
Scopus	4676	81	61

Figure 2 presents the classification according to content of the works selected in the initial bibliographic review using four categories: TE materials, TEG modules, applications and sustainability. The classification was applied by grouping the articles into different themes and subtopics after reading the titles, abstract and keywords. From this selection, it was found that 10 articles were not relevant to the topic of TE materials and, out of the 51 relevant articles, some were classified in two of the categories.

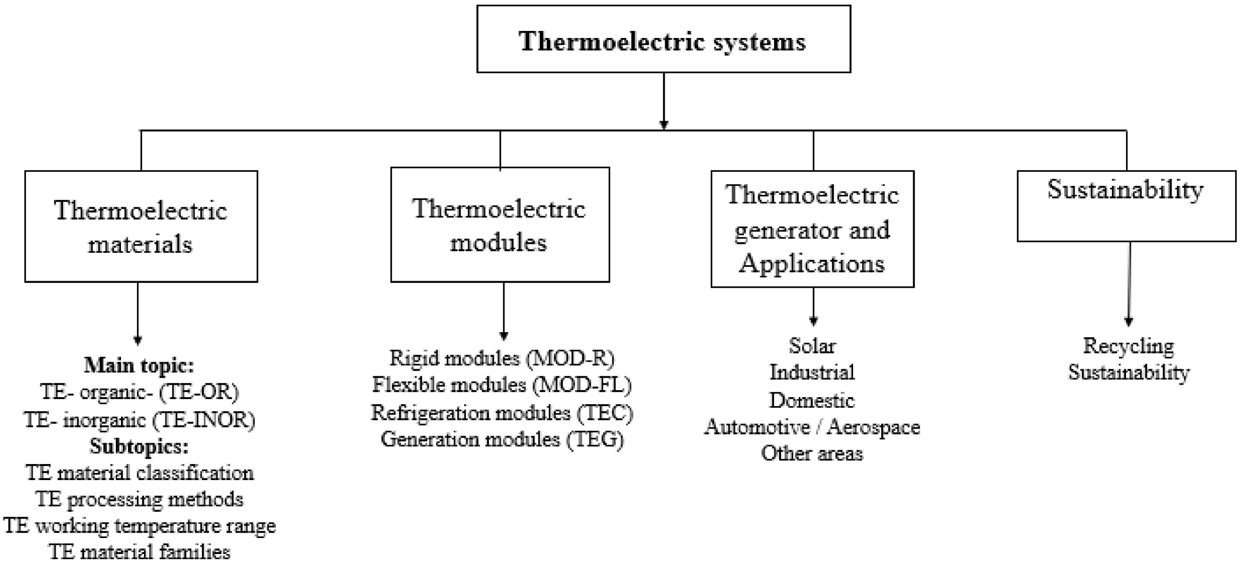

Figure 2. Topics identified in the articles of the initial systematic search.

In this initial systematic search, a knowledge gap was found in the sustainability category in which themes of profitability, cost/efficiency ratio and recycling, among others, were developed. For this reason, it was decided to focus on this area in the present analysis.

Regarding the development of this review, six basic themes related to the sustainability of these types of materials were considered: efficiency, cost, recycling, sustainability, life cycle and profitability. For this, after the removal of duplication in articles, four filters were applied. The first filter was the year of publication in which only the articles published from 2010 to 2020 were considered. The second filter was the type of document; in this case only the research articles were considered. The third filter was based on reading the title and abstract, and finally, a thorough reading of the selected articles was carried out, which resulted in a total of 36 relevant articles, from which the analysis presented below was developed. Table 2 shows the search strings used and the results of their systematic debugging. Likewise, Figure 3 shows the results of the search carried out, where

Figure 3a shows the data obtained by percentage for each search string. Figure 3b shows a classification of the publications analyzed by year, revealing the growing interest in TEG. In Table 3, the publications and topics covered by each of these strings are presented according to the topics to be addressed in this study.

Table 2. Search strings and filters used in the systematic search.

Search Strings	Number of Articles Found				
	Unfiltered	Filter 1	Filter 2	Filter 3	Filter 4
"Thermoelectric" AND "Modules" AND "Recycling"	37	23	22	6	5
"Thermoelectric" AND " Modules" AND "Cost" AND "Efficiency"	211	115	105	25	12
"Thermoelectric generators" AND "Cost" AND "Efficiency"	298	249	143	9	5
"Thermoelectric generators" AND "Sustainability"	25	24	13	5	4
"Thermoelectric" AND "Modules" AND "Life cycle"	25	19	11	3	2
"Thermoelectric modules" AND "demand"	43	38	24	5	1
"Thermoelectric" AND "Materials" AND "Modules" AND "Cost" AND "efficiency"	92	77	45	22	7

Filter 1: year of publication of the article from 2010 to 2020, Filter 2: type of document "article type", Filter 3: reading of title and summary, Filter 4: reading the article.

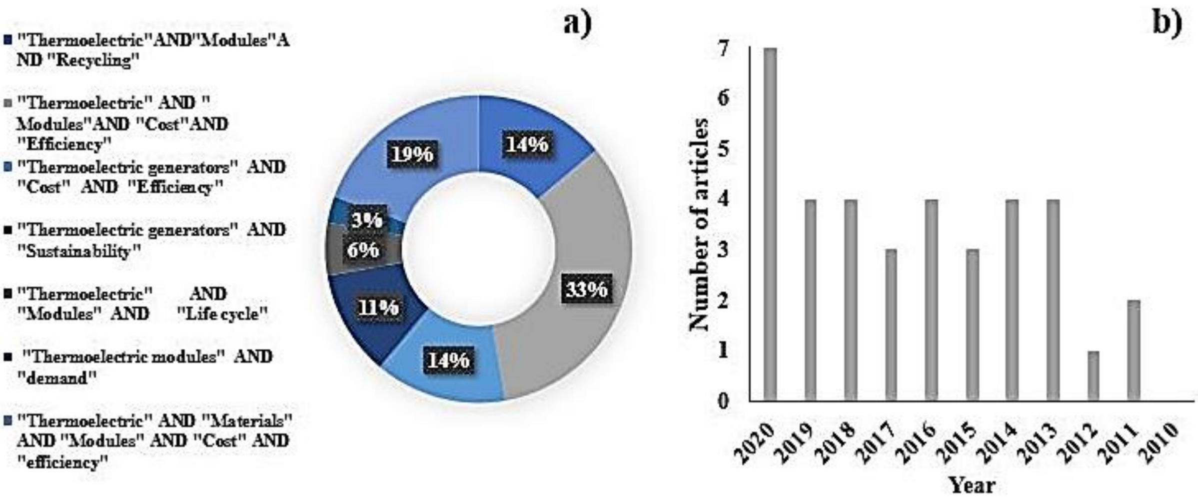

Figure 3. Systematic search results for (**a**) number of articles found per search string, and (**b**) number of articles per year.

Table 3. Articles found in the systematic search with their classification by subject matter.

Ref.	Year	MAT	MOD	TEG	COST	EFF	MDL	RE
[27]	2020	X	X			X		
[28]	2020		X			X	X	
[22]	2020		X		X	X		
[21]	2020		X	X	X	X	X	
[16]	2020			X	X	X	X	

Table 3. *Cont.*

Ref.	Year	MAT	MOD	TEG	COST	EFF	MDL	RE
[29]	2020		X	X		X		
[18]	2020		X					X
[30]	2019	X				X	X	
[31]	2019		X		X	X	X	
[32]	2019		X			X	X	
[23]	2019		X					
[33]	2018	X			X	X		
[34]	2018		X			X	X	
[35]	2018		X			X	X	
[36]	2018	X	X		X	X	X	
[37]	2017		X					X
[38]	2017		X					X
[39]	2017	X	X			X		
[40]	2016	X	X		X	X	X	
[41]	2016		X			X	X	
[42]	2016			X	X		X	
[43]	2016			X	X	X	X	
[44]	2015	X	X			X		
[45]	2015		X	X		X	X	
[46]	2015	X	X			X	X	
[47]	2014		X	X		X	X	
[48]	2014		X			X		
[49]	2014		X				X	
[50]	2014		X					X
[51]	2013		X		X	X		
[52]	2013	X	X	X		X		
[53]	2013		X					X
[54]	2013			X	X	X	X	
[55]	2012		X		X		X	
[56]	2011	X	X			X		
[57]	2011					X		

MAT: TE materials, MOD: thermoelectric modules, TEG: thermoelectric generators, EFF: efficiency, MDL: modeling, and RE: recycling.

3. Results and Discussion

3.1. Thermoelectric Materials

It is estimated that between 50% and 80% of the studies related to thermoelectric generation systems (TEG) focus on the TE materials themselves. Therefore, the development of materials that have a low cost and high efficiency is fundamental to achieving commercial profitability in these systems [17,18].

In the systematic search carried out, 10 articles were found in which the TE material was studied in depth in order to increase its efficiency, decrease costs or develop viable materials in both environmental and sustainability aspects by analyzing factors such as the material's abundance and its degree of toxicity among others. Table 4 presents a classification of these items according to the cost, efficiency and sustainability of the TEG modules. In this table, the cost column expresses the materials cost reduction with a minus sign (−), and the materials cost increase with a plus sign (+). In this aspect, the elements that compose in some cases the type of processing are taken into account. Note that in most of the articles reviewed, the cost of the material was not stated explicitly. For efficiency, the minus sign refers to materials with low efficiencies (ZT < 1) and the plus sign (+) for materials with significant efficiencies (ZT > 1). Finally, the sustainability of the materials is assessed using measures such as the availability, toxicity and useful life; therefore, a minus sign indicates that the material is unsustainable and a plus sign that it is sustainable.

Table 4. Classification of the articles found according to the results obtained in the development of thermoelectric materials.

Ref.	Cost	Efficiency	Sustainability	Year
[27]	−	+	−	2020
[30]	−	−	+	2019
[33]	−	+	−	2018
[36]	+	+	+	2018
[39]	−	−	+	2017
[40]	−	+	+	2016
[44]	−	+	−	2015
[46]	−	−	+	2015
[52]	−	+	+	2013
[56]	−	+	+	2011

Cost: −, Low cost; +, high cost. Efficiency: +, high efficiency; −, low efficiency. Environmental aspects and sustainability: −, unsustainable; +, more sustainable.

Among the articles shown in Table 3, three case studies were found [19,58,59] that examined cost of TE materials, including their cost effectiveness and the processing technique used to develop them.

Among the cost-effective materials, the use of oxides as raw materials stands out. Hung et al. [43] studied different oxides (Na_2CoO_4, $Ca_3Co_4O_9$, ZnO, $SrTiO_3$ and $CaMnO_3$) in order to decrease costs in the manufacture of TEG modules since the cost of manufacturing the oxides is approximately 1.1 $/kg, which is equivalent to only a quarter of composite materials composed of metals and rare earths. On the other hand, Lee et al. [39] studied the potential of $TiO_{2\text{-}x}$ for TE materials manufactured by plasma deposition. Ozturk et al. [30] studied two types of oxides: $Ca_{2.5}Ag_{0.3}X_{0.2}Co_4O_9$ type n and $Zn_{0.96}Al_{0.02}Y_{0.02}O$ type n, where X and Y are different dopants manufactured using the sol-gel method. In these works, the benefits of using oxides as raw materials are highlighted. Among these benefits are low cost, abundance, resistance to high temperatures, as well as simplified manufacturing processes not requiring controlled atmospheres. However, it can be seen that the purpose of these studies is to improve the efficiency of materials. In Table 5 it can be seen that the oxides present the least merit (efficiency). According to the review, the modules' oxide-based TEGs can increase their efficiency through the use of special processing [37] or doping techniques [28], which makes their use more viable.

Table 5. Thermoelectric materials type, merit rating, and temperature.

Material	Type	ZT	T (°C)	Ref.	Material	Type	ZT	T (°C)	Ref.
SnSe	p	2.5	627	[36]	$Pb_{0.93}Sb_{0.05}S_{0.5}Se_{0.5}$	n	1.7	627	[5]
FeNbSb	p	1.5	927	[44]	$Si_{80}Ge_{20}$	n	1.5	727	[36]
Bi_2Te_3	p	0.9	100	[35]	$AgPbmSbTe_2$+m	n	2	527	[36]
Na_2CoO_4	p	0.75	527	[46]	$Cu_xBi_2Te_{2.7}Se_{0.3}$	n	1	127	[36]
$Ca_{2.5}Ag_{0.3}Eu_{0.2}Co_4O_9$	p	0.57	800	[30]	$Mg_2(Si_{0.4}Sn_{0.6})_{0.99}Sb_{0.01}$	n	0.8	327	[29]
$Ca_{2.5}Ag_{0.3}Er_{0.2}Co_4O_9$	p	0.54	800	[30]	$Mg_2(Si_{0.53}Sn_{0.4}Ge_{0.05}Bi_{0.02})$	n	1.4	527	[29]
$Ca_{2.5}Ag_{0.3}Nb_{0.2}Co_4O_9$	p	0.52	800	[30]	ZnO	n	0.6	902	[46]
$Ca_{2.5}Ag_{0.3}Sm_{0.2}Co_4O_9$	p	0.51	800	[30]	$SrTiO_3$	n	0.4	827	[46]
$Ca_{2.5}Ag_{0.3}Lu_{0.2}Co_4O_9$	p	0.50	800	[30]	$CaMnO_3$	n	0.3	902	[46]
$Ca_3Co_4O_9$	p	0.5	827	[46]	$Zn_{0.96}Al_{0.02}Ge_{0.02}O$	n	0.04	800	[30]
$Mm_{0.28}Fe_{1.52}Co_{2.48}Sb_{1.2}$	p	0.5	477	[41]	$Zn_{0.96}Al_{0.02}Ga_{0.02}O$	n	0.17	800	[30]
Ca2.5Ag0.3Yb0.2Co4O9	p	0.47	800	[30]	$Zn_{0.96}Al_{0.02}In_{0.02}O$	n	0.12	800	[30]
$MnSi_{1.75}Ge_{0.01}$	p	0.4	527	[29]	$Mg_2Si_{0.4}Sn_{0.6}$	n	1.2	450	[35]
$MnSi_{1.81}$	p	0.3	400	[35]	Bi_2Te_3	n	0.8	100	[35]
$Ca_3Co_2O_6$	p	0.25	877	[35]	TiO_{2-x}	n	0.132	477	[28]
H-SnSe	p	2	427	[36]	Ni	n	0.020	477	[28]
BiBaCuSeO	p	1	527	[35]	$Yb_{0.09}Ba_{0.09}La_{0.05}Co_4Sb_{12}$	n	1.2	477	[41]
PbTe-SrTe+Te 2%	p	1	350	[33]	$Pb_{0.93}Sb_{0.05}S_{0.5}Se_{0.5}$	n	1.7	627	[27]

On the other hand, the use of cheap and abundant materials such as lead-based materials or silicides has also been the subject of recent studies. Han et al. [33] studied the feasibility of PbTe-SrTe base materials doped with 2% Te. They concluded there was a cost reduction through a low-cost processing method such as stable screen printing, although one of the base materials and the tellurium are high cost and low abundance elements. Jiang et al. [27] proposed PbS as an alternative to the base material PbTe, arguing that by doping with Sb and Se, efficiency can be considerably improved, in addition to them being abundant and low-cost materials.

Fu et al. [44] and Salvador et al. [52] developed materials such as FeNbSb (Half-Heuslers) and $Yb_{0.09}Ba_{0.09}La_{0.05}Co_4Sb_{12}$ (skutterudite), respectively, which are composed of low-cost and abundant elements, and by doping techniques are able to increase their efficiency. However, Ouyang et al. [36] evaluated some of the latest generation materials and recommended that materials such as skutterudites and half-heuslers could only be used in applications where cost is not of concern, due to the high manufacturing costs of these materials. On the other hand, Skomedal et al. [40] suggested the use of magnesium silicides as a favorable TE material, due to their low cost, abundance and low toxicity, despite their low efficiency when doping with elements such as Sn and Sb. They concluded that materials based on magnesium silicides are recommended for applications where low cost or low weight are more important than efficiency.

In addition, Homm et al. [56] analyzed some TE materials such as SiGe, PbTe, Bi_2Te_3, $FeSi_2$ and ZnO. The authors classified them according to selection criteria for different applications that required certain specifications for temperature, efficiency and cost, but taking into account the environmental aspects that each one presented.

According to the present review, it is observed that there is a conflict between the aspects of cost, efficiency and sustainability. Figure 4 presents a classification based on these aspects of recent studies addressed in this analysis. Three articles were found involving costs and efficiency in zone A, [27,29,44]; three articles between costs and sustainability in zone B [30,39,46]; one article involving efficiency and sustainability in zone C [36]; and tree articles involving all aspects, costs, efficiency and sustainability in zone U [40,52,56]. From this classification it is concluded that the oxides are inexpensive TE materials with important advantages. In particular, they are abundant, do not require high-cost processing, and resist high temperatures, which prevents premature degradation of the TE material. Moreover, they enable the formation of robust materials with a longer useful life, and have a good cost-sustainability ratio. However, their efficiency is reduced with respect to the commercially used TE materials, which prompts us to think about the different research approaches to improve them, such as nano-structuring, electronic band engineering, quantum confinement, as well as strategies such as crystal electron glass phonon, doping, and introduction of defects, among others. However, the use of any of these techniques requires specialized and complex processes, which would be reflected in the final cost of the product and would probably mean that the cost-efficiency ratio is not viable for developments in a commercial environment. Therefore, the development of this research is of utmost importance for providing not only a better future perspective of TE materials, but also because there are few investigations that specifically address the economic component of these materials.

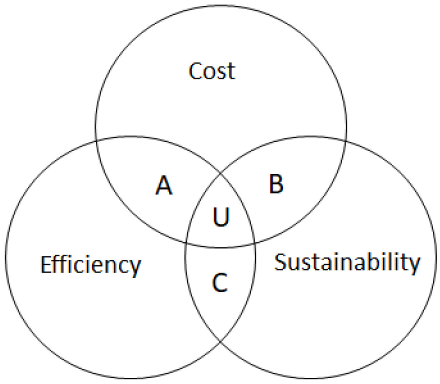

Figure 4. Classification of the articles found according to the results obtained for TE materials.

Furthermore, from a sustainability perspective, little information is available on commonly used TE materials. An example of this is the use of toxic materials such as lead, tellurium and bismuth in their fabrication. Therefore, an important aim of research is to explore is the environmental risks that these materials can present at different stages of the useful life of TEG modules and to search for abundant and low-cost elements.

Interest in certain thermoelectric materials is based on a combination of their characteristics and performance. Figure 5 shows the trends in the number of publications in recent years in relation to some representative thermoelectric materials, according to a survey carried out in the Scopus database. The figure shows the growing research interest in these thermoelectric materials.

Recent Development of New Materials for Thermoelectric Applications

The increases in the ZT values are produced especially by the decrease in the thermal conductivity of crystal lattices, and the recent advances in the development of new TE materials are based on the search for mechanisms that make it possible to minimize the thermal conductivity in the crystal structures of TE materials. Advances in TE materials provide measurable improvements in ZT values through the use of nanotechnology-based techniques. Nanophonon metamaterials provide special local resonance states in semiconductor materials for suppression of thermal conductivity. According to Ouyang et al. [60], nascent theories are being forged in the field of TE materials. Among the most promising are the coherent phonon theories (https://www.nature.com/articles/nmat3826, aacessed on 23 August 2021), the nanophonon metamaterial [61], the rattling effect [62], the topological phonon [63,64] and the topological electron [65]. Likewise, the synthesis of low-dimensional materials would allow the separation of related thermoelectric parameters to optimize thermoelectric performance. Among the advances in this field, the 1D Nanowires stand out [66,67], as well as the 2D Materials [68,69] and the Nanomesh Structures [70,71]. Finally, it should be noted that given the recent advances in computing, artificial intelligence and machine learning in combination with atomic simulation techniques, the development of new tools to predict new structures and characteristics of novel materials is envisioned, and these will provide accurate forecasts of the inherent properties of TE materials.

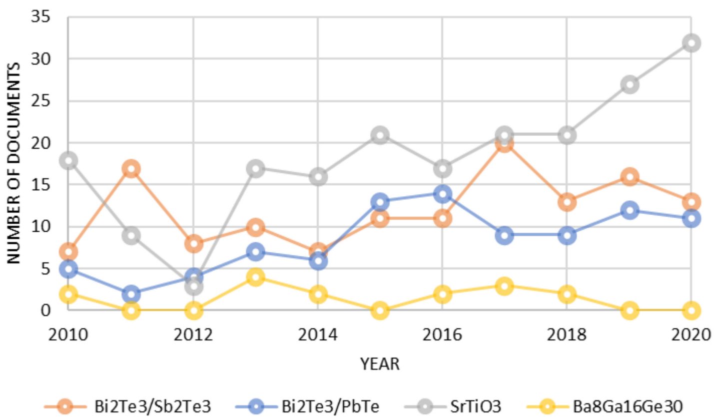

Figure 5. Research in thermoelectric materials in recent years.

3.2. TEG Modules

In the systematic search carried out, 29 articles related to TEG modules were found. We observed that one of the most commonly addressed topics is the efficiency/cost relationship presented by TE materials. This aspect is usually approached from several points of view, such as cost reduction, varying the TE material, or through a design of the TEG module that preserves its efficiency. However, another important factor that must be taken into account is the sustainability of the modules, which ranges from the analysis of their useful service life to the study of their final disposal. In Figure 6, the classification of the articles according to their content can be observed. These were classified by costs, sustainability, efficiency and modeling. In this graph, one article as found classified involving costs [42]; seven involving efficiency [27,39,44,49,52,56,57]; one in modelling [48]; and six in sustainability [18,23,37,38,50,53]. In the common areas D, E, F, G, H, it was not found papers between costs and sustainability and costs and modelling in zones D and G respectively. Between costs and efficiency, zone E, three articles were found [18,22,55]; while for zone F, between efficiency and modelling, seven were found [28,34,35,41,45–47]. Finally, ten references were found in thermoelectric modules, zone H [16,21,30–32,36,40,41,43,55].

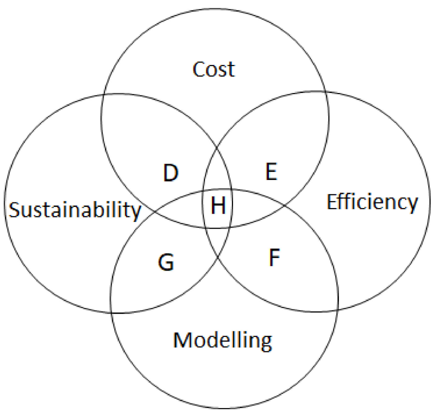

Figure 6. Classification of the articles found according to the results obtained for thermoelectric modules.

3.3. Cost/Efficiency

Currently, TEG modules are not in such high demand as could be expected as they are a clean source of energy that does not require exhaustive maintenance. However, the high cost/efficiency ratio that TEG modules present means that their introduction into the industry is difficult and still limits their viability.

3.4. Module Manufacturing

With the aim of decreasing the production costs without affecting the efficiency of the modules, some authors focus their research on the design of TE modules, the search for new materials in order to decrease costs, or increasing their efficiency. The use of oxides, half-heusler materials, skutterudites, and composite materials (organic/inorganic) can be a solution to overcome the limitations of the TEG modules. It is important, however, that these studies be developed in a holistic context oriented to applicability.

Studies such as those by Salvador et al. [52], on skutterudite encapsulated modules with a ZT of 1, enable this type of module to compete directly with the efficiencies of commercial PbTe modules. However, to date, commercial modules with this type of materials are scarce and costly due to their current manufacturing methods. Fu et al. [44] evaluated the potential of doped half-heusler materials with an efficiency of 6.2% and a power density of 2.2 W/cm^2. These modules can resist high temperatures (~927 °C) and are an economical alternative to commercially used TE materials.

In order to reduce costs in TEG modules and to improve their coupling to any surface, Lee et al. [39] studied the manufacture of TEG devices and basic electronics using titanium oxides (which operate at temperatures ~500 °C), and deposited them by plasma sintering, thus obtaining an assembly of thermocouples connected in series and in parallel with an efficiency of 0.85% and an electrical power of 2.43 mW at 450 °C. The authors did not conduct an economic analysis on the assembly, only referring to its easy and low manufacturing cost through the elimination of many of the parts that make up the commercial TEG modules. On the other hand, Yazawa et al. [54] proposed the use of flexible modules that incorporate organic/inorganic composite materials and reduce costs but with a reduced performance (ZT between 0.01 and 0.25), a performance that is relatively low compared with commercial TE materials.

Anderson et al. [23] carried out a techno-economic analysis on the total cost of TEG devices, finding that the use of impure TE materials such as oxides or other types of cheaper TE materials are not the most feasible option at a cost level, since TE material only represents 15% of the total value of a TEG module.

Table 6 presents some characteristics of commercial modules such as base material, dimensions, power output, open circuit voltage, operating temperature range and cost. Currently, the most common TEG modules are those manufactured from bismuth telluride. Among these, a great variety can be found in which characteristics such as their configuration, output power and circuit voltage might vary. Most modules can work at maximum temperatures between 320 °C and 350 °C, but their optimal operating conditions are around 250 °C. These types of modules can be found in the market with prices ranging between $10 and $28 US.

Likewise, commercially it is possible to find modules that resist higher temperature ranges, such as TEG PbTe-BiTe modules. These TEG modules can work at maximum temperatures of around 360 °C, and commercially they can be found with better output powers than the BiTe based ones. Naturally, the improvement of these characteristics is reflected in their cost.

Table 850 °C reaching 6% efficiency, being attractive for the recovery of residual heat at high temperatures. However, this type of component is up to seven times the cost of traditional BiTe-based modules. This makes them less attractive due to their cost-efficiency ratio.

Table 6. Technical characteristics and cost of commercial modules.

Commercial Modules	Materials	Dimensions (W × L × H) (mm^3)	Output Power (W)	Open Circuit Voltage (V)	Operating Temperature Range	Cost ($ US)	Ref
Hz-1	BiTe based	29.21 × 29.21 × 5.08	2.3	6.1	50° to 250°	$10.00	[72]
Hz-9	BiTe based	62.74 × 62.74 × 6.63	9	6.1	50° to 250°	$15.00	[72]
Hz-14	BiTe based	62.74 × 62.74 × 6.63	14	3.1	50° to 250°	$25.00	[36]
Hz-14HV	BiTe based	61.05 × 71.05 × 7.87	14	8	50° to 250°	$25.00	[17]
Hz-20	BiTe based	74.68 × 74.68 × 5.08	20	4.5	50° to 250°	$50.00	[13]
Hz-20HV	BiTe based	74.50 × 68.00 × 5.00	20	10.8	50° to 250°	$50.00	[72]
TEP1-1264-3.4	BiTe based	40 × 40 × 5	5.4	10.8	30° to 300°	$40.89	[11]
TEG1-126610-5.1	BiTe based	40 × 40 × 4	5.1	7.8	30° to 300°	$28.41	[73]
TEM 070-6006	SnTe based	40 × 40 × 4	16.8	-	70° to 600°	-	[12]
TEG1-PB-12611-60	PbTe-BiTe	56 × 56 × 7.7	21.7	9.2	30° to 350°	$69.00	[14]
CMO-32-62S	BiTe cold site-Calcium	64.5 × 64.5 × 8.3	12.30	12.8	50° to 800°	$375.00	[73]
CMO-25-42S	manganese oxide hot site	42 × 42 × 7	7.5	10.0	50° to 800°	$330.00	[73]

3.5. Module Design

Commercial modules, such as those from Table 6, generally have a pre-designed configuration from the supplier, which means that for some applications, they are not suitable or do not show their optimal performance. On the other hand, the design is an important factor since by means of the design parameters, the efficiency, the cost and the useful life of the modules can be improved. Likewise, most of the studies related to TEG components or systems are carried out using simulation tools [15], due to the complexity of their manufacture, as well as the cost that these would generate for their development if they were carried out exclusively by experimental means. Some of the most relevant studies in this regard are listed below.

Segmented modules represent one of the most viable alternatives from the view point of design. In these, various TE materials are used to manufacture the module legs, seeking to increase the working temperature, minimize the thermal effects, and increase the efficiency of the TEG modules. Recent work related to the manufacture of segmented modules includes the work of Hung et al. [46], who implemented commercial TE materials in cold areas of the cell and oxides in hot areas, in order to increase the working temperature of the modules. The viability of these TEG modules was analyzed using numerical modeling, which found that the oxide-segmented modules have an efficiency of around 10%. Ouyang et al. [36] carried out a study in which high-ZT TE materials were evaluated by finite element analysis. A systematic model was achieved for the segmented modules, finding a cost-performance ratio of ~0.86 $/W with an efficiency of 17.8%. Similarly, Jiang et al. [27] evaluated a TEG module composed of np Bi2Te3 and np PbS/PbTe, which exhibited an efficiency of 11.2% with a $\Delta T = 317$ °C. In addition, they optimized the ratio of the legs at low and high temperatures, determining the optimal ratio to be 7:17. The maximum power obtained in this TEG module was 0.53 W for a $\Delta T = 312$ °C.

During the design of TEG modules, the optimization of parameters such as the length of the legs or the number of thermocouples of the TE materials helps to reduce the amount of material used, without compromising the efficiency of the modules. According to Rezania et al. [49], the temperature differences at the n- and p-type junctions of TE elements are not identical. Such temperature differences are lower in n-type TE elements, compared with those in p-type TE elements, due to the higher thermal conductivity in the n-type material. Consequently, the footprint size of the n-type element must be larger than the footprint of the p-type TE element, due to the higher thermal conductivity in the n-type material. Therefore, the optimal ratio of footprint areas to achieve the maximum generation and the best cost-efficiency ratio in thermoelectric modules must satisfy that An/Ap < 1, where An and Ap are the footprint areas of the the n-type and p-type junctions, respectively. Brito et al. [41], found that when the thickness of the TE elements is smaller, the electrical resistance is reduced, but this will impact the ΔT of the TE module, because a lower thermal resistance will increase the thermal output and attenuate the temperature difference between the hot and cold sources. However, this will only occur if the usable hot source is low and the other thermal resistances are high enough to significantly affect the ΔT of the TE module. In their study, Dongxu et al. [31] found that the thickness of the TE legs can be reduced to 1.1 mm, which is 4 mm less than commercial modules, while still achieving the same efficiency. In all these works, simulation tools were successfully used to find the relationships between the different parts of the modules and their respective powers.

3.6. Useful Life

The evolution in the design of TEG modules has also contributed to increasing their useful life. Moreover, the thermal stresses to which the TEG modules are subjected, affect them adversely by the formation of microcracks and the expansion and contraction of TE materials. To address this problem, Skomedal et al. [40] incorporated spring-supported contacts in the legs of TEG modules to dampen thermal expansion and contraction in the modules; these authors concluded that good diffusion barriers and possible coatings can reduce oxidation of the hot side electrodes and interconnections. Likewise, they noted that the TEG module could generate 1 to 3 W/cm^2. Furthermore, Ming et al. [45] studied, via numerical analysis, how the non-uniform flow causes the junctions of the TEG modules to be damaged and also decreases their output power. In addition, Merienne et al. [32] investigated the effect of thermal cycles on commercial TEG modules, finding that when rapid temperature changes are applied, their output power can decrease by up to 61%, compared with a module in which the heat flow is constant and the temperature changes are minimal. Therefore, it is of utmost importance to analyze in detail the design of TEGs and the operating conditions to which they will be subjected in order to establish applications that allow them to extend their useful life.

The current commercial TEG modules have still not reached the economic feasibility, nor the efficiency required for thermal energy recovery applications. Therefore, as mentioned above, the characterization of commercial TEG modules, in the specific conditions to which they are going to be subjected, requires the use of expensive control equipment, or manual processes, which leads to difficulty in making long-term measurements. Therefore, Elzalik et al. [28] developed a characterization method that allows precise and inexpensive estimation of the maximum power point and the dynamic parameters of the TEG module. The proposed procedure can be used with different sources of residual thermal energy and under different operating conditions.

In relation to the sustainability of TEG modules, authors such as Khanmohammadi et al. [22] and Heber et al. [21] state that the amortization period and the power ratio of commercial TEGs make them not viable to be used exclusively as a generation method. Therefore, they recommend using them as a complement to other methods of recovering residual thermal energy, thus generating an integrated system, or the development of segmented TEG modules that allow greater efficiency and profitability.

3.7. Thermoelectric Generators

As previously mentioned, the main limitation for the use of TEG is its cost-efficiency ratio; therefore, this type of generator still does not compete with conventional mechanical thermodynamic systems or the electrochemical systems of Rankine, Stirling, Brayton, expansion devices or fuel cells. These types of conventional generation systems have a wide application ranging from heat utilization with a lower output power range to greater than 10 kW. For their part, TE materials are usually utilized for applications with an output power of less than 10 kW [51], which makes them suitable for specific power requirements.

Yazawa et al. [40], focused their research on the economic viability that TEGs can achieve with respect to other types of electricity generation systems. The authors found that TEG modules with a ZT of 0.8 can achieve a cost-performance ratio of 0.86 $/W. This makes TEG systems competitive with other power generation systems. It was also found that TEG systems have commercial profitability if they have a cost-performance ratio of less than $1/W [34].

In the manufacture of TEG systems, the main component is the TEG module, and the other components such as heat sinks, and supports help the system to perform better. Mori et al. [42] estimated that the TEG modules only represent 60% of the cost of whole systems and focused the design of the system on heat concentration structures that would help to double the efficiency of power generation and that could reduce the total cost in the system by half. Likewise, Hendricks et al. [43] warned that the cost of the heat exchanger, which is the element of the system that most frequently increases the total costs of TEG modules, should be further investigated to achieve profitability of the system.

Lately, the automotive industry has initiated investigations into TEG systems due to the considerable losses of caloric energy that arise from the combustion process, which could be used to power other vehicle systems. Indeed, the incorporation of these energy recovery systems, could significantly reduce CO_2 emissions into the atmosphere.

Arsie et al. [47] proposed the incorporation of a TEG system in the exhaust of a car using commercial 14 Hz TEG modules. In their research, the temperature gradient was guaranteed using refrigerant on the cold side of the module; the system was connected directly to the battery and alternator of a vehicle, and using a longitudinal model, it was possible to determine that the system displaces the energy of the alternator by between 15 and 20%, having an average saving of ~1 g/km of CO_2 in standard driving cycles.

Fernández et al. [34] used commercial Bi2Te3 TEG modules for heat recovery from light duty diesel engines as they produce ~386 W of recoverable power under common vehicle driving conditions. In their research they found that in commercial TEG modules, it is only possible to recover about 37.6 W, if no additional improvements, such as advanced heat exchangers, are applied. The authors also found that when a cooling system is able to maintain the cold side of the TEG module at 50 °C (less than the engine system coolant temperature), up to 75 W of power can be obtained.

Heber et al. [21] in 2020, manufactured a TEG system for natural gas heavy vehicles, in which they used 168 commercial modules based on SnTe. The cost of the TEG was EUR 1811, and a maximum power of 1507 W was achieved with a power density 50 W/kg, a reduction in CO_2 emissions of 4.9 (9.4) g (CO_2)/km, and a cost-efficiency ratio of 1.2 EUR/W, which suggests that the system is profitable.

Likewise, the use of TEGs as complementary systems has been investigated to compensate the cost-efficiency ratio in different applications to reduce CO_2 emissions. Thus, Bellos et al. [74] investigated the efficiency of a solar energy-induced TEG using commercial Bi_2Te_3 TEG modules, and carrying out a financial analysis, found that the cost of the investment would be 1 EUR/W, with a payback period of 4.55 years and a leveled cost of electricity of 0.0441 EUR/kWh, indicating that this system would be unprofitable.

3.8. Sustainability (Circular Economy)

As noted above, commercial TE materials are not yet sufficiently cheap, and high efficiency materials are not yet mass produced. Until now, the most commonly used commercial TE materials are Bi2Te3-based alloys because they have advantages such as easy bulk processing. However, although they are precursors, high energy expenditure and expensive techniques are used in their processing both for power generation and for cooling at temperatures close to ambient levels. On the other hand, TE materials use elements such as bismuth, tellurium, antimony, selenium, and lead, among others, which are expensive, scarce, and sometimes toxic. As mentioned earlier, from the point of view of the circular economy, the recycling of TEG modules could generate great economic benefits since it would allow obtaining raw materials for the manufacture of new TEG modules or other electronic devices, generating a reduction in the consumption of scarce elements. Moreover, they also generate environmental advantages because the improper disposal of these materials is avoided, which can benefit both the environment and human health.

At the time of writing this paper, the scientific publications on recycling TEG modules are still quite sparse. However, currently there are different ways to recycle TEG modules based on tellurium bismuth, from which three approaches can be differentiated based on the separation techniques: (i) chemical, (ii) thermal, and (iii) bacterial methods. On the other hand, in some cases only some parts of the TEG modules are recycled or only the elements of the semiconductors are recovered. According to the bibliographic review carried out, approaches have been proposed for the recycling of commercial TEG modules based on bismuth tellurium, by taking advantage of the differences in melting temperature of the constituent materials. In this way, the separation of the different constituents of commercial modules (plastics, Cu, Bi_2Te_3 and Al_2O_3) in an efficient way might be achieved by mechanical processing which relies on the entropy changes of these materials [27].

The TE materials have been separated by thermal processes followed by chemical separation processes, in which the characterization of the materials of the TE modules was conducted by techniques such as differential scanning calorimetry (DSC), X-ray diffraction (DRX) and field emission scanning electron microscopy (FESEM), which give information on the material types, melting temperatures and the distribution of the materials. This allows their separation based on the differences in thermal and chemical properties. This type of separation is initially carried out by means of thermal treatments such as hot oil baths at 250 °C for the removal of solder from the -n (Bi_2Te_3) and the p-type ($Bi_{0.5}Sb_{1.5}Te_3$) semiconductors. Later they are subjected to a mixed acid solution (HCl and HNO3 in a 3:1 ratio) at room temperature. At this point, the Sb of the semiconductors precipitates. Then, the solution is then filtered, washed and sintered in order to obtain nano-powders of Bi_2Te_3 -n type with a particle size of ~15 nm purity [25,36].

None of the previous works reports a characterization of the thermoelectric properties of the recovered TE materials. The characterization would be of great importance in order to know if the processes used for their separation in any way affect the properties of the recovered products and their possible use in future applications. Table 7 lists some recent works in relation to the final disposal of thermoelectric modules.

Table 7. Bibliographic review of the final disposal of thermoelectric modules in recent years.

Year	Ref	Number of Articles	Country
2020	[9]	1	Finland
2017	[26]	2	France
	[27]		Korea
2014	[39]	1	Korea
2013	[42]	1	Korea

It is clear then that there is a global need for sustainable technologies [75–77], and the circularity of materials and processes are areas where TE can have a significant impact as the main, partner, or complementary technology since it is a particularly adaptable technology [78].

4. Remarks and Conclusions

Between 2010 and 2020, there have been few studies concerning thermoelectric generator (TEG) modules that examine the larger holistic picture from mass manufacturing, profitability (from the economic point of view), efficiency, life cycle, and competitiveness to final disposal. Despite this, in the systematic search, an approach towards circular economy and sustainability has been considered indirectly since the articles analyzed covered topics ranging from raw thermoelectric TE materials, manufacturing of TEG modules, energy efficiency, and also the applicability and recycling of materials and modules.

The cost of manufacturing the modules is a little explored topic since in most works, only the relationships concerning the cost of the constituent TE materials are analyzed, and other factors that contribute to the high commercial cost of these modules are not examined.

In the present review it is highlighted that the efficiency of TEG modules depends, in addition to the TE materials, on external agents such as refrigeration systems, connections between the TEG modules, and the environment in which they will be operated. Therefore, these are important research topics to improve the cost-efficiency ratio of TEG systems.

Modeling is a valuable tool in predicting the efficiency of TEG modules at different stages of their development and implementation.

Additionally, the present research evidences the lack of work oriented towards the circular economy and sustainability of TEG modules, highlighting this aspect as an extremely important area for the development of TEG systems.

Taking into account the limitations that current TEG modules present in relation to the efficiency-cost relationship and the lack of research from circular economy and sustainability perspectives, this review proposes an approach for new studies that would allow an improved understanding of the processes used and their useful life. The research approach would aim to balance the cost-efficiency ratio from a sustainability perspective, not only addressing the base material, but also provide a more general approach to understanding the entire life-cycle of the modules from their manufacture to their final disposal. This would provide more information on the economic viability and sustainability of TE materials and modules.

5. Implications and Final Reflection

The circular economy represents a development opportunity that would lay the foundations for a sustainable recovery. Responding to the problem of the large accumulation of waste will in turn allow progress in solving the climate crisis and thus avoid the loss of valuable habitats.

The present study was developed with the aim of promoting the development of alternative generation systems to reduce dependence on the consumption of oil and other minerals. This will lead to a reduction in the generation of high levels of emissions, which will help the restoration of the battered ecosystems of our planet. Here is an opportunity that the authors believe should be seized as soon as possible.

As shown in this study, there are few development initiatives, particularly in the field of thermoelectric materials, that implement sustainable models. This is striking since what has sparked interest in the development of this technology has been precisely the need to counteract an existing problem. However, there is no awareness of the implications of not adopting an environmental perspective and transforming our way of thinking in order to build a better world for all.

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