

University of Wales Trinity Saint David
School of Engineering

Mechanical testing of an environmentally sustainable
alternative to ceramic tiles: a quantitative study
Major Project ACMA6013

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Declaration of Authenticity

This thesis is the result of the author's original research. It has been composed by the author and has not been previously submitted for examination which has lead to the award of a degree.'

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Abstract

Ceramic tiles are a popular choice for wall and flooring coverings, resulting in mass production around the world. The production of ceramic products has a significant environmental impact, and with the majority of ceramics being produced in China, the carbon footprint from importation is significant. There is a need for a sustainable, locally produced alternative to ceramic tiles. Art of Eco are a company who endeavour to produce such a product with their eco slab stone. However, the mechanical properties require quantification. Additionally, Art of Eco have started to use graphene in the hypothesis to enhance the properties of their products, but the effect is yet to be determined. The aim of this study is to determine the mechanical properties of Art of Eco's products, both with and without graphene, and compare them to two types of traditional ceramic tiles. To achieve this, a three-point bend test was conducted following the ISO 10545-4:2019 standard.

Test results showed that the graphene-enhanced Art of Eco samples had an increase in various parameters, including an average of 19.49% increase in modulus of rupture, 23.73% increase in Young's modulus and up to 60.81% increase in the breaking load. Both types of traditional ceramic tile displayed higher mechanical properties than the Art of Eco samples tested.

The data obtained shows an increase in mechanical properties in the graphene-enhanced samples. However, due to multiple inconsistencies in both the tile dimensions and the tile structures, it is not clear if these results are a true representation of the effect of graphene on Art of Eco's product, and further work would benefit multiple aspects of this study.

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Introduction

Ceramic materials are a common choice in the construction industry for flooring and wall tiles due to their durability, versatility, availability, and aesthetic appeal. However, like all materials, the production of ceramic tiles has an environmental impact.

In 2021, world ceramic tile production reached 18,339 million m², an increase of 7.2% on that recorded in 2020 (CeramicWorldWeb, 2022). The majority of production occurred in Asia, particularly in China and India, representing 74% of total global production. Europe produced just 11.6% of world production. In 2020, China was the leading manufacturer of ceramic tile in the world with a total production 8,474 million m².

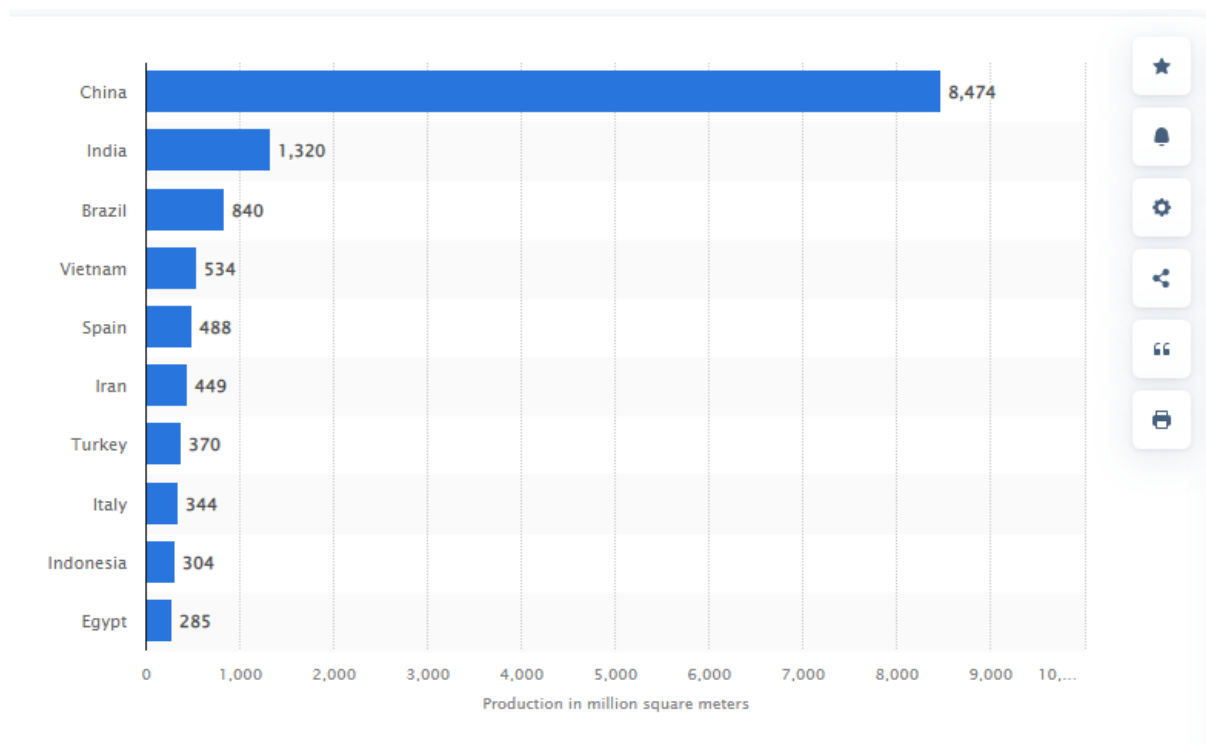


Figure 1 Ceramic tile production by country (Garside, 2021)

The ceramic tile industry in Europe is a significant contributor to the economy, employing a considerable number of people and generating a significant annual turnover. In 2018, the industry engaged 2,000 companies, produced over 1,304 million m² of tiles per year, and contributed to a business turnover of 30 billion euros (Rio et al., 2022). However, the UK's contribution to Europe's total production of ceramic tiles is relatively small, and the majority of tiles used in the UK are imported from large producers in countries such as China and

India. As a result, the environmental impact of ceramic production is further increased by transportation, adding to the carbon footprint of the tiles.

Ceramic tiles are produced using raw materials such as clay and silica (Tilemag, 2019), which are processed and fired at a high temperature to produce a hard and dense material. This process consumes significant amounts of energy and produces greenhouse gas emissions, which contribute to climate change. In a study carried out by Tikul, (2014), it was found that a small ceramic factory produced 15.89 (kg CO₂-eq) per m² of ceramic tile manufactured and 1.14×10^{-5} (kg CFC 11-eq), which contributes to ozone layer depletion, whilst a medium plant was found to produce 15.89 (kg CO₂-eq) per m² of ceramic tiles produced, and 2.38×10^{-7} (kg CFC 11-eq) (Tikul, 2014). Ceramic tiles feature some desirable properties, such as high hardness, good chemical resistance, and low water absorption. However, they display low fracture toughness and are usually brittle, which limits their use in some applications such as in a high temperature environment where they can crack, or in heavy-duty industrial applications where high loads or repeated impacts are expected.

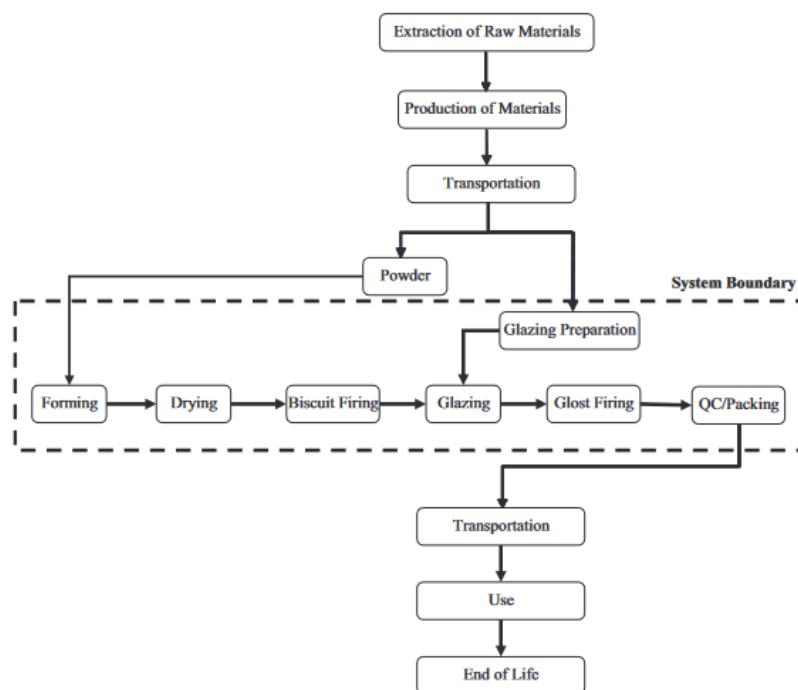


Figure 2 Ceramic tile lifecycle (Tikul, 2014)

Figure 2 above shows the life cycle of ceramic tile production.

There are several environmental impacts of ceramic tile production. Air pollution is created by production of dust and other pollutants that are released during the cutting of the raw materials, causing respiratory issues. Water pollution is another issue as ceramic tile production generates significant volumes of wastewater, which can contain a high level of

heavy metals, suspended solids and other pollutants. As previously mentioned, ceramic tile production consumes a high amount of energy due to the high temperatures required during the firing stage, producing greenhouse gas emissions.

The UK relies heavily on imported ceramic tiles, which creates a high carbon footprint due to transportation. This, coupled with the environmental impact of the ceramic production itself, highlights the urgent need for a sustainable, locally produced alternative.

Art of Eco

Art of Eco is a company that is producing a sustainable alternative to ceramic tiles. Their slab-stone is created using a cold cure process, using 93% recycled stone aggregate sourced from local Welsh facilities and quarries. A binding agent is mixed with the other materials to ensure that they bind efficiently. After mixing, it is poured into moulds of up to 2.2m x 1m, compressed, and left to dry. After drying, the slabs are cut to the required measurements. The process is shown below in figure 3. By using a cold cure process, the energy-intensive kilning process typically used in the ceramics industry is eliminated.

Art of Eco (AoE) is based in Ammanford, Wales, and has ambitious growth plans, including expanding into mainland Europe. To support this expansion, they need to upscale and plan for the layout, storage, and equipment required at their new facilities. It is currently unknown whether graphene enhances the mechanical properties of their product, therefore identifying these properties would provide key information for determining the necessary equipment for future production.

Art of Eco state that their current cost is roughly £30/ m² under current manufacturing conditions, including printing, topcoat, and packaging. Their target cost is £17/m². In comparison, the average cost of ceramic tiles can range between £15/m² (myjobquote, 2023) for basic tiles and up to £40/m² for some tiles with a more intricate design (Tile Mountain, nd). This makes Art of Eco a competitive choice considering the supposed lower environmental impact and the choice of customization offered. In addition to the production of their regular tiles, Art of Eco found that the addition of graphene may enhance the mechanical properties of their product. The addition of graphene costs less than £0.50/m² which, when the hypothesized benefits that graphene might add are considered, is a small price to pay.

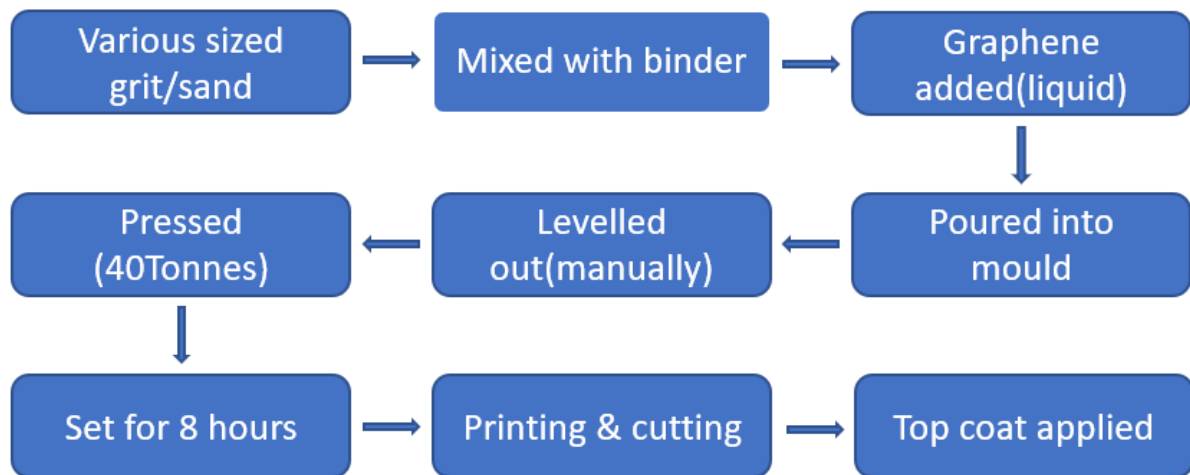


Figure 3 Art of Eco production process

The importance of sustainability

With the increasing rate of global warming caused by human activity, it is more important than ever to create methods to reduce the effect that humans have on the environment. Traditional manufacturing processes often involve the extraction of finite resources and cause the emission of harmful pollutants. By adopting more sustainable methods, it is possible to reduce the harmful effects of outdated materials and manufacturing processes. Art of Eco's ambition is to provide a product that eliminates these traditional processes and provides an environmentally friendly alternative. Energy intensive industries such as the iron and steel, oil refining, cement and ceramics industries globally produce roughly two thirds of industrial greenhouse gas emissions (Sangwan et al., 2017), and it is estimated that 70% of industrial energy consumption use thermal processes, such as those used to produce ceramic tiles, which in turn produce waste heat and cause heat pollution.

The EU's goal is to become climate neutral by 2050. In order to achieve this, energy intensive industries such as the ceramics industry must reduce their consumption to help mitigate climate change (Monteiro et al., 2022).

The addition of graphene as an additive or nanofiller has the potential to significantly enhance the properties of materials, such as their strength and durability. This can result in a stronger and longer-lasting product that requires less material to create, thereby addressing some of the environmental concerns caused by human activities. While the production of graphene itself requires energy and can generate emissions, the benefits gained from its

addition to materials often outweigh these costs, although the method used to produce graphene can significantly impact this.

Industries need to move towards reusing and recycling more materials to reduce waste and conserve natural resources. Art of Eco has adopted a circular economy approach by using recycled quarry aggregate amongst other materials to manufacture their tiles. A circular economy focuses on being restorative and regenerative, by keeping resources in use for as long as possible and therefore extracting the maximum value from them (Ellenmacarthurfoundation,2022).

Art of Eco's aim is to implement a circular economy approach in the manufacturing of their products, sourcing the majority of their materials from local companies, and thus reducing transport-related carbon emission. Additionally, the aggregate used in their products is waste aggregate that would otherwise not be used and is sourced from a quarry less than 5 miles away. Their aim is to be able to re-use their tiles after reaching end of life by crushing them down and using the material to produce new tiles. However, the graphene used in their tiles is produced by Firstgraphene, who source their raw material, graphite, in Sri Lanka. This is then transported to Australia for manufacturing the graphene. This has a significant impact on the carbon footprint of the graphene produced, and a full lifecycle analysis should be undertaken to evaluate the overall environmental impact of using graphene in their products and is suggested as further work.

Aims and objectives.

The primary aim of this study was to quantify the mechanical properties of Art of Eco's proposed sustainable alternative tile, and whether the addition of graphene improves the mechanical properties. The secondary aim of this study was to determine whether these tiles were superior in strength to a standard ceramic tile.

Although, in theory, the production and manufacturing process of Art of Eco's product is expected to be significantly less environmentally harmful than that of traditional ceramic tiles, the mechanical properties of their product have not yet been quantified. Without this information, it is difficult to accurately compare the strengths and weaknesses of their product to traditional ceramic tiles.

In order to achieve these aims, the following objectives were established:

Objective 1: Identify relevant literature on the use of graphene to enhance mechanical properties of materials to form a hypothesis.

Objective 2: Establish what method of testing is appropriate, and how to undertake such testing.

Objective 3: Carry out mechanical testing of Art of Eco tiles without graphene.

Objective 4: Carry out mechanical testing of Art of Eco tiles with graphene.

Objective 5: Carry out mechanical testing of common ceramic wall tiles.

Objective 6: Carry out mechanical testing of common ceramic floor tiles.

Objective 7: Compare test results for all tests carried out.

Objective 8: Discuss the possible reasons for the test results recorded.

Objective 9: Discuss whether the addition of graphene to the tile exhibited any mechanical behaviour similar to that identified in the literature.

Objective 10: Identify areas for future research.

Timing plan

A timing plan provided a method to identify and prioritise project tasks and use time effectively. Through the use of a Gantt chart, it became clear which tasks needed to be carried out and when, to track progress, identify where any delays might occur, or any other issues.

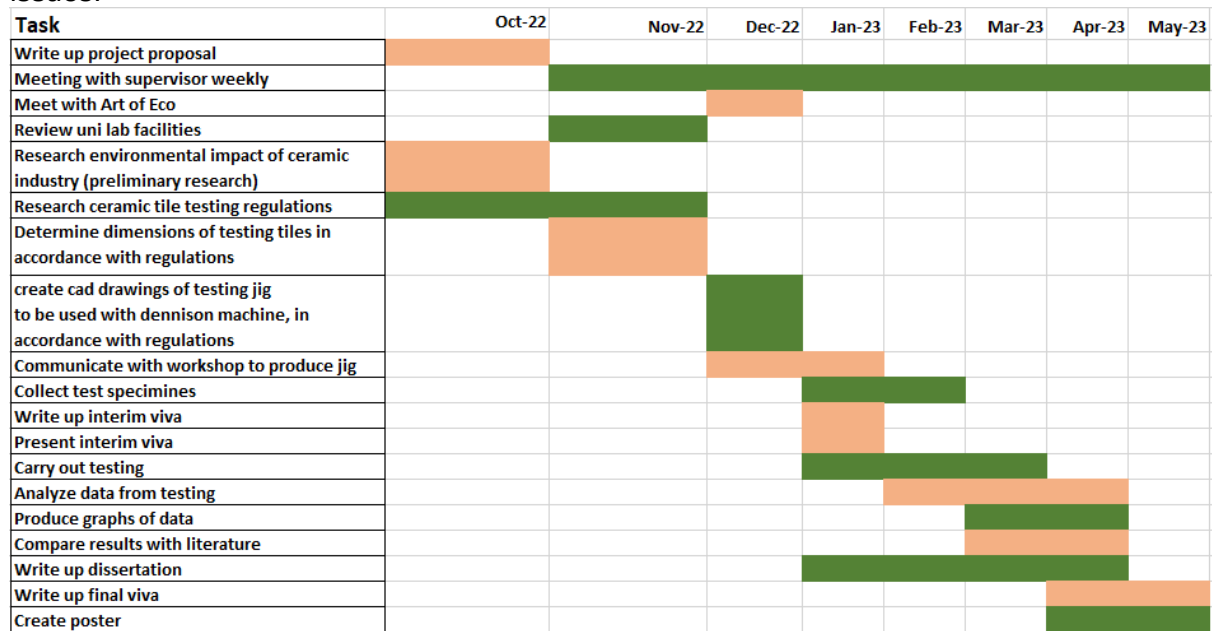


Figure 4 Initial Gantt chart of project

Figure 4 above shows the initial timescale for the project. Some issues occurred with the tile dimensions provided for testing, which delayed the testing. Additionally, an error made during measuring the dimensions to produce the jig to fit the Dennison machine resulted in delays, as new measurements had to be taken and then holes redrilled in the jig (as seen in

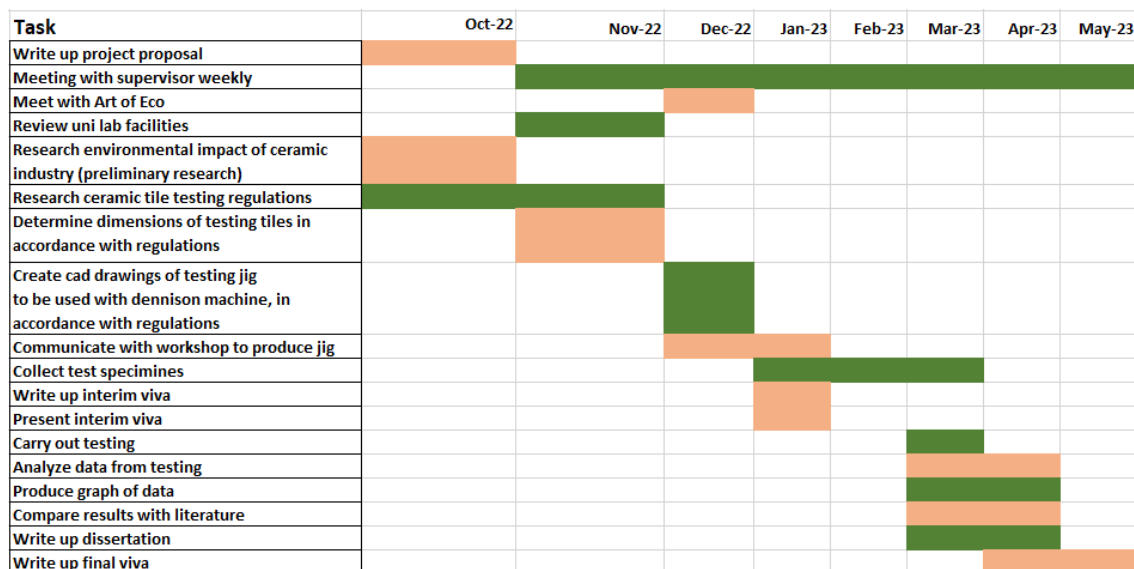


Figure 5 Revised Gantt chart of project.

Figure 30). The Gantt chart in Figure 5 shows the updated timing plan to reflect the delays that occurred.

Mechanical properties and practical application of graphene

Art of Eco have been using graphene in the hope that it will enhance the mechanical properties of their products. Graphene has been used successfully to improve various products, such as ceramic tiles, concrete, and some forms of plastic. The section below describes the history of graphene, why it is being considered and the various production processes that are used to produce it, some of which can be environmentally damaging.

Graphene is a material made up of a single two-dimensional layer of carbon atoms arranged in a tightly bound hexagonal lattice structure. It is the building block for graphite, which is essentially millions of layers of graphene, and for graphitic materials of all other dimensionalities (Geim & Novoselov, 2007).

Graphene was first isolated in 2004 at the University of Manchester (Cooper et al., 2012), and has since seen a significant increase in popularity in recent years due to its remarkable mechanical, electrical and thermal properties. Unlike graphite, which is a naturally brittle material, graphene is the strongest material discovered to date due to the strength of the carbon bonds, which are just 0.142 nm in length. It displays a Young's modulus of up to 1 TPa and ultimate tensile strength of up to 130 GPa in ideal conditions (Shen & Oyadiji, 2020). In comparison, structural steel has an ultimate tensile strength of 400 MPa (La Fuente, 2022). Not only is graphene extremely strong, it is also very light, weighing just 0.77 milligrams per square metre (La Fuente, 2022). In addition to the strength of graphene, it exhibits elastic properties, being able to maintain its initial size after being exposed to strain (La Fuente, 2022). Graphene also exhibits extremely high crystal and electronic quality. These properties have resulted in a wealth of potential applications of graphene, including a promising reinforcement or filler material for composites (Geim & Novoselov, 2007).

Since its discovery in 2004, due to the extraordinary properties of graphene and the potential it shows, there has been a permanent growth in the number of papers published, particularly on the subject of ceramic/graphene composites, as seen in adapted figure 6 below. Another study shows that the trend in publications on graphene has increased from 90 articles in the year 2000 to almost 45,000 in 2015 (Markandan et al., 2016).

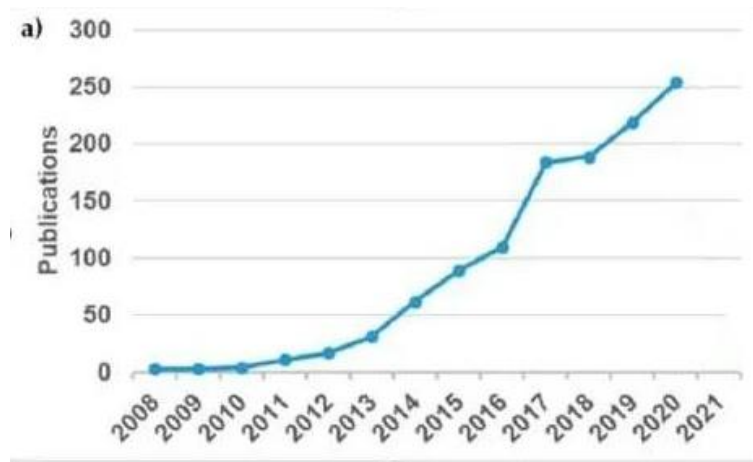


Figure 6 Ceramic/graphene papers published 2008-2021 (Ramírez et al., 2021)

These statistics indicate that graphene has a promising future in many different applications, with materials science being the most researched area (Markandan et al., 2016).

Creating graphene

Graphene can be produced in two ways. The most common method is the top-down approach of exfoliating graphite to produce graphene. However, this process often requires large amounts of environmentally damaging solvents and oxidants, as well as high-energy consumption during the mixing, shearing or electrochemical treatment. Additionally, the graphene can be left with a defective perforated structure after the reduction processing step (Luong et al., 2020). Bottom-up synthesis is the second approach, using methods such as the chemical vapour deposition method. This method can provide high quality graphene, however, only in small quantities, or, if produced in larger quantities, the end product is prone to defects. (Luong et al., 2020).

The first successful method of producing mono-layer graphene was by mechanical exfoliation. This was achieved by Geim and Novoselov (2007), using the scotch tape method. Since then, there have been many other processes developed to produce either few-layer or single-layer graphene (Cooper et al., 2012). However, there are still serious

barriers to large-scale production of high quality graphene, especially by methods that are environmentally friendly (Wu et al., 2020) .

Mechanical exfoliation: This method was developed by Geim and Novoselov and involves using sticky tape to peel off individual graphene layers from a graphite crystal. The process requires patience and precision, and the resulting graphene flakes are relatively small and difficult to scale up to industrial production (Cooper et al., 2012).

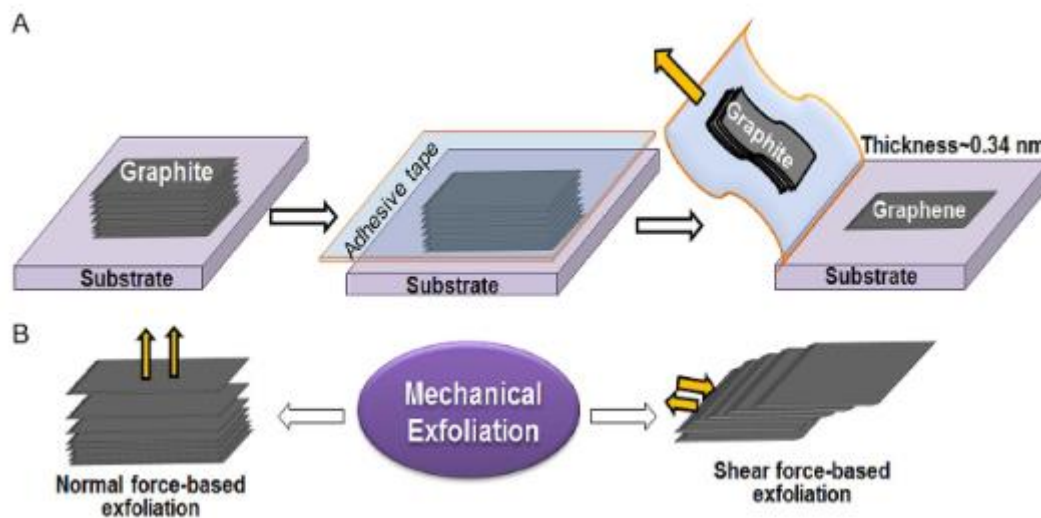


Figure 7 Mechanical exfoliation diagram (Agrawal & Yi, 2020).

Chemical vapour deposition (CVD): This method involves growing graphene on a substrate by introducing a carbon-containing gas (such as methane or ethylene) into a high-temperature furnace. The carbon atoms in the gas settle on the substrate and form a graphene layer. This process can produce high-quality, large-area graphene sheets. The process is relatively inexpensive but requires a controlled environment (Saeed et al., 2020).

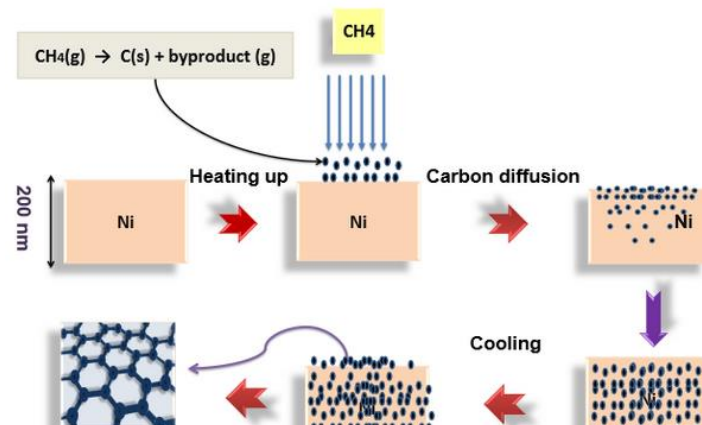


Figure 8 Chemical vapor deposition diagram (Cunningham, 2014).

Liquid-phase exfoliation: This method involves using solvents to exfoliate graphene layers from graphite. The graphite is mixed with a solvent, such as N-Methyl-2-pyrrolidone, which is currently the optimum exfoliating agent due to its chemical properties facilitating the dismantling of the Van der Waals bonds holding the stacked layers of graphene together. The mixture is then sonicated to break apart the graphite layers. The resulting solution contains small flakes of graphene that can be used for various applications. Liquid-phase exfoliation is a scalable and cost-effective method but may result in lower quality graphene compared to other methods. Another issue with this method is that N-Methyl-2-pyrrolidone is a toxic substance and therefore not eco-friendly. *Morton et al. (2023)* carried out a study to provide an eco-friendly solution and found that a mixture of deionised water and ethanol showed promising results (Morton et al., 2023) .

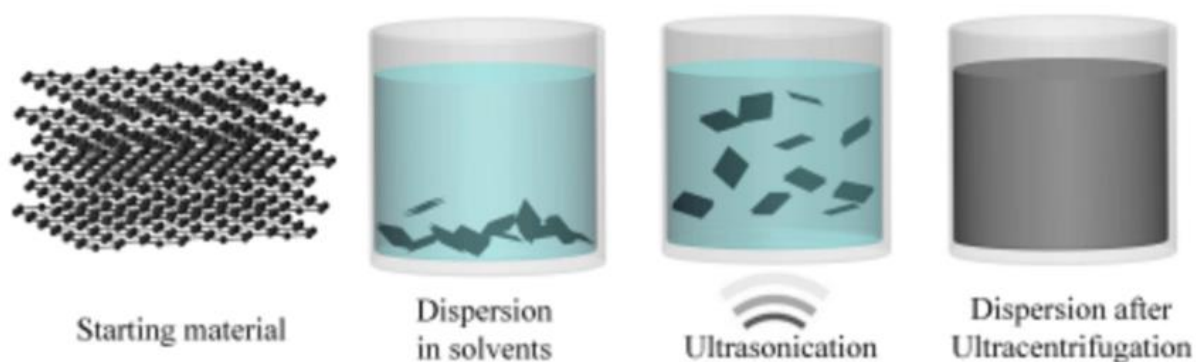


Figure 9 Liquid-phase exfoliation diagram (Hogan et al., 2017).

Electrochemical exfoliation: This is an alternative method to mechanical exfoliation and involves using an electrical current to exfoliate graphene from graphite. The graphite is immersed in a solution containing an electrolyte, and an electrical current is applied to the solution. The current causes the graphite layers to separate and exfoliate into graphene sheets. Electrochemical exfoliation is a relatively new method that shows promise for producing high-quality graphene at a low cost (Achee et al., 2018). It is also a method that is not as environmentally harmful as other graphene production methods, which rely on oxidants that are often toxic. Instead, electrochemical methods take advantage of the conductive properties of graphene, allowing intercalation of molecules between graphene layers. Using graphite itself as an electrode, a charge can be applied (positive or negative). Oppositely charged ions are then attracted to the graphite, and are intercalated between the graphene layers, which facilitates exfoliation (Yu et al., 2015). Compared to other exfoliation methods, such as mechanical and chemical vapour deposition, this approach has a lower cost for mass production of graphene.

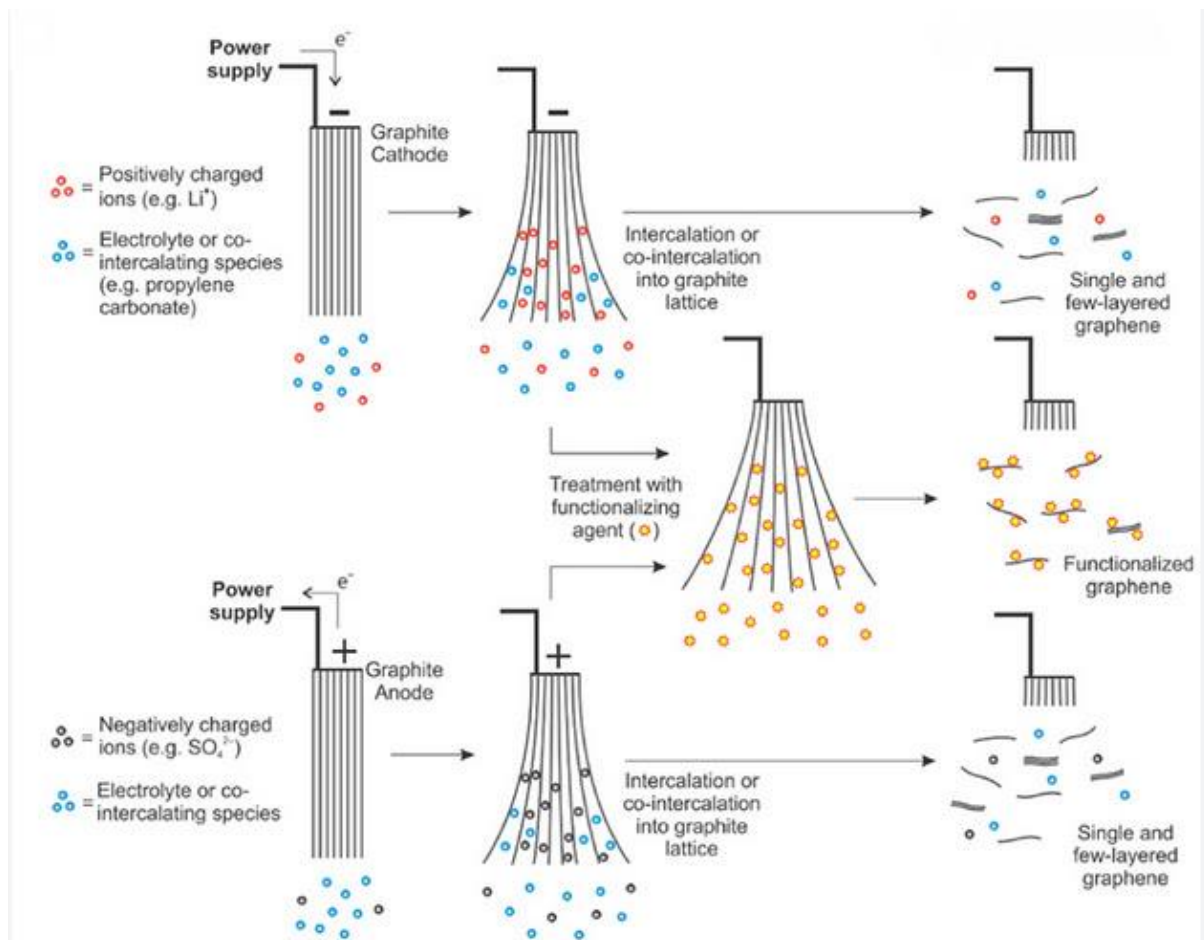


Figure 10 Electrochemical exfoliation of graphene diagram (Liu et al., 2019).

Flash Joule heating: This process uses end-of-life ground up plastic and a coke additive, which is used for conductivity, packed between electrodes in a tube, which is then blasted with a high voltage. The intense heat vaporises other elements and leaves behind turbostratic graphene. This process appears to be promising as it does not require the use of solvents and is not energy intensive (Wyss et al., 2022). The yield of graphene depends on the carbon content of the source. A wide range of waste can be used, be it organic, plastic, household, or other carbon-based waste, and be turned into graphene that is free of contaminants (Critchley, 2021). A study undertaken by *Luong et al. (2020)* showed that the graphene produced by this method contained the lowest concentrations of defects reported so far. Figure 11 shows the process of flash joule heating, including the reduced environmental impact of over 92% compared with chemical oxidation.

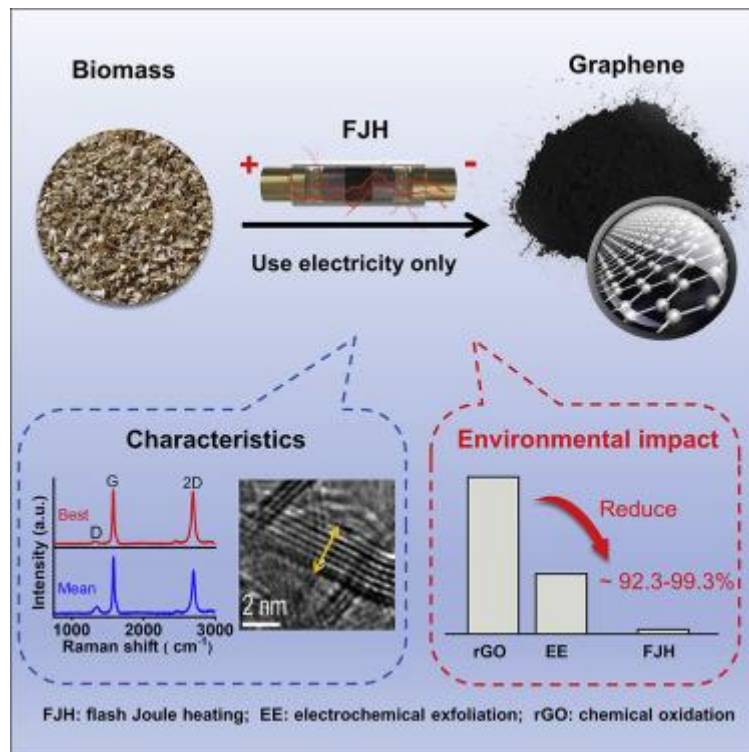


Figure 11 Flash joule heating of graphene, including environmental impact (Jia et al., 2022)

Art of Eco obtain their graphene from Firstgraphene. This company uses a proprietary single-step electrochemical exfoliation process. Firstgraphene claims this method enables them to manufacture large volumes of high purity graphene under controlled conditions (FirstGraphene, 2021). This method is considered to be relatively environmentally friendly when compared to other production methods, such as chemical exfoliation or some forms of mechanical exfoliation. However, the environmental impact can depend on other factors such as the source of the graphite used and the type of electrolyte solution used, and how it is disposed of. Other factors include how the electricity used in the method is produced. As previously outlined, the graphene produced by Firstgraphene carries a very high carbon footprint.

Graphene improving sustainability.

Studies show that the integration of graphene into a variety of materials highlights the sustainable potential of graphene as an additive. Due to the strength of graphene, when it is integrated into another material, it reduces the amount of the host material required as a result of the increase in physical properties (Critchley, 2021).

A prime example of this is using graphene to enhance the properties of cement and concrete. The addition of graphene into cement means that the overall amount of material to perform the same structural requirements is far less (Critchley, 2021). The production of concrete is responsible for roughly 8% of the world's CO₂ emissions (Nature, 2021). Studies show that adding 0.03wt% of graphene to concrete, the carbon footprint is reduced by up to 33% and the global carbon emissions by 2% (Critchley, 2021).

Literature review

There are many studies investigating the use of graphene as a nano filler, including its use in ceramics, concretes, and polymers.

A study by *Yin et al. (2019)* on the mechanical and ballistic resistance of graphene platelets within boron carbide (B₄C) ceramic armour found that the addition of graphene platelets to the B₄C ceramic improved the mechanical properties of the material, including its fracture toughness, elastic modulus and hardness. The study showed that there was a 23% improvement in fracture toughness in comparison with B₄C ceramic without the addition of graphene platelets. This was attributed to the inhibition of crack propagation of the graphene platelets. The penetration depth of the B₄C ceramic armour compared to commercial silicon carbide (SiC) was found to be the smallest, with the graphene improving the ballistic performance by up to 50% (Yin et al., 2019). This study also showed that the network structure formed by the graphene within the ceramic matrix enhances the interface bonding and thus results in increased crack bridging and deflection.

The results of a study by *Wang et al. (2019)* showed that the addition of graphene oxide (GO) to alumina significantly improved the toughness and strength of the composite material. *Wang et al. (2019)* stated that “graphene oxide (GO) is one of the most important

ceramic- toughening candidates due to its outstanding physical and chemical properties” (Wang et al., 2019).

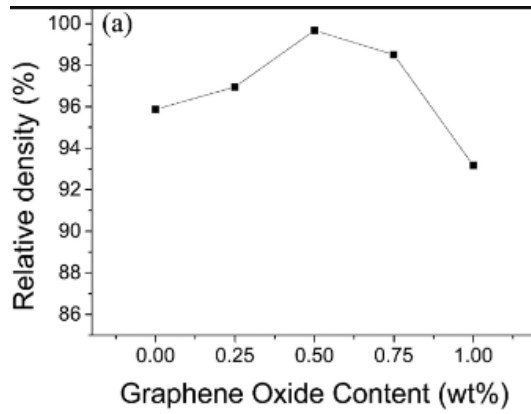


Figure 13 Relative density graph (Wang et al., 2019)

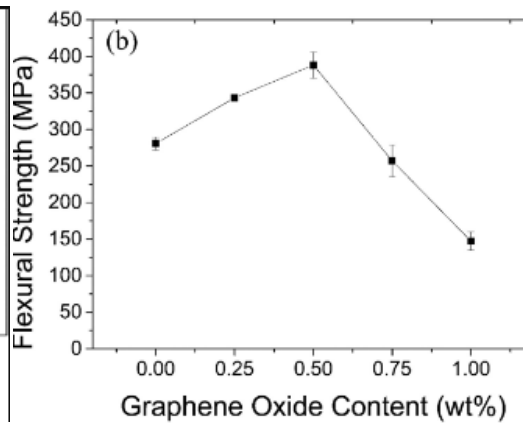


Figure 12 Flexural strength graph (Wang et al., 2019)

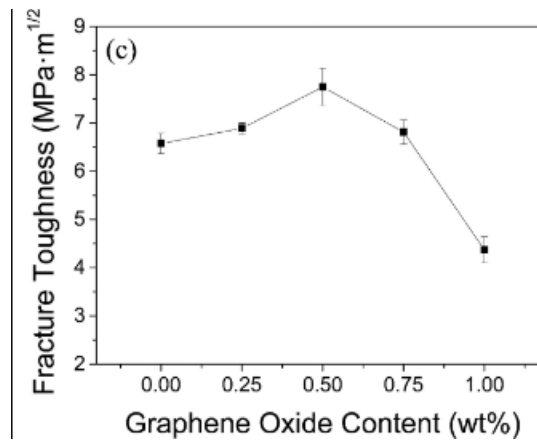


Figure 14 Fracture Toughness graph (Wang et al., 2019)

Figure 12-14 above show that a 0.5% graphene oxide content exhibits the best characteristics, with all three properties showing a decrease in performance with any further quantity added. Wang et al. (2019) states that this is likely due to the graphene oxide agglomerating at higher content levels, and therefore acting as defects and decreasing the mechanical properties and also potentially being the source of cracks. Upon closer inspection using scanning electron microscope (SEM), various toughening mechanisms were observed. This included crack deflection, bridging, and branching. Figure 15 below shows micro-cracks zigzagging the crack propagation path, and increased amount of work of the fracture.

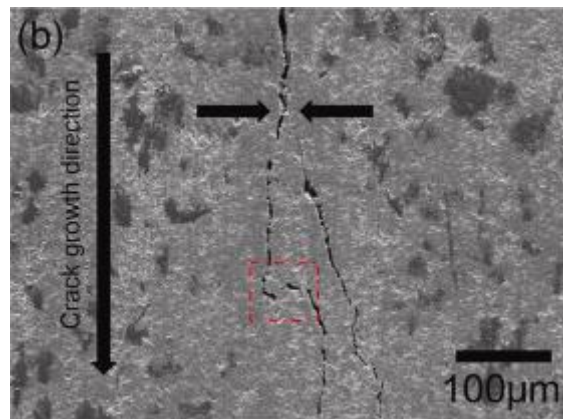


Figure 15 crack propagation path (Wang et al., 2019)

The main toughening mechanism observed in this study was crack bridging. The graphene oxide sheets in the composite bridged the cracks and prevented them from propagating further. However, this mechanism was found to be dependent on the distribution and orientation of the graphene oxide sheets in the composite.

A study by *Chen et al. (2021)* showed that the addition of graphene to the alumina matrix can significantly improve the fracture toughness and inhibit crack propagation in the composite ceramics. It was also found that the size and content of the graphene particles played a crucial role in determining the crack propagation path and the resulting mechanical properties of the composites, with larger diameter graphene providing a stronger interface and an improved toughening effect. A stronger interfacial bonding force showed hindered crack propagation. The study also found that optimum graphene content is 0.5% volume, with a higher content causing the graphene sheets to agglomerate, leading to decreased crack deflection and increased brittleness of the ceramic composite. A low graphene content, however, did not provide enough bridging effect to effectively inhibit crack propagation (Chen et al., 2021).

Walker et al. (2011) found that the introduction of graphene platelets in a silicon nitride ceramic composite increased the fracture toughness by 235% at 1.5% graphene volume. The graphene platelets were observed to act as barriers to crack propagation and also to promote crack deflection and bridging, thereby enhancing the toughness of the material. It was also found that the uniform dispersion of the graphene platelets had a significant effect on the improvement of the fracture toughness of the composite. The uniform dispersion ensures that the platelets are present during crack initiation and can effectively strengthen the material. Non-uniform dispersion of the graphene platelets showed a significant reduction in the fracture toughness compared to uniform dispersion. Figure 16 below shows

the presence of the toughening mechanisms as a result of the addition of graphene.

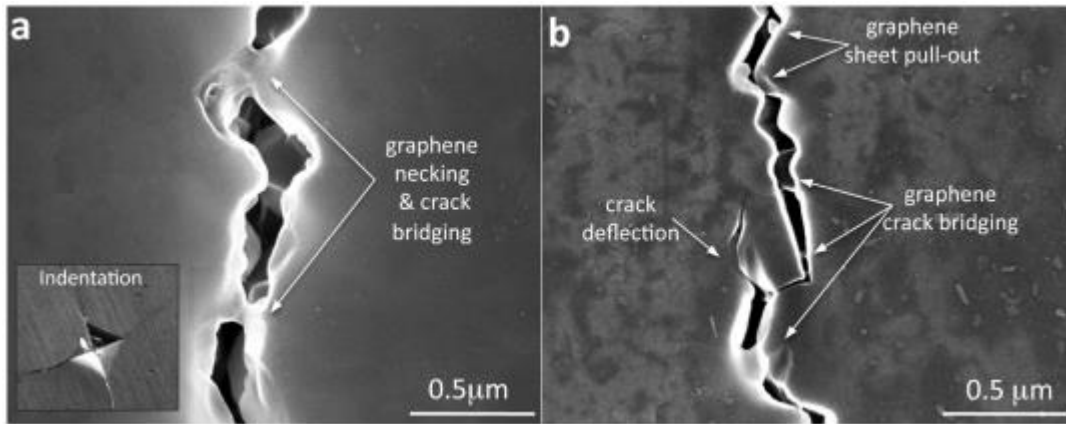


Figure 16 Toughening mechanisms found in a silicon nitride ceramic composite (Walker et al., 2011)

Similarly, Wang et al. (2019) found that a $\text{Al}_2\text{O}_3\text{-WC-TiC}$ ceramic composite reinforced by graphene had improved mechanical qualities, including increased fracture toughness, flexural strength, and hardness. Figure 17 shows that the flexural strength and Vickers hardness were highest at 0.5% graphene volume, after which the improvements were either lost or the graphene actively decreased the mechanical performance of the composite (Wang et al., 2019).

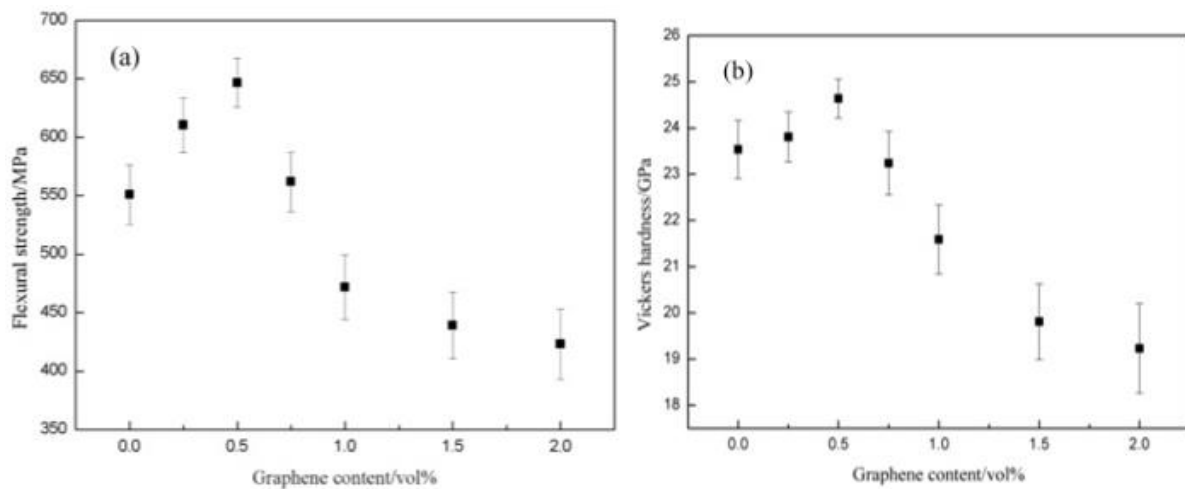


Figure 17 Flexural strength and Vickers hardness of ceramic composite found by Wang et al. (2019)

In agreement with the previous studies, toughening mechanisms such as crack deflection, crack branching and pull-out of graphene platelets were observed. Crack deflection is also observed in figure 18. The large specific area of graphene allows it to contact with different ceramic grains, thus increasing the amount of weak bonding surfaces. These surfaces tend to dissipate more fracture energy and were found to improve the fracture toughness. It was

also found that an abundance of weak interfaces negatively impacts the structure of the composite, and decrease the mechanical performance, as shown in the flexural strength graph above.

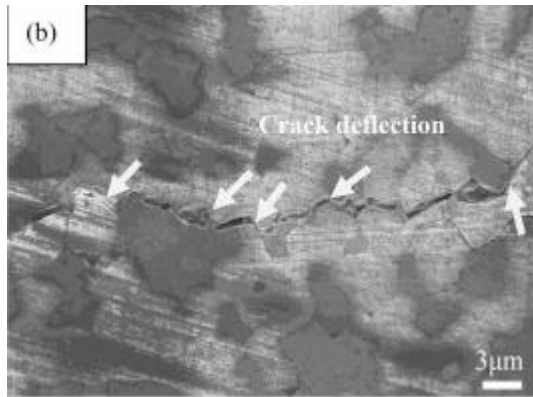


Figure 18 Crack deflection in a ceramic composite found by Wang et al (2019)

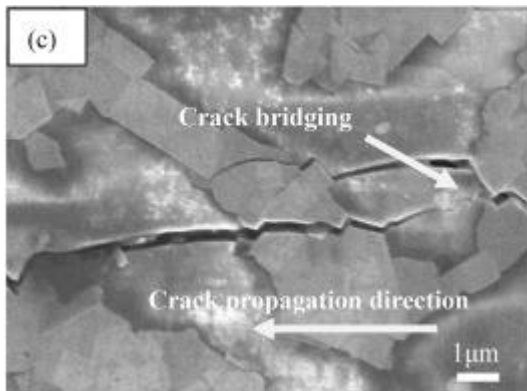


Figure 19 Crack bridging found by Wang et al. (2019)

It was observed that the crack bridging in figure 19 shows that, once a crack reaches an area containing graphene, which has a high tensile strength and Young's modulus, the stress concentration around the crack tip is disrupted and the crack changes direction and propagates along the path of weaker interface which is between the graphene and ceramic. This results in crack bridging and will consume more fracture energy, and therefore improve fracture toughness. This study provides conclusive evidence of the toughening mechanisms that graphene can provide when added to ceramic composite.

Pan et al. (2015) carried out a thorough study on the addition of graphene oxide to ordinary cement. They reported that the addition of 0.05wt% graphene oxide improved the mechanical properties and microstructure of the composite considerably. The results of their testing showed significant improvement in compressive strength, which was increased by 15-33%, and flexural strength, which was improved by 41-59%. The strength of cementitious

materials is related to the age of it, as seen in figure 20 below. These improvements were thought to be caused by higher interaction between the cracks and the graphene due to the 2D geometry and high aspect ratio of graphene, and the formation of strong interfacial forces due to chemical reactions within the material (Pan et al., 2015).

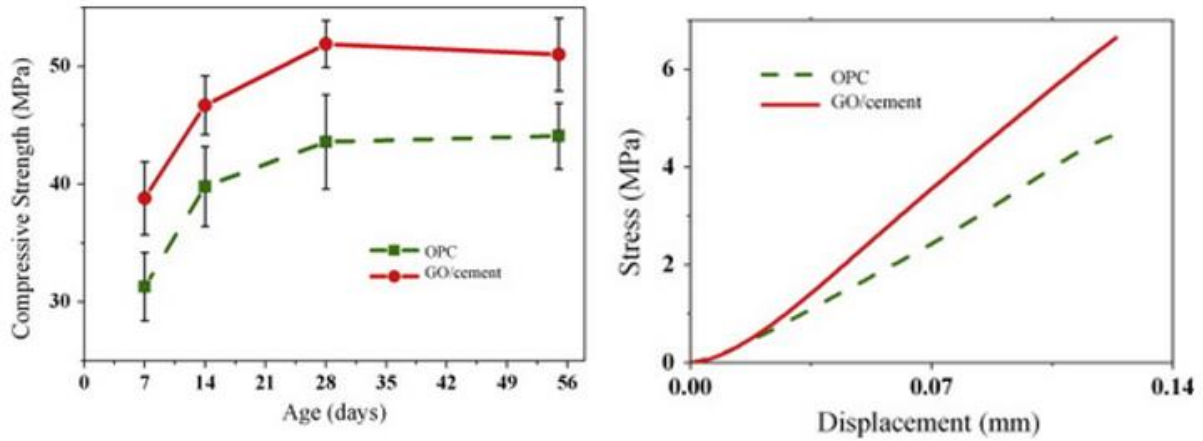


Figure 20 compressive strength and displacement charts (Pan et al., 2015).

Figure 20 also shows the stress/displacement curve, clearly displaying the strengthening effect the graphene oxide had on the material. Scanning electron microscope analysis was performed (figure 21) showing cracks passing as normal through the plain cement example (a), whilst the sample with graphene oxide, example (b), showed much finer cracks with considerable discontinuity, showing evidence of crack bridging.

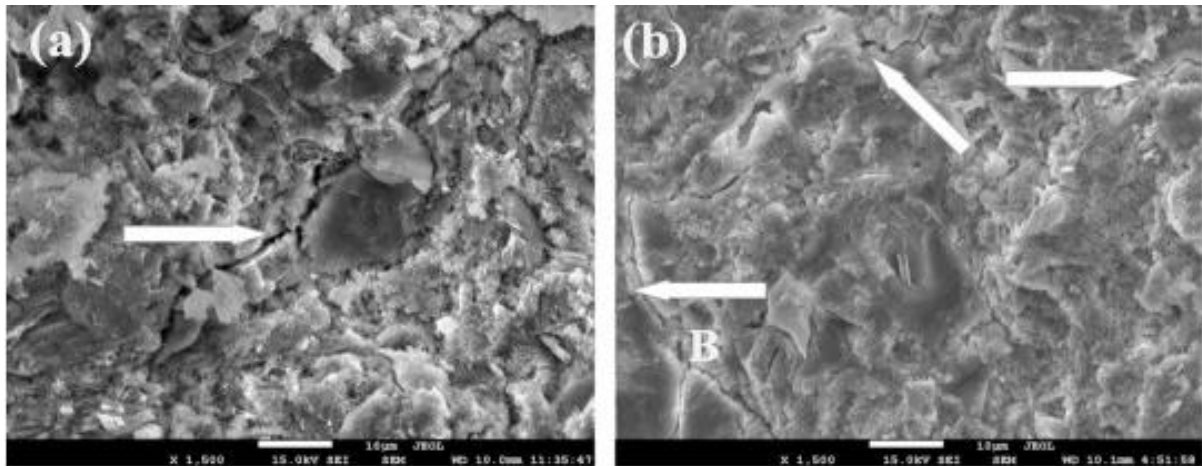


Figure 21 example (a): plain cement sample. Example (b): sample with graphene (Pan et al., 2015)

Lv et al. (2013) also studied the effect of graphene oxide on cement composites, carrying out tests with varying graphene oxide contents. Their results showed that the addition of graphene oxide had a strong toughening effect on the cement composite, with the highest

increase in properties occurring at 0.03% graphene content, beyond which the improvements were not as great. At this content, the tensile strength increased by 78.6%, flexural strength by 60.7%, and compressive strength by 38.9%, when compared to samples without graphene. It was concluded that these results were due to the graphene effectively regulating the microstructure of the cement, reducing its brittleness, whilst also improving the toughness. Figure 22 shows that, as seen in the study by *Pan et al. (2015)*, the strength improved with the age of the composite.

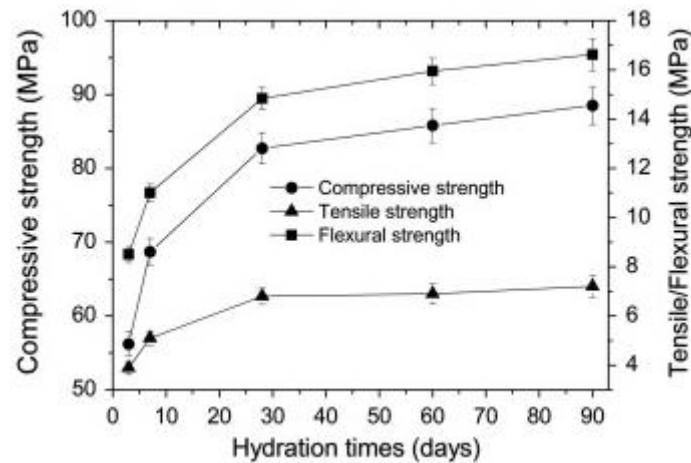


Figure 22 Compressive strength against hydration time (Lv et al., 2013)

Gholampour et al. (2017) also found that the addition of graphene oxide to cement mortar improved the mechanical properties, revealing that graphene oxide helped prevent crack propagation. Eight different dosages of graphene oxide were added, shown in figure 23, and found that the highest increase in mechanical properties were with a 0.1% graphene oxide content. Any further increase resulted in detrimental effects such as micro-cracks, which were related to the restacking and aggregation of graphene oxide sheets in the cement matrix. After 28 days, the tensile and compressive strength were increased by 37.5% and 77.7% respectively. It was also noted that the growth rate of the strength after three days was generally higher than that of 28 days, indicating that there is a more significant strengthening effect during the early stages (*Gholampour et al., 2017*). This study validates the positive influence of graphene oxide as a nano reinforcement method.

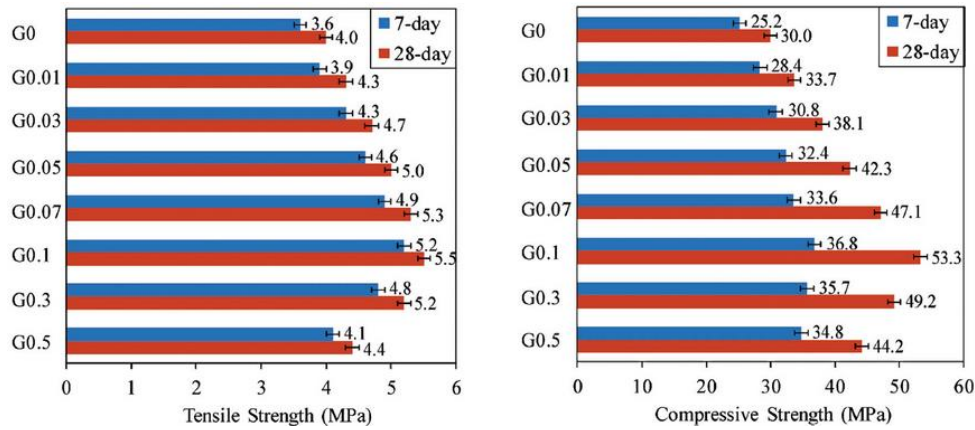


Figure 23 Tensile strength and compressive strength of samples found by Gholampour et al. (2017)

Heng et al. (2019) investigated the effect of the incorporation of graphene nanoplatelets to polyurethane elastomer and found that there were significant improvements in the mechanical properties of polyurethane, with an improvement of 81.1% in tensile strength at 1% graphene volume. Their studies revealed that there was strong interaction between the graphene nanoplatelets and polyurethane, allowing the effective load transfer between the two materials. Various volumes of graphene nanoplatelets were tested, with the most significant improvements made by 1% graphene nanoplatelet volume. However, it was also found that there was an inverse relationship between the volume of graphene nanoplatelets and the elongation at break, with a 31% reduction at 1% graphene volume. This was said to be the result of a reduction in elastic deformation due to the covalent bonding between the graphene nanoplatelets and the polyurethane matrix (Heng et al., 2019).

Literature summary

The above studies demonstrate that the addition of graphene to various composite materials, including ceramic, concrete and polymers, improves their mechanical properties. These studies found that the addition of graphene platelets to ceramic materials such as boron carbide (B^4C), alumina, and silicon nitride can improve their fracture toughness, elastic modulus, and hardness. The toughening mechanisms observed in these studies include crack deflection, bridging, and branching, which are dependent on the distribution and orientation of the graphene sheets in the composite. It was also found that the size and content of the graphene particles play a crucial role in determining the crack propagation

path and the resulting mechanical properties of the composites. Additionally, the uniform dispersion of the graphene is essential for improving the fracture toughness of the composite, as non-uniform dispersion showed a significant reduction in the fracture toughness. The optimum graphene content in these composites is typically around 0.5% volume, with higher or lower contents decreasing the effectiveness of the graphene in improving the mechanical properties.

In conclusion, the addition of graphene has been proved to enhance the mechanical properties of various materials and should theoretically benefit Art of Eco's product. However, the distribution and reaction with the materials can have a significant impact on the effectiveness of the graphene (Wang et al., 2019), and further studies to determine the relationship between graphene and the materials used in Art of Eco's products are suggested as further work. Additionally, Further studies to investigate the impact of varying volumes of graphene would be beneficial. Currently Art of Eco use roughly 0.2% volume graphene in their products, which may not be optimal.

Many of the studies used microscopic methods to analyse the effect of graphene on the structure of the materials. This was not possible in the experiment carried out due to lack of availability of equipment and it would be highly beneficial to carry out further work using such methods.

Methods

In order to test the hypothesis that the addition of graphene would strengthen the tiles, a three-point bend test was carried out following the standard ISO 10545-4:2019, which determines the modulus of rupture and breaking strength of the test subjects. There are several other standards relating to the bending strength of ceramics, such as ASTM C1161 and ASTM C1499, which are both American standards to obtain the flexural strength of a sample. However, these standards are aimed at advanced ceramics and were not considered as relevant as the ISO 10545-4:2019 standard used. The ISO 10545-4:2019 was also chosen as it could be replicated easily for the Art of Eco samples.

Type of bend test

Initially, there were two different types of tests considered for this experiment: a three-point bend test and a four-point bend test. Three-point bend testing is easier to carry out, as it requires a simpler testing jig. The simple nature of the test jig makes it less prone to errors in the geometry and alignment of the jig. It is the more common of the two tests and is usually used by industrial laboratories for quality assurance purposes. Although four-point bend tests require a higher degree of attention to detail, it is becoming more common due to a larger area of the test sample being subject to a more uniform stress (Morrell, 2007). The three-point test was chosen due to the simple nature of the jig required, and it also complies with the ISO 10545-4:2019 standard followed.

Three-point bend test

A three-point bend test is a mechanical test commonly used to evaluate the flexural strength of material. It involves applying a load at the centre of a usually rectangular or cylindrical sample that is supported at either end by fixed points or supports. As the load is applied, the sample bends and undergoes compression on the top side, and tension on the bottom side. Figure 24 below depicts a standard three-point bend test.

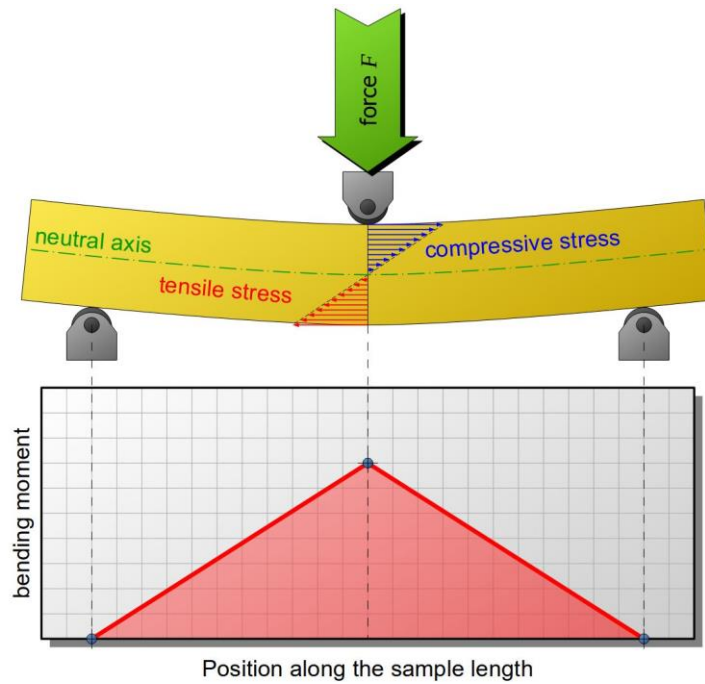


Figure 24 Three-point bend test diagram (Tec-Science, 2022)

Stress and strain forces also act on the sample, which can then be measured. In this report, the three-point bend test is used to test ceramic tiles and samples from Art of Eco, with and without graphene, in accordance with the ISO 10545-4:2019 standard. During testing the sample will undergo elastic and plastic deformation. In the elastic deformation stage, the tile will bend and deform, but will return to its original shape once the load is removed. In the plastic deformation stage, the tile will bend beyond its elastic limit, and it will be permanently deformed. There are various parameters that can be determined from a three-point bend test.

Both the ceramic tiles and Art of Eco samples are classed as a non-ductile material. A property of materials of this type is that any small cracks or flaws in the material will act as stress concentrators, with the material around being unable to sufficiently flow to blunt the local stress. The result of this is that, once the tensile or shear stress reaches a certain level, the area with the highest stressed crack or flaw begins to propagate and is then difficult to stop spreading further (Morrell, 2007).

In theory and as described in the literature, the addition of graphene should enhance both the modulus of rupture and breaking strength of the tiles produced by Art of Eco by enhancing the mechanical properties and providing toughening mechanisms such as crack deflection, bridging, and branching. Art of Eco's tiles were tested against two types of ceramic tiles: ceramic wall tiles and ceramic floor tiles. The ceramic floor tiles are

manufactured using a higher temperature kilning process and should therefore exhibit stronger mechanical properties. Art of Eco have no previous information on whether their tiles are stronger or weaker than traditional ceramic tiles. The experimental methods and procedures are described below.

Preparation of materials

Firstly, a jig was designed to carry out the bend testing. This required the size of the testing sample to be determined.

Due to the nature of the production process used by Art of Eco, the tiles produced are of large size (2.2mx1m) and then cut down to the required size. ISO 10545-4:2019 standard states that when L (the long side of the tile) is >600mm, the tiles should be cut to maintain their original aspect ratio. Due to the limited test area of the Dennison machine, the tiles were cut to 500x200mm.

Once the tile dimensions were determined, the jig dimensions were set in accordance with the standard, which states that if the long side of the tile is over 95mm, then the diameter of the support rods must be $20\pm 1\text{mm}$, the rubber thickness $5\pm 1\text{mm}$ and the overlap of the tile beyond the edge of the supports $10\pm 5\text{mm}$. The dimensions for the jig produced was a 500x220mm plate, with the support rods 480mm apart from the centre of the support rods.

The jig was then modelled using Solidworks 2022. Figures 25 and 26 show the various angles of the model created in Solidworks.



Figure 25 Side view of the lower part of the jig in Solidworks

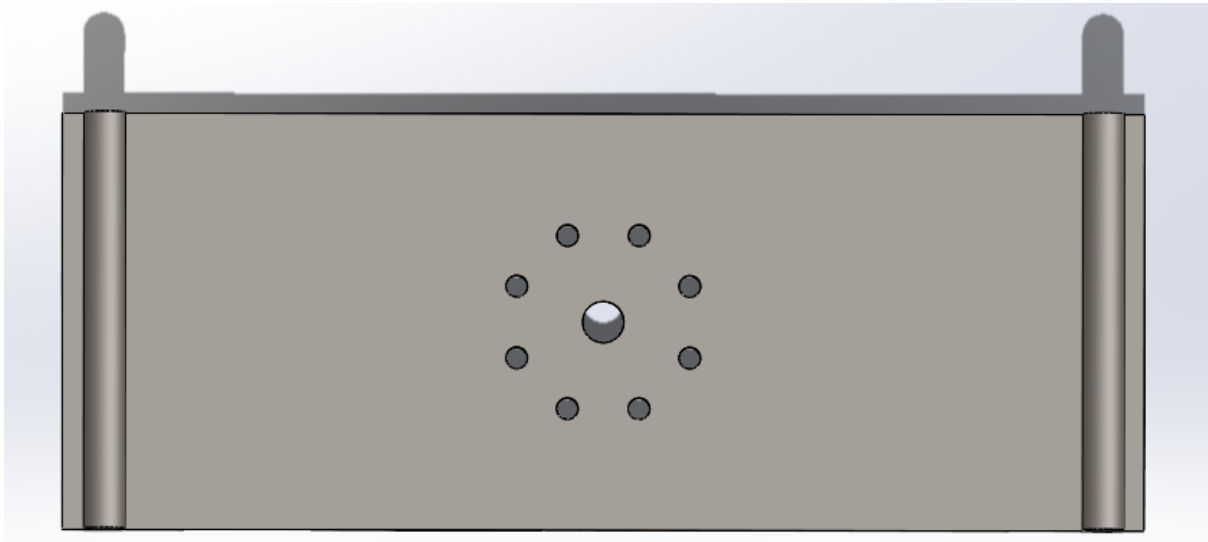


Figure 26 top view of the lower part of the jig in Solidworks

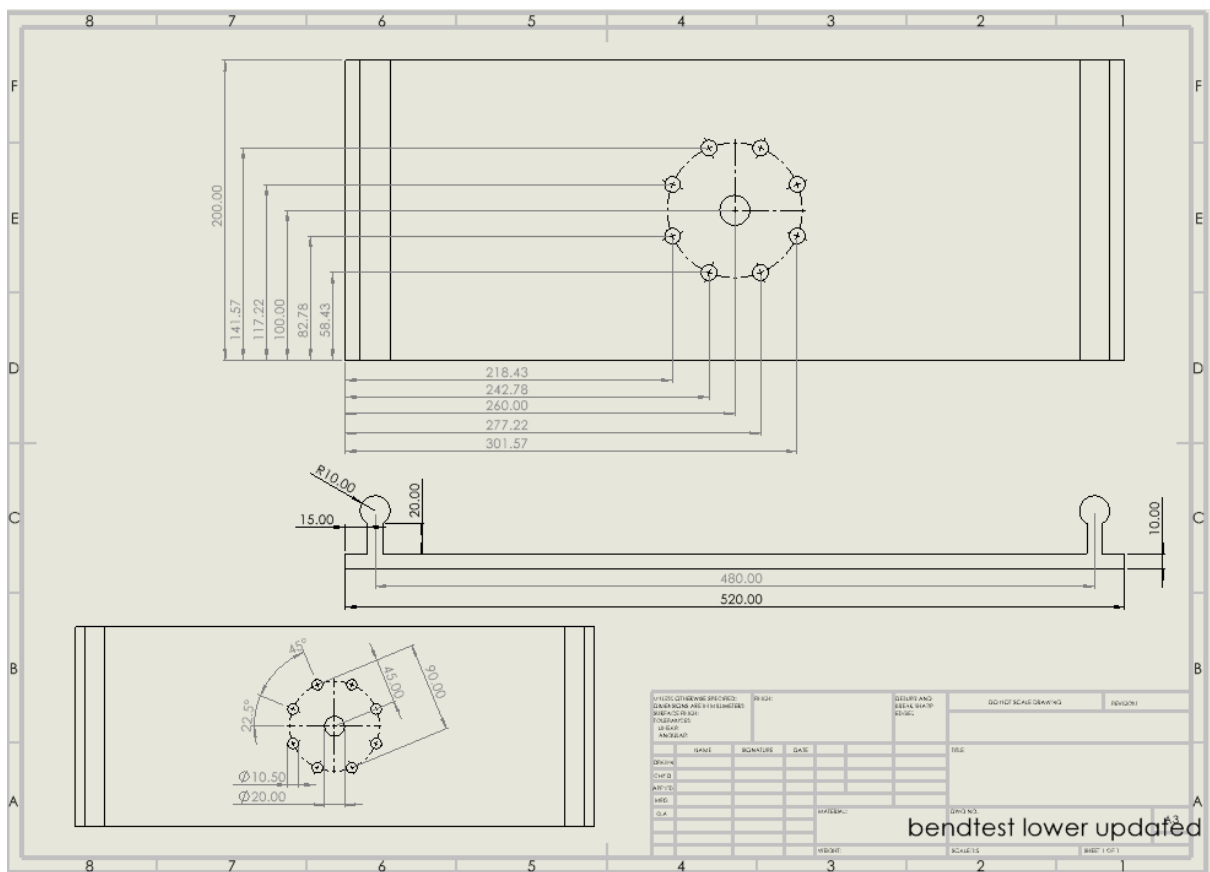


Figure 27 Technical drawing of jig lower

Figure 27 shows the technical drawing which details the dimensions of the lower part of the test jig. The supports were placed 480mm apart on a base of 520mm. The supports were

raised by 20mm to allow for the tiles to bend without any possibility of tiles bottoming out on the bolts or any other part of the jig. The support rods were given a 10mm radius. The holes seen in the middle of the jig enabled it to be bolted to the Dennison machine. Eight holes were used in the same pattern as on the Dennison; the holes were 10.5mm in diameter to allow for a clearance fit for the M9 bolts used to attach it to the Dennison. The holes were arranged in a 90mm PCD from the centre of the jig, allowing the holes to be 25° apart, matching the layout of the Dennison. The base of the jig was 10mm thick to ensure the structure did not deform in any way under load.

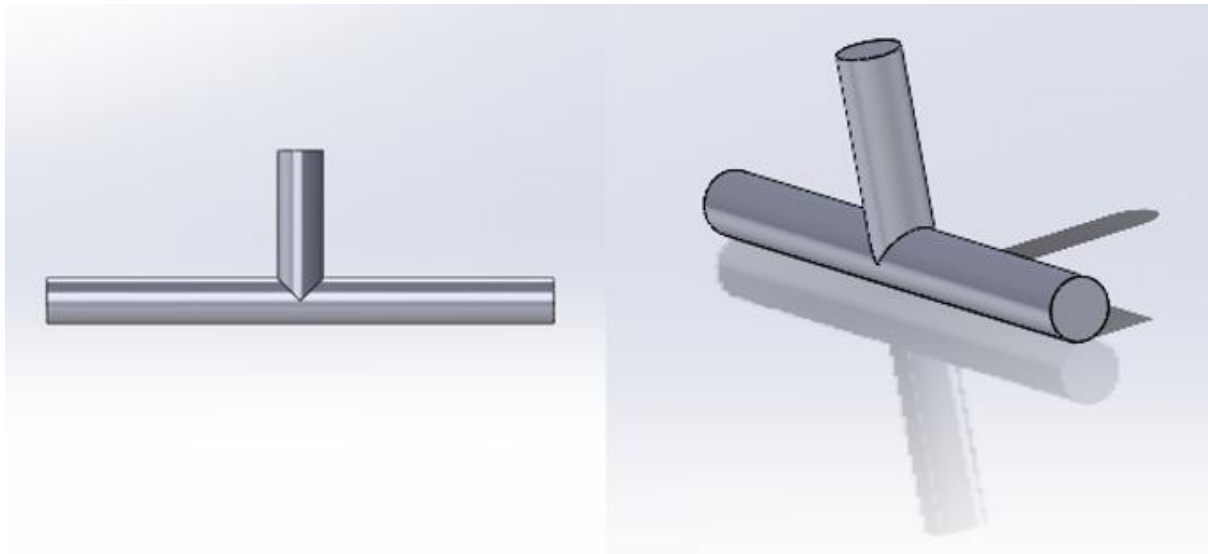


Figure 28 Solidworks model of top part of the jig

Figure 28 shows model of the top part of the jig in Solidworks.

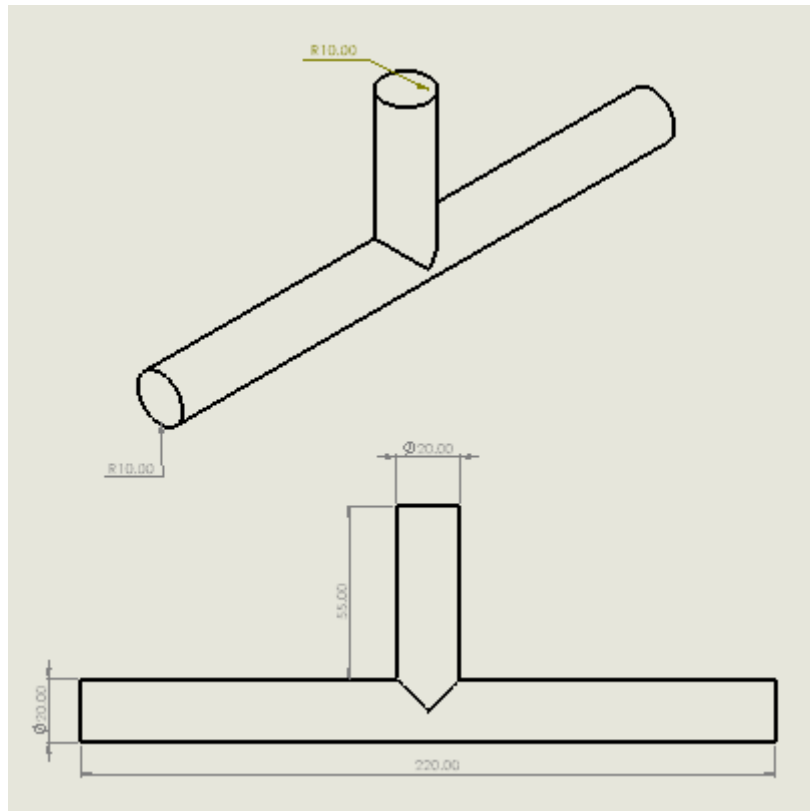


Figure 29 Technical drawing of the top part of the jig

Figure 29 shows the technical drawing produced from the Solidworks model for the top part of the jig. The width of the part was 220mm to match the bottom part of the assembly, with a 20mm diameter to satisfy the standard. The vertical section was also set to 20mm diameter to fit the Dennison. The height of the vertical was set to 55mm to allow maximum contact area with the Dennison, also with a diameter of 20mm.

Once the technical drawings were completed, they were sent to the university workshop where the parts were manufactured using mild steel.

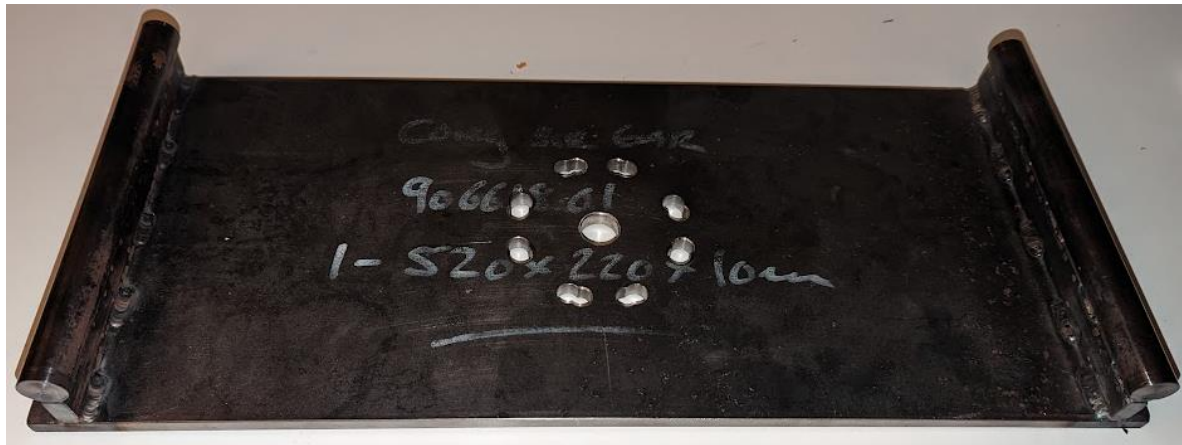


Figure 30 completed bottom part of the jig

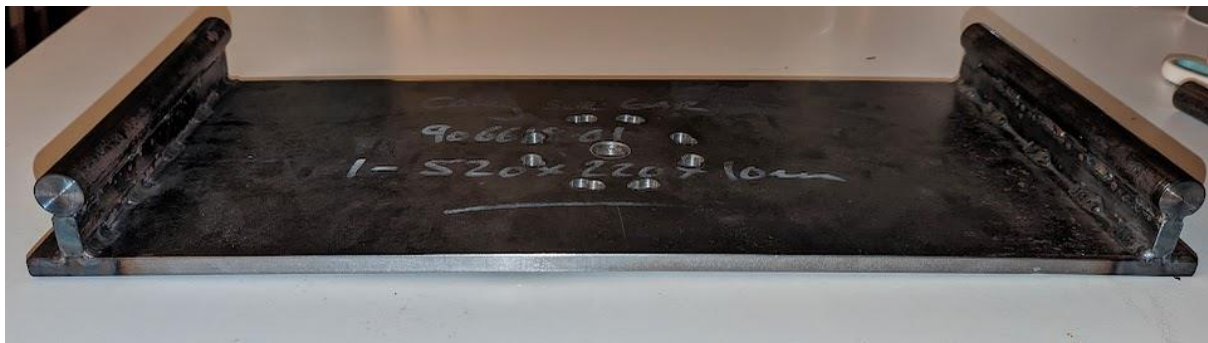


Figure 31 Side view of completed bottom part of the jig

Figures 30 and 31 above show the finished lower part of the jig. Figure 30 shows the slot-like holes which were a result of having to redrill the holes due to an error when measuring the layout on the Dennison machine. Once the jig was finished, rubber strips were applied to the contact surfaces as required by the standard. However, the hardness was not measured to the same requirements as the standard, as stated in the limitations. The rubber is applied to reduce the likelihood of high stress occurring at the contact points (due to uneven surface of the samples or other contaminants). Figure 32 shows the rubber in place on the lower jig support.



Figure 32 Jig support with rubber applied.

The number of tiles to be tested was determined by Table 3 from the standard, as shown below in Table 1. The Art of Eco tiles had an area of 100,000mm² and therefore a total of 7 of each tile were tested. However, one standard Art of Eco sample (sample 1) was damaged during transport and two additional samples were produced (samples 8 and 9) to replace it. This resulted in 8 standard samples being tested and 7 graphene samples. Due to a lack of availability, a total of only 5 ceramic floor tiles were tested. This quantity still complied with the standard, which states a minimum of 5 samples should be tested for tiles within this size group.

Table 1 Number of specimens required.

Table 3 — Minimum number of test specimens for tiles with a work size thickness <7,5 mm

Maximum area A mm ²	Number of specimens to be tested	Total number of tiles	Total number of specimens	Minimum number of acceptable results
$2\,500 < A \leq 40\,000$	1	7	7	5
$40\,000 < A \leq 360\,000$	1	7	7	5
$A > 360\,000$	3	3	9	8

The thickness of the tiles was measured before the testing, due to the large variation in size. Four measurements were taken from each of the long sides of the tiles and an average

calculated for each, shown in the table 2 below. The width of each Art of Eco tile was also measured before testing. The average width of both types of tiles was 200mm.

Table 2 Average thickness of Art of Eco standard and graphene samples

Average tile dimensions AoE standard				Average tile dimensions AoE graphene			
Sample no.	Thickness (mm)			Sample no.	Thickness (mm)		
2	8.89			1	15.075		
3	9.55			2	17.97		
4	12.6			3	15.73		
5	13.34			4	14.87		
6	14.74			5	12.46		
7	15.925			6	12.72		
8	17.55			7	13.11		
9	15.49						
Mean:	13.51063			Mean:	14.56214		
				Avg % thickness increase	7.78%		

The dimensions shown above display a high degree of variation, with a range of 9mm between both sample types. This is likely a result of the manufacturing process, where the screening method used to sort the small grain aggregate in the tiles uses a square mesh grid to eradicate larger particles of aggregate. However, particles can also pass through the mesh diagonally allowing larger particles through. A refined screening process would allow for less variation in particle size, and therefore more consistent results.

Both types of ceramic tile were very uniform, showing a variation of less than 0.2mm across each sample. The average for the ceramic wall tile was measured at 9.2mm, whilst the floor tile was measured at 8.9mm

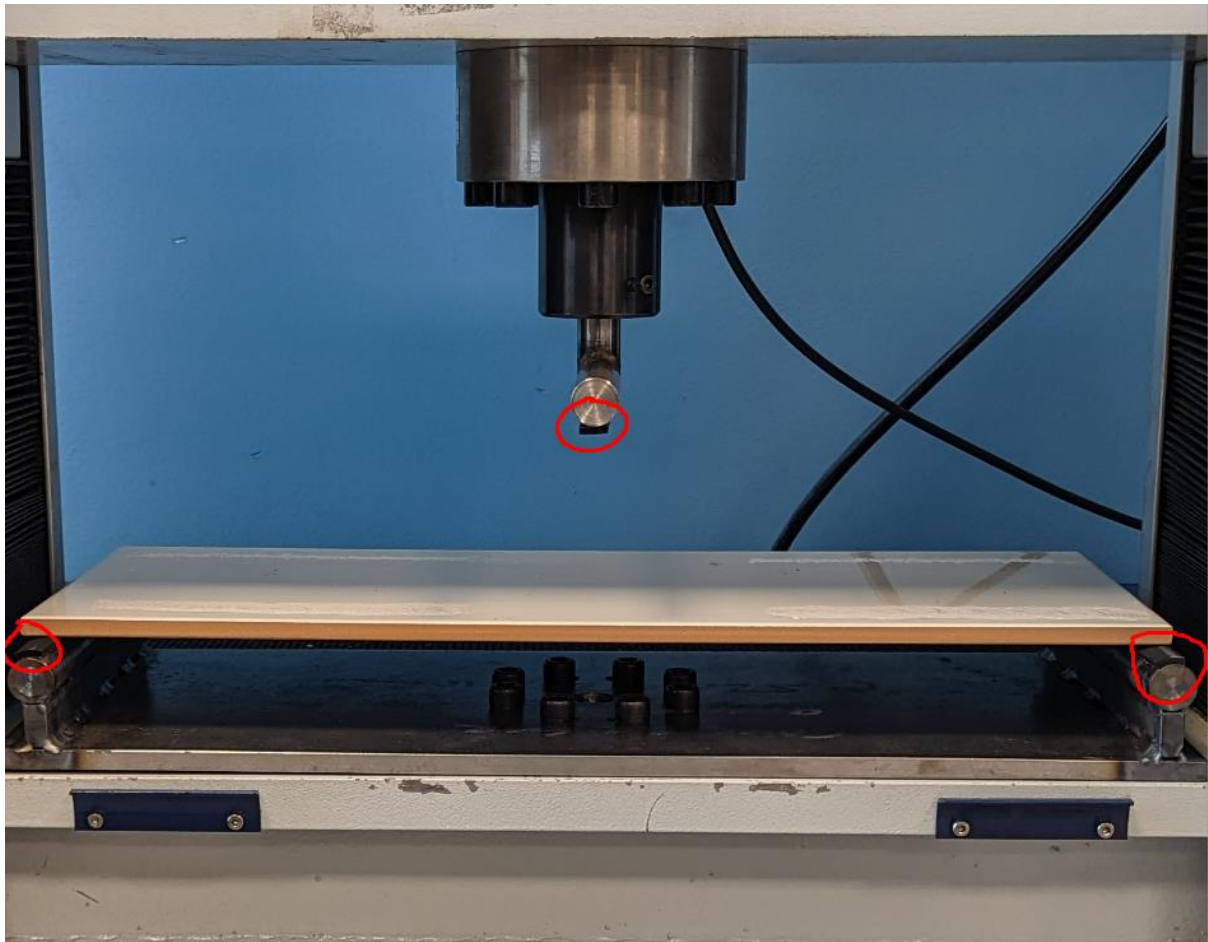


Figure 33 Test setup ready with ceramic wall tile in place

Figure 33 shows the experiment setup with a ceramic floor tile; the red circles show the rubber strips in place.

Experimental procedure

The experimental procedure followed that of the standard ISO 10545-4:2019 whereby the following steps should be taken:

- 1 - Remove any loose particles with a stiff brush
- 2 - Place the test specimen on the supporting rods, with the glazed or proper surface uppermost, with the tile projecting by the specified amount beyond each support rod
- 3 – Position the central rod equidistant between the support rods and apply the load evenly in such a way as to obtain a rate of increase in stress of 1N/mm^2

To calculate the correct load rate, equation (1) was used, where σ = Stress, F = Force and A = area.

Equation 1 Stress formula

$$\sigma = \frac{F}{A}$$

By transposing equation (1), the force required can be calculated.

$$\therefore F = \sigma * A$$

Where $\sigma = 1$

And A= cross-sectional area of specimen.

The area used to calculate the load rate was obtained from a ceramic tile sample with a cross sectional area of 200mm width and 9mm thickness, giving an area of 1800mm².

The force can now be calculated using the above numbers. $F = 1 * 1800$

$$F = 1800N$$

This equates to a load rate of 1.8kN/s or 1N/mm² as per the standard.

Once the tile was in place, the top support rod was lowered manually to create a small gap above the tile but without applying any load.

The Dennison was then set to perform the test at a load rate of 1.8kN/s.

The Dennison automatically records the following parameters: time taken, displacement of the top arm and force applied.

Once the specimen failed, the recorded data was saved, and the top support returned to the starting position. Due to the varying sizes of the Art of Eco tiles, the top support was adjusted manually for each tile.

Error

In any experiment it is important to realise the errors that are introduced during any testing, be it human error or errors that are introduced by measuring equipment. In this experiment, human error was reduced as much as practically possible for activities such as transporting



Figure 34 Graphene sample with imperfections

the tiles and the placement of the tiles on the jig. For example, the ISO 10545-4:2019 standard followed in this experiment stated that there should be 10 ± 5 mm overhang at either end of the tile. However, this was not directly measured and rather measured by eye, which could have introduced errors. The same is true for how square the samples were on the supports, which were also placed by eye. The Art of Eco tiles were measured using Vernier callipers at 4 different points on each of the tile samples due to the high variability of the samples. However, this may still not have been an accurate measurement, with the possibility of bumps on the surface of the tile (such as those shown in figure 34) producing inaccurate readings.

The Dennison machine used was calibrated a few months previously by the manufacturer and should therefore have taken measurements accurately. However, due to the exact accuracy of the measurement system being unknown, it was assumed that the accuracy was to plus or minus one half of the smallest increment of measurement. As the force was measured by the Dennison in 5N increments, this would produce an error of ± 2.5 N.

Table 3 shows the maximum and minimum breaking load for the Art of Eco standard samples, using the error figure above.

Table 3 Minimum, maximum and recorded breaking loads of standard samples

Sample no.	Minimum breaking load (N)	Recorded breaking load (N)	Maximum breaking load (N)	Error %
AoE std 2	127.5	130	132.5	1.92%
AoE std 3	87.5	90	92.5	2.78%
AoE std 4	232.5	235	237.5	1.06%
AoE std 5	122.5	125	127.5	2.00%
AoE std 6	392.5	395	397.5	0.63%
AoE std 7	317.5	320	322.5	0.78%
AoE std 8	387.5	390	392.5	0.64%
AoE std 9	142.5	145	147.5	1.72%

The standard Art of Eco tiles were used here as they had the lowest breaking load, which produced the highest percentage of error at $\pm 2.78\%$. This is a relatively small percentage and, for this test where there were many other variables, that introduced errors and inconsistencies. It was not considered worthwhile to substitute this error into all parameters calculated.

Results

Once the testing was completed, the data recorded were exported into Excel for analysis.

The maximum load applied to each tile was recorded and is shown in the table below, including the average breaking load for each type of tile.

Table 4 Breaking load of all samples

Sample breaking loads				
Sample no.	Breaking load(N)		Sample no.	Breaking load(N)
ceramic wall 1	390		Ceramic floor 1	565
ceramic wall 2	420		Ceramic floor 2	580
ceramic wall 3	420		Ceramic floor 3	605
ceramic wall 4	415		Ceramic floor 4	565
ceramic wall 5	395		Ceramic floor 5	570
ceramic wall 6	395			
ceramic wall 7	410			
Mean:	406.4		Mean:	577.0
Sample no.	Breaking load(N)		Sample no.	Breaking load(N)
AoE std 2	130		AoE Graphene 1	240
AoE std 3	90		AoE Graphene 2	820
AoE std 4	235		AoE Graphene 3	305
AoE std 5	125		AoE Graphene 4	395
AoE std 6	395		AoE Graphene 5	110
AoE std 7	320		AoE Graphene 6	345
AoE std 8	390		AoE Graphene 7	360
AoE std 9	145			
Mean:	228.8		Mean:	367.9
			Increase %	60.81%

Once the maximum force for each tile was calculated, the formula provided in the standard for breaking strength (S) can be used to calculate the breaking strength of each tile.

The formula in the standard states that the breaking strength can be calculated using equation 2 below:

Equation 2 Breaking strength formula.

$$S = \frac{Fl_2}{B}$$

Where: F is the breaking load, expressed in N;

l₂ is the span between the support rods in mm;

B is the short side of the test specimen under testing in mm.

so, for ceramic tile 1:

$$S = \frac{390 * 480}{200} = 936N$$

Therefore, the breaking strength (S) of ceramic tile 1 is 936N.

The same calculation was then performed on every sample. The results are shown below in table 5, showing the breaking strength of each sample in Newtons.

Table 5 Breaking strength of all samples

Breaking strength (S)				
Sample no.				
	Ceramic wall tile	Ceramic floor tile	AoE standard	AoE graphene
1	936	1549.714	312	576
2	1008	1590.857	218.1818	1977.88945
3	1008	1659.429	555.665	728.358209
4	996	1549.714	304.5685	952.763819
5	948	1563.429	948	266.666667
6	948		760.396	840.609137
7	984		936	864
8			348	
9				

The modulus of rupture can also be calculated and is provided in the standard, as it states that:

The modulus of rupture, R, which is expressed in Newtons per square millimetre, is calculated using the equation 3 below.

Equation 3 Modulus of rupture formula

$$R = \frac{3Fl_2}{2Bh^3}$$

where:

F is the breaking force, in Newtons

l_2 is the span between the rods, in mm.

B is the short side of the tile, in mm

h is the minimum tile thickness at the breaking point, in mm

Calculation for ceramic wall tile 1:

$$R = \frac{3 * 390 * 480}{2 * 200 * 9.1^2} = 7.133$$

Thus, the modulus of rupture for ceramic tile sample 1 is equal to 7.133N/mm²

The table below shows the calculated modulus of rupture for each sample, in N/mm²

Table 6 Modulus of rupture (R) in Nmm² of all samples

Modulus of rupture (R) Nmm²				
Sample No.	AoE standard	AoE graphene	Ceramic wall tile	Ceramic floor tile
1		5.11	16.95	29.02
2	7.13	8.22	18.26	29.79
3	4.64	4.86	18.26	31.07
4	8.33	5.58	18.04	29.02
5	2.79	3.19	17.17	29.28
6	7.80	9.37	17.17	
7	4.87	7.67	17.82	
8	4.19			
9	2.32			

Figure 35 below shows the applied force until breaking point, against time, for the ceramic wall and floor tiles. The wall tiles are plotted in red, whilst the floor tiles are plotted in green.

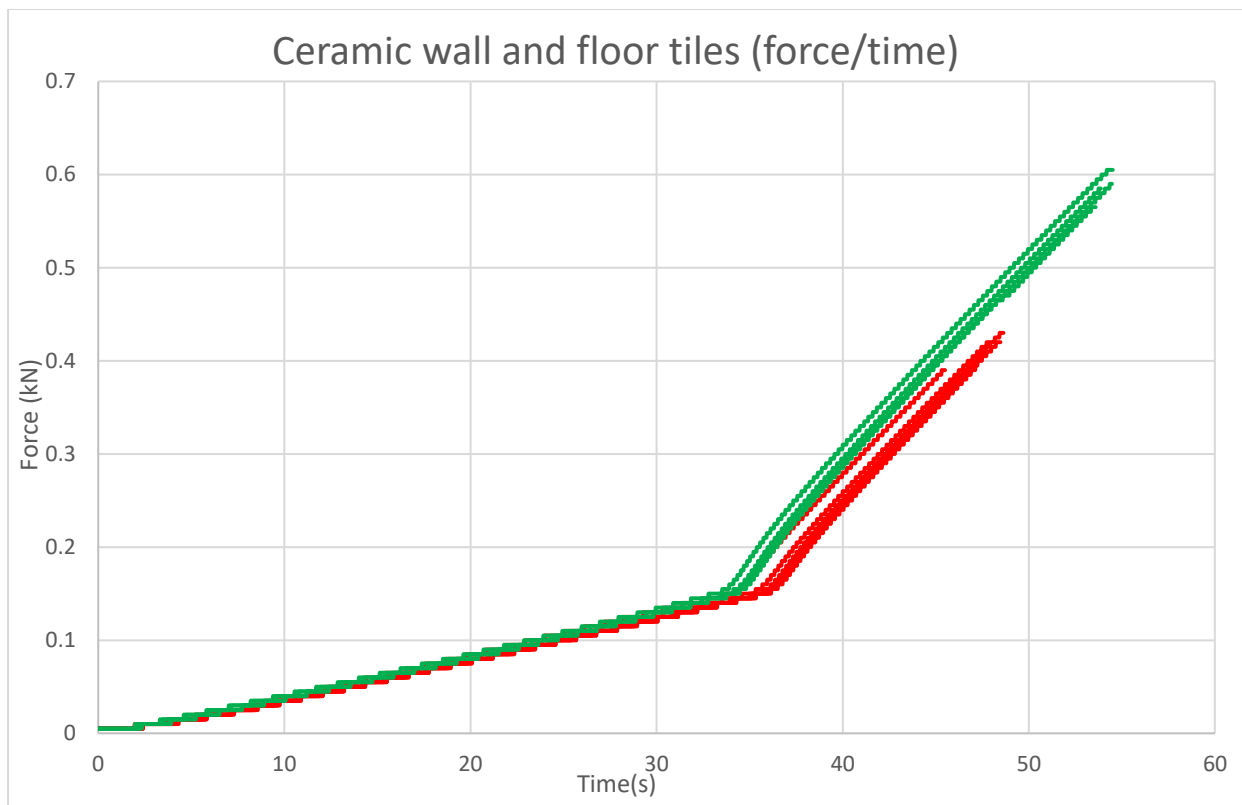


Figure 35 Ceramic tiles force against time graph

The graph in figure 35 shows that the floor tiles were stronger than the wall tiles. This is due to the manufacturing process; floor tiles are manufactured using higher heat during the kilning process and are denser as a result. The graph shows a bilinear pattern to the loading. The shallower gradient is the result of the rubber that was applied to the test jig being compressed. Once the rubber is fully compressed the gradient steepens. The gradient of the graph relates to the stiffness of the material: the steeper the gradient, the stiffer the material. The maximum values for each tile are shown below. The graph shows that both types of ceramic tile demonstrated the same behaviour during testing, as well as breaking in a consistent manner.

The breaking force for each tile is shown below in Table 7, including the mean breaking force.

Table 7 Maximum ceramic tile breaking loads

Sample no.	Maximum load (kN)		Sample no.	Maximum load(kN)	
Ceramic wall tile			Ceramic floor tile		
1	0.390		1	0.565	
2	0.420		2	0.58	
3	0.420		3	0.605	
4	0.415		4	0.565	
5	0.395		5	0.57	
6	0.395				
7	0.410				
Mean:	0.406		Mean:	0.577	

The values in table 7 above show that both types of ceramic tile are consistent in breaking force, showing only a small amount of variation between them (7%). This implies that the structure of the tiles is also consistent, with few weak points.

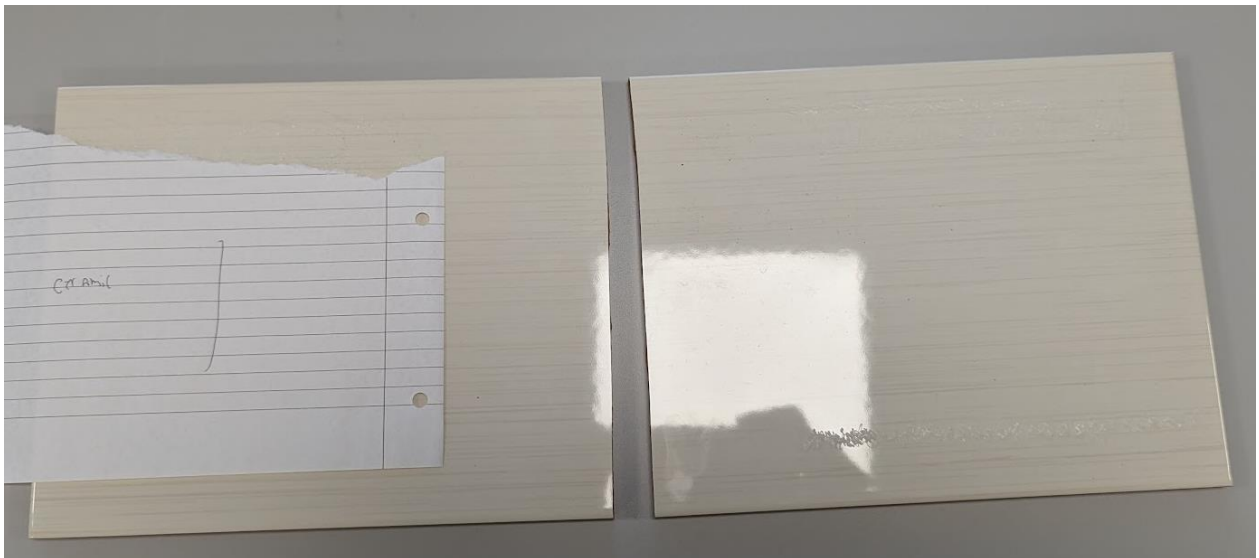


Figure 36 Ceramic wall tile after failure



Figure 37 Breaking edge of ceramic wall tile.

Figure 36 above shows ceramic wall tile sample 1 after failure.

Figure 37 shows the breaking edge of the tile; the uniform dense structure of the tile can be observed.

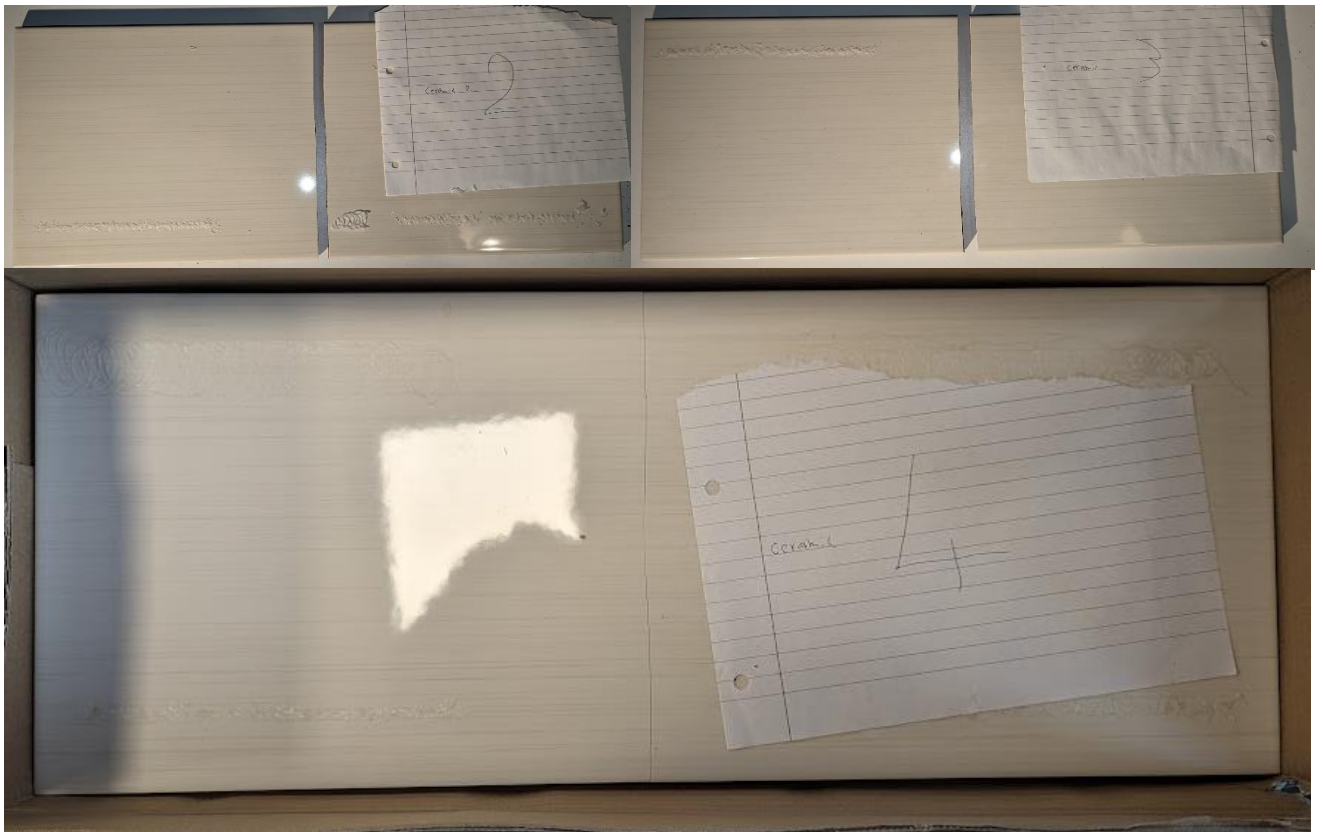


Figure 38 Fracture patterns of ceramic wall tile 2-4 after testing

Figure 38 shows ceramic wall tiles samples 2-4 after failure; the uniform breaking pattern can be observed here.



Figure 39 Ceramic floor tile after failure

Figure 39 shows a ceramic floor tile sample, showing the same uniform breaking pattern as the ceramic wall tile samples. Three of the five floor tiles broke in the same manner, whereas

the remaining two failed in a different manner, producing a fractured breaking edge, as seen in figure 40 below.



Figure 40 Ceramic floor tiles after failure

It is possible that these tiles failed in this manner due to an imperfection in the structure in this area, or due to the top arm of the jig not being able to rotate, applying an increased load at this point, resulting in a stress concentration.

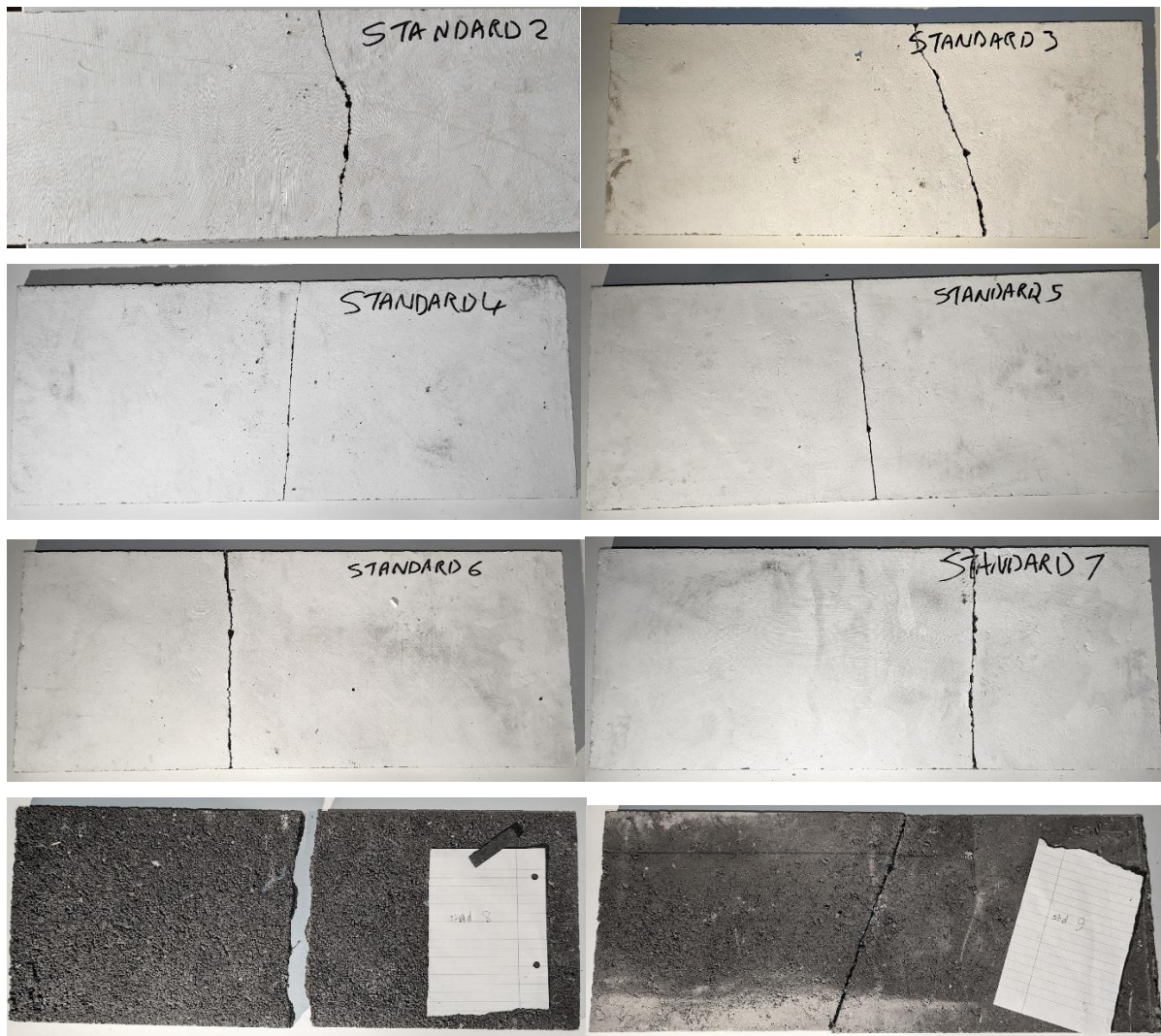


Figure 41 Art of Eco standard samples after failure

Figures 40 and 41 above show the standard Art of Eco tiles after failure, demonstrating a high degree of variation in the location of the failure. The cause is likely to be weaknesses in the material structure at those points. Samples 2-7 were produced at the same time and painted white. Samples 8 and 9 were produced at a different time, without being painted.

Since the tiles were manufactured at different times, there may have been variations in their structural properties. Art of Eco claim that the binding agent used in the tiles strengthens the tiles to 97% of their final strength within 8 hours, and the remaining 3% may take up to 100 years to fully develop. Although this percentage is small, it should still be taken into consideration in further work. Additionally, the binder reacts with the moisture content in the materials, therefore different moisture levels could produce tiles with varying strengths. Another factor that may affect the tiles' strength is the use of different materials during production. In order to minimise the possibility of such variations, it would be beneficial to

produce all samples at the same time, using the same materials. This is suggested as an area for further work.

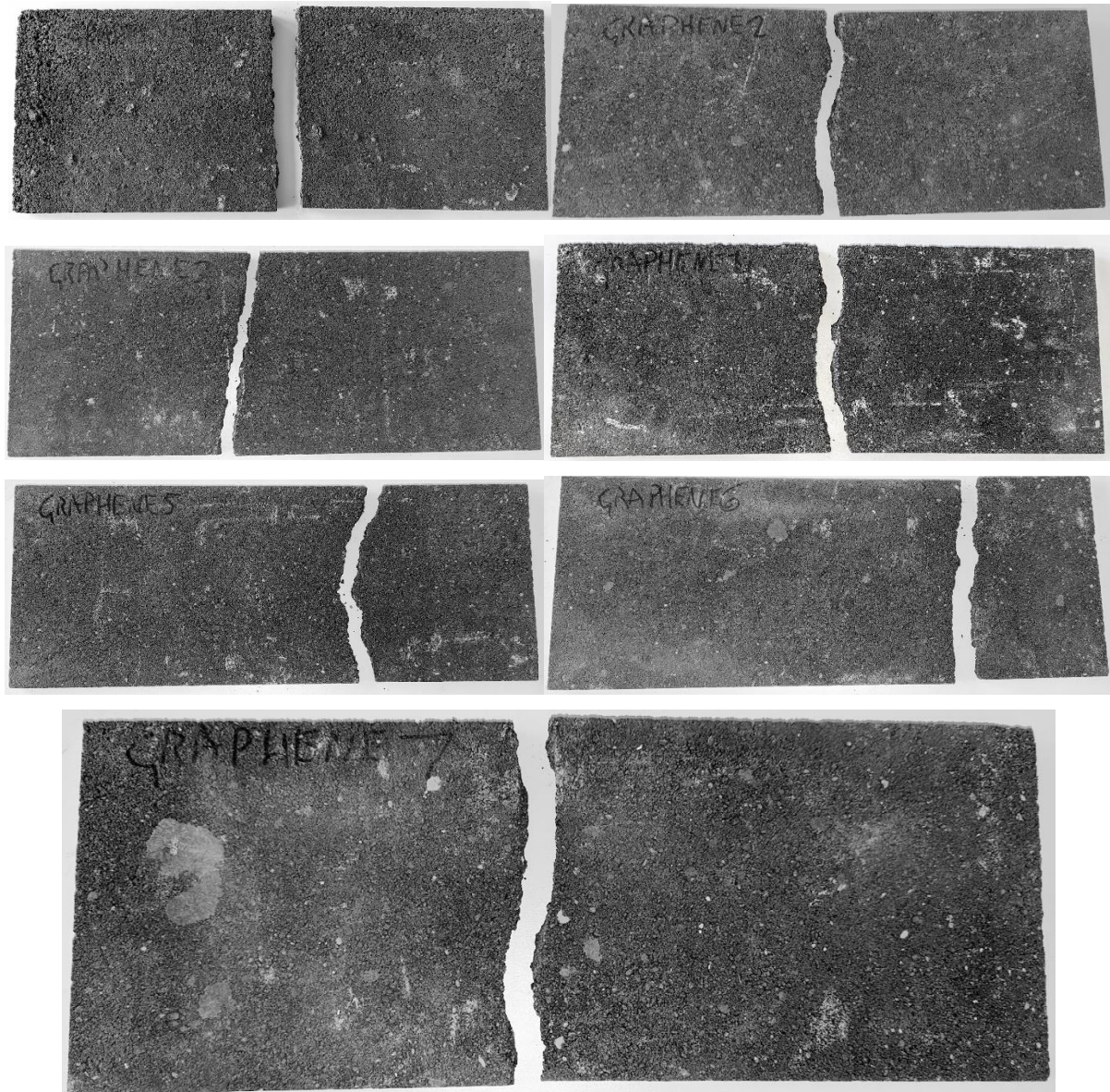


Figure 42 Art of Eco Graphene samples after failure

Figure 42 shows the graphene-enhanced Art of Eco tiles after failure. From Figure 42 it is evident that, as with the standard samples shown in Figure 41, there was also a high degree of variation in the location of failure in the graphene samples. Samples 2, 4 and 7 showed a similar fracture pattern. When comparing the fractures of the standard samples to those with graphene, it would appear that the graphene sample fracture patterns are not as straight as those in the standard samples. This could be caused by the toughening mechanisms observed in the literature, where graphene may have resulted in crack bridging,

causing the crack path to be diverted from the initial straight line fractures seen in both the ceramic tiles and a number of the standard samples. However, it is also possible that there were larger particles of aggregate or other materials in these areas, and the cracks followed weaker areas in the tile structure.



Figure 43 Crack developing in a standard Art of Eco sample.

Figure 43 shows one of the standard Art of Eco samples during testing, with a crack propagating from the convex side of the sample, which would be under maximum tension, hence the crack propagating in this location.

Figure 44 below shows the standard Art of Eco tiles at the fracture point from the side.

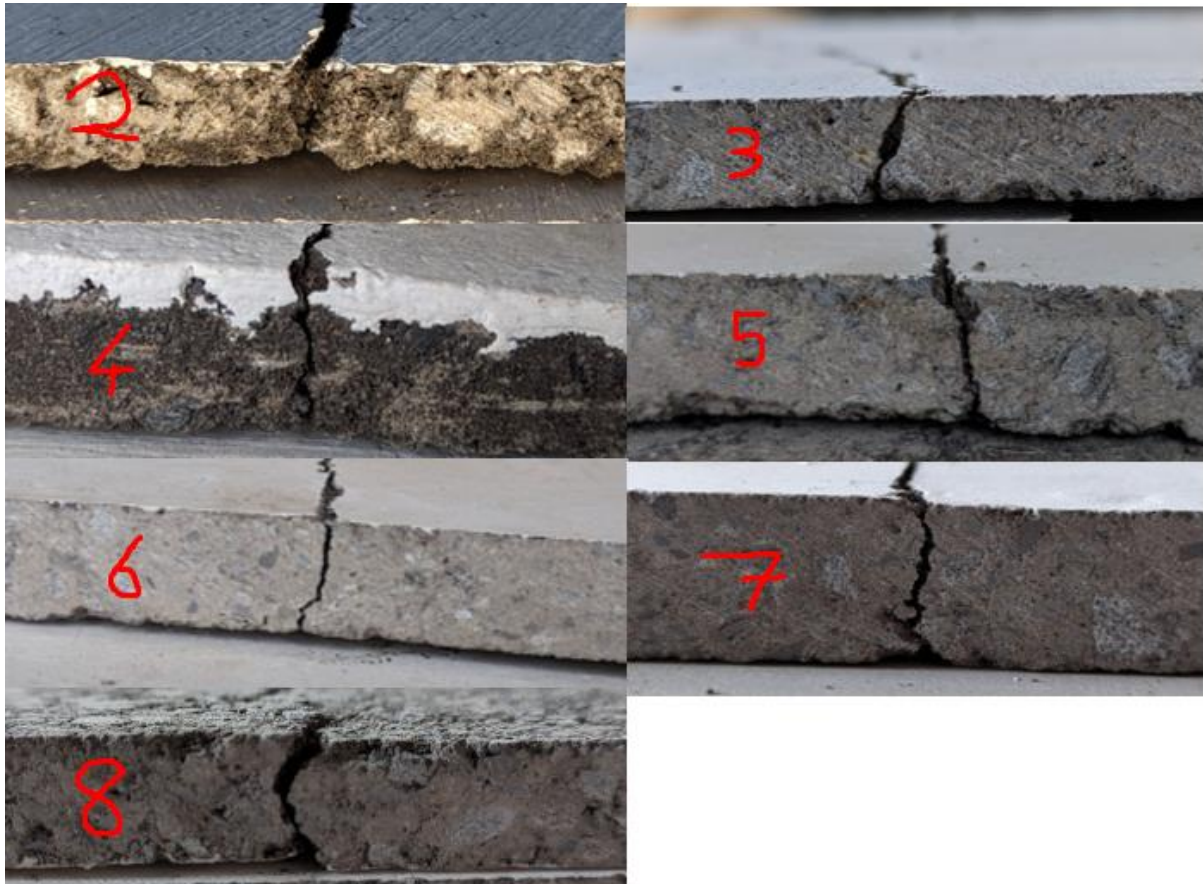


Figure 44 Side view of fracture path of standard Art of Eco samples

Unfortunately, sample 9 was not photographed and is therefore not included.

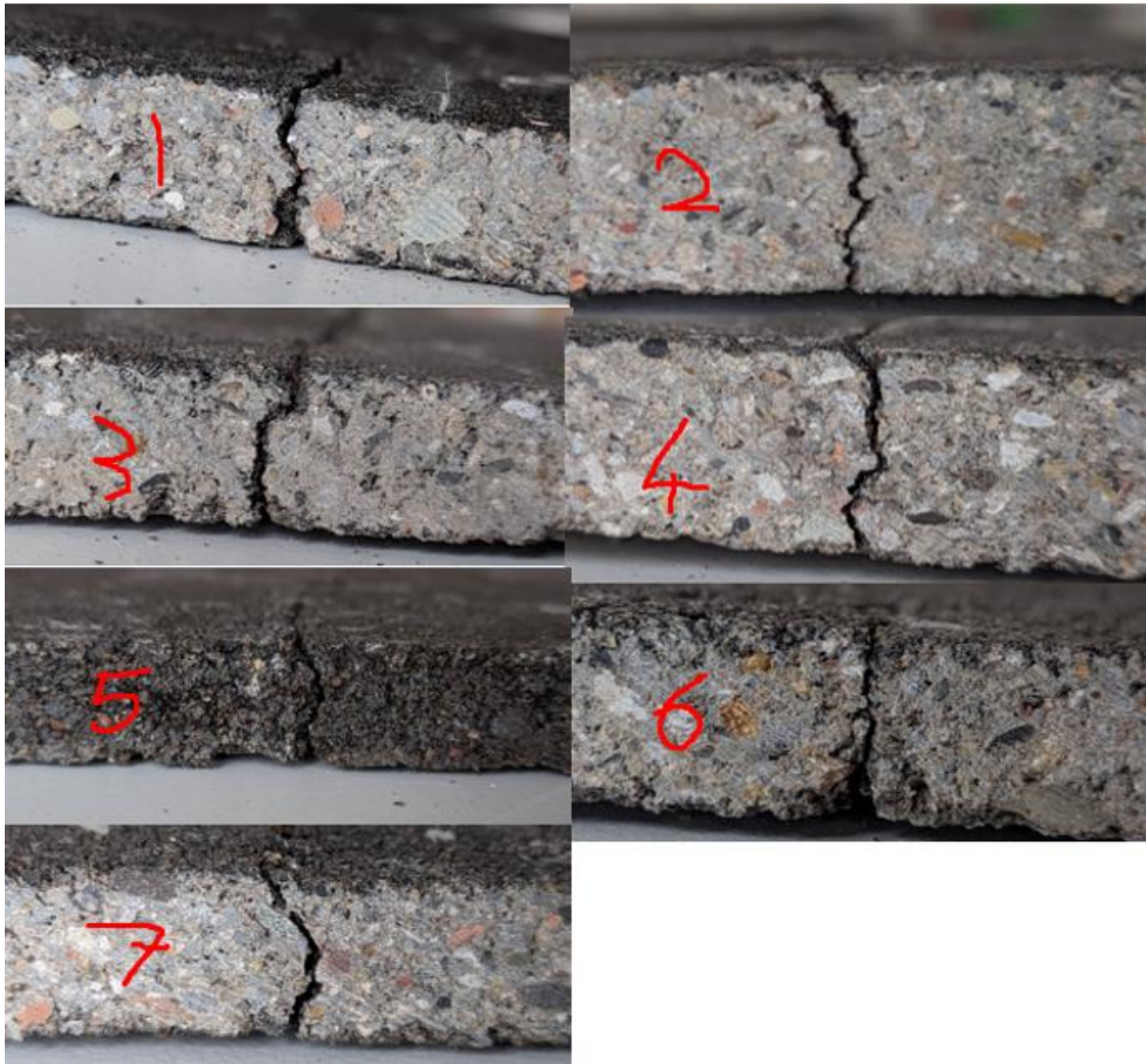


Figure 45 Side view of fracture path of graphene Art of Eco samples

Figures 44 and 45 above show the side view of both the Art of Eco standard and graphene samples. When compared, it appears that the fracture patterns in the graphene samples were more varied, whereas the standard samples had a more linear fracture. This may have been a result of toughening mechanisms of the graphene, such as crack bridging, as previously discussed. It may also have been caused by different materials used during the production of the tiles, or other unknown imperfections in the tile structure.

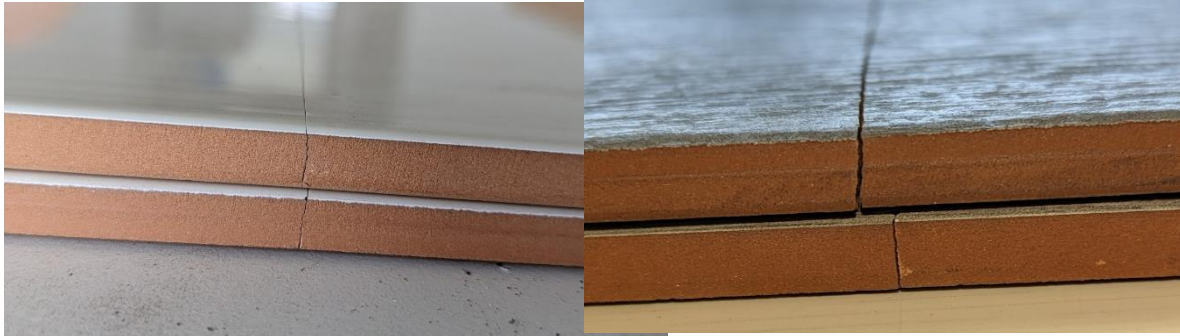


Figure 46 Side view of fracture path of ceramic wall tile samples (left) and ceramic floor tile samples (right)

Figure 46 shows that the fracture path in both ceramic types is very uniform and is likely due to the small particle size and dense structure of the tiles.



Figure 47 Breaking edge of graphene Art of Eco sample.



Figure 48 Breaking edge of standard Art of Eco sample.

Figures 47 and 48 show the breaking edges of a graphene sample and standard sample respectively. The graphene sample appears to have a higher variation in particle size and type. This could have resulted in increased interaction between the graphene and the structure of the tile, increasing the strength of the tile.

Tables 8 and 9 show the average overall thickness of the tiles sorted in ascending order, including the thickness at the breaking edge and the breaking strength of the standard and graphene Art of Eco samples respectively.

Table 8 Art of Eco standard samples average overall thickness (mm), breaking edge thickness (mm) and breaking load (N)

Sample no.			
AoE standard	Average overall thickness (mm)	Thickness of breaking edge (mm)	Breaking load (N)
2	8.89	9.025	130
3	9.55	8.65	90
4	12.6	10.7	235
5	13.34	12.925	125
6	14.74	13.95	395
7	15.49	15.7	320
9	15.925	15.35	145
8	17.55	18.575	390

Table 9 Art of Eco graphene samples average overall thickness (mm), breaking edge thickness (mm) and breaking load (N)

Sample no.			
AoE graphene	Average overall thickness (mm)	Thickness of breaking edge (mm)	Breaking load (N)
5	12.46	11.7875	110
6	12.72	11.9	345
7	13.11	13.575	360
4	14.87	16.225	395
1	15.075	14.7	240
3	15.73	15.8875	305
2	17.97	19.8	820

Figure 49 below shows the plots for each tile of both the Art of Eco standard tiles and the tiles with graphene added. The graph shows the force on the tile plotted against the time it took to fail.

The red lines show the standard tiles whilst the blue lines show the graphene-enhanced tiles.

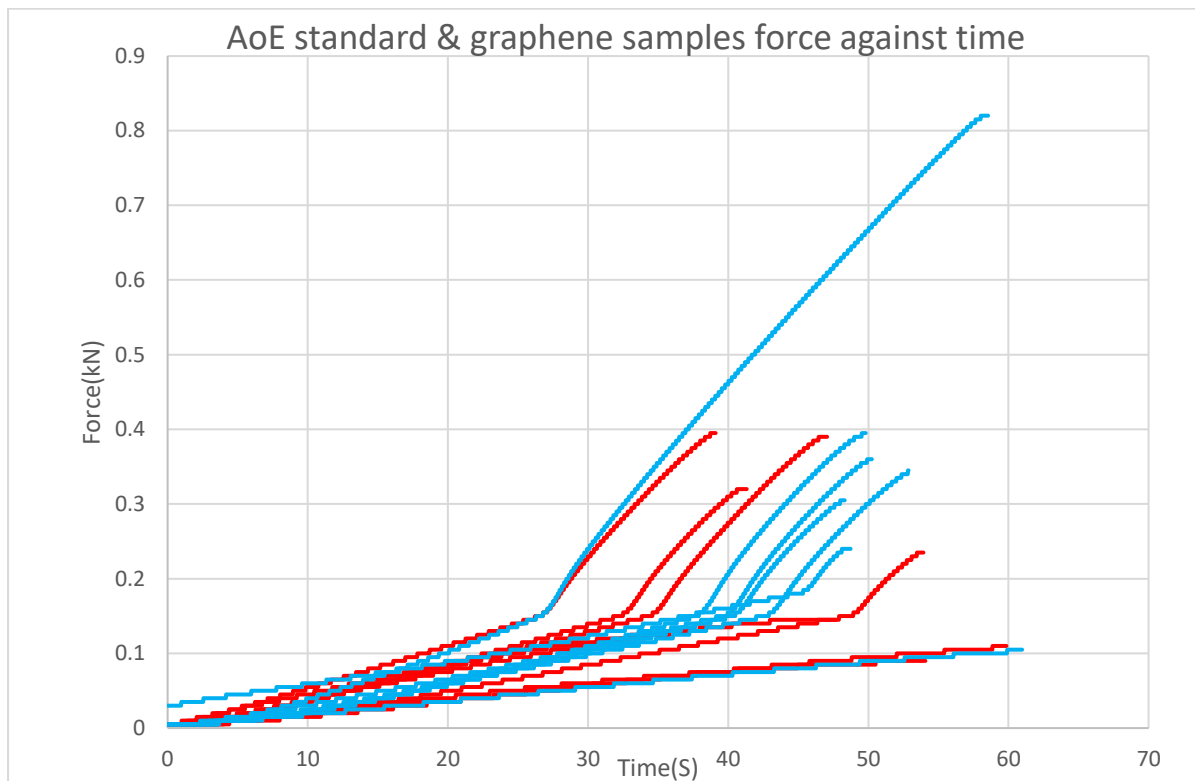


Figure 49 Force (kN) against time (s) graph of Art of Eco standard and graphene samples

Figure 49 above shows that, although the Art of Eco tiles display the same bilinear behaviour, both the gradient and breaking force vary significantly. This is likely due to the large amount of variation in the tile dimensions. The gradient of the plots also varies significantly. In general, when compared to the ceramic tiles, the gradient of the Art of Eco tiles is shallower than the ceramic tiles, indicating the tiles are not as stiff. The graph above shows that all but one sample with graphene had a breaking point of over 0.2kN, whilst just 4 of the standard Art of Eco tiles did.

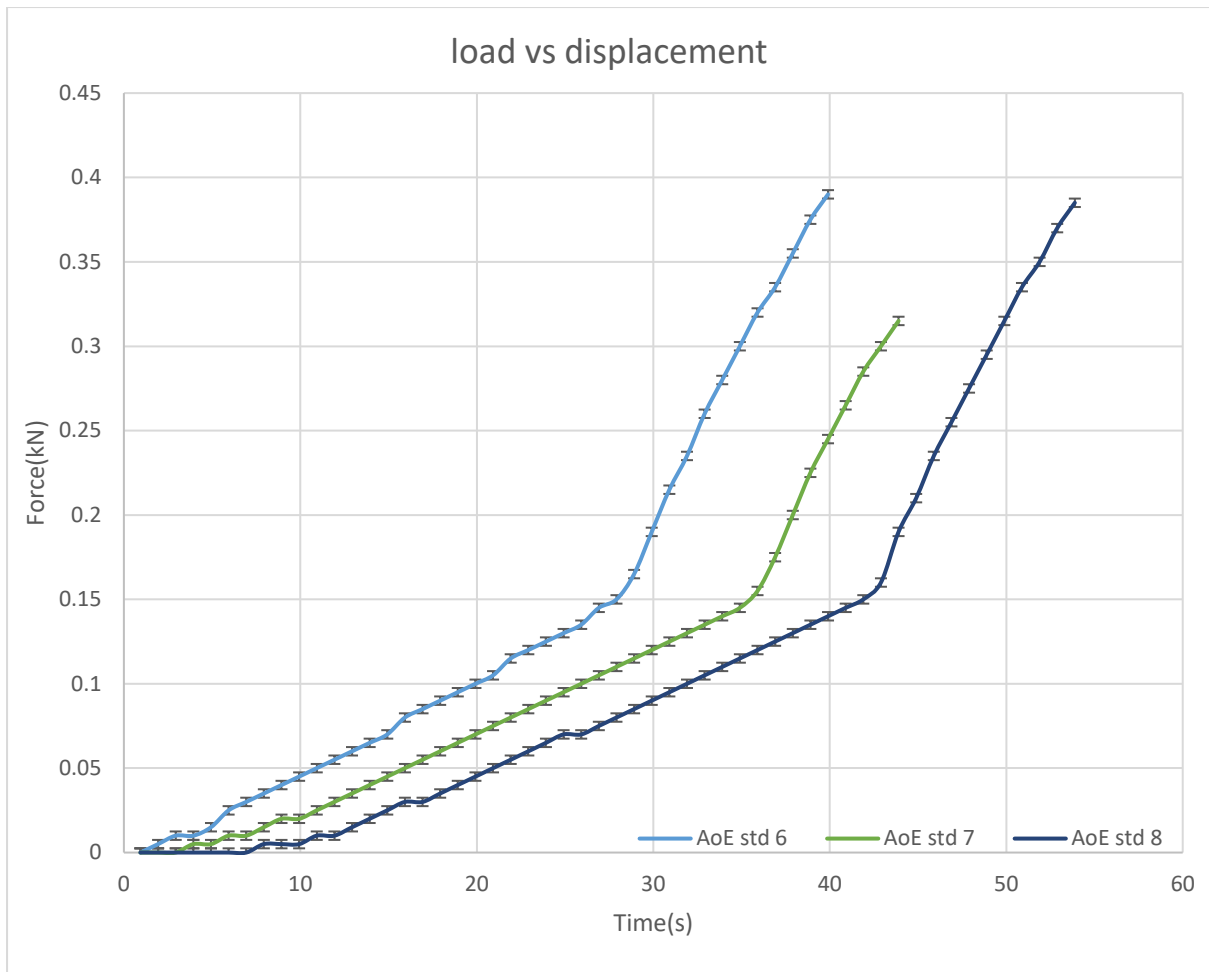


Figure 50 Adjusted force (kN) against time(s) graph including error bars for 3 standard Art of Eco samples.

Figure 50 shows a sample of the load vs displacement graph for three of the standard Art of Eco samples, with error bars of $\pm 2.5\text{N}$, as discussed previously. This shows that there were minimal errors in the data.

The graphs in figure 51 show the Art of Eco standard and graphene samples separately for easier viewing. The graphs show that the bilinear behaviour only occurred when the force reached roughly 0.155 kN. The results show that, whilst all but one (the thinnest) of the graphene samples displayed this behaviour, just 4 of the standard samples (displayed in green) did due to their breaking point being below 0.155 kN.

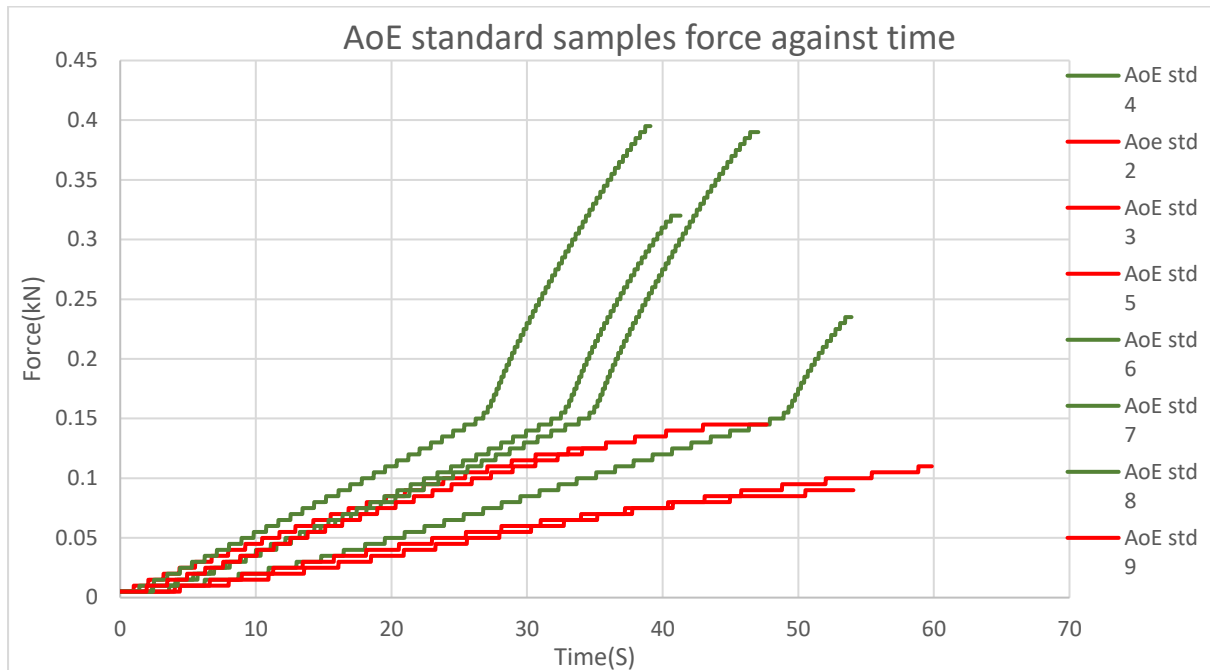


Figure 51 Art of Eco standard samples graph showing force (kN) against time (s)

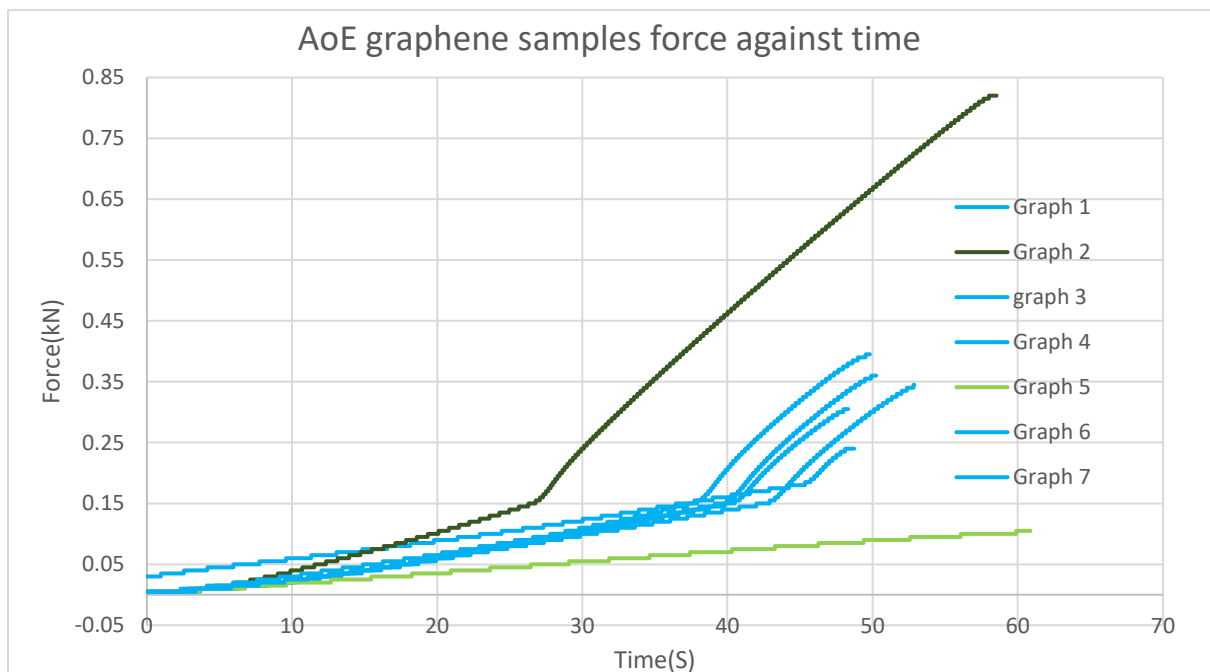


Figure 52 Art of Eco graphene samples graph showing force (kN) against time (s)

The graph in figure 52 above shows that, apart from graphene sample 2 and 5, the graphene showed a higher degree of consistency compared to the standard samples. Sample 5 had the lowest breaking force out of the graphene samples by a considerable margin, however, it was also the thinnest sample. Conversely, graphene sample 2 had the highest breaking force, taking 820N to fail. This sample was the thickest of all the samples tested. The above graphs show that a total of 5 tiles failed before the rubber was compressed, and as such, did not display the bilinear behaviour shown by the other samples. This meant that the calculation of the Young's modulus was not possible for these samples, as the equation uses the steeper linear part of the slope during the calculation. It is suggested as further work that the rubber is tested in accordance with ISO48-2 as stated in ISO 10545-4:2019.

Table 10 Art of Eco standard and graphene sample maximum loads (kN)

Sample no.	Maximum load (kN)	Sample no.	Maximum load(kN)	
AoE standard		AoE graphene		
2	0.130	1	0.24	
3	0.090	2	0.82	
4	0.235	3	0.305	
5	0.125	4	0.395	
6	0.395	5	0.11	
7	0.320	6	0.345	
8	0.390	7	0.36	
9	0.145			
Mean:	0.229	Mean:	0.3678	

The data in table 10 shows that there was an increase in the mean breaking force of 60.81% for the graphene samples, although graphene sample 2 was unusually thick. Calculating the increase without this sample shows an increase of 27.87%. When compared to the ceramic sample breaking forces, both types of Art of Eco's tiles have a lower breaking force. Due to the large amount of variation in both the breaking force, the dimensions of the tiles and the variation in the location of failure (shown in figures 41 and 42), the results imply that the structure of the Art of Eco tiles have a much less uniform structure than that of ceramic tiles. They also displayed a higher frequency of weak points.

The average thickness was also measured along the breaking edge of the tiles and an average was taken from 4 measurements across the breaking edge. The measurements are shown in table 10. Due to both types of ceramic tile being very consistent, the average was taken from 2 of each tile.

Table 11 Average thickness (mm) of breaking edge for all samples

Avg thickness (mm) of breaking edge							
AoE standard 2	14.7		AoE Graphene 1	14.7		Ceramic wall	9.1
AoE standard 3	19.8		AoE Graphene 2	19.8			
AoE standard 4	10.7		AoE Graphene 3	15.88		ceramic floor	8.9
AoE standard 5	12.92		AoE Graphene 4	16.22			
AoE standard 6	13.95		AoE Graphene 5	11.79			
AoE standard 7	15.7		AoE Graphene 6	11.3			
AoE standard 8	18.57		AoE Graphene 7	13.57			
AoE standard 9	15.35						

The Young's modulus was calculated using the equation (4) as used by Sasmita et al. (2018).

Equation 4 Young's modulus formula

$$E = \frac{mL^3}{4bh^3}$$

Where:

m is the slope of the linear portion of the force displacement curve

L is the span of the supports

b is the length of the short side of the tile

h is the thickness of the tile

Table 12 shows the Young's modulus for each sample.

Table 12 Young's modulus (GPA) for each sample

Young's modulus (E)			
Sample no.	E(GPA)	Sample no.	E(GPA)
cwall 1	11.146	floor 1	18.05
cwall 2	11.95	floor 2	18.97
cwall 3	12.104	floor 3	19.975
cwall 4	12.303	floor 4	19.255
cwall 5	12.093	floor 5	19.26
cwall 6	12.333		
cwall 7	12.273		
sample no. (AoE)	E(GPA)	Sample no. (AoE)	E(GPA)
std3?	3.97	graphene1	1.666
std 4	2.35	graphene 2	3.935
std 5?	2.5	graphene 3	1.945
std 6	3.296	graphene 4	3.13
std 7	2.18	graphene 5?	1.408
std 8	1.56	graphene 6	3.254
std 9?	1.447	graphene 7	3.49
mean:	2.3465	mean:	2.903333
		avg increase (%)	23.73%

Young's modulus is a measure of stiffness of a material and shows how much a material will stretch or compress in response to an applied force. This method is a more accurate measurement of the sample's strength in this experiment as the sample's dimensions are taken into consideration in the formula. Table 12 shows that there was an increase of 23.73% in Young's modulus of the graphene samples compared to that of the standard samples. However, this did not include the samples that failed before the rubber compressed, which are marked with a "?" next to the sample number. The Young's modulus of these samples was calculated using the entire portion of the displacement/load curve. This resulted in the standard samples 3 and 5 being unusually high and should not be considered accurate. When compared to the ceramic tiles, the Art of Eco tiles have a significantly lower Young's modulus. This could be due to a number of reasons, such as the binder not working effectively and inconsistencies in the structure. When compared to the study by Sasmita *et al.* (2018), the Young's modulus for the tiles tested in their report ranged from 18-60GPA, which shows that the formula provides an accurate result for the ceramic tile samples used, which ranged from 11-20GPA.

Discussion

The results show that there is a very high degree of inconsistency in both the physical structure of the samples, and subsequently a high degree of variability in the test results of the Art of Eco samples.

Looking at tables 8 and 9 showing the thickness of the tiles in ascending order, it would be expected that, as the thickness increases, so should the strength, and therefore the breaking point. The standard Art of Eco tiles show very inconsistent breaking points, with sample 9 being one of the thickest samples, yet having a breaking strength of just 145N. These results show that the standard Art of Eco tiles contain a great degree of variability and therefore also inconsistencies, resulting in weak points that will cause premature failure.

The tile samples with graphene demonstrate a relationship between thickness and breaking strength, with the exception of samples 1 and 3, which despite being some of the thickest samples, showed poor performance comparatively. It is possible samples 1 and 3 failed due to weak points within the structure of the tile. Overall, the strongest tile was graphene sample 2, with a breaking load of 820N. If this result is ignored (due to the abnormally thick nature of the tile), the highest breaking force of both types of Art of Eco tiles is 395N. Interestingly, graphene sample 4 was thicker than the standard sample 6. However, if standard sample 4 and graphene sample 6 (which have an overall thickness difference of 0.12mm) are compared, the graphene sample has a 46% higher breaking load than that of the standard sample 4.

The results from the ceramic tiles show a good level of consistency, in both the manner in which the tiles failed and the test results clearly showing the difference in strength between the wall and floor tiles. This proves that the experimental setup and procedure was a reliable test.

The results show that, although the Art of Eco graphene samples were 7.78% thicker overall and 13.51% thicker on the breaking edge, the average breaking load was 60.81% higher in the graphene samples than that of the standard samples. This shows that despite the thicker structure, the average breaking load was significantly higher. The average modulus of rupture was also higher in the graphene samples by 19.49%. This is a larger increase than the difference in thickness, and it could be assumed that if the samples were the same dimensions, the graphene tiles would still display superior structural properties.

Conclusion

There is a clear need for more sustainable products globally in all industries, and the ceramics industry is no exception.

Graphene shows promise as an additive/nanofiller in composite materials, including ceramic tiles. The numerous studies highlighted in this report provide clear evidence that graphene can enhance the mechanical properties of the various materials studied. The method in which graphene is produced can have a substantial influence on the environmental impact that graphene can have, as well as on the quality of the graphene. Therefore, it is essential to consider the advantages and disadvantages of the use of graphene overall.

Art of Eco's product showed potential improvement with addition of graphene. However, it is difficult to ascertain the true scope of this, considering there was a large variation in sample dimensions and quality, as well as imperfections being present in the samples. The data showed that there was an increase in strength of up to 60.81% compared to the standard Art of Eco samples. It must be noted that this figure was due to an unusually thick sample. If this sample were excluded from the calculation, there was an increase of 27.87% in breaking force. When compared to the 7% increase in sample thickness, this shows the increase in breaking force was larger than the increase in thickness. The Young's modulus was also calculated, which includes the sample dimensions in the calculations. The results showed that there was an increase of 8.81% in the graphene samples, which could have been a result of toughening mechanisms provided by the graphene. Additionally, there was a 19.49% increase in the modulus of rupture in the graphene samples when compared with the standard samples.

Ceramic tile samples used in this test proved to withstand a higher breaking force than the Art of Eco samples, with the floor tiles displaying the highest strength with an average breaking force of 577N, as well as having a higher Young's modulus value of 11 GPA for the wall samples and 19 GPA for the floor samples.

There is currently limited information on the carbon footprint of Art of Eco's products. This is a factor that should be considered, especially with the inclusion of graphene, which may have a significant effect on this. A full life cycle analysis should be carried out and is suggested as further work.

Overall, Art of Eco's product shows promise as an environmentally sustainable alternative to ceramic tiles but requires a higher accuracy of test results to successfully quantify both the mechanical properties and the effect of graphene on their products. If the production process

were to be further refined, more reliable results may be obtained, and a better indication on whether the addition of graphene is enhancing the mechanical properties of Art of Eco's products. In view of this, further work should be carried out.

Limitations

The tile specimens varied greatly in size, with a minimum thickness of 7mm in some tiles, and a maximum thickness of 20mm. This creates a large margin of error. The size variation may be caused by either the material used (it is likely that the recycled quarry aggregate has a high degree of variation), or the moulding process used not being reliable.

The graphene distribution is critically important for a strong interface between the other materials. If the graphene is not homogeneously distributed it will create weak points within the structure, resulting in failure. Additionally, the material mix itself may have inconsistencies in it, such as large particles of rock that have not been removed in the screening stage.

The jig produced to perform the three-point bend testing was fixed without any rotation. This could have led to uneven load distribution and caused pressure points, resulting in failure. However, the results from the ceramic tiles proved to be consistent despite this.

The tile may not have been placed directly in the centre of the supports, causing inconsistent loading.

The Standard states that the rubber should have a hardness of 50 ± 5 IRHD, measured in accordance with ISO48-2. It was not possible to measure the hardness of the rubber in this manner.

The sample tiles were produced at different time periods. As the binding agent used causes the tiles to get stronger over time and different materials may have been used for different batches, some tiles may have been stronger as a result of this.

It was not possible to fully observe the tiles whilst they were under load or at the point of failure. It is therefore harder to determine the type of failure that occurred in each sample and what the cause was.

Further work

It would be of interest to vary the quantity of graphene added to the tiles. According to the studies highlighted in this report, this has a large impact on the strength, with a certain percentage of graphene being optimal and any further increase having a detrimental effect on the strength of the tile.

Carrying out other tests, such as impact testing (ISO 10545-5:2019 covers impact testing of ceramic tiles) and weather testing, would be beneficial. Art of Eco's material can be used for many different applications and as such various testing methods would be beneficial.

Further studies on the interaction between graphene and the various materials used in Art of Eco's products should be undertaken. Some studies, including that of *Zhao et al. (2020)*, have shown that some materials can cause agglomeration of graphene, resulting in poor dispersion and mechanical properties.

Further testing using tiles that are of equal dimensions and uniform in structure with no imperfections would provide greatly improved accuracy in the results and, as such, it would be clearer what effect graphene has on the properties of the tiles. Improved quality control in the production process would provide this.

Samples produced using an exact quantity of each material would greatly improve the accuracy of the results.

Using a high speed camera to record the tiles whilst under load and recording the point of failure would provide a much higher degree of accuracy in determining the type of failure that occurs in the sample, and what the cause is.

Further investigation into the cellular makeup of the tiles using microscopic methods such as scanning electrode microscope (SEM), as used in other studies shown in this report, would be beneficial, as it would show the failure mechanisms and what effect the graphene is having, how it is interacting with the other materials and what toughening mechanisms are present.

Water ingress testing could prove of value as some of the applications of Art of Eco's products require them to be water resistant.

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