Final Report

FlexFund Project [FF2021-1036]: Submerged bi-axial fatigue analysis for flexible membrane Wave Energy Converters

Aims and objectives: The aim of this project was to enhance the fatigue behaviour of polymeric materials used in energy harvesting from ocean waves, through the synthesis of a synergy of fillers with natural rubber (NR) and characterised by bi-axial submerged fatigue tests. To achieve these aims, during the project period, we invested efforts in the three sectors as follows:

1. **Bespoke Biaxial Fatigue Rig for Soft Materials: ADMET eXpert 8000 Planar Biaxial Fatigue Testing System**

A significant portion of efforts has been directed towards developing a state-of-the-art bespoke biaxial fatigue machine suitable for soft polymers. This is a totally customised fatigue machine, unique in the UK that can test a wide range of filled and unfilled polymers and is being developed during the project period. The machine has three separate units as shown in Fig.1a:

1- The biaxial rig with two independent axes (1 kN maximum load capacity on each axis).
2- Temperature control unit for the submerged test (10 °C to 30 °C).
3- DIC (2D- Digital Image Correlation) for strain measurement and crack propagation monitoring.

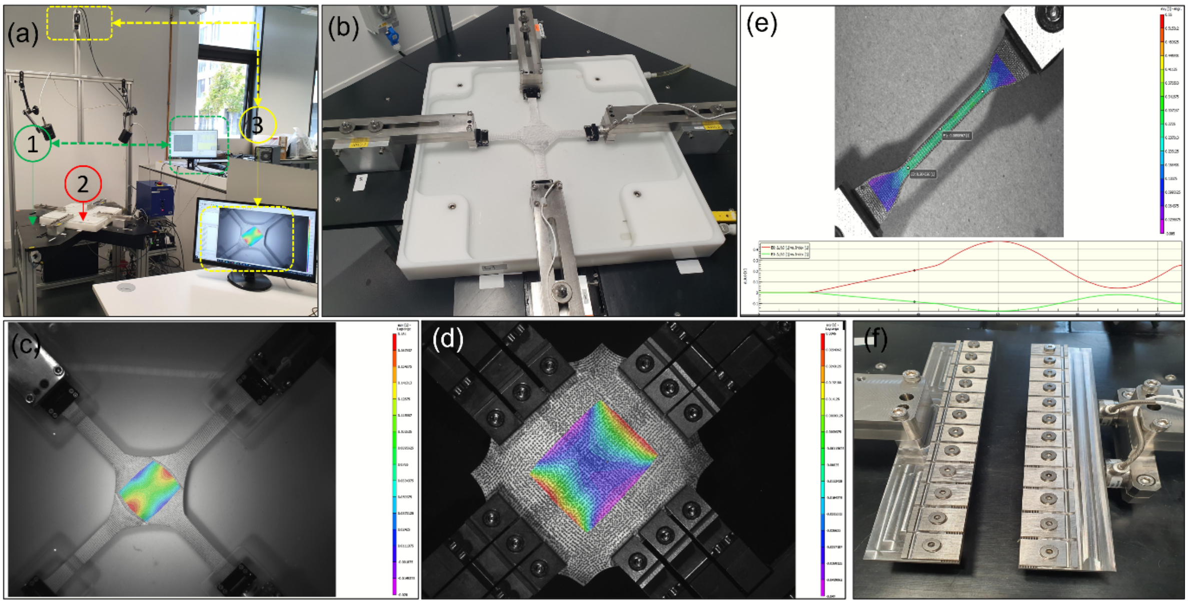

Figure 1. Biaxial machine: (a) three units of the machine, (b) submerged test setup using cruciform sample, (c) Strain distribution within the cruciform sample using DIC, (d) strain distribution within the square sample using DIC, (e) strain distribution within the dog-bone sample using DIC, (f) the grip for testing 12 samples at a time.

The machine is designed for high cyclic uniaxial and biaxial (equi-biaxial and nonequi-biaxial) fatigue tests for soft polymers in dry and submerged conditions. It is capable of testing with minimum and maximum speeds of 0.05 mm/sec and 500 mm/sec, respectively. The maximum distance between opposing crossheads can be reached up to 590 mm (i.e. high strain amplitude up to 500 % for small samples). The machine is equipped with a temperature bath (Fig.1b) and temperature control system for the submerged test (Fig.1b-c) which is one of the main features of the machine for testing flexible membranes being used in wave energy converters. The machine is equipped with different grips for testing various samples including dog-bone, rectangle, square, and cruciform samples in which 12 samples can be tested at a time (Fig.1 f). A lateral sliding pinching grip system for a square planar biaxial sample was provided for the machine (Fig.1d). It can be programmed in either displacement or load control modes. Moreover, the machine can perform a variety of waveform patterns including sawtooth, square, triangle, and sinusoidal up to 8 Hz. Two different load cell capacities were provided including 1 kN and 100 N so that extremely soft elastomers can be thoroughly tested using the latter one. A 2D Digital Image Correlation (DIC) System analysis software includes post-processing features such as minimum/maximum, mean and standard deviation, time-slice extraction, stress-strain curve generation, and data extraction. The package contains algorithms to provide full-field displacement and strain data for mechanical testing on planar specimens and includes the ability to graphically display the data as an overlay over test specimen images (Fig.1c-e). The operator can easily identify strain distributions from the image-overlay contour plots or contour plots independently. Optical surface deformation measurements can be used for but not limited to material testing including fracture mechanics, biaxial tension tests, Finite Element (FE) validation such as strain localization around cracks and notches, and general component testing. A 5.0 Megapixel (2448 x 2028 @ 75 fps) digital camera was provided with a resolution of 10 microns when used with the largest Field of View in the range of 300mm x 300mm.

2. Material Manufacturing

Many efforts have been carried out on tailoring the mechanical properties of Natural Rubber (NR) reinforced with Carbon Nanotubes (CNTs) [1,2]. However, it was strongly dependent on the state of CNT dispersion and interfacial bonding of NR/CNTs, especially for better fatigue life, energy dissipation, and energy harvesting efficiency for wave Energy Convertors (WEC). Achieving a homogenous CNTs dispersion within the matrix is challenging due to large surface energy and intensive van der Waals forces amongst CNTs. This creates many agglomerations which result in deteriorating mechanical and fatigue properties of the NR. Therefore, it is essential to develop an optimum compounding approach. In this project, two different approaches were used as follows: one batch is prepared using solution mixing (Fig.2a) assisted by ultrasonication in the initial stage, accompanied by further compounding using a twin roll mill to remove the entrap bubbles efficiently. The other one is subject to only mechanical mixing in dry conditions (Fig.2b). Although, a better CNT dispersion can be reached in the former (Fig. 2c), solvent removal is quite challenging as many black spots can be seen in Fig.2c [3,4]. In addition, improper CNTs dispersion is the main drawback of the mechanical/melt technique as manifested in Fig. 2d. Moreover, different CNTs loadings were used to find out the optimum CNTs content. This was done using the microstructural (Fig.2c-d), rheological, and dynamic shear characterization (Fig.2e-g). As it is showed in Fig.2h-i, both solution and melting methods manifest no significant differences in the dynamic shear properties and hysteresis losses at various strain amplitudes. In addition, solvent-free nature of the latter and its low cost made it the optimum compounding approach. Regardless of the CNT content used, the incorporation of CNTs into the NR improved the dynamic shear properties of the nanocomposite compared to the pristine rubber as shown in Fig.2 j-k. The storage modulus and the loss modulus of filled rubbers are enhanced to 201 % and 1232 %, respectively, for 10 phr CNTs loading at 200 % strain amplitude compared to the control. Finally, CNT increased energy dissipation of the nanocomposites compared to the NR in which an increase of 1040 % in energy dissipation for 10 phr MWCNTs with respect to the control at a strain amplitude of 200% was achieved. The loading-unloading curve shows more nonlinearity at higher strain for higher CNT contents (Fig.2k), which was related to more pronounced Mullins and strain-induced crystallization effects, i.e. the nonlinearity was more remarkable at higher CNTs loadings.

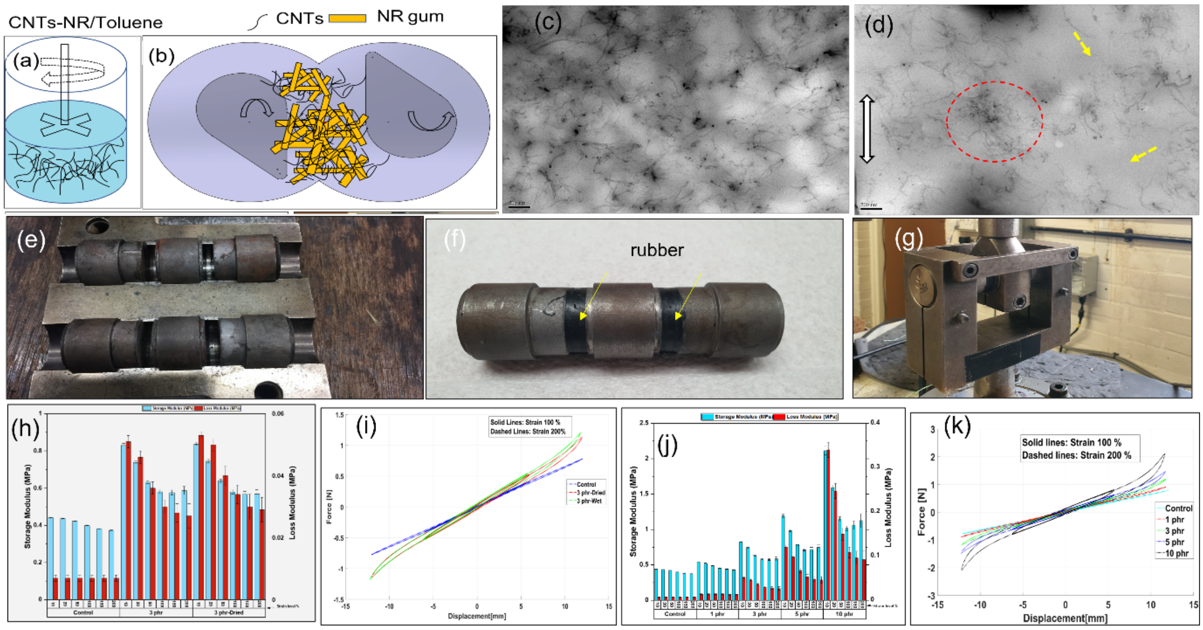

Fig. 2. (a-b), Solution and mechanical/melting methods, respectively, (c-d) TEM image of solution and melting methods respectively, (d-e) Preparation of double bonded shear test piece, (f) double bonded shear test, (g-h) comparison of dynamic shear properties different compounding approaches, (i-j), dynamic shear properties of different CNT loadings.

3. Fatigue Tests of CNT-filled Natural Rubbers

In the previous section, microstructural and dynamic shear properties of different compounding approaches as well as various NR/CNT contents subjected to few cyclic shear stress were analysed. It was concluded that 3 phr CNT content can reach better mechanical properties. In addition, although solution approach manifests relatively a more homogenous CNT dispersion compared to dry technique, no significant differences were distinguished between them in quasi-static and low cyclic shear conditions. Since the main application of NR developed in this study is for flexible membrane used in WECs which is subjected to high alternating loading conditions, it is essential to compare fatigue life of the NR/CNT prepared by solution and mechanical methods. Therefore, a uniaxial fatigue test has been running in our lab in both dry and submerged conditions to mimic NR behaviour in both dry and subsea conditions (Fig. 3a). A dog-bone sample according to ISO 37-type 2 was used for the uniaxial fatigue characterization (Fig.3b). Strain amplitude in the range of 50 % to 200 % with the interval of 50 % was used to plot S-N curve of the sample. A fixed 3 phr CNT was used for both compounding approaches to find out which methodology reaches to a better fatigue life. Then, they will be compared with the neat natural rubber in terms of quasi-static mechanical properties and hysteresis loss and uniaxial and biaxial properties. A sinusoidal wave form at 2 Hz frequency and approximately similar R-ratio, the ratio of minimum strain to maximum strain, were applied to all samples. A positive ratio was used to obtain better fatigue life due to strain induced crystallization of rubber [5]. The strain was measured using Digital Image correlation (DIC) in which a homogenous strain is achieved throughout the gauge length as shown in Fig.3c. [5]. A crack nucleation approach was used in this study instead of crack growth approach which we believe can reach more reliable results of fatigue behaviour of the NR. This is because nucleation of many tiny cracks and their extensions with respect to time are real phenomena taking place in NR (Fig.3d) rather than formation of individual critical crack which is the fundamental in crack extension approach. Fig.3e shows the fatigue life of dry (mechanical/melting) and wet (solution technique) compounding approaches obtained in dry condition at three different strain amplitudes. The former can reach much better fatigue life, approximately three times larger, compared to the wet condition at all strain levels. This is mainly attributed to the presence of many tiny pores and voids within the samples prepared by the wet method resulting from the toluene remained in the sample. However, no significant pores and porosities were noted for the samples prepared by dry method. This is a significant finding obtained in this study as many of the published works in the literature indicated that wet dispersion approach can be a better method in NR/CNTs synthesis. However, our fatigue test outcomes indicate dry method is ideal approach for membrane applications as they require high fatigue life. It can be concluded that although wet dispersion approach can achieve better CNT dispersion state, it sacrifices the fatigue life of the rubber; therefore, it cannot be necessarily a good approach for many NR applications which require high fatigue life. Two journal papers were published so far related and we are expecting to submit few more soon upon the completion of fatigue test [6,7].

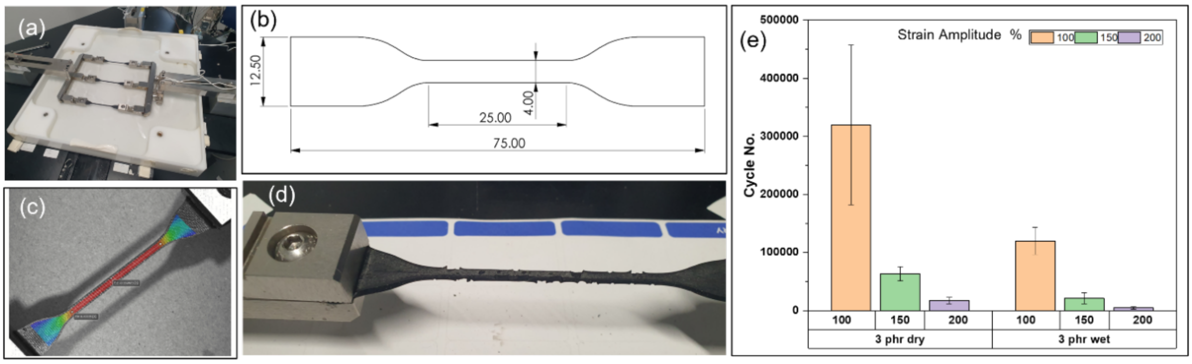

Fig. 3. (a) Uniaxial fatigue test setup in submerged test, (b) dog-bone sample geometry, (c) strain distribution within the sample using DIC, (D) formation of several tiny crack within the gauge length during fatigue test, (e) Comparison of fatigue life of two different compounding approaches at different strain amplitudes.

Published papers during/acknowledging this project [2021-2023]:

<u>Published:</u>

- Esmaeili, I. Masters I, M. Hossain. Comparison of two compounding techniques for carbon nanotubes filled natural rubbers through microscopic and mechanical characterizations, Materials Letters, 133:133786, 2023

- Z. Liao, J. Yang, M. Hossain, G. Chagnon, X. Yao. The time and temperature dependences of the stress recovery of Ecoflex polymer, International Journal of Non-Linear Mechanics, 149:104338, 2023

- Esmaeili, I. Masters, M. Hossain. A novel carbon nanotubes doped natural rubber nanocomposite with balanced dynamic shear properties and energy dissipation for wave energy applications, Results in Materials,17:100358, 2023

- Collins, M. Contino, C. Marano, I. Masters, M. Hossain. The influence of time-dependency on the behaviour of elastomeric membranes for wave energy harvesting using experimental and numerical modelling techniques, European Journal of Mechanics-A/Solids, 98:104895, 2023

- J. Yang J, Z. Liao, M. Hossain, X. Yao. Thermo-mechanical properties of digitally-printed elastomeric polyurethane: experimental characterization and constitutive modelling using a nonlinear temperature-strain coupled scaling strategy, International Journal of Solids and Structures, 267:112163, 2023

- Collins, M. Hossain, W. Dettmer, I. Masters. Flexible membrane structures for wave energy harvesting: A review of the developments, materials and computational modelling approaches, Renewable and Sustainable Energy Reviews, 151:111478, 2021

- A. K. Bastola, M. Hossain. The shape-morphing performance of magnetoactive soft materials performance, Materials and Design, 211:110172, 2021

Under review :

- K. V. S. Gurjar, A. S. Sadangi, A. Kumar, D. Ahmad, K. Patra, I. Collins I, M. Hossain. Dielectric Elastomer Generators: Recent Updates on Materials, Electronic Circuits, Prototype Developments, and Performance Evaluation, Renewable and Sustainable Energy Reviews, In Review, 2023

- Esmaeili, D. George, I. Masters, M. Hossain. Biaxial experimental characterizations of soft polymers: A critical review, Polymer, In Review, 2023

References:

1. Sethulekshmi, A. S. S., Saritha, A. & Joseph, K. A comprehensive review on the recent advancements in natural rubber nanocomposites. *Int. J. Biol. Macromol.* **194**, 819–842 (2022).

2. Medupin, R. O., Abubakre, O. K., Abdulkareem, A. S., Muriana, R. A. & Abdulrahman, A. S. Carbon Nanotube Reinforced Natural Rubber Nanocomposite for Anthropomorphic Prosthetic Foot Purpose. *Sci. Rep.* **9**, 1–11 (2019).

3. Ismail, H., Ramly, F. & Othman, N. Multiwall carbon nanotube-filled natural rubber: The effects of filler loading and mixing method. *Polym. - Plast. Technol. Eng.* **49**, 260–266 (2010).

4. Kueseng, K. & Jacob, K. I. Natural rubber nanocomposites with SiC nanoparticles and carbon nanotubes. *Eur. Polym. J.* **42**, 220–227 (2006).

5. Diani, J. & Gall, K. Finite Strain 3D Thermoviscoelastic Constitutive Model. *Society* 1–10 (2006) doi:10.1002/pen.

6. Esmaeili, A., Hossain, M. & Masters, I. Comparison of two compounding techniques for carbon nanotubes filled natural rubbers through microscopic and dynamic mechanical characterizations. *Mater. Lett.* **335**, 133786 (2023).

7. Esmaeili, A., Masters, I. & Hossain, M. A novel carbon nanotubes doped natural rubber nanocomposite with balanced dynamic shear properties and energy dissipation for wave energy applications. *Results Mater.* **17**, 100358 (2023).