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作品名稱　HYBRID COMPOSITE FROM X-RAY WASTE

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關鍵詞　kenaf、x-ray film waste

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## Abstract

This study considered the tensile and flexural characterization of new lighter and cheaper hybrid composite materials to replace the existing insert panel for the currently available bulletproof vest. The materials chosen included a natural fibre, *i.e.,* kenaf fibre, chemically treated with sodium hydroxide solution, and, as a means of recycling, used x-ray films with a surface treatment. Using the traditional hand lay-up method, the materials were fabricated into seven layers of different configurations, which were then subjected to tensile and flexural tests. The findings showed that one of the configurations that consisted of both treated materials had a tensile strength of 396.9M Pa, which is quite strong, and a flexural modulus of 6.24G Pa, which makes it flexible enough to be made into wearable equipment. This configuration was then chosen to be the base design for the specimen subjected to impact test. The interfacial bond between the two distinct materials proved to be a major issue, even with the help of fibre treatment. Therefore, some improvements need to be made for the material to be comparable to existing materials performance-wise hence making this configuration suitable for ballistic application.

## 1.0 Introduction

As the world economy ascends to a new stage, demand for wood will increase proportionally. Current statistics show that the timber trade in the world market has exceeded 1500 million. The demand for good-quality timber will lead to non renewable logging of tropical hardwood forest in many developing countries, and give rise to serious global concern, especially in Asian countries. Indonesia's current dominance of the export market is expected to end within 20 years at present rates of logging, while Thailand has banned all commercial logging in its hardwood forests.

Therefore, a sharp rise in the cost of natural timber products is expected in the near future. With timber substitutes available in the form of wood chipboard, the demand for wood has been alleviated to some extend, but the raw material is still mainly wood, and the mechanical properties are not very satisfactory

### 1.1 Natural fibers

Natural fibers or natural fibres are fibres that are produced by plants, animals, and geological processes.They can be used as a component of composite materials, where the orientation of fibers impacts the properties[2]. Natural fibers can also be matted into sheets to make products such as paper, felt or fabric.[3,4 ]

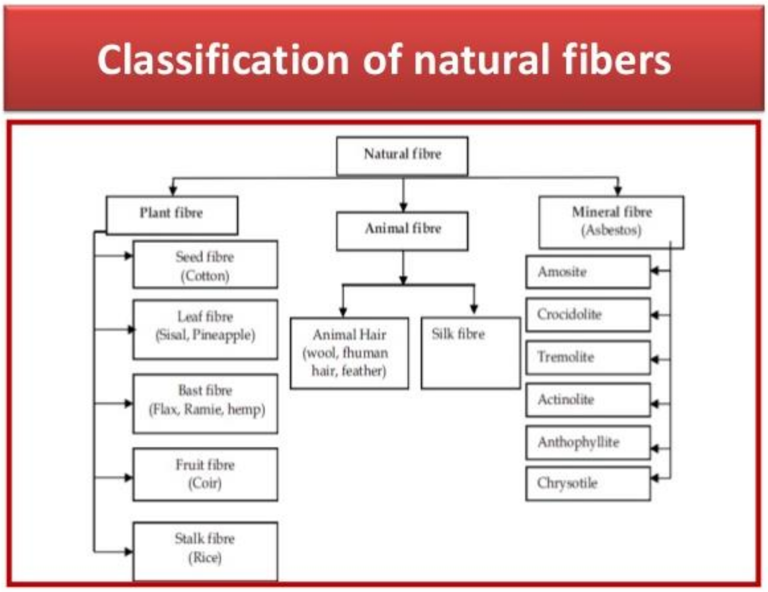
The earliest evidence of humans using fibers is the discovery of wool and dyed flax fibers found in a prehistoric cave in the Republic of Georgia that date back to 36,000 BP. Natural fibers can be used for high-tech applications, such as composite parts for automobiles. Compared to composites reinforced with glass fibers, composites with natural fibers have advantages such as lower density, better thermal insulation, and reduced skin irritation. Further, unlike glass fibers, natural fibers can be broken down by bacteria once they are no longer in use.

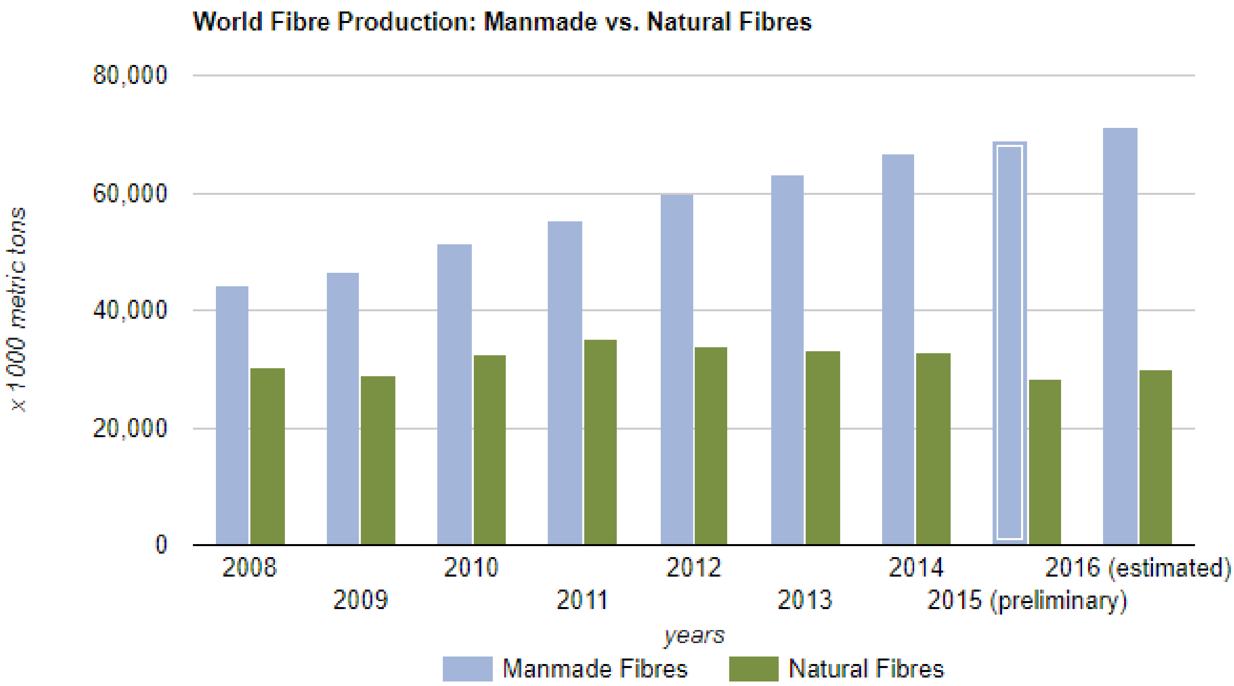
Natural fibers are made from plant, animal, and mineral sources. Natural fibers can be classified according to their origin. The example of natural fibre from plant are cocnut fibre, jute, kenaf, bamboo, straw. The example of natural fibers from animal are sheep's wool, feather fiber,

Natural fibres can be considered as naturally occurring composites consisting mainly of cellulose fibrils (fibres) embedded in lignin matrix (resin). These cellulose fibrils are aligned along the length of the fibre, irrespective of its origin, i.e. whether it is extracted from bark or stem, leaf or fruit. It appears that such an alignment renders maximum tensile and flexural strengths, in addition to providing rigidity in that direction of the fibre as observed in the case of bamboo.

Fibres such as cotton, banana and pineapple are also used in making cloth in addition to being used in the paper industry. However, in recent times many of the conventional applications of these natural fibres are threatened by synthetic products (plastics and synthetics such as glass and nylon fibres). This poses the problem that limiting the utilization of natural fibres in the fibre industry, which is basically a rural/cottage industry, leads to displacement of labour. Hence, there is an urgent need for finding diversified uses of these fibres. In addition, natural fibres have the following advantages

(i) These fibres, though they have poor strength properties due to low density, can lead to
high specific strength properties. Wood flour, for example, used as 50% filler in thermosetting phenolic resin, is found to improve strength and impact resistance of the resin
(ii) They are abundantly available resources, having low cost (Table 1) and low energy consumption compared with synthetic fibres
(iii) Natural fibres are non-toxic to work with.



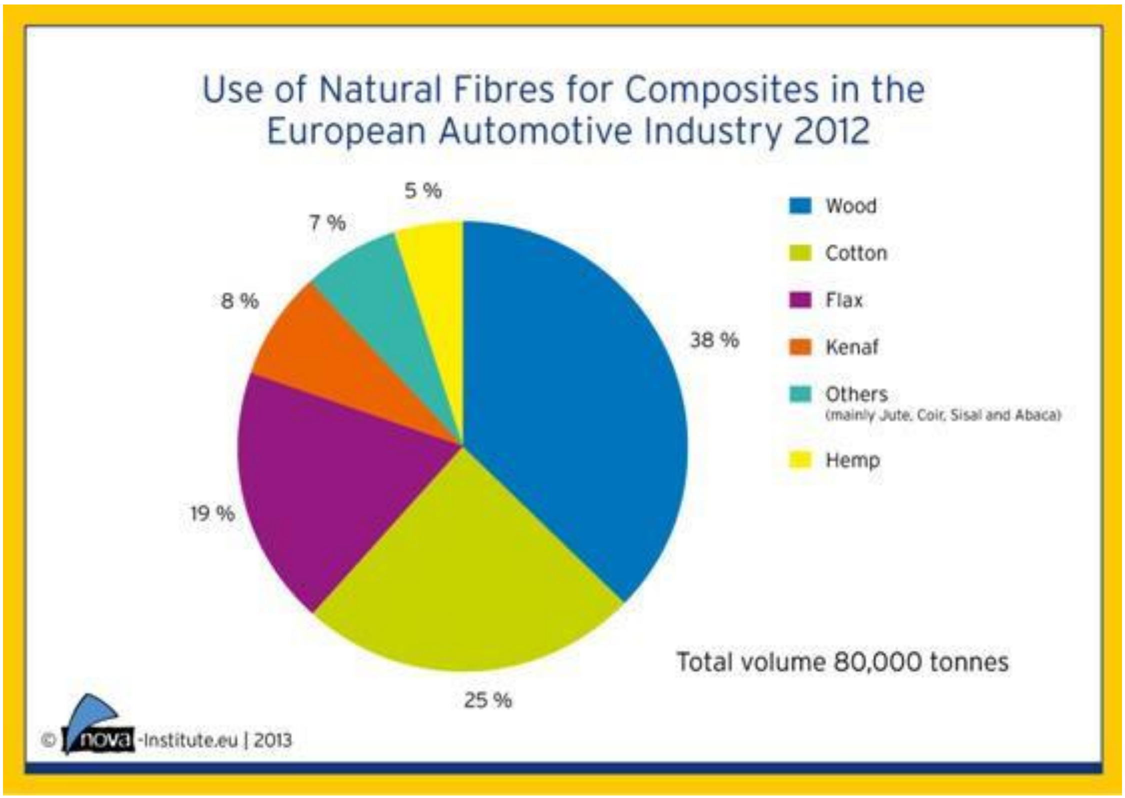




Malaysia is rich with natural fibres such as kenaf, palm oil fruit, pineapple and coconut fibre. Million tons of these fibres had been produced and these fibres can be transforming to useful materials. Composite manufacturing industries have been looking for plant based natural fibre reinforcements, such as flax, hemp, jute, sisal, kenaf, banana as an alternative material which in order to replace solid wood.

Natural fibres have the advantage that they are renewable resources and have marketing appeal. Natural fibre reinforced polymer composites have been used for many applications such as automotive components, aerospace parts, sporting goods and building industry (Rowell, 2008).

Natural fibres have been used as reinforcing materials for over 3000 years, in combination with polymeric materials. In the course of nature life, composite materials were subjected to both mechanical loading and exposed to severe environmental conditions. The natural fibre reinforced composite are lightweight and free from health hazard, reasonably strong, and hence it's have the potential to be used as material for strong components such as building materials, shipping, and automotive. Despite the advantages, they suffer from some limitations such as poor moisture resistance especially absorption and low strength compared to synthetic fibre such as glass (Abdu Khalil et.al., 2009).

The strength of the fibre reinforced composites is dependent on the properties of fibre, the aspect ratio of fibre content, length of individual fibre, orientation of fibre, extent of intermingling of fibres, fibre to matrix interface bonding and arrangement of both the fibres and also on failure strain of individual fibres. Maximum results are obtained when the fibres are highly strain compatible (Sreekala, George, Kumaran, & Thomas, 2002).

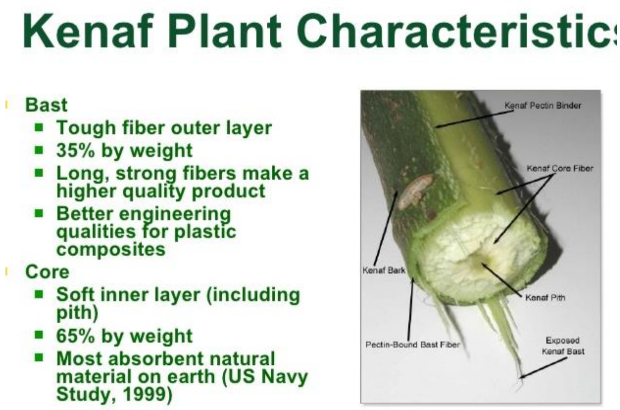


### 1.2 Kenaf Fibre

Kenaf is an old crop with roots in Africa. A member of the hibiscus family (Hibiscus cannabinus L), it is related to cotton and okra, and grows well in many parts of the world. Kenaf grows quickly, rising to heights of 16-20 feet in as little as 4 to 5 months. U.S. Department of Agriculture studies show that yields of 6 to 10 tons of dry fiber per acre per year are generally 3 to 5 times greater than the yield for Southern pine trees, which can take from 7 to 40 years to reach harvestable size [1]. Kenaf has two fibers: the outer fiber called "bast" and comprises roughly 40% of the stalk's dry weight and the whiter, inner fiber called "core". Upon harvest, the whole kenaf plant is processed in a mechanical fiber separator, similar to a cotton gin. The separation of the two fibers allows for independent processing and provides raw materials for a growing number of products including paper, particle board, animal bedding and bioremediation aids [2]. The stem, leaves and flowers of the plant are shown in


Figure 1. Figure 1 Kenaf Plant LEGEND: 1. Flower, 2. Stalk-Outer Fiber = Bast (40%), 3. Stalk-Inner Fiber = Core (60%)

Kenaf has a cellulosic source with both economic and ecological advantages. It is a warm season annual fibre crop closely related to cotton and jute. Kenaf has been used as a cordage crop to produce twine, rope and sackcloth. Kenaf has good mechanical properties and can grow quickly as it takes only 150 days to harvest. The kenaf comprises 35-40% bast fibre and 60-65% core 6 fibres by weight of the kenaf’s stalk. Kenaf contains approximately 65.7% cellulose, 21.6% lignin and pectin and other composition. It could grow under wide range of weather condition, to a height of more than 3m and a base diameter of 3-5cm. Kenaf has a single, straight, unbranched stem consisting of two parts, namely outer fibrous bark and inner woody core (Mohanty et. al., 2002).



Nowadays, there are various new applications for kenaf including paper products, building materials, absorbents and animal feeds. In Malaysia, realizing the diverse possibilities of commercially exploitable derived products from kenaf, the National Kenaf Research and Development Program has been formed in an effort to develop kenaf as a possible new industrial crop for Malaysia. The government has allocated RM12 million for research and further development of the kenaf-based industry under the 9th Malaysia Plan (2006–2010) in recognition of kenaf as a commercially viable crop (Salleh et. al., 2012).

Whole stalk kenaf can also be used in corrugated medium. The whole stalk plant material can also be used in non-pulping products such as building materials such as particleboard and within injection molded and extruded plastics. Unlike the pulping process with whole stalk plant material which yields fewer than 46% by weight, the use of non-pulped whole stalk material yields nearly 100% usable materials.

The difference is the result of the intentional removal of non-fibrous materials such as lignins and sugars during the pulping process, whereas the removal of these intercellular materials is not required for the non-pulped products (Anuar and Zuraida, 2011). When bast material is mechanically pull out from the core, it was chemicallypulped without the core, it produce a 57% yield of bast fibre. On a whole stalk dry weight basis, the bast comprises 17.4% to 28.6%. The individual bast fibres are up to 5.0 mm long averaging 2.6 mm in length and 20 mm in width. Chemical bast pulp is well suited for specialty papers, such as high quality stationery or filter paper. Bast pulp, compared to softwood pulp, has a similar tensile strength, but greater tear strength and bulk fibre, thus it could serve as a replacement for softwood pulp. Pulping kenaf bast and core fibres can benefit the environment because the process requires fewer chemicals and less energy compared to standard pulping processes for wood fibres (Nishino et.al., 2003).

The kenaf fibres can also serve as a virgin fibre for increasing recycled paper quality and paper strength. Although the kenaf bast fibre strands were once only considered for use as a cordage fibre in such products as rope, twine, carpet backing and burlap. A variety of additional uses has developed for the bast fibre strands. These include use in automobile dashboards, carpet padding, corrugated medium as a substitute for fibreglass and other synthetic fibres, textiles and as fibres for injection moulded and extruded plastics. Kenaf bast fibre strands are presently in commercial use in other environmentally friendly products such as fibre lawn mats impregnated with grass seed and spray on soil mulches for use along highway rights of way or construction sites to prevent soil erosion from water and wind (Karimi et. al., 2014).

Chemical pulping of the woody core will yield about 41% core fibre from the original woody portion of a kenaf stalk. The core fibres make up from 20% to 40% of the entire stalk by weight. The core pulp, compared to hardwood pulps, has lower tear strength but greater tensile and burst strength. Due to the high absorbency of the woody core material, researchers have investigated the use of kenaf as an absorbent, as a poultry litter and animal bedding, as a bulking agent form sewage sludge composting and as a potting soil amendment. In addition to the above core products, which are all now available in the market place, several kenaf core products are available which are successfully used for toxic waste clean-up, oil spills on water and the remediation of chemically contaminated soils (Abdul Khalil et. al., 2010).

The stock of kenaf can be used almost entirely. Kenaf leaves and stems have a potential as livestock feed. Dried leaves contain 30% crude protein and are used as vegetables in some part of the world (Ochi, 2008). In recent years, with increasing concerns for environmental protection, kenaf has found more applications. The breakthroughs and advances in environmental technology have resulted from intensive testing and research in the kenaf industry. Kenaf fibre/plastic compounds based on kenaf can replace glass reinforced plastics in many applications such as automotive industry, construction and housing industry, food packaging industry, oil and chemical absorbents, animal bedding and poultry litter, and soil free potting mix. The compounds have the mechanical and strength characteristics of glass filled plastics but are less expensive and in many instances are completely recyclable (Ochi, 2008).

(i) The Automotive Industry

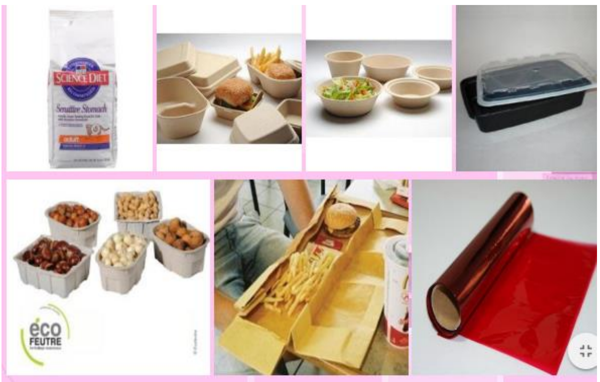
The 1996 Ford Mondeo which sold abroad features interior automobile panels made of kenaf fibre. Kenaf international supplies the fibre, which is processed by the supplier to Ford. The company expects that sales to European automobile manufacturers will steadily increase as the industry becomes comfortable with the product and the kenaf products from automotive group are capable of meeting required demand (Fuqua et. al., 2012).

(ii) Construction and Housing Industry

Kenaf/plastic compounds molded into lightweight panels can replace wood and wood based products in many applications. This product has the potential to be the first economically priced plastic lumber that can be engineered for use as building materials in housing industry. In some cases, emphasis has centered in the utilization of core of the plant. Kenaf core has been used as packaging material, animal bedding, oil absorbents and poultry litter (Akil et. al., 2011).

(iii) Food Packaging Industry

Pellets made from a kenaf/plastic compound can be molded into commercial food storage containers and virtually any other product now made of plastic. Non-food related packaging opportunities are also numerous including bulk chemical and pharmaceutical packaging, parts packaging in the electrical and electronics industries and disposable packaging for large consumer appliances. In every instance, fibre composites have distinct technical and pricing advantages over plywood and cardboard and are recyclable as well (Karimi et. al., 2014).



(iv) Oil & Chemical Absorbents

The core of kenaf fibre is very absorbent and one of its main uses is to clean up oil spills and similar chemicals. The kenaf fibre product is also non-toxic, non-abrasive and is more effective than traditional remediants like clay and silica. This product is distributed for use in oil fields but the product also absorbs gasoline, diesel, transmission fluid and coolant spills. In addition to use by individuals for personal garages, bulk applications include clean-up operations in refineries, utility companies, land and sea spills, oil rigs, industries that handle bulk storage terminals and for military field refuelling applications (Fuqua et. al., 2012).

(v) Animal Bedding and Poultry Litter

Kenaf bedding is sold in bags to farm and ranch supply stores and in bulk to large buyers such as stables, zoos and poultry farms. This product has superior absorbency, requires fewer changes, is cost competitive with most traditional litter and bedding products comprised of wood shavings, saw dust or shredded paper (Coetzee et. al., 2007).

(vi) Soil free Potting Mix

This product competes with commercial potting soils and can also be custom mixed for different horticultural applications. Kenaf has a long term supply arrangement with a nursery products wholesale business. The products are blended and mix with peat moss, compete with commercial mixes containing mostly peat moss or pine bark (Akil et. al., 2011).

**1.3 X-ray film**

X-ray films are a synthetic fibre made from polymer which are a non-biodegradable waste and needs an innovative way to be recycled. An x-ray film shows a radiographic image and is produced from either a single or double emulsion of silver halide, usually silver bromide, which produces a silver ion ($Ag^+$) and an electron when exposed to light. The electrons attract the silver ion when they become attached to the sensitivity specks. Subsequently, clumps of metallic silver are formed when the silver ions attach (Curry *et al.* 1990; Mosby and Bushong 2009).

The base, the emulsion, and the protective coating are the three main parts that make up an x-ray film. The base is where the other materials are applied and exists in all x-ray films. Usually, the base is made from a clear, flexible plastic such as cellulose acetate. The base's main purpose is supporting the emulsion. The softer layers of the gelatin coating are called the emulsion. An emulsion holds something in suspension. It is this material that is sensitive to radiation and forms the latent image on the film in suspension. Lastly is the protective layer, whose main function is to protect the softer emulsion layers below. In simpler terms, it is a very thin skin of gelatin protecting the film from scratches throughout handling. To film manufacturers, it has very important properties that include shrinkage (during drying this forms glassy protective layers) and dissolution in warm water. If it is dissolved in cold water, it will absorb the water and swell (NDT Resource Center 2001).

### 1.4 Problem Statement

Do kenaf / x-ray film hybrid as alternative in production of composite material ?

### 1.5 Objective

The specific objectives of this research were to :

i. To utilize x-ray film waste to kenaf / x-ray film hybrid composite material
ii. To investigate the effect of a kenaf / x-ray film hybrid composite
iii. To investigate the mechanical properties kenaf / x-ray film hybrid composite

## 2.0 Methodology

2.1 Preparation of material

The materials chosen as the test specimens were x-ray films and a kenaf fibre-reinforced polymeric composite. The x-ray films (Hospital Universiti Kebangsaan Malaysia (HUKM), Kuala Lumpur, Malaysia) were a flexible and transparent blue-tinted base coated on both sides with an emulsion-gelatine that carries radiation sensitive silver halide crystals, usually either silver bromide or silver chloride.

The surfaces of some of the x-ray films were punctured by consistent holes 2 cm apart, which is considered as a surface treatment. The epoxy resin (ZKK Sdn. Bhd, Selangor, Malaysia) was reinforced with kenaf fibre (ZKK Sdn. Bhd, Selangor, Malaysia) which was in woven mat form, producing a fibre-reinforced polymeric composite.
Part of the kenaf fiber was treated using a sodium hydroxide solution with 6% concentration for 3 h immersed in a water bath at 95 *°C* (Edeerozey *et al*. 2007).

2.2. Preparation of composite kenaf

As shown in Table 3, the specimens were fabricated using the traditional hand lay-up method into eleven different configurations with five layers. The configurations were based on the intention to study the interfacial bond between the two distinct materials. A configuration consisting of fully treated x-ray films was intended, but the specimen immediately delaminated when attempts were made to cut them into the respective specimens.

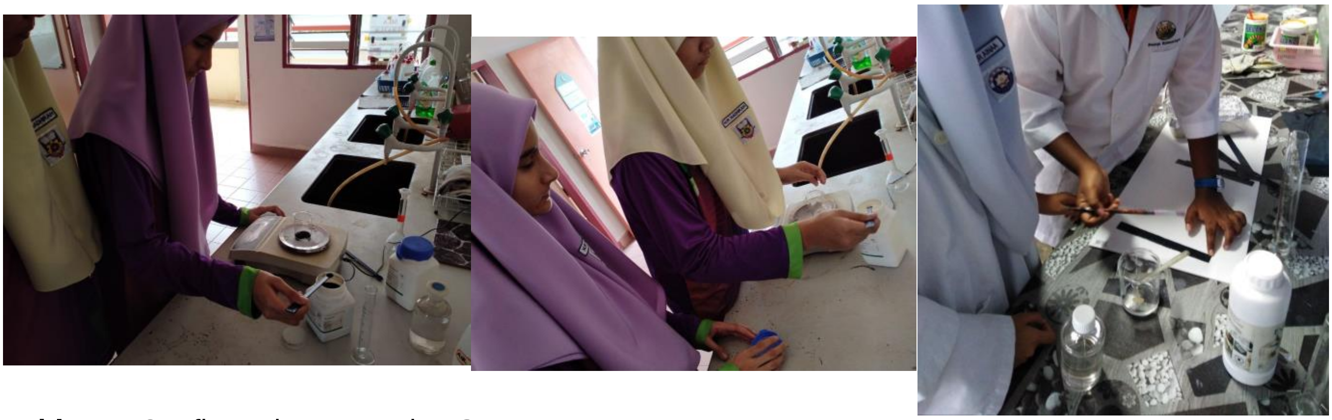


**Table 1 :** Configurations Layering Sequence

| Configuration | Name | Layers |
|---|---|---|
| 1 | Alternate (treated x-ray) | K-TX-K-TX-K-TX-K |
| 2 | Sandwich (treated x-ray) | K-K-TX-TX-TX-K-K |
| 3 | Sandwich (treated kenaf) | TK-TK-X-X-X-TK-TK |
| 4 | Full kenaf (treated) | TK-TK-TK-TK-TK-TK-TK |
| 5 | Sandwich (fully treated) | TK-TK-TX-TX-TX-TK-TK |
| 6 | Full kenaf (untreated) | K-K-K-K-K-K-K |
| 7 | Full x-ray (treated) | TX-TX-TX-TX-TX-TX-TX |
| 8 | Sandwich (untreated) | K-K-X-X-X-K-K |

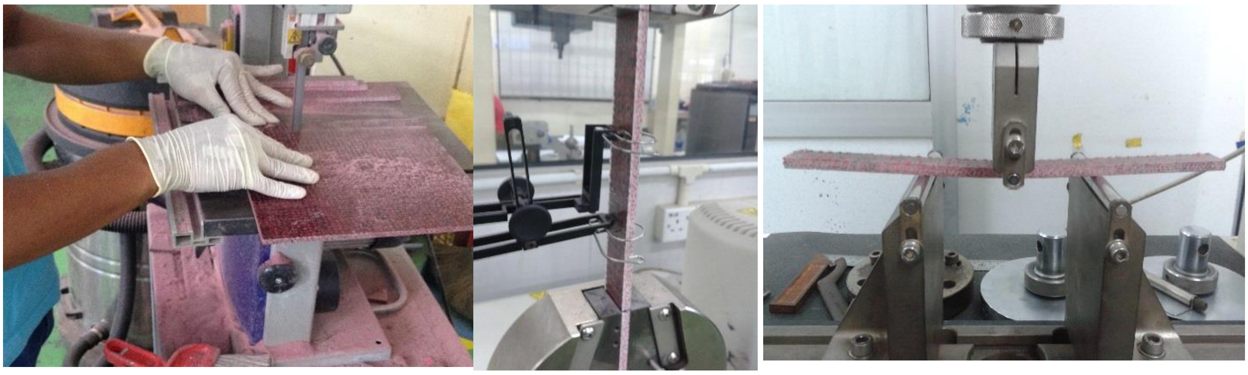
Note:

K- kenaf fiber reinforced composite; TK- treated kenaf fiber reinforced composite;

X- x-ray film; TX- treated x-ray film

## 2.3 Testing

Figure 1 below shows the process done prior to the testing, which is the cutting and shaping of specimen as well as the testing setup itself or tensile and flexural test.



a b c

**Fig. 1**. Testing process: a) specimen cutting; b) tensile test setup; c) 3-point bending setup

The specimens were cut to size according to the ASTM D3039 (2014) standard, which is 250 mm × 25 mm for the tensile test, and the ASTM D790 (2017) standard, which is 250 mm × 20 mm for the flexural test. The tensile test was conducted using a Shimadzu AG-IS ultimate testing machine (Selangor, Malaysia), and the flexural test was a 3-point bending test conducted using an Instron 4204 flexural testing machine (Selangor, Malaysia). For the flexural tests, the mean thickness and span separations can be seen in Table 4.

## 3.0 Result and Discussion

### 3.1 Surface Treatment

The treatment of kenaf with the NaOH solution increased the unit break of the fibre bundle by removing impurities and making the fibre stronger, thus providing an easier surface to work with. The concentration of the solution, which was 6% NaOH, and the condition of the treatment, in which the fibres were immersed in a water bath at 95 *°C,* were decided based on the results as shown in Fig. 2 below. The 9% treated kenaf fibre showed significant decrease in the unit break of fibre bundle due to the chemical being too strong, whereby damaging the fibres and making them weaker.

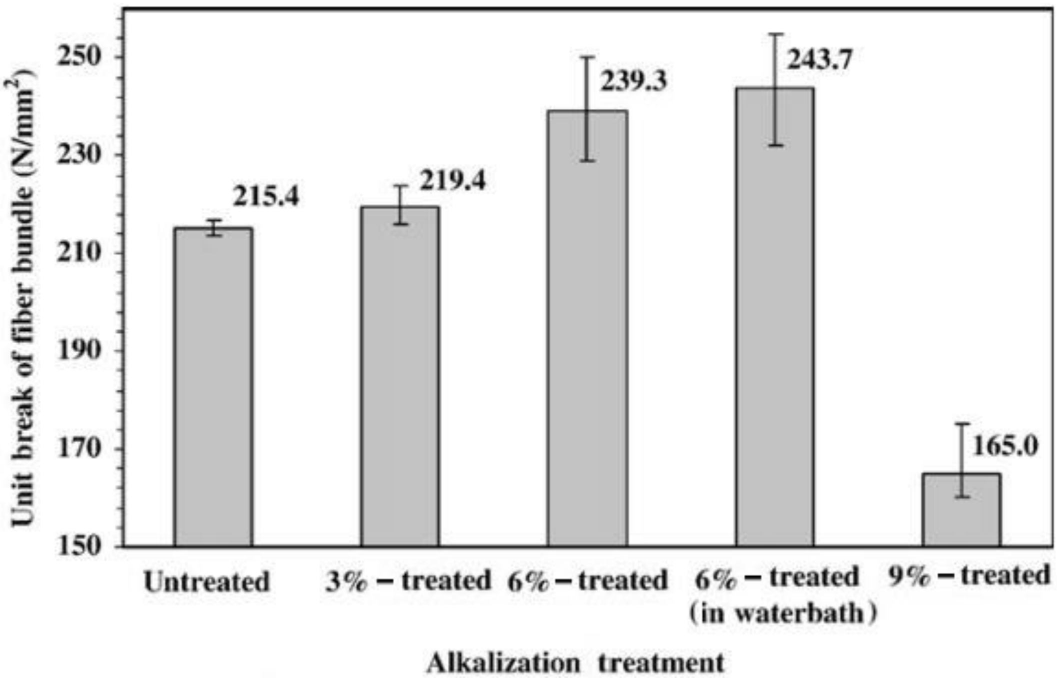
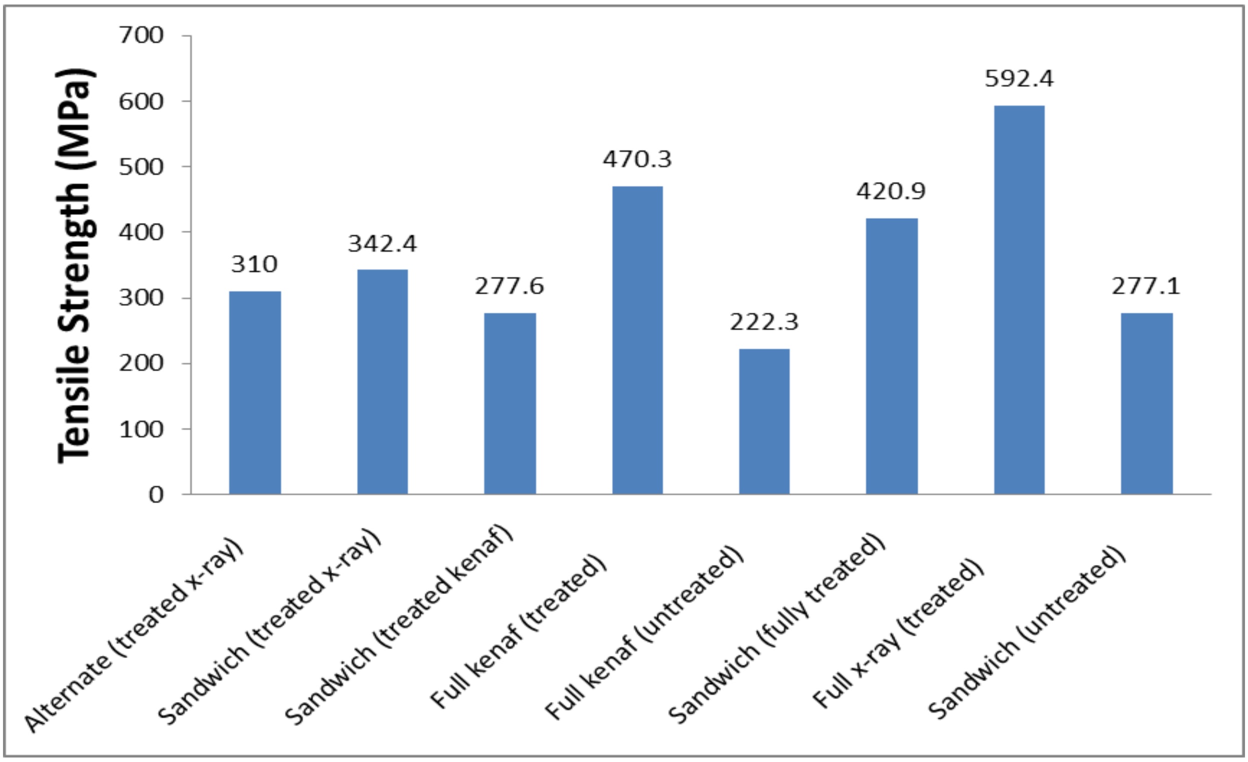



Fig. 2. Average unit break of kenaf fibre bundles (Edeerozey *et al*. 2007)

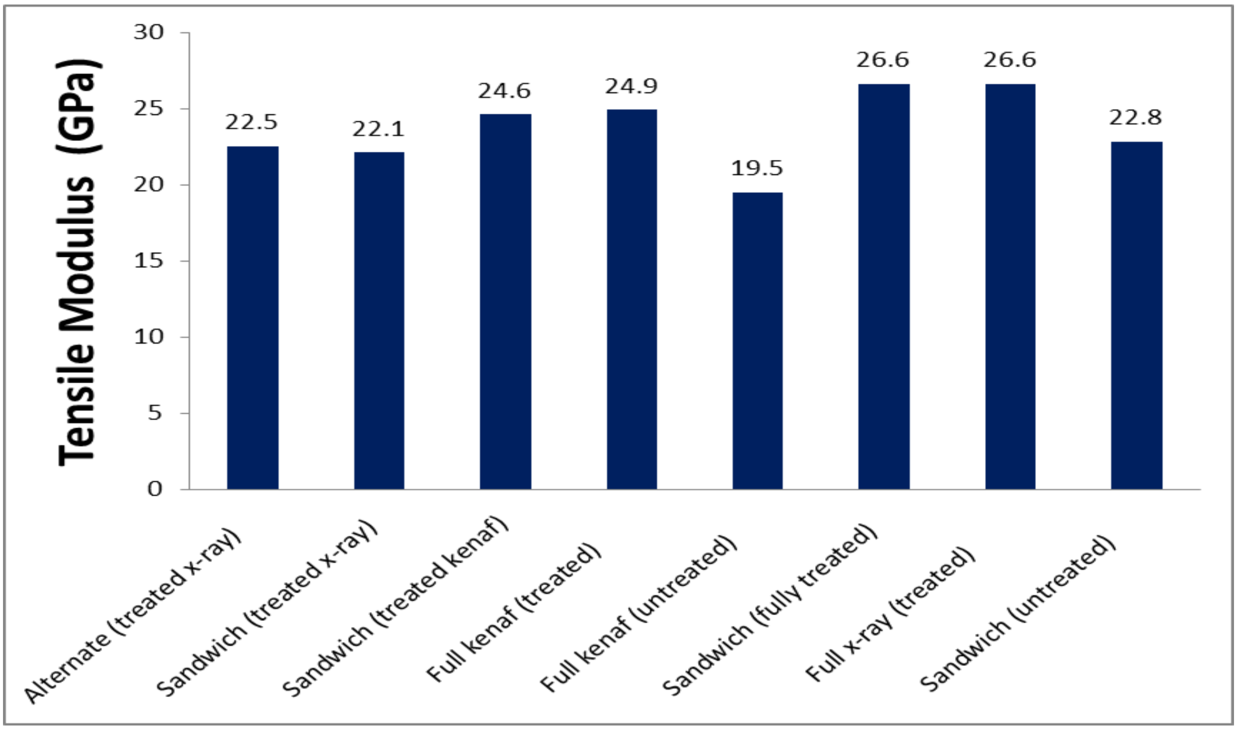
### 3.2 Mechanical properties

The tests were conducted with 5 specimens for each configuration. Table 5 below shows the mean results, which were derived from the average value of these tests.

| Sample | Configuration | Ultimate Tensile Strength (MPa) | Tensile Modulus (GPa) | Flexural Modulus (GPa) | Maximum Flexural Stress (MPa) |
|---|---|---|---|---|---|
| 1 | Alternate (treated x-ray) K-TX-K-TX-K-TX-K | 310.0 | 22.5 | 3.99 | 10.76 |
| 2 | Sandwich (treated x-ray) K-K-TX-TX-TX-K-K | 342.4 | 22.1 | 5.64 | 61.00 |
| 3 | Sandwich (treated kenaf) TK-TK-X-X-X-TK-TK | 277.6 | 24.6 | 5.47 | 27.45 |
| **4** | **Full kenaf (treated) TK-TK-TK-TK-TK-TK-TK** | 470.3 | 24.9 | **8.83** | **90.59** |
| 5 | **Sandwich (fully treated) TK-TK-TX-TX-TX-TK-TK** | **420.9** | **26.6** | **7.24** | **58.08** |
| 6 | Full kenaf (untreated) K-K-K-K-K-K-K | 222.3 | 19.5 | 5.33 | 74.10 |
| 7 | **Full x-ray (treated) TX-TX-TX-TX-TX-TX-TX** | **592.4** | **26.6** | 1.21 | 34.89 |
| 8 | Sandwich (untreated) K-K-X-X-X-K-K | 277.1 | 22.8 | 1.32 | 24.41 |



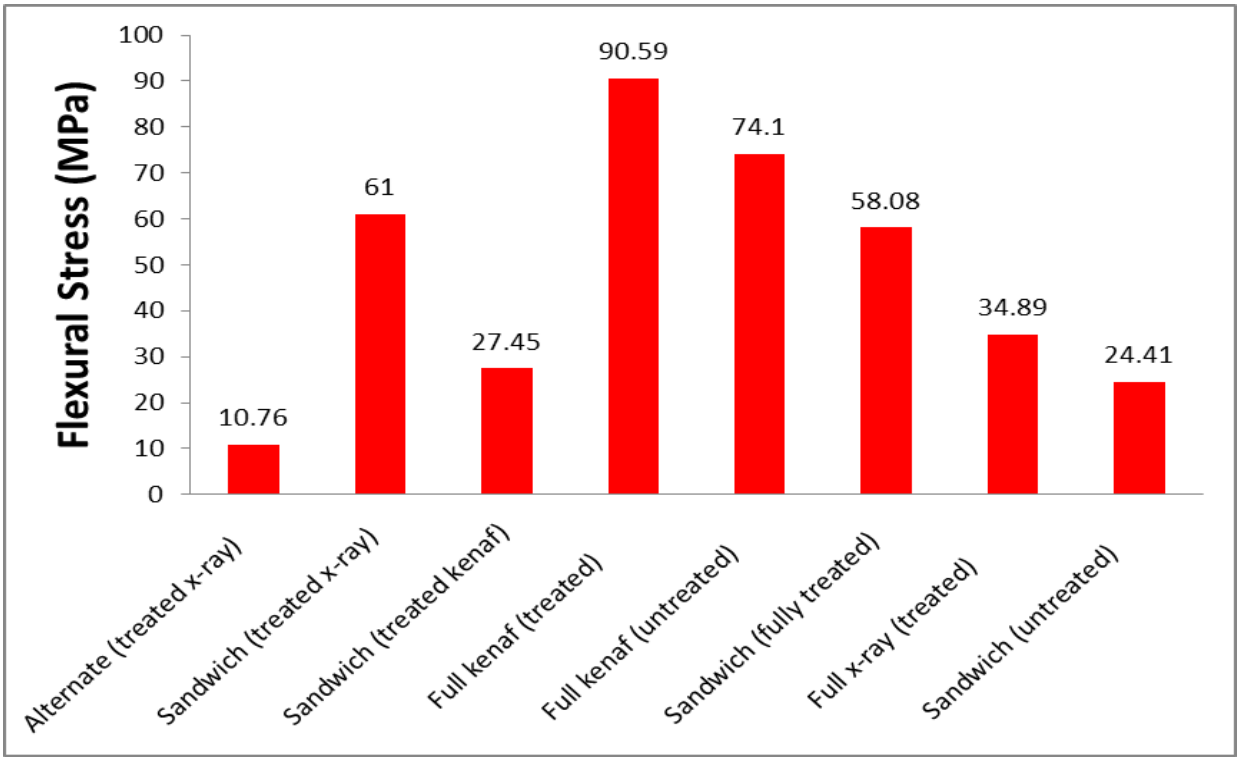

Graph 1 : Tensile strength of composite kenaf and x-ray film waste



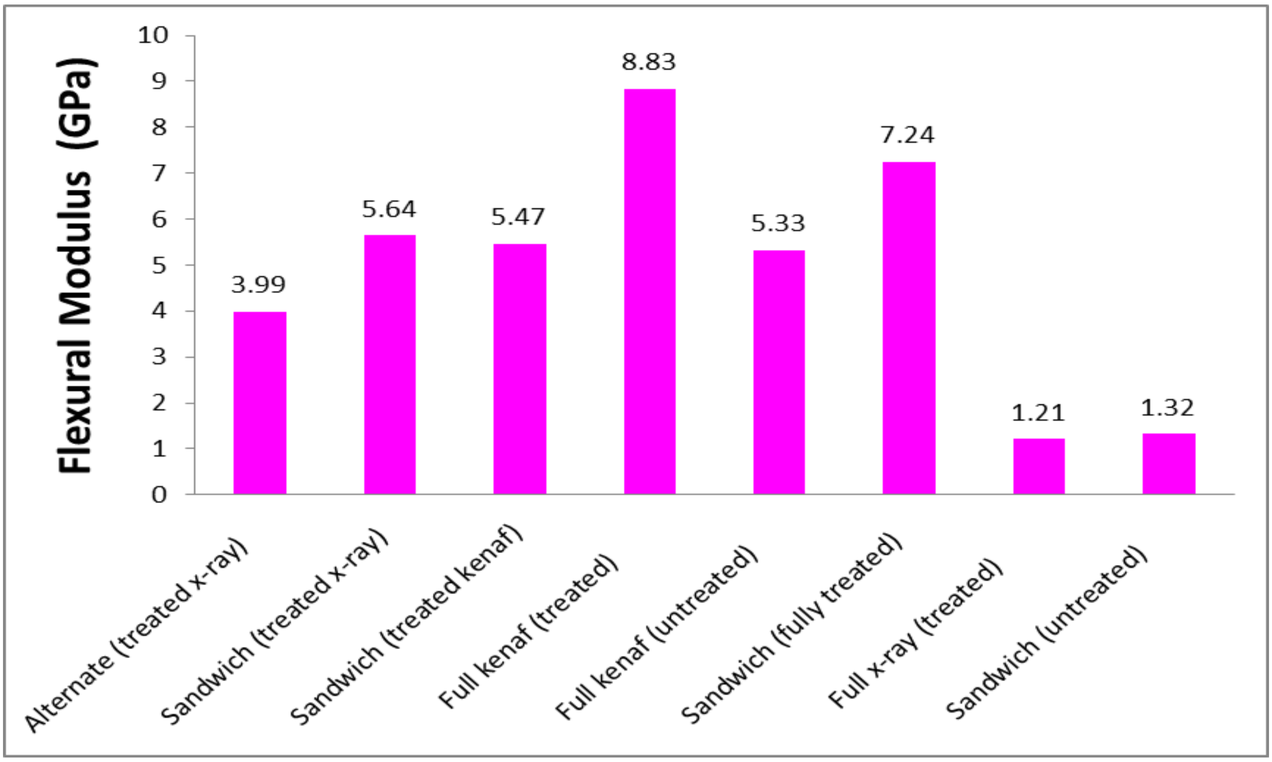

Graph 2 : Tensile modulus of composite kenaf and x-ray film waste

The table shows that sample 7 (fully treated x-ray film layers) had the highest tensile strength, followed by sample 4 (fully treated kenaf layers) the highest flextural stress and sample 5 (three layers of surface-treated x-ray films sandwiched in-between two layers of kenaf fiber treated with NaOH solution on the top and bottom of the layers).

The reason for sample 7 high strength was its plastic-based nature, and both sample 4 and 5 had no interfacial bond issues, in contrast to the other configurations that consisted of 2 different materials. Therefore, with the aim of producing a specimen with hybrid composite properties, Configuration 7 was chosen.




Graph 3 : Flexural stress of composite kenaf and x-ray film waste




Graph 4 : Flexural modulus of composite kenaf and x-ray film waste

The table shows sample 7 with the lowest flexural modulus and sample 4 with the highest. However, for bulletproof armour application, materials that are not too soft, thereby making it weak, and not too hard, thereby making it uncomfortable for the wearer, are desired. Hence, set 5 was again chosen for its moderate flexural modulus.

**Table 6.** Mechanical Properties of Natural Fibre Composites (Pickering *et. al.* 2016)

| Fibre | Ultimate Tensile Strength | Tensile Modulus (GPa) | Flexural Strength (MPa) | Flexural Modulus (GPa) |
|---|---|---|---|---|
| Sisal | 330.0 | 10.0 | 290.0 | 22.0 |
| Flax | 160.0 | 15.0 | 190.0 | 15.0. |
| Harakeke | 223.0 | 17.0 | 223.0 | 14.0 |
| Hemp | 165.0 | 17.0 | 180.0 | 9.0 |

Based on Table 6, the mechanical properties of other natural fibre reinforced polymeric composite have quite similar tensile strength to kenaf fibre. However, they were still not as strong as set 5. On the other hand, their flexural modulus was higher than kenaf fibre, meaning that they are rigid and not suitable for ballistic application.

Based on Fig. 3, when set 5 was compared with the other sandwich-based configurations (namely set 2 that uses only treated x-ray films, Configuration 3 that uses only treated fibre, and set 1 that uses non-treated layers), the maximum tensile stress of Configuration 5 considerably exceeded that of the other configurations.

It can also be observed that even though the maximum flexural stress of set 5 was lower than that of set 2 , the curve was steeper than the other configurations, which made it the most flexible. This quality favors manufacturers because they can produce more flexible equipment that fits its wearers more comfortably.

## 4.0 Conclusion

1. Even though treating the kenaf fibre with sodium hydroxide solution and the x-ray films with a surface treatment substantially improved the interfacial bond between the two materials, the interfacial bond was still lacking.
2. Configurations with full kenaf fibre layers gave a high tensile strength. However, they were very rigid, and this would make them very uncomfortable for the human body.

3. The chemical and surface treatments performed on the specimen only improved the interfacial adhesion between the two different materials; they did not improve in any way the interfacial adhesion between the same materials.
4. Even though the chemical treatment improved the bonding strength for kenaf to the x-ray film, it unfortunately weakened the strength of the fibre itself. This effect may be due to either the chemical thinning out of the fibres, or just an error in the treatment process.

## 5.0 References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This work is aimed to recycle X-ray film in making the kenaf / x-ray film hybrid composite material, which holds many promising properties. Note that there are also quite a few reports regarding such composite material. A search with keywords "kenaf x-ray film" would generate similar works also carried out in Malaysia.
(https://www.sciencedirect.com/science/article/pii/S2238785418307300,
https://www.sciencedirect.com/science/article/pii/B9780081022924000096?via%3Dihub)
The authors are encouraged to elucidate the uniqueness of their work, especially when compared with the published ones.