PREPARATION OF FLEXIBLE DIELECTRIC NANOCOMPOSITES USING NANOCELLULOSE AND RECYCLED ALUM SLUDGE FOR WEARABLE TECHNOLOGY APPLICATIONS

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Abstract

With the rapid development of wearable technology, flexible dielectric materials with environmental-friendly, low-cost and high-energy efficiency characteristics are in increasing demand. In this work, a flexible dielectric nanocomposite was developed by incorporating two components: cellulose nanofibrils (CNF) and recycled alum sludge, as the reinforcement phase and for enhancing the dielectric properties, in a bio-elastomer matrix. CNF and alum sludge were processed from waste materials which would otherwise be disposed to landfills. CNF were derived from water hyacinth, an invasive aquatic weed. Dried water hyacinth was treated using a simple and less energy-intensive process to obtain CNF. The alum sludge raw material was collected from a water treatment plant of Scottish Water and heat treated and refined before being used in preparing the composites. A biodegradable elastomer polydimethylsiloxane was used as the matrix and the nanocomposites were processed by casting the materials in petri dishes. The processed composites were characterised using scanning electron microscopy (SEM), thermogravimetric (TGA/DTG) and X-ray diffraction (XRD) analysis. The SEM micrographs illustrated CNF of approximately 20nm in diameter and alum sludge particles of approximately 200um in size. The TGA/DTG analysis results showed that a total mass of 46% has been removed when the sludge sample was heated up to 900°C. The XRD result showed that both quartz SiO_2 and cubic γ-Al_2O_3 structures can be found in the sample that was heat treated up to 800°C. Other experiments also showed that the composites exhibit comparable mechanical and dielectric performances with other works in the literature. The work depicts that it is a sustainable practice of reusing such wastes in preparing flexible, lightweight and miniature dielectric materials that can be used for wearable technology applications.

Keywords: *Nanocellulose, Biodegradable, Sustainable, Alum sludge, Nanocomposite, Wearable technology, Dielectric*

1. INTRODUCTION

As automation and data exchange help define manufacturing in the new Industry 4.0 era (Radanliev et al., 2021), portable electronic technology, wearable technology and big data technology are all in a rapid expansion stage, sharing common natures of flexibility, integration, miniaturisation and intellectualisation (Xu et al., 2021). Wearable technology are lightweight, smart electronic devices that are worn close to and/or on the surface of the skin, where they detect, analyse and transmit information from the user to the external devices (Düking et al., 2016). This can facilitate substantial applications including health monitoring (Memon et al., 2020) and human-machine interaction (Lin et al., 2022). In order to fulfill flexibility and integration requirements of wearable technology, researchers have paid more attention on processing novel materials for this type of devices, with flexible, low-cost, multifunctional properties, good mechanical performance and high-energy-efficiency characteristics (Xu et al., 2020, Yin et al., 2021). For example, Zhao *et al*'s work showed how flexible dielectric materials are critical in wearable technology (Zhao et al., 2020).

On the other hand, as the escalation of environmental issues such as climate change, severe environmental pollution and the depletion of fossil fuels and other resources, it has never been more urgent to explore more sustainable, environmental-friendly and biodegradable materials as alternatives to those with adverse impacts to the planet. Cellulose, as one of the naturally occurring polymers, has gained significant popularity as a candidate for the above campaign. Cellulose is a polysaccharide consisting of linear chains of D-glucose units (Updegraff, 1969). It is a key structural component in many plant-based sources, for example tree, hemp, wheat straw, bamboo and so on. In cellulose structure, elementary units align to form larger bundles of microfibrils and ultimately a cellulosic fibre matrix, within which hemicellulose and lignin are bound to the surface and entrapped within the microfibrils (ROBERTS, 2003). The extraction of cellulose nanomaterials using various methods has been widely reported (Istirokhatun et al., 2015, Mahardika et al., 2018); nanocellulose can be used as a key component for preparing nanocomposites because of the high modulus, strength and lightweight properties, as well as the inherent biodegradable and biocompatible characteristics (Phanthong et al., 2018). The work presented in this paper uses a pervasive aquatic plant, water hyacinth (Eichhornia crassipes) as the feedstock to derive the cellulose nanofibrils (CNF) as the key component for forming nanocomposites. Water hyacinth, which is considered as a pest plant, is very adaptable to the environment and can invasively grow in a rapid pace to destroy the surrounding environment (Mitan, 2018). Huge sums of money are being spent worldwide to selectively remove the weeds by manual harvesting and use them as animal feed or for energy production by direct combustion (Gunnarsson and Petersen, 2007). However, it is worth investigating the feasibility of using water hyacinth as feedstock for nanocelluloes processing due to their high cellulose content.

Another key component used for nanocomposite forming in this work is alum sludge, with a similar idea of waste management as for the water hyacinth material. Alum sludge is a by-product of water treatment process in which aluminium sulphate is prevalently used as the primary coagulant (Babatunde and Zhao, 2007). According to the data from a project partner-Scottish Water: there are approximately 10k tons of alum sludge generated from the water treatment work to be landfilled every year in Scotland. The industry has been facing escalating financial and environmental pressure to develop more sustainable strategies to deal with alum sludge wastes. In the available literature, some work of utilising alum sludge has been reported (e.g. aluminium recovery or agriculture and land reclamation (Dassanayake et al., 2015)). However, little work can be found of applying it in processing energy materials for enhanced energy density and efficiency. Therefore in this work, the recycled alum sludge was prepared and used in forming nanocomposites and its effect on the composites' properties will be investigated.

In order to process flexible nanocomposites, a biodegradable elastomer acetoxy-polysiloxane (PDMS) was used as the matrix. PDMS can be found in wide applications including biomedical engineering (Ceseracciu et al., 2015), energy harvesting (Jang and Oh, 2018) and micro-manufacturing (Chen et al., 2014), with its distinctive advantages of flexibility, biocompatibility, optical transparency, chemical and thermal stability and ease of manufacturing (Pereira Sales et al., 2022). In this work, the two key component, CNF and recycled alum sludge have been incorporated with PDMS to form flexible nanocomposites. The processed materials were characterised to investigate whether it can be a sustainable practice of reusing such waste materials in preparing flexible, lightweight and miniature dielectric materials that can be used for wearable technology applications.

2. MATERIALS AND METHODS

2.1. Materials

Water hyacinth (WH) stems were harvested from a local lake in Taman Tasik Seri Aman, Puchong, Malaysia. The WH stems were carefully separated from the roots and leaves, washed and air dried for 3 days before being ground into smaller size as raw materials (**Figure 1a**). Dewatered alum sludge cake was obtained directly from a local water treatment plant of Scottish Water (the Scottish Water Rosebery Water Treatment Works, Midlothian, Scotland). The average water content of the dewatered sludge was found to be approximately 76% (**Figure 1b**).

(a) **(b)**

Figure 1. (a) dried and ground water hyacinth material; **(b)** dewatered alum sludge cake sample collected from a treatment plant of Scottish Water.

Sodium hypochlorite solution (NaClO, 6-14% active chlorine), heptane ($CH_3(CH_2)_5CH_3$, 99%) and sodium hydroxide (NaOH) were received from Sigma-Aldrich, UK. Glacial acetic acid (CH_3COOH) was received from Fisher Scientific, UK. The biodegradable elastomer of acetoxy-polysiloxane (PDMS, Eastosil E43) was received from Wacker Chemicals Ltd., UK. All chemicals were reagent grade and used without further purification.

2.2. Processing Methods

The major experimental procedures involved in this work can be categorised into three parts: the processing of CNF, heat treatment and refinement of alum sludge and the forming of cellulose/alum sludge composites in PDMS matrix. **Figure 2** shows the schematic summary of the above processes.

2.2.1. Heat treatment of alum sludge

The dewatered sludge was firstly dried in an oven over a period of 24 hours at 105°C before being further processed. Once dried, the resulting crumb particle size was reduced using a planetary ball mill. Sieve separation was used and all material smaller than 250µm was retained; any particles larger than this were returned to the ball mill for further processing until all material passed through a 250µm mesh. The sieved material was then heat treated in a box furnace for 7 hours at 800°C using a heating rate of 50°C/min and left to cool overnight before removing from the furnace. The refined and heat treated powder was then used to prepare the composite samples.

2.2.2. Cellulose nanofibril processing

A simple and less energy intensive method was used to process CNF: dried water hyacinth was soaked in water to form slurry before being subjected to a delignification process using NaClO solution. Additionally, the material was processed in 1% NaOH solution to continue the extraction process. Both reactions were completed at room temperature and the material was washed using ultrapure water after each chemical reaction. Upon the completion of the above, a 0.5% cellulose slurry was prepared. To obtain nanofibrils, the cellulose slurry was further treated in a high shear homogeniser (PSI-20, Adaptive Instruments, UK) by passing it through a 200µm Z-shape interaction chamber for 1 pass, at 700bar. The obtained nanocellulose was eventually solvent exchanged from water into heptane to aid the composite forming process.

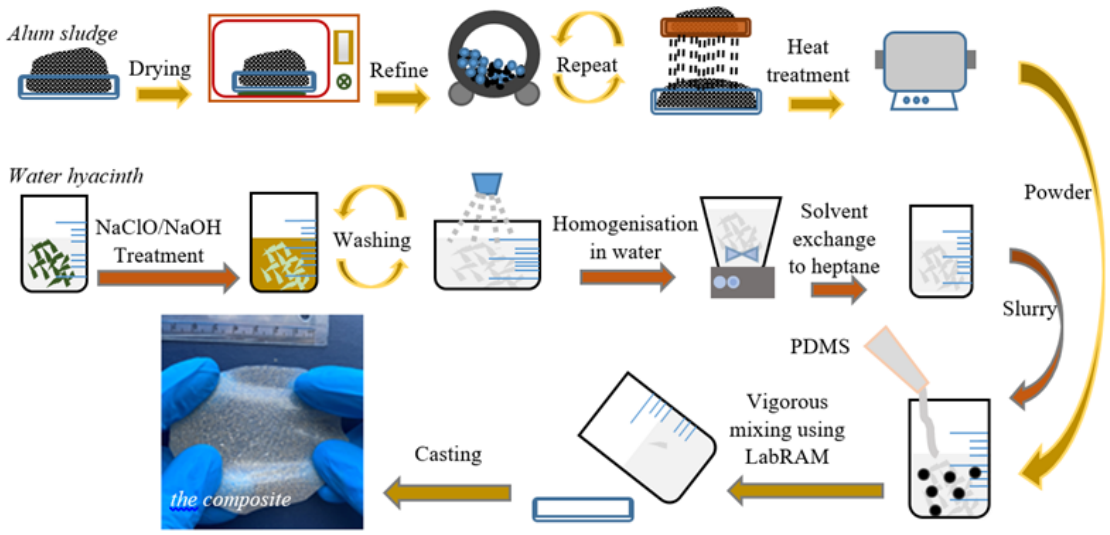


Figure 2. Schematic presentation of processing methods for alum sludge heat treatment, cellulose nanofibrils extraction and composite forming.

2.2.3 Composite forming

The components obtained from the above two works were mixed with PDMS with the presence of heptane solvent. The use of heptane solvent has necessitated a flowable condition of the mixture during the composite forming process. Vigorous mixing using an acoustic mixer (LabRAM, Resodyn, USA) and a rotor stator mixer (T 25 digital ULTRA-TURRAX, IKA, UK) was introduced to make sure all the components were uniformly dispersed. The mixed compound was subsequently poured in a petri dish and left to cure at the ambient condition. The dry weight of the formed composites is approximately 3g containing 10wt% of CNF and 10wt% of alum sludge powder.

2.3 Characterisation

The morphological features of the raw materials and the composite were examined using scanning electron microscopy (FESEM, Hitachi, UK). Thermogravimetric analysis (TGA/DTG) was conducted on the raw alum sludge crumb to determine its thermal properties. The solid structural analysis of the alum sludge samples with and without heat treatment were studied by X-ray diffractometer (XRD) at room temperature using Bruker D2 benchtop system.

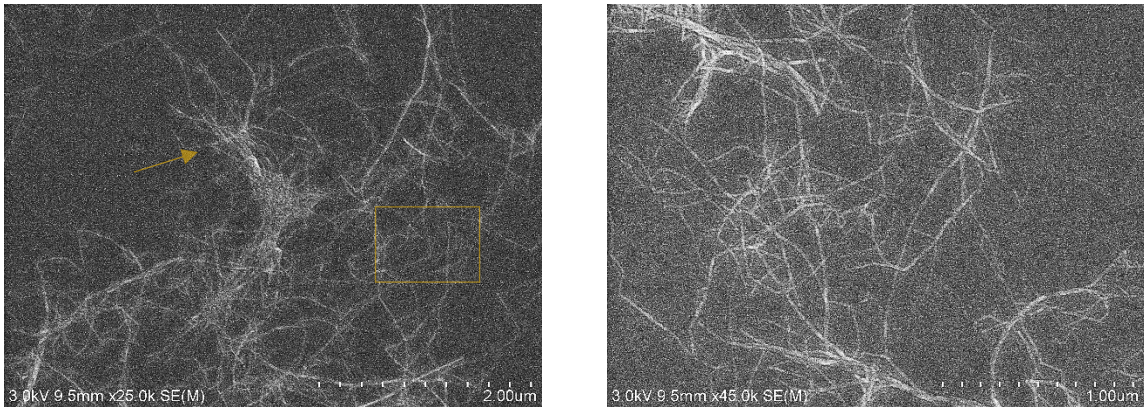
The dielectric properties were examined using a network analyzer (ZVA40; Rohde & Schwarz, Inc., Germany) with a split-post dielectric resonator (QWED, Poland) at 1.9 GHz. Each of the measurements was repeated five times.

The mechanical properties of the composites were also investigated in this work. A universal testing machine was used in which the material was pulled to break at a rate of 100mm/min.

3. RESULTS AND DISCUSSION

CNF were successfully obtained using both chemical and mechanical steps. In the first step, lignin and other organic substance was removed in the acidified NaClO solution, followed by the removal of hemicellulose in the NaOH solution. This can be observed by the change of material's colour (from dark brown to white) in the process. The resultant material was then placed in a Z-shape interaction chamber of approximately 200 µm in diameter, where a significant amount of shear force was generated to fibrillate the material into nanofibrils. **Figure 3** indicates the microstructure of CNF obtained from the above process. It is obvious that the long nature of the fibrils in addition to the dying process before the

SEM imaging, resulted in the fibril aggregation and a formation of a web-like network structure. This observation is similar with the ones in some other published works (Chen et al., 2011). The enlarged image in **Figure 3(b)** clearly shows the morphology of individual fibrils, which can be estimated as approximately 20nm in diameter. Due to the long nature of fibrils, it is not possible to measure their length accurately; however, it can be estimated from **Figure 3(a)** that the length of fibrils can be in the order of several micrometers.

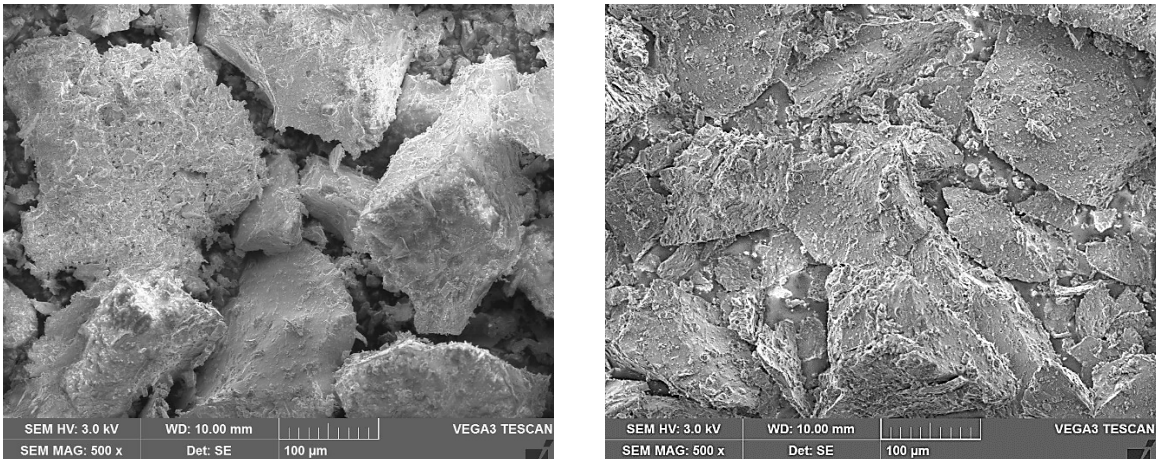


(a) **(b)**

Figure 3. (a) FESEM images of cellulose nanofibrils showing the web-like network structure; the arrow highlights a unfibrillated fibre bundle; and **(b)** the enlarged image of the rectangular area in **(a)**, showing the morphology of individual nanofibrils.

Nevertheless, it is also quite evident that a small amount of large, unfibrillated fibre bundles were present in the images, having a diameter of approximately several hundred nanometers. This could be the result of the incomplete fibrillation of material during the homogenisation step. The previous work has proven that the degree of fibrillation is determined by various aspects including the nature of the feedstock, processing time and the amount of shear force applied on the materials; the latter two aspects are directly correspondent to the amount of energy consumed in the process. In this work, the material was processed using 200 µm for 1 pass (1 cycle), which is considerably less than the one used in previous work where the material is passed through a 100µm chamber up to 5 cycles. The subsequent experiments in this work have identified that the processed material is still suitable for the composite forming process.

Both the dried raw alum sludge crumb and heat treated samples were characterised using SEM which are shown in **Figure 4 (a)** and **(b)**, respectively. As seen, the sludge particle size and shape are irregular with a rough surface texture. This concurs with observations reported elsewhere (Alves et al., 2014, Soleha et al., 2016). No significant difference in the morphology can be observed when comparing the dried raw sludge and the heat treated samples, although the surface of the latter looks slightly compact. It is necessary to investigate the surface area of the two samples using porosimetry, which is planned as a future work.



(a) **(b)**

Figure 4. Microstructure of alum sludge crumb without **(a)** and with **(b)** heat treatment at 800°C.

The TGA/DTG thermogram elucidating the thermal stability of the raw alum sludge crumb is shown in **Figure 5**. It is seen that a noticeable amount of weight (approximately 46%) has been lost when the sample was heated up to 900°C. Three steps can be seen in the weight loss profile: i) up to 100°C, approximately 4% of weight, which could mainly be associated to the physically absorbed moisture (remaining free/unbound water), has been gradually eliminated. ii) There is a major weight loss between 105°C and 500°C. This loss could possibly be because of the removal of the remaining bonded water and the decomposition of some organic compounds (e.g. polyaluminium chloride hydroxide sulfate, one of the key components in the sludge, usually decomposes at about 200°C (Soleha et al., 2016)). It is reported that the decomposition of polyacrylamide, an anionic organic polymer (e.g. Magnafloc®) that is commonly used to improve the dewatering of alum sludge in the water treatment process, can occur in between 350°C and 550°C (Bache and Zhao, 2001); this could be another aspect for the above weight loss in our work. iii) Above 500°C, the curve started plateauing, showing insignificant weight loss in this range. All these observations are correlated with the thermal event shown in the DTG curve in the same temperature range. In all, it is notable that the alum sludge used in this work contains various compounds that are thermally unstable; thus it is necessary for them to undergo some heat treatment to remove the volatile substances before being used for forming the composites. A TGA/DTG test for the heat treated samples were not conducted in this work but is believed to be necessary for a comparison with the one of untreated samples to see the efficiency of the volatile substance removal. This is planned as the future work.

The XRD patterns of the samples characterised by SEM (**Figure 4 (a)** and **(b)**) are shown in **Figure 6**. The spectrum for the raw alum sludge crumb exhibits a hexagonal quartz SiO_2 structure. This could be attributed to the presence of clay and sand particles suspending in the treated water during the purification process. Other compounds in the sludge do not exhibit any detectable crystal structure.

The XRD spectrum for the sample that has been heat treated at 800ºC shows extra peaks in addition to those for quartz SiO_2 and similar features can be found in other works too (Owaid et al., 2013, Awab et al., 2012). It is believed that these peaks are associated to cubic γ-Al_2O_3 which was formed in the oxidation process at elevated temperatures. Since sharper and more intense peaks indicate greater crystallinity, it is apparent that the aluminium oxide is of relatively low crystallinity, even after heat treatment at 800°C. Therefore, it is worth trying to investigate whether this can be improved by using higher temperatures in the heat treatment and how this would impact on the dielectric properties, which can be one of the future works.

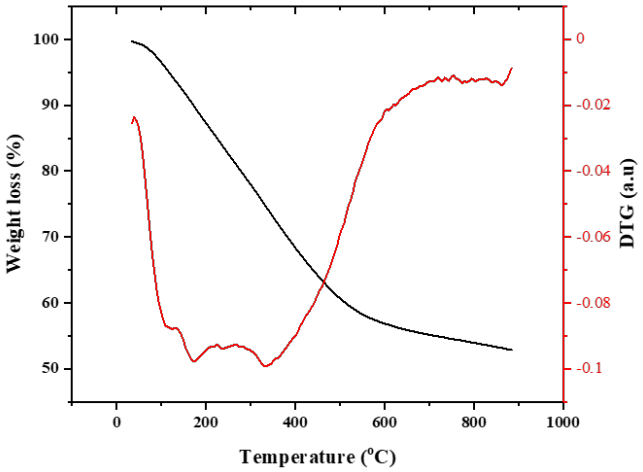


Figure 5. TGA/DTG thermograms of the raw alum sludge crumb.

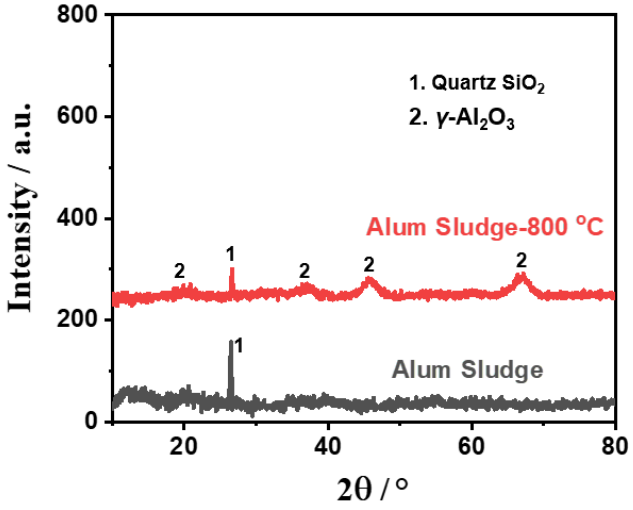


Figure 6. XRD spectra for the raw and 800°C heat treated alum sludge samples.

A SEM micrograph showing the cross-sectional structure of the composite film can be found in **Figure 7**. It can be estimated from the image that the average thickness of the sample is approximately 450µm, which also coincides with the value measured using a digital micrometer. The thickness is mainly determined by the dry weight of the formed composite, which in this case is approximately 3g. It is pronounced that alum sludge particles are embedded in the matrix with an even distribution. It is not possible to observe cellulose nanofibrils in the micrograph of small magnifications as in **Figure 7** because of their size in nanometre scale and high embedment in the PDMS matrix.

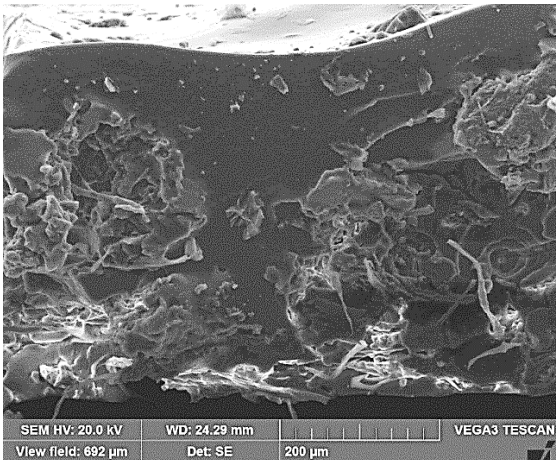


Figure 7. SEM micrograph of cross-sectional structure of the composite film.

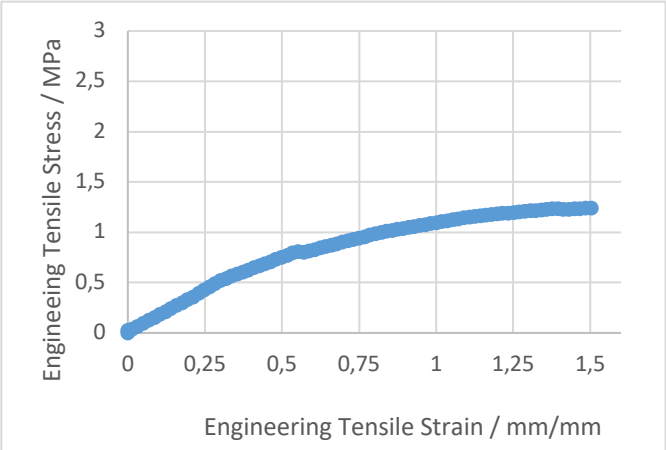


Figure 8. Engineering tensile stress v.s. engineering tensile strain curve for the composite film.

The mechanical tests indicates that the average tensile strength of the composite is 1.1 ± 0.12MPa and the percentage elongation is 108% ± 25%. The deviation of the above values may be because of the small flaws present in the films that could affect the mechanical properties to some extent. The engineering stress and strain curve of a representative specimen is shown in **Figure 8**. The percentage elongation value is comparable with the one of neat PDMS film in Johnston *et al*'s work (Johnston et al., 2014), although the tensile strength value is relatively lower. The flaws (e.g. cracks or voids) generated during the composite forming could be a reason but the compatibility of the reinforcement components (CNF and alum sludge) with the polymer matrix also plays an important role of determining the mechanical properties of the composite. These will be further investigated in the future work.

The dielectric property measurement indicates that the film with a thickness of approximately 450µm has a permittivity of 2.77 ± 0.01 and a loss tangent of 0.0231 ± 0.005 at 1.9 GHz. Its quality factor (Qf value) was also obtained as 11685 GHz. These data are comparable to the dielectric properties of cellulose-based materials in terms of permittivity and loss tangent near 1 GHz (Khouaja et al., 2021), indicating a promising candidate for sustainable composite substrates for the next generation of telecommunications.

4. CONCLUSIONS

A work demonstrating the use of waste materials to process nanocomposites as dielectrics was successfully completed. Two waste materials, dried water hyacinth and alum sludge were used to process key components for the composites as reinforcements and for enhancing the dielectric properties. A simple and less-energy intensive method was used to extract CNF from dried water hyacinth and the as received alum sludge was heat treated and refined to be suitable for the composite forming process. More experimental works are needed for optimising the heat treatment of alum sludge and studying mechanical properties of the composites. Nevertheless, the work has shown strong evidence that it could be a sustainable practice of reusing such waste materials in preparing flexible, lightweight and miniature dielectric materials that can be used for wearable technology applications.

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