A Review of the Production, Implementation, and Disposal of Graphene Based Phononics and Thermoelectrics

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ABSTRACT: In this paper, a literature review and socio-technical system are examined to find the best suitable production process for thermal and graphene-based thermoelectric rectifiers. The literature review is divided into three parts. Namely, production of graphene and graphene rectifiers, implementations of graphene, and disposal of graphene-based thermoelectric devices. Liquid-phase exfoliation is found to be the most useful form of graphene synthesis in the current market. Secondly, focused ion beam modification of the graphene is found to be most suitable for producing rectifiers as the rectification ratio is considerably higher than other discussed method.

Key words: Graphene; Thermoelectrics; Thermal Rectifier; Synthesis; Socio-Technical System

1 INTRODUCTION

Thermoelectrics is not the combination of the phononic and electric field of study but the conversion. Thermoelectrics use the thermoelectric effect to convert electric current into thermal energy and vice versa. This phenomenon can be described by the collective of the Seebeck, Peltier, and Thompson effects[1]. Nevertheless, thermoelectrics, phononics, and electronics go hand in hand when developing thermoelectric devices.

In a previous work, research was performed towards the thermal rectification effects in graphene based nanoribbons[2]. Thermal rectifiers, also known as thermal diodes, have existed in literature since the early 20th century[3]. Afterwhich, scientific interest faded till the early 21st century. In the early 21st century, the field of study surrounding thermal rectifiers became more accessible through advanced simulation software and a better understanding of molecular dynamics. Nowadays, multiple papers have been published on thermal rectification using software such as the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS)[4, 5, 6]. Thermal rectification on its own is not applicable in the market. However, it is a fundamental concept for creating more complex phononic and thermoelectric products.

Graphene, a monolayer carbon structure in honeycomb layout, looks promising for further developing the thermal rectification ratio as graphene possesses high thermal conductivity, is widely available, and easily modified[7, 8]. Hence to compliment the previous research, the production, implementation, and disposal of graphene is examined for phononic and thermoelectrics purposes.

2 BACKGROUND

Thermal rectification involves an asymmetry in the heat transfer through a lattice or ribbon. This implies that the heat transfer propagates easier in one direction and is inhibited in the opposite direction. The thermal rectifier, also known as thermal diode, is the practical implementation of the thermal rectification effect. There are several ways of obtaining the asymmetry in heat transfer. Such methods include the use of different materials and defect/geometry engineering. G. Papageorgiou et al., state that ”defect-free, mono-layer graphene is considered to be the strongest material ever tested”[8]. Consequently, this allows the material to remain relatively strong while only one atom thick. In addition, the honeycomb like structure of graphene allows for a variety of defects to be used by defect engineering without causing severe changes to the graphene structure. Hence, this makes it a prime candidate for tunable thermal rectifiers[9].

Similar to how thermal rectification works in thermal diodes, thermoelectric rectifiers rely on an asymmetry in the thermal to electric conversion ratio[10]. In addition, the rectification effect can also be obtained through modifying the geometry of the nanoribbon. The study by Kaushal et al.,[11] propose an example graphene based thermoelectric rectifier to convert the thermal energy into useful electric energy. Their design shows how the asymmetry of the thermoelectric properties can be achieved in a graphene based design.

Throughout the years, there have been several materials used for the production of thermal and thermoelectric rectifiers. Although carbon-based materials have existed for a long time, the older carbon-based materials never had the thermal and electrical properties to be interesting to the industry. Only when in 1993 it was discovered that the one and two dimensional forms of the materials possessed high thermal conductivity, became it of interest to researchers[12]. Consequently, researchers started to develop better carbon based materials. Such as graphite, graphene nanotubes, and mono-layer graphene. As mono-layer graphene is one of the newer carbon-based materials, it is still a very present in current day literature[11, 13, 14]. Figure 1 shows the increasing trend on graphene papers published till halfway of the year 2017.

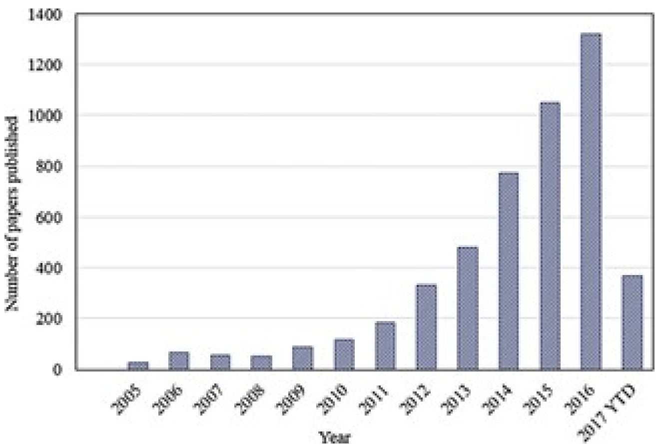

Figure 1: Yearly published papers about graphene.[15]

2.1 *Research Question*

All things considered, graphene is showing great promise for thermal and thermoelectric rectifiers. To optimise its integration into the current thermoelectric market, its production, implementation, and disposal will need to be taken into account. Therefore, it is of great interest to figure out what impact the changes in the production process have on the integration and disposal of the product. In other words:

What graphene-based rectifier production process is most optimal for better integration with the thermoelectric market?

3 METHODOLOGY

The main bulk of the research is performed through the use of a literature review. The literature review is divided into three sections discussing the production, implementation, and disposal. The second part of the research depicts the socio-technical system[16] of an example thermoelectric product. The socio-technical system is examined twice, once before integration of thermoelectric devices, and once after. Subsequently, changes to the socio-technical system can be observed to evaluate the impact of implementing thermoelectric devices.

4 LITERATURE REVIEW

4.1 *Production*

The production process is important to the product, as it will affect both the implementation and disposal. The production will be divided into two topics. Namely, the production of graphene and the modification of graphene to acquire the desired shape of a thermal rectifier.

4.1.a Synthesis Methods for Graphene

Two main branches exist for producing graphene. The first method is a ”bottom-up” approach in which the graphene is constructed from individual carbon atoms. On the contrary, the second method is considered to be ”top-down” approach. The ”top-down” approach deconstructs multi-layer graphene (graphite) into individual layers[17]. Within each of the branches several specialised sub-methods exist. Figure 2 shows the segmentation of the different graphene synthesis sub-methods based on the publications and granted patents. The graphene produced mainly consists of carbon atoms. However, different methods produce graphene fragments of different purity. In general, the higher the carbon percentage, the better the material properties will be. The study from Mahanta et al.,[19] shows that higher oxide concentrations in graphene will cause a lower thermal conductivity. They experimentally obtained a thermal conductivity of 18 W/Km for a graphene sample consisting of 46% carbon. Which is considerably lower than 2275 W/Km obtained for a 99% carbon-based graphene sample. Similarly to the change in thermal conductivity, higher oxide concentrations lead to higher electrical resistance[20].

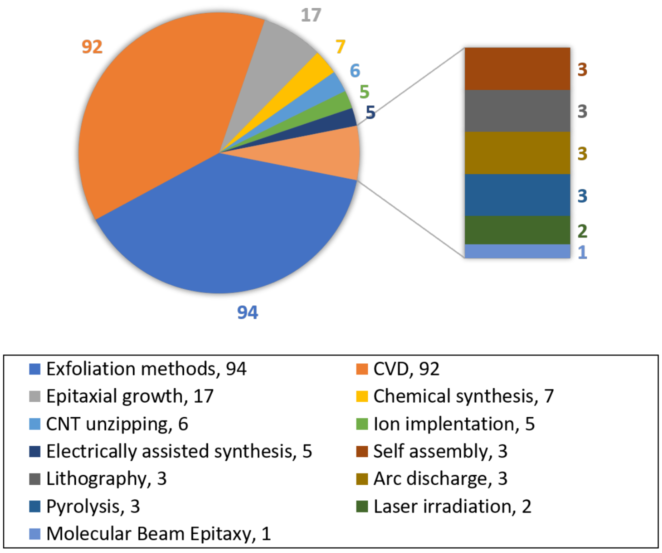

Figure 2: Segmentation of different graphene synthesis sub-methods by Somani et al.[18]

Four of the more interesting synthesis methods for the production of graphene-based thermal rectifiers are discussed. Namely, Liquid-phase exfoliation, oxidative exfoliation, chemical vapour deposition, and the electrochemical method.

Liquid-phase exfoliation of graphite uses ultrasonic treatments in solvents to separate the graphite layers[21]. Unlike liquid-phase exfoliation of graphite oxide, this method produces graphene with low concentrations of defects and oxygen functional groups. Therefore, producing the extremely high quality graphene required for thermal rectifiers.

The oxidative exfoliation method produces graphene-oxide through deconstructing graphite-oxide. In many regards, it is very similar to the liquid-phase exfoliation of graphite[17]. Due to the oxygen functional bonds in graphite-oxide, the inter-layer distance is increased from 0.335 to 0.625nm resulting in a weakened van der Waals force between layers. Consequently, it does not require the use of ultrasonic treatments to break off the graphene layers. Nevertheless, it produces graphene with low carbon purity. To further increase the material quality, reduction of the oxygen functional bonds is required. This can be achieved through chemical, electrochemical, thermal, and hydrothermal reduction.

Chemical vapour deposition is a "bottom-up" approach using hydrocarbon gasses to grow the graphene sheet. The hydrocarbon gasses are heated from 650 to 1000°C. At high temperatures, the carbon and hydrogen atoms split when contacting a metallic catalyst material[17]. Hereafter, the carbon atoms form the graphene sheet atop the metal catalyst. Chemical vapour deposition produces high quality graphene with little to no defects. However, disadvantages include low throughput and high operating costs. Additionally, it requires additional purification to remove residual catalyst material from the graphene. Even so, there is a lot of development in the chemical vapour deposition sector to reduce operating costs and increase yield. An even more interesting development is the study by Molina-Jiron et al.[22] They present a feasible way to convert carbon-dioxide into mono-layer graphene using the chemical vapour deposition method.

Unlike most graphene exfoliation methods that rely on oxidants, the electrochemical discharge mathod relies on the graphite's conductive properties to deconstruct the graphite into mono-layer graphene. By using the graphite as a electrode, it can be given either a positive or negative charge. This will cause graphite exfoliation due to the oppositely charged ions inside the material[23]. Although the method can produce high quality graphene, its complex and high precision process as well as its high electricity demand make it cost ineffective[17].

4.1.b Graphene-based Thermal Rectifiers

The next step in the production is to make the graphene attain the desired form for a thermal rectifier. In the study by Wang et al.,[5] two methods were used to produce the rectification effect. The first method makes use of a focused ion beam to produce small nanopores in the graphene surface. In the study by Ibrahim et al.,[25] they achieved nanopores with a diameter of 30nm for a milling time of 0.1 second.

The second production method used in the study of Wang et al.,[5] used an electron beam induced deposition. The method uses hydrocarbons in collaboration with the electron beam to contaminate a local area on the surface of the graphene. In a study by Xu et al.,[26] they were able to create a deposition on the graphene with a diameter of under 2nm in an operation time of 12 seconds. Additionally they stated, that the method can also be used as a means of carbon based adhesive to join or shape features of the graphene. Nevertheless, it still requires the use of hydrocarbons such as methane. Making it less environmentally friendly than the focused ion beam.

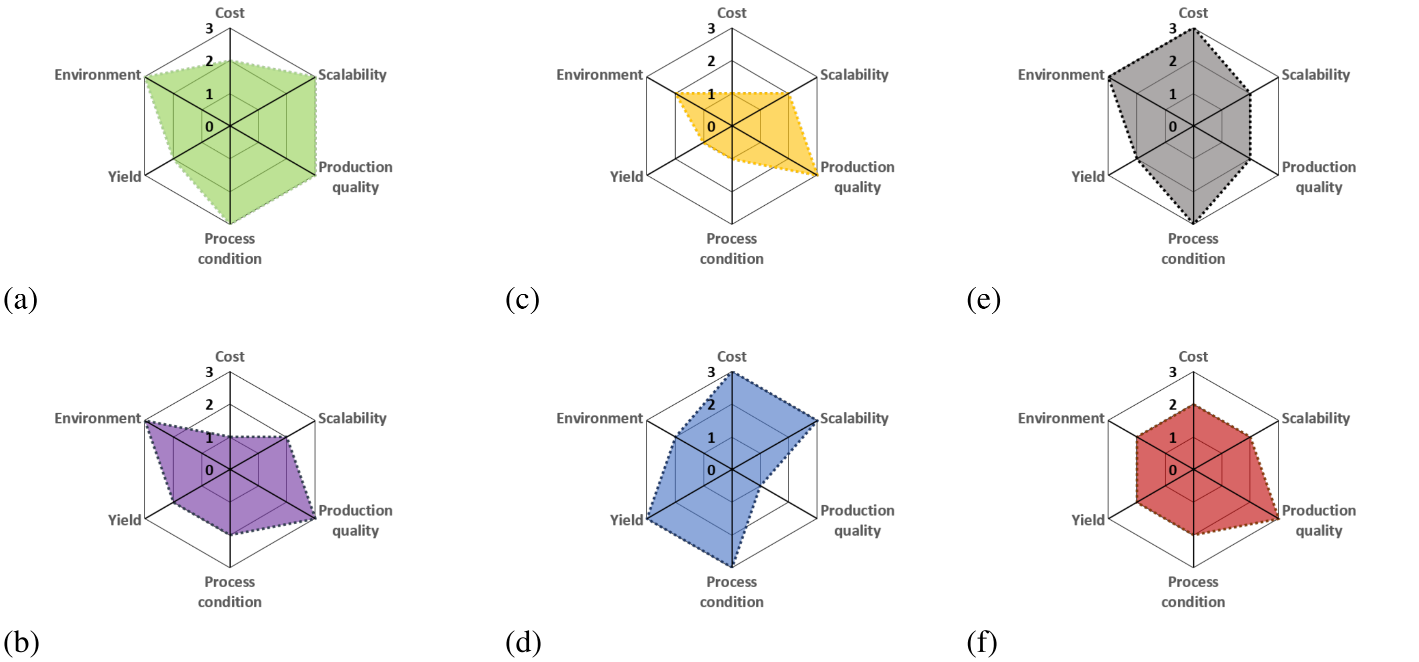

Figure 3: Process evaluation for both the synthesis and modification of graphene. Based on the evaluation model by Lee et al.,[17] and supplemented by additional papers. (a) Liquid phase exfiliation.[21] (b) Chemical vapour deposition.[22] (c) Electrochemical.[23, 17] (d) Oxidation exfoliation.[24] (e) Focused ion beam.[25] (f) Electron beam deposition.[26] Preference score: 0 = none or not available, 1 = low, 2 = average and 3 = high.

Finally, the electron beam induced deposition method in the study by Wang et al., was able to obtain a rectification ratio of 10%. This is relatively low compared to the 26% obtained for the sample that was modified by the focused ion beam.

4.2 Implementations

Figure 4 shows the industrial applications of graphene. It displays the electronic industry as the biggest sector. The data behind the figure is based on several patents and papers published in the year 2010. Meaning that the percentages will not accurately represent the current market. Nevertheless, the figure does show the significance of the electronic and energy storage market. As thermal and thermoelectric rectifiers can be integrated in the electronic, energy storage, and energy generation sectors. This observation and Figure 1 show that there is probably a high demand for graphene-based rectifiers in the current market.

4.3 Disposal

The disposal of graphene-based rectifiers is a more complex topic since there are little to no papers on this topic. Therefore, it is assumed that graphene-based rectifiers are disposed of in three ways. The first method disposes of the graphene by incineration for thermal energy recovery. This is done in waste-to-energy plants such as Twence B.V.[27] The second method involves re-using the product for a second life time. This is possible when the product can be fully disassembled. However, it is very unlikely that this is realistic since adhesives are used in the product assembly process. The third method reduces the thermal or thermoelectric parts to small granular material that can be recycled.

The burning of graphene has been explored by the paper from Ermakov et al.[28] Through the use of molecular dynamics simulations they showed that burning multi-layer graphene can cause partial burning on underlying graphene layers. This resulted in the release of carbon-monoxide and carbon-trioxide (Figure 5). Carbon-monoxide is responsible for many deaths as it is toxic to humans[29]. It is for that reason of great importance to limit the carbon-monoxide emissions. The application of water vapour filters can be used to turn carbon-monoxide into hydrogen and carbon-dioxide[30]. Thus, greatly reducing the risk to human health. Most thermoelectric devices consist

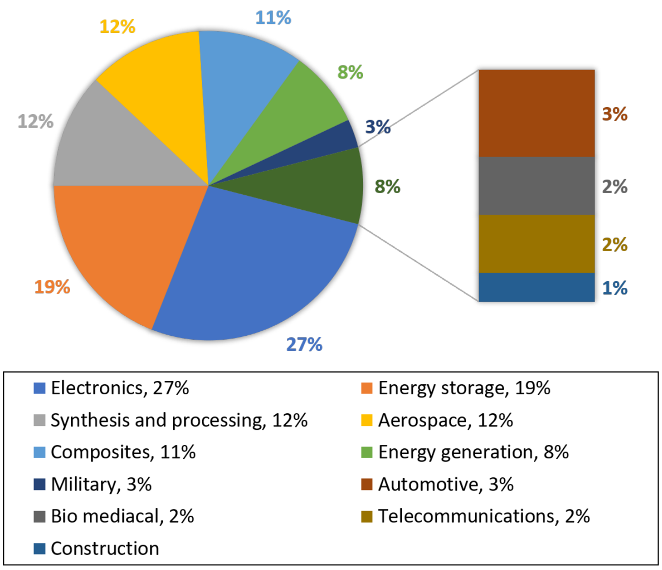

Figure 4: Applications of graphene[15]

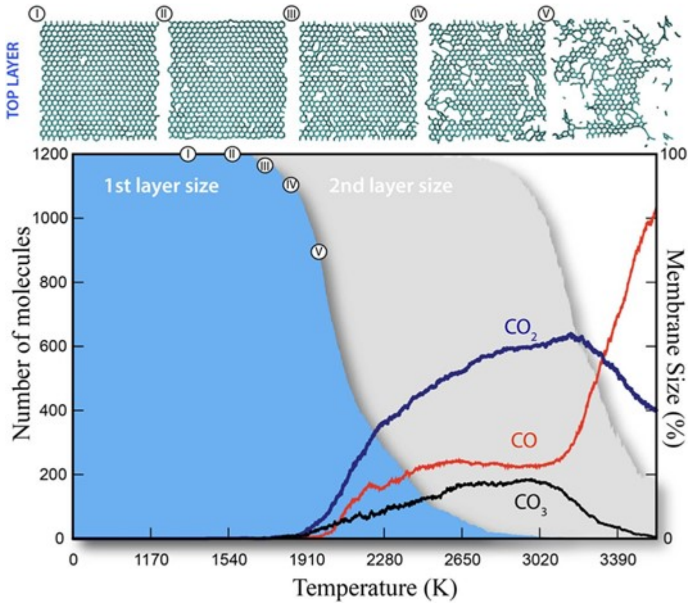

Figure 5: MD simulation of burning multi-layer graphene. [28]

of a single solid-state material enveloped by a ceramic substrate. The ceramic substrate is cut open to reveal the inner connectors and conductors which can be in turn be removed. For a graphene-based thermoelectric device, the inner connectors and conductors are made from graphene. The inner components are then milled down into smaller pieces[31]. The study from Jiang et al.,[32] focuses on producing fibre-reinforced epoxy resin from recycled graphene capacitors. Similar to the thermoelectric devices, the device is first cut open to reveal the capacitors. The graphene capacitors are then heat-treated at 200° to get rid of any solvents inside the device. Afterwards, the temperature of the heat treatment is further increased to reduce the polymer bindings keeping the graphene in place. Finally, the graphene is removed and shredded in epoxy resin. The shredded graphene now functions as reinforcement fibre for the epoxy resin, strengthening the material.
Nevertheless, the recycling and milling of graphene brings it own complication concerning human health. The study by Fadeel et al.,[33] discusses the possible health effects of being exposed to graphene nanoparticles. The research mainly showed the lack of understanding surrounding the biological interaction that graphene-based materials have with the human body. Even so, they flagged multi-layer graphene nanotubes as possibly carcinogenic and toxic to the human body.

5 SOCIO-TECHNICAL SYSTEM

The socio-technical systems design method is used to show the possible impact that thermoelectric rectifiers can have on the product environment. Moreover, the method takes the human, social, organisational, and technical factors of the system into account. The introduction of solid-state thermoelectric heat pumps is used as an example product environment (Figure 6).
Conventional heat pumps systems are common in everyday usage. They keep your fridge cool, are used in liquid-cooled pc systems, and maintain a steady temperature for your house. Generally, these systems contain a liquid loop with a compressor, valve, and heat exchange surfaces. The introduction of the graphene-based thermoelectric rectifier has the potential to innovate and disrupt the industry. It does not only eliminate the need for compressors and expansion valves, but also the need for coolants. Conventional systems require the use of coolants' compressing nature to either heat or cool the liquid. Solid-state heat pumps do not rely on this mechanic. Other interesting changes to the socio-technical configuration are the fabrication, maintenance, and disposal section. As solid-state thermoelectric parts are more complex than a pump or valve, their production process will also require more complex machinery. Such machinery is discussed in Section 4.1.b. The maintenance of the product is highly simplified. To illustrate, the elimination of liquid loops within the product allows

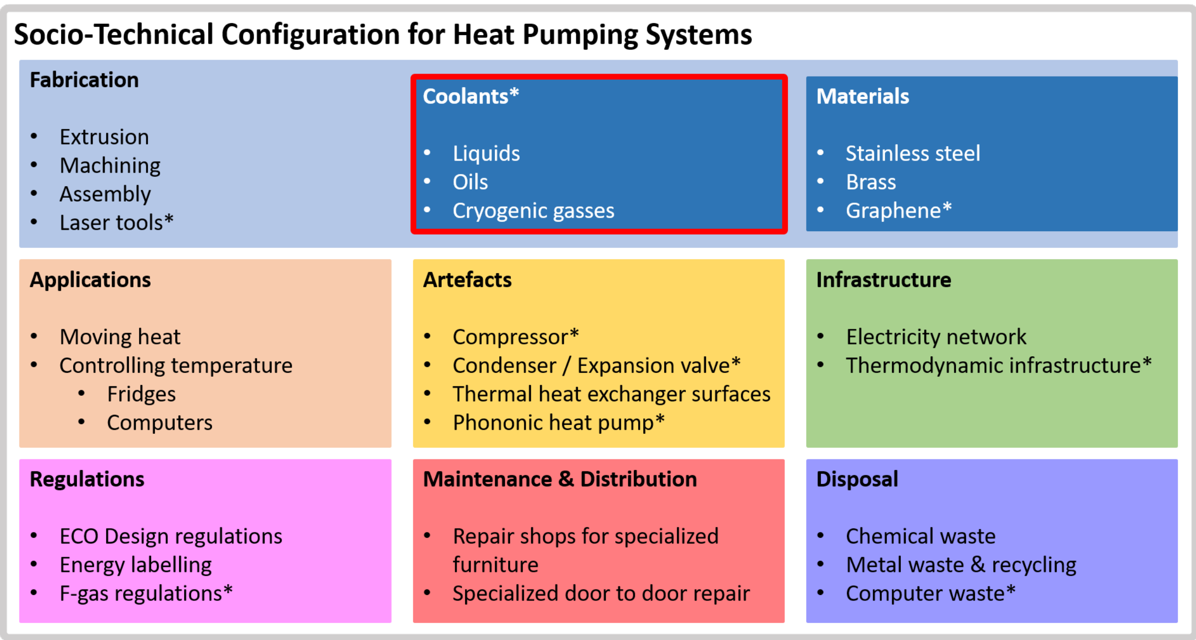

Figure 6: Overview of the socio-technical configuration for heat pump designs. *impacted by the addition of the solid-state thermo-electric heat pump technology. [16]

for individual parts to be replaced without having to empty possible toxic coolants first. This allows for parts to be reordered if broken instead of sending a technician or sending an entirely new product. Finally, disposal becomes less toxic due to the elimination of possibly toxic coolants. In its place come the complex recycling and disposal of graphene thermoelectric devices.

6 DISCUSSION

The reviewed data suggests that graphene production is still developing at a rapid pace. New and better production methods are still being published in the last couple years[22, 20]. These methods mainly focus on improving the quality, scalability, and environmental impact of the graphene synthesis. On top of that, with the increase of interest in graphene (Figure 1) in the last 10 years surrounding papers and patents, it can only be assumed that practical applications are increasing accordingly.

Out of all the graphene synthesis methods, liquid-phase exfoliation and chemical vapour deposition performed best when taking into account the high graphene purity requirements (Figure 4.1.a). This is in accordance with the patents and published papers provided in Figure 2. However, it can be assumed that this segmentation of synthesis methods have changed a lot over the past couple years. The study also supports that the quality of graphene produced for ”top-down” configurations is mainly dependent on the variant of graphite used to obtain the graphene. Processes that use graphite-oxide have considerably worse material properties[19].

While previous research mainly focused on providing better synthesis methods, the results demonstrate that there is a clear shortage of papers on the disposal of graphene. Not only was there little information on graphene based thermoelectric disposal but also a lack of understanding on the possible health implications due to graphene exposure[33]. Consequently, it would be beneficial to focus on disposal and health implications before continuing the rapid development of graphene synthesis.

7 CONCLUSION

The research aimed to identify the best production and disposal processes for better product integration with the current thermoelectric market. In conclusion, the liquid-phase exfoliation method proved to be the best practice for graphene synthesis. It produces high quality graphene and is a widely used practice. Nevertheless, chemical vapour deposition would have been the better recommendation if it was not for its high operating temperatures involved with using carbon-dioxide as carbon source. Yet, it is a good alternative in the near future as it is being developed.

The secondary processing step required to produce graphene-based thermal and thermoelectric rectifiers show the best promise using the focused ion beam method. The focused ion beam is best capable of producing high thermal rectification. However, loses on the front of precision and tunability.

The most important thing when it comes to disposal of graphene is the production. There is no saying in advance how graphene will be disposed. Disposal is regulated differently everywhere on the globe. Thus, making sure the material is optimised for any type of disposal is the biggest concern. This is achieved by using as little disposable chemicals as possible. As well as producing high quality mono-layer graphene and preventing the use of multilayered graphene.

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