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Effect of accelerated weathering on the performance of natural fibre reinforced recyclable polymer composites and comparison with conventional composites

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ABSTRACT

This work investigated the long-term performance of unidirectional (UD) flax fibre reinforced recyclable polymer composites (FFRPs), after accelerated weathering for a total of 56 days, in laboratory environment. The employed recyclable polymer matrices were a bio-based epoxy and a liquid thermoplastic acrylic resin. The performance of the developed composites, were compared with traditional UD glass fibre composites (GFRPs), employing a standard petro-based epoxy resin, in terms of flexural and viscoelastic properties, while visual inspection and Scanning Electron Microscopy (SEM) were employed to investigate further the effects of ageing after exposure. It was revealed that the reinforcing fibres dominated the performance of the composites after ageing, whilst the choice of polymer matrix exhibited less influence.

Introduction

Recent decades have been challenging for the humanity due to global warming and the consequent climate change. To tackle that, from the side of engineering, there is a global endeavour towards circular economy by employing circular and bio-based materials [1–3]. At European level, strategies like the EU Green Deal and Circular Economy action plans have recently emerged [3,4], whilst there is an expectancy of new legislations related to sustainability, that signifies further the need for bio-based and recyclable materials [5]. In the field of composite materials, natural fibre reinforcements and bio-based and/or recyclable polymer matrices are of primary importance [6–10]. Natural fibre reinforcements possess advantageous properties in comparison to glass fibre reinforcements, such as: (i) low density, (ii) comparable specific stiffness, (iii) biodegradability, (iv) renewability, and (v) low energy consumption [10–12]. However, their highly hydrophilic character and therefore low durability impedes their usage in high-performance structural applications [6,13].

Flax is a dominant natural bast fibre that has extensively been used in composite materials, as a natural fibre reinforcement [14,15]. The fibre itself possesses 62–72% cellulose, 18.6–20.6% hemicellulose, 2–5% lignin, 2.3% pectin, and 8–12% moisture content. The density of flax fibre is 1.4–1.5 g/cm^3, tensile strength 343–2000 MPa, and stiffness 27.6–103 GPa [16]. Flax fibres are lighter in weight and have higher specific stiffness than glass fibres [16]. Flax fibre composites are used in automotives, maritime, household, sports, recreations, and other engineering structures [14,15,17], and have potentiality to employ them in aeronautics [18–20], wind energy [21–23] and even in space applications (satellites) [24,25]. From the environmental impact point of view, flax fibre composites contribute less impact than their glass fibre counterparts for the same applications [26,27].

Contrary to their lower environmental impact, flax fibre composites exhibit poor performance in the long-run, in comparison with glass fibre composites, mainly as aforementioned due to hydrophilicity [28,29]. Long-term performance of flax fibre composites has long been studied using accelerated ageing, under different conditions such as hygrothermal [28,30,31], salt-fog spray [32], weathering or UV/spray [29,33,34], freeze/thaw [35], soil environment [36], etc. In general, significant deterioration of mechanical performance has been reported, after accelerated weathering, of which the magnitude is largely dependant on the weathering conditions such as the intensity of UV irradiance, temperature levels, etc., as well as ageing duration. It has also been reported that specific chemical treatments may alleviate ageing degradation [34]. Since flax is a lignocellulosic plant fibre which are highly sensitive to moisture, and obviously also prone to degradation by the synergistic effects of UV radiation, moisture, and heat, as reported by Refs. [29,33,

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34]. On the other hand, glass fibre reinforced composites, are more resistant to weathering due to the (relatively greater) hydrophobic nature of glass fibres and better interfacial bonding between glass fibres and polymer matrix [37,38].

With respect to the polymer matrix, circular/recyclable and bio-based polymer matrices are also currently being studied for composite applications, to replace petroleum derived polymer matrices, that are usually non-renewable, non-recyclable, non-reusable, non-biodegradable, contributing to fossil fuel depletion and CO_2 emissions, negatively affecting the environment [39–43]. The incorporation of recyclable and/or bio-based polymer matrices in composites may allow for recyclability, non-toxicity, and lower environmental impacts [40–45]. Given the market need for circular polymers, there is at the moment a number of off-the-shelf bio-based epoxy resins, under the brand-name of Polar Bear, Sei Whale, Green Turtle, Spinner Dolphin, Plankton, Super Sap® 305 system, One epoxy resin, SR GreenPoxy 56, SR Greencast 160, SR InfuGreen 810, SR Surf Clear Evo, SR Firegreen 37, PB 410 GS, Super Sap INR, RSF 816R-G, etc., as well as new liquid acrylic thermoplastic resins (i.e., Elium®). Bio-based and recyclable epoxy resin systems are processed similarly to conventional synthetic resin systems, although liquid thermoplastics brought a breakthrough in the composites industry, since they allow processing with Out-of-Autoclave manufacturing methods, without compromising recyclability [45,46] and performance [31].

The combination of flax fibre reinforcements with sustainable (recyclable) polymer matrices, is a potential solution to ensure circularity and tackle End-of-Life (EoL) composites waste [3,4]. However, the understanding of the behaviour of such composites, in the long-term perspective requires more attention, due to the fact that both natural fibre reinforcements and recyclable resin systems are found to be more prone to environmental ageing, compared to traditional synthetic composites [6,33,47]. Long-term performance assessment, is of primary importance, in order to allow for their adoption in engineering applications. It is also noted that there is a lack of substantial research work on the long-term performance assessment under accelerated weathering of unidirectional (UD) flax fibre reinforced polymer composites. Also, there is a lack of research works on the comparative long-term performance between UD flax fibre composites (also, NFCs – Natural Fibre Composites) and UD glass fibre composites under exposed in accelerated weathering conditions. Hence, the objective of the current research is to study the long-term performance of flax and glass fibre reinforced polymer composites by incorporating recyclable polymers (a bio-based epoxy thermoset and an acrylic based liquid thermoplastic) in a lab-scale accelerated weathering chamber (QUV/spray) for a total of 56 d For reference purposes, the performance of these composite materials was compared with a traditional non-recyclable petro-epoxy based flax and glass fibre composites. The performance of all the unaged and aged composite specimens was assessed via flexural, and viscoelastic (DMA), microscopy (SEM), and visual analysis. The knowledge obtained from this research work will provide new insights and a better understanding about the long-term performance of NFCs undergoing service environmental ageing facilitating their employment as candidate materials in applications like automotives, sports, marine, wind energy, etc.

Materials and methods

Materials

In this study, dry UD flax fabric (FlaxDry UD 180, areal density 180 g/m^2, fibre density 1.45 g/cm^3), and dry UD glass fabric (areal density 220 g/m^2, fibre density 2.5 g/cm^3) were used, supplied by EcoTechnilin, France, and Haufler Composites, Germany, respectively. Three types of matrix materials were employed which are (i) a petroleum-based epoxy resin system (SP 106 resin and SP 106 slow hardener, mix ratio 100:18) supplied by Gurit, UK; (ii) a bio-based epoxy resin system, having 23% bio-based carbon content (Polar Bear, with Recyclamine hardener which makes the composites recyclable, mix ratio 100:22) supplied by R*Concept, Spain; and (iii) a recyclable acrylic liquid thermoplastic resin (Elium® 188 XO) provided by Arkema, France.

Methods

Fabrication of composite laminates

For this work, samples were machined from composite laminates, that were fabricated using a combination of wet lay-up, followed by compression (applied pressure of 6 tons), at room temperature as shown in Fig. 1. After a 24 h cold-curing process, post-curing followed, at (i) 80 °C for 5 h for the petro-epoxy resin-based composites, (ii) 100 °C for 3 h for the bio-based recyclable epoxy-based composites, and (iii) 60 °C for 24 h + 80 °C for 1 h, for the acrylic thermoplastic resin-based composites. For the case of flax laminates, 10 layers of flax fibre reinforcement were used, while 15 layers of glass fibre reinforcement were used for the case of glass composites. The fabricated composite laminates for this work were: (i) flax/petro-epoxy (FFRP1), (ii) flax/bio-based recyclable epoxy (FFRP2), (iii) flax/acrylic thermoplastic (FFRP3), (iv) glass/petro-epoxy (GFRP1), (v) glass/bio-based recyclable epoxy (GFRP2), and (vi) glass/acrylic thermoplastic (GFRP3). After post-curing, the composite laminates were machined, into desired coupon size for experimentation. The physical properties of the fabricated composite laminates are presented in Table 1.

Accelerated weathering

The accelerated weathering was conducted in a simulated environment at lab-scale, using an accelerated weathering tester (Model QUV/spray, Q-Lab, USA) (Fig. 2), according to ASTM G154–16 standard. The programmed weathering cycle lasted for 12 h. One cycle consisted of 6 h of irradiation (0.8 W/m^2 UVA radiation, wavelength ≈ 340 nm) at 60 °C, 4 h of water condensation at 40 °C, and 2 h of water spray at room temperature (Fig. 3). Fluorescent lamps type UVA 340 were used for irradiation. Two weathering cycles were performed per day (24 h). As such, after 7, 14, 28, and 56 d of weathering exposure, test coupons were removed from the weathering chamber, for characterization. After removal from the ageing chamber, coupons were stored in sealed plastic bags, in a refrigerator (4 °C), until they were mechanically tested.

Testing

Flexural testing. The flexural tests of the unaged and weathered composite coupons were conducted according to the ISO 14,125, on a Universal Mechanical Testing Machine (Instron 5966, maximum capacity 10 kN) using a crosshead speed of 2 mm/min at a span distance of 80 mm (Fig. 4a). The dimensions of the flexural test coupons were 100 (l) × 15 (w) mm^2. Flexural strength and flexural modulus were calculated using the following formulas:

$$\text{Flexural strength, } \sigma_f = \frac{3PL}{2bd^2} \quad (1)$$

$$\text{Flexural modulus, } E = \frac{L^3 m}{4bd^3} \quad (2)$$

where, P = the maximum applied load, L = the length of support span, m = the slope of the tangent, b = the width of the specimen, and d = the thickness of the specimen. Five coupons were tested for each case.

Dynamic mechanical analysis (DMA). DMA testing was performed to following the ASTM D 5023, on a DMA 850, Discovery, USA as presented in Fig. 4(b). The dimensions of the test coupons were 64 (l) × 13 (w) mm^2. For all DMA tests, the temperature increased at a rate of 3 °C/min from room temperature to 140 °C at 1.0 Hz and 30.0 μm amplitude. Four coupons were tested for each case. From the DMA testing, the storage modulus (E') was calculated from the E' vs. temperature curves at 30 °C, and the glass transition temperature (T_g) was calculated from the peak of

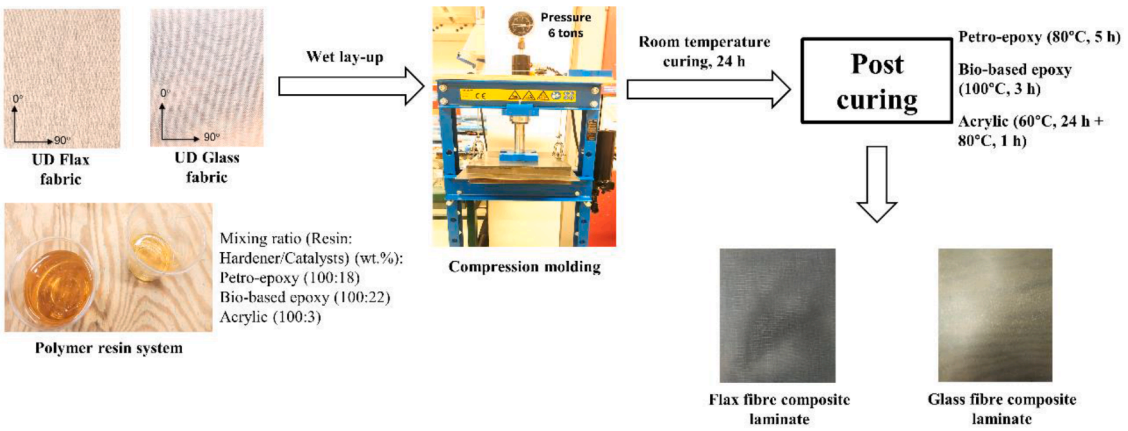


Fig. 1. Fabrication process of composite laminates.

Table 1
Physical properties of the fabricated composite laminates.

Symbol	Composite laminates	Average thickness (mm)	Average fibre volume fraction (%)	
			Flax	Glass
FFRP1	Flax/petro-epoxy	2.62±0.10	47.41 ±1.91	–
FFRP2	Flax/bio-based recyclable epoxy	2.73±0.13	45.58 ±2.24	–
FFRP3	Flax/acrylic thermoplastic	2.50±0.07	49.62 ±1.40	–
GFRP1	Glass/petro-epoxy	2.25±0.08	–	58.81 ±2.12
GFRP2	Glass/bio-based recyclable epoxy	2.21±0.17	–	60.10 ±4.41
GFRP3	Glass/acrylic thermoplastic	2.17±0.14	–	60.93 ±3.60

Fig. 2. Accelerated weathering tester (Model QUV/Spray).

tan δ curves.

Scanning electron microscopy (SEM). Surface damage on coupons, due to weathering, was assessed using a Scanning Electron Microscopy (SEM). As such, micrographs of the unaged and 56 d weathered coupons were recorded, using a Zeiss-Supra 55VP-FEG-SEM and a Zeiss-Ultra 55-FEG-SEM.

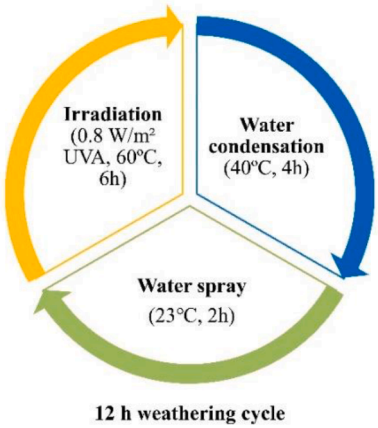


Fig. 3. Weathering cycle (1 cycle consists of 12 h).

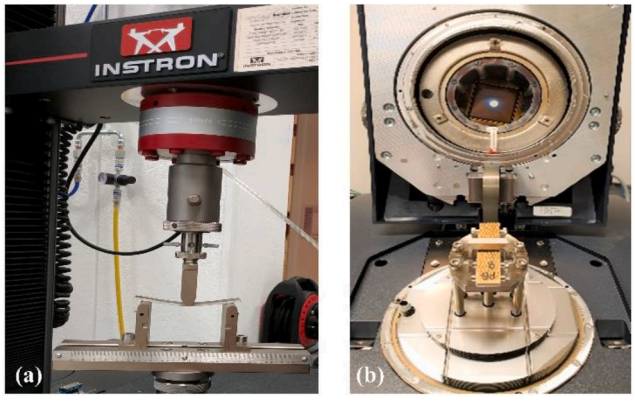
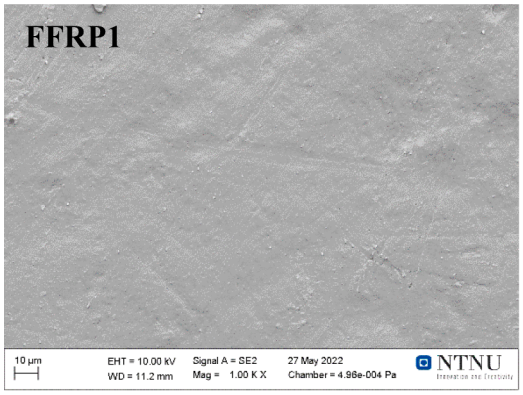


Fig. 4. (a) Universal mechanical testing machine setup, and (b) DMA setup.

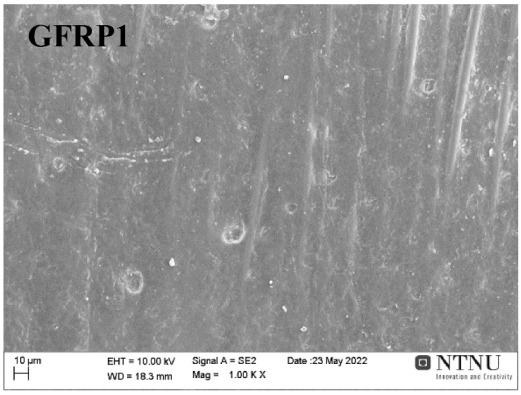
Results and discussion

Scanning electron microscopy (SEM)

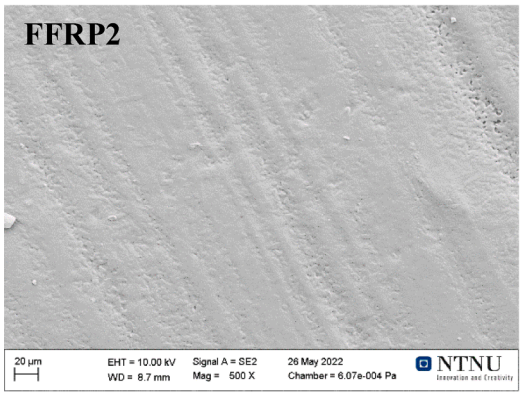
Changes in surface morphology of unaged and aged composite samples, were analysed using Scanning Electron Microscopy (SEM). The surface morphologies of unaged flax fibre composites such as FFRP1, FFRP2, and FFRP3, and unaged glass fibre composites such as GFRP1, GFRP2, and GFRP3 are exhibited in Fig. 5. In the case of unaged composite specimens, there is almost clear appearance of composite surfaces without voids or cracks. However, there is also the presence of tiny



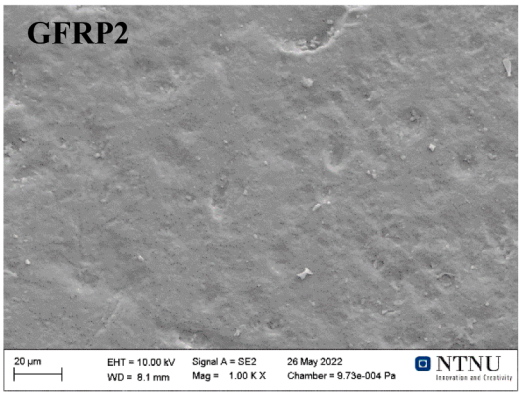
(a) FFRP1 (Flax/petro-epoxy)



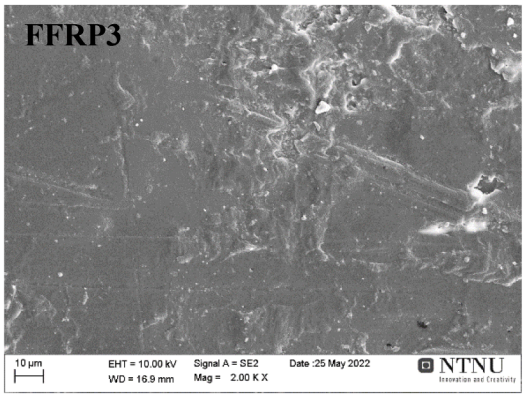
(b) GFRP1 (Glass/petro-epoxy)



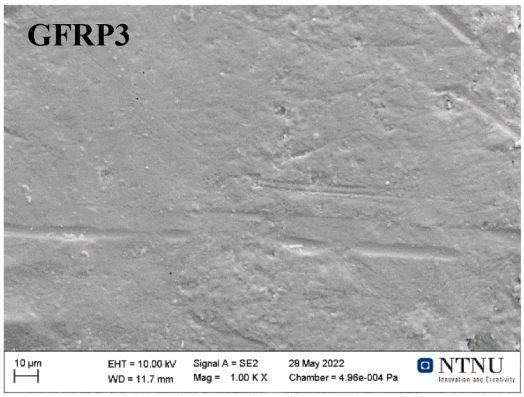
(c) FFRP2 (Flax/bio-based recyclable epoxy)



(d) GFRP2 (Glass/bio-based recyclable epoxy)



(e) FFRP3 (Flax/acrylic thermoplastic)



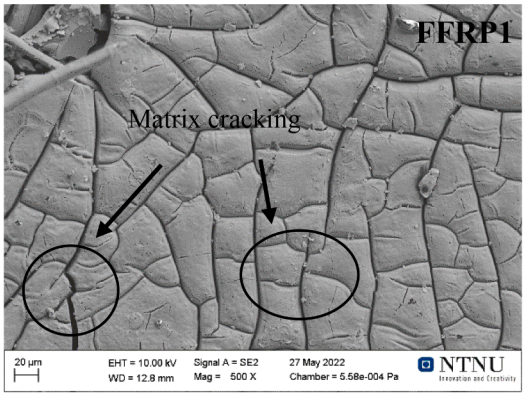
(f) GFRP3 (Glass/acrylic thermoplastic)

Fig. 5. SEM images of surfaces of unaged flax fibre composites: FFRP1 (a), FFRP2 (c), and FFRP3 (e), and surfaces of unaged glass fibre composites: GFRP1 (b), GFRP2 (d), and GFRP3 (f).

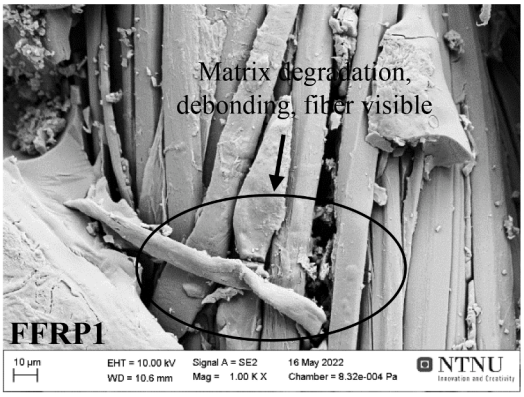
fractures or voids, pits, and marks, which may be created during the processing of the composite laminates.

Fig. 6 shows the SEM images of surfaces of 56 d weathered (aged) flax fibre composites such as FFRP1 (a, b), FFRP2 (c, d), and FFRP3 (e, f). In the case of exposed flax fibre composites, severe degradation on the surface of polymer matrices and flax fibres had occurred due to the synergistic actions of accelerated weathering (UV rays, heat, condensation, and water sprays) [33,48,49]. The surface of the composites became rougher or crazing, matrix cracking, and flax fibres were visible on the composite surfaces due to matrix degradation, fibre-matrix debonding occurred.

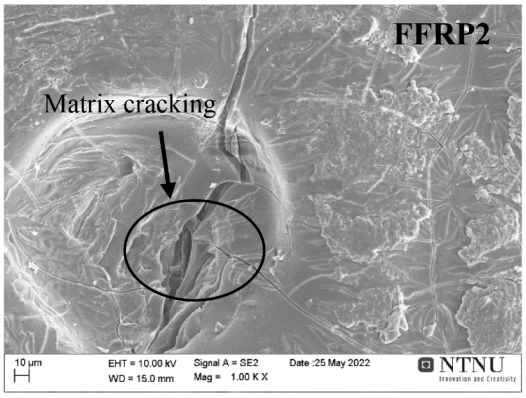
Fig. 7 shows the SEM images of surfaces of 56 d weathered (aged) glass fibre composites such as GFRP1 (a, b), GFRP2 (c, d), and GFRP3 (e, f). In this case, the polymer matrix degradation was also observed, however, damage of fibres did not visible since glass fibres is highly hydrophobic in nature [50,51].



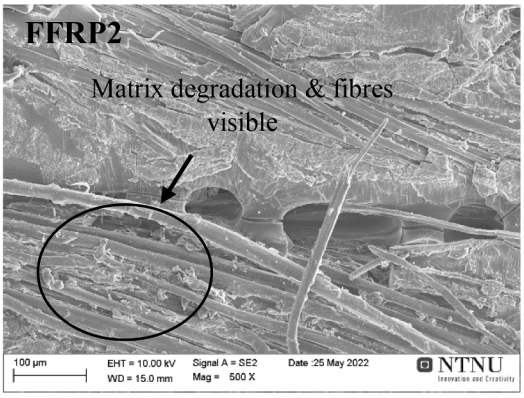
(a) Aged FFRP1 (Flax/petro-epoxy)



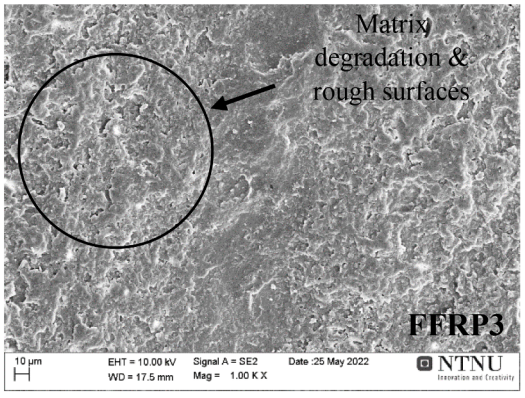
(b) Aged FFRP1 (Flax/petro-epoxy)



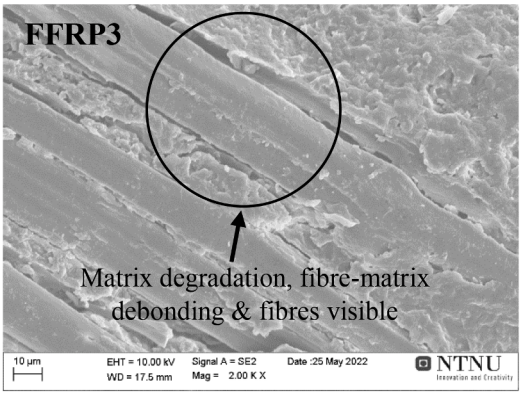
(c) Aged FFRP2 (Flax/bio-based recyclable epoxy)



(d) Aged FFRP2 (Flax/bio-based recyclable epoxy)



(e) Aged FFRP3 (Flax/acrylic thermoplastic)



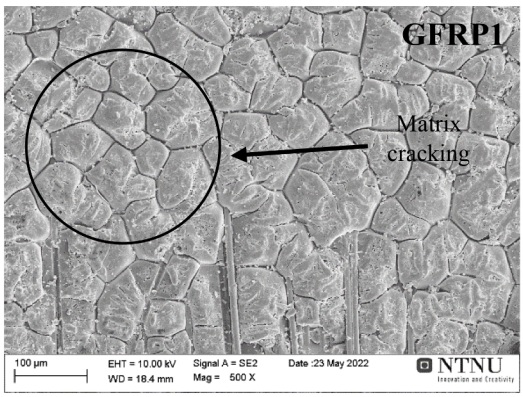
(f) Aged FFRP3 (Flax/acrylic thermoplastic)

Fig. 6. SEM images of surfaces of 56 d weathered (aged) flax fibre composites: FFRP1 (a, b), FFRP2 (c, d), and FFRP3 (e, f).

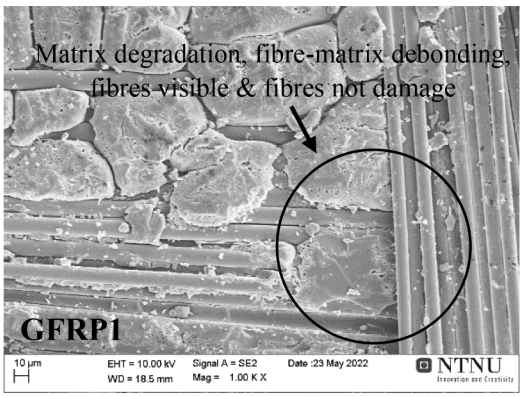
From the SEM images, it is seen that the degradation of flax fibre composites was more pronounced than the glass fibre composites which is also reflected in the degradation of flexural properties (Section 3.1), and viscoelastic behaviors (Section 3.2). Due to the hydrophilic nature of flax fibres, it was easily affected by moisture during the weathering cycles, creating swelling stress and interface damage, adsorption/desorption occurred due to the repetitive wet and dry stages during weathering cycles. The UV radiation at elevated temperature cause polymer and flax fibre damage by photo-oxidation or thermo-oxidation. Due to moisture absorption, matrix plasticization also occurred. All these synergistic actions during accelerated weathering make the surface of the matrix rougher, crazing, or cracking [33,48,50–52].

Visual appearance

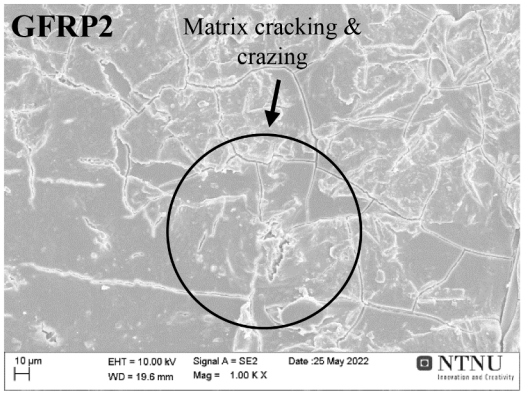
Visual appearance changes of coupons, due to ageing, was examined. Fig. 8 represents the snapshots of unaged and aged specimens, after accelerated weathering for 56 d As can be seen, the original colour of all composite laminates has been altered, due to ageing. The surface colour of FFRP1 and FFRP2 coupons changed into yellowish after 56 d of



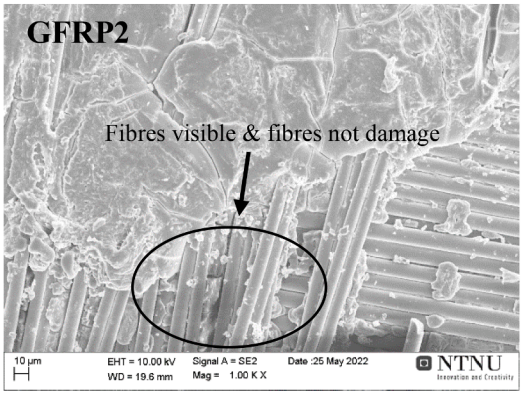
(a) Aged GFRP1 (Glass/petro-epoxy)



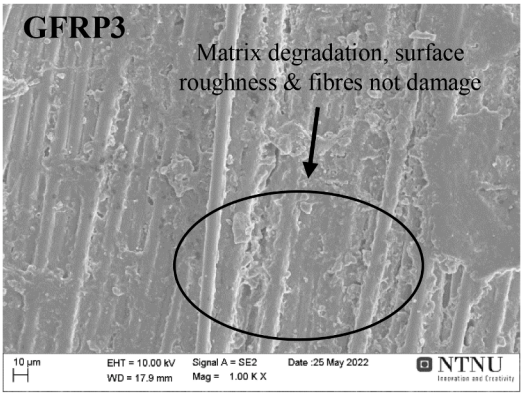
(b) Aged GFRP1 (Glass/petro-epoxy)



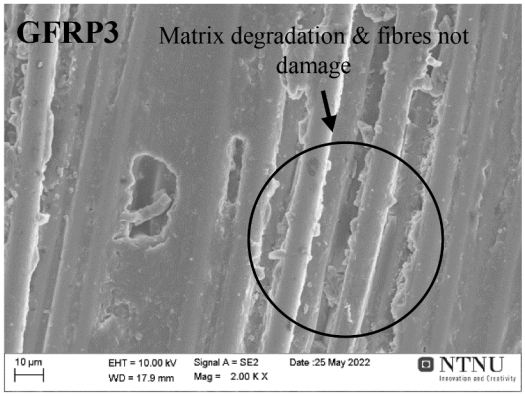
(c) Aged GFRP2 (Glass/bio-based recyclable epoxy)



(d) Aged GFRP2 (Glass/bio-based recyclable epoxy)



(e) Aged GFRP3 (Glass/acrylic thermoplastic)



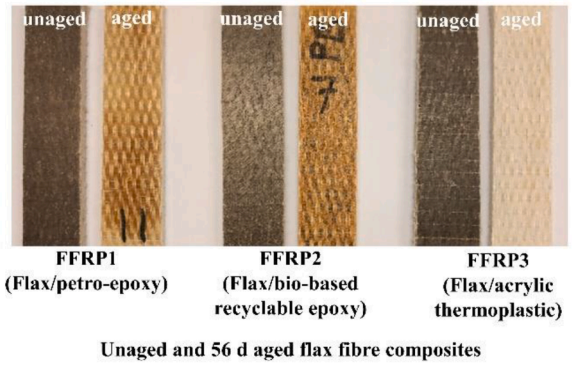
(f) Aged GFRP3 (Glass/acrylic thermoplastic)

Fig. 7. SEM images of surfaces of 56 d weathered (aged) glass fibre composites: GFRP1 (a, b), GFRP2 (c, d), and GFRP3 (e, f).

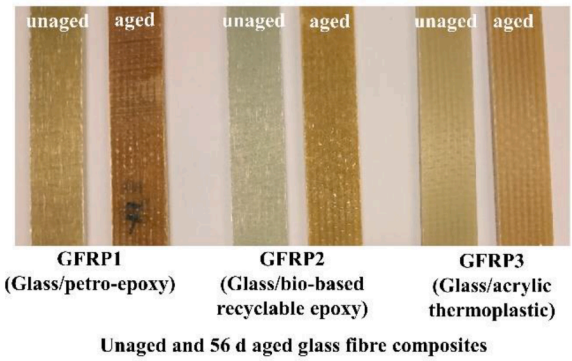
weathering, whilst the FFRP3 coupon changed into beige colour. In the case of glass fibre composites, the GFRP1 exhibited more yellowish colour than the GFRP2 and GFRP3 specimens, which is a matrix dependant change. For flax composites the discoloration of the exposed composite coupons after accelerated weathering is due to photo-degradation of both the reinforcement and the polymer matrix [33,53]. The lignin component present in the case of flax fibres is highly sensitive to UV rays which may breakdown or oxidize during weathering [33,47, 53,54]. According to Yan et al. [33], the possible reason of colour fading on the flax/epoxy composite case, are the repetitive process of erosion of polymer matrix, voids formation, and photo-oxidation during weathering cycles with the combined action of UV light and water spray. As result of accelerated weathering up to 56 d, the exposed composites surface is severely influenced resulting in colour change, rough surface increase, surface micro-cracking, chalking, and leaching [33,54]. These phenomena are also well observed from the SEM images in the previous section (Section 3.1).

Flexural properties

Table 2 tabulates the flexural properties of unaged and aged composites after 7, 14, 28 and 56 d of weathering, together with the



(a)



(b)

Fig. 8. Snapshots of test specimens after 56 d accelerated weathering: (a) unaged and 56 d aged flax fibre composites, and (b) unaged and 56 d aged glass fibre composites.

respective mass changes due to ageing, while Fig. 9 displays the typical stress-strain curves for the unaged and aged coupons.

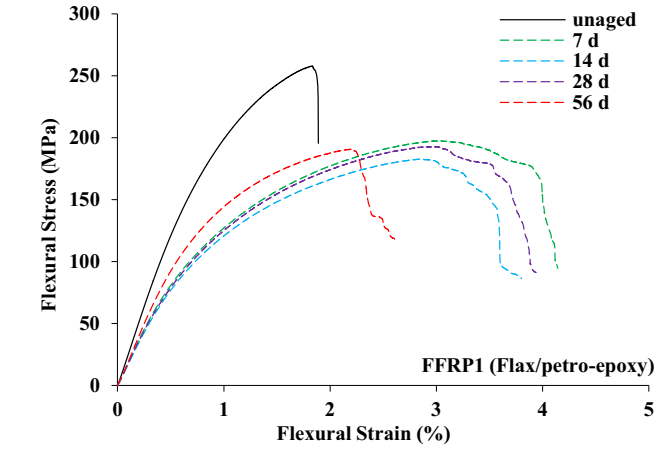
The mass increase in the flax fibre composites was very low (approximately 1%) during the weathering period except FFRP3 at 56 d (approximately 5% weight gain). The weight gain of flax composites was mainly attributed to the moisture uptake. On the other hand, the negative values of mass of glass fibre composites reported due to the weathering which was mainly attributed to the erosion degradation of polymer matrices in the exposed composite specimens as a function of weathering (given the fact that the composites were water-sprayed during weathering cycles) [50]. Signify moisture uptake of flax masque the erosion of flax fibre composites. However, erosion from weathering is obvious in the case of GFRPs (glass fibre composites).

In terms of polymers used in composites, it was exhibited that petro-epoxy, and bio-based recyclable epoxy matrix-based composite samples experienced the similar behaviour of mass change for flax composites (FFRP1 and FFRP2) and glass composites (GFRP1 and GFRP2). The acrylic thermoplastic based flax composites (FFRP3) showed comparatively lower mass gain than other two epoxy based flax composites up to 28 d, however, after 56 d, unexpectedly, the FFRP3 exhibited higher mass gain (approximately 5%) than other two flax composites in similar conditions, which may be due to the generation of micro-cracks in the matrix. GFRP3 showed slightly higher weight loss than other two epoxy-based glass composites (GFRP1 and GFRP2). These phenomena may be due to the slightly higher UV resistance properties of both petro- and bio-epoxy resins.

Fig. 9 depicts typical flexural stress vs. strain curves, of unaged and weathered composite laminates: (a) FFRP1, (b) GFRP1, (c) FFRP2, (d) GFRP2, (e) FFRP3, and (f) GFRP3. As can be seen from Fig. 9, flax composites exhibit a more ductile behaviour, compared to an expected brittle behaviour of glass composites. Percentage (%) retention curves of flexural strength and modulus as a function of time, are presented in Fig. 10, and Fig. 11, respectively. The flexural strength of unaged FFRP1, FFRP2 and FFRP3 composites was 261.83, 277.56, and 220.35 MPa, respectively. After accelerated weathering of 56 d, the flexural strength of the flax composites dropped to 196.05, 194.73, and 109.12 MPa,

Table 2
Mean values of mass changes (%) of weathered flexural test samples and mean values of flexural properties of unaged and weathered (aged) composite samples.

Composite laminates	Time (d)	Mass change (%) of test samples	Flexural Strength (MPa)	Flexural Modulus (GPa)	Strain (%)
FFRP1 (petro-epoxy)	0	–	261.83 ±1.18	26.65 ±0.46	1.78 ±0.03
	7	0.60±0.05	186.92 ±7.76	17.94 ±0.63	2.93 ±0.13
	14	1.10±0.52	185.38 ±5.72	18.23 ±0.79	2.61 ±0.16
	28	1.02±0.31	198.76 ±4.58	18.52 ±0.51	2.80 ±0.11
	56	0.89±0.43	196.05 ±3.55	20.82 ±0.30	2.11 ±0.08
FFRP2 (bio-based recyclable epoxy)	0	–	277.56 ±5.77	26.45 ±0.74	1.76 ±0.11
	7	0.80±0.13	197.78 ±4.06	17.91 ±0.24	2.86 ±0.13
	14	1.23±0.19	198.77 ±5.09	18.41 ±0.62	2.85 ±0.09
	28	0.97±0.23	192.60 ±4.05	17.50 ±0.50	2.89 ±0.10
	56	0.95±0.09	194.73 ±10.10	20.16 ±0.93	2.14 ±0.21
FFRP3 (acrylic thermoplastic)	0	–	220.35 ±4.19	24.89 ±2.26	1.85 ±0.09
	7	0.07±0.17	152.28 ±9.30	18.69 ±0.99	2.25 ±0.13
	14	0.05±0.34	144.49 ±8.92	16.24 ±1.52	2.38 ±0.14
	28	0.45±1.09	119.87 ±8.60	14.54 ±1.35	2.12 ±0.24
	56	4.74±0.71	109.12 ±18.07	13.36 ±1.54	2.09 ±0.24
GFRP1 (petro-epoxy)	0	–	876.92 ±53.33	44.07 ±2.43	2.23 ±0.30
	7	−0.01 ±0.05	925.68 ±70.56	44.54 ±1.34	2.31 ±0.25
	14	−0.01 ±0.03	973.90 ±37.16	44.43 ±2.20	2.45 ±0.11
	28	−0.08 ±0.03	981.49 ±32.55	43.87 ±2.48	2.51 ±0.06
	56	−0.23 ±0.03	969.88 ±41.59	43.96 ±2.25	2.50 ±0.11
GFRP2 (bio-based recyclable epoxy)	0	–	989.89 ±27.80	46.02 ±1.84	2.41 ±0.13
	7	−0.05 ±0.02	999.72 ±26.55	44.76 ±0.81	2.51 ±0.07
	14	−0.01 ±0.03	1000.39 ±41.26	44.26 ±0.90	2.54 ±0.11
	28	−0.12 ±0.02	1004.24 ±39.02	44.55 ±0.99	2.56 ±0.11
	56	−0.18 ±0.07	969.94 ±54.58	42.93 ±2.80	2.54 ±0.07
GFRP3 (acrylic thermoplastic)	0	–	1016.49 ±33.64	46.17 ±1.42	2.48 ±0.08
	7	−0.38 ±0.17	994.40 ±51.35	45.55 ±1.64	2.47 ±0.14
	14	−0.52 ±0.28	995.64 ±20.37	45.05 ±1.15	2.48 ±0.10
	28	−0.53 ±0.27	990.37 ±57.77	45.03 ±0.88	2.45 ±0.18
	56	−0.42 ±0.31	999.63 ±25.74	44.97 ±1.31	2.58 ±0.06



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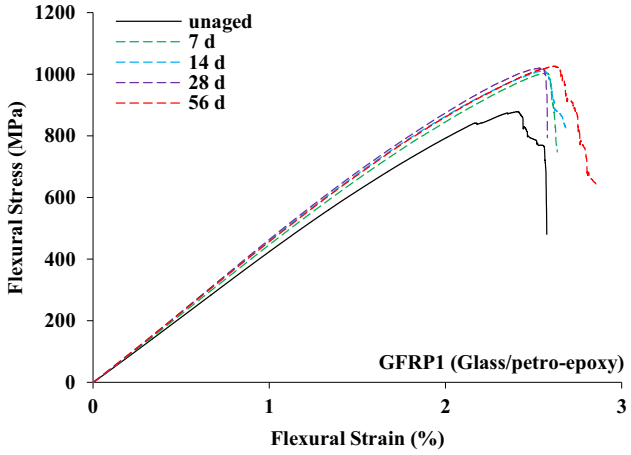

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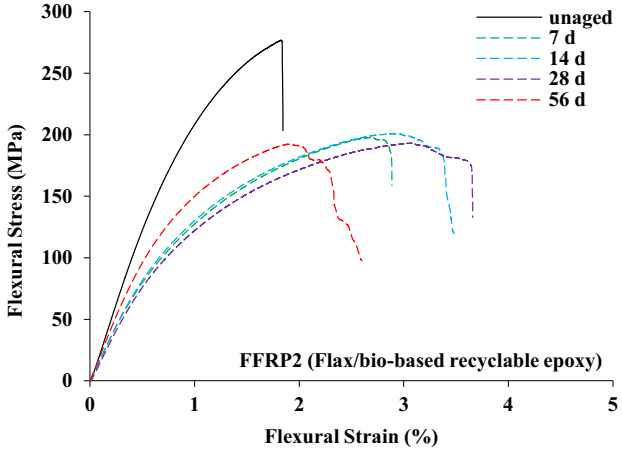

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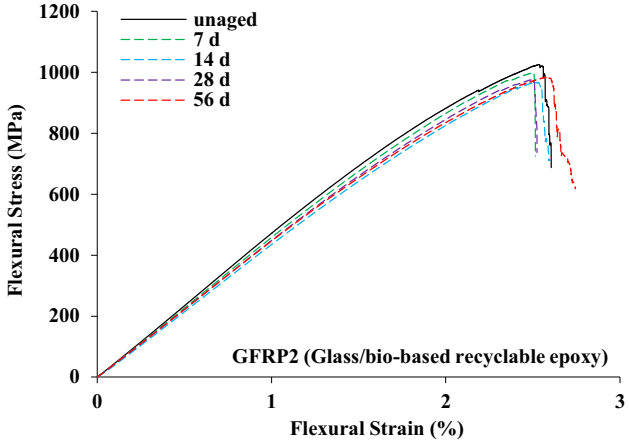

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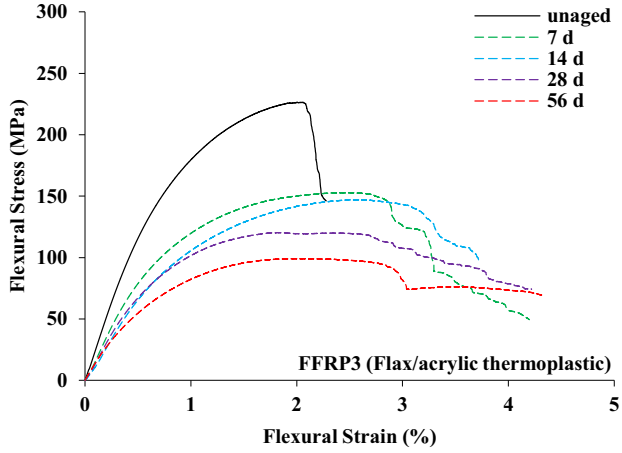

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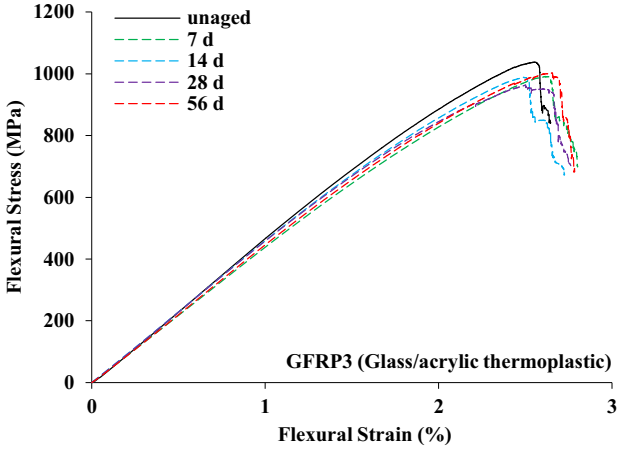


(f)

Fig. 9. Typical flexural stress vs. strain curves of unaged and weathered (aged) composite laminates: (a) FFRP1, (b) GFRP1, (c) FFRP2, (d) GFRP2, (e) FFRP3, and (f) GFRP3.

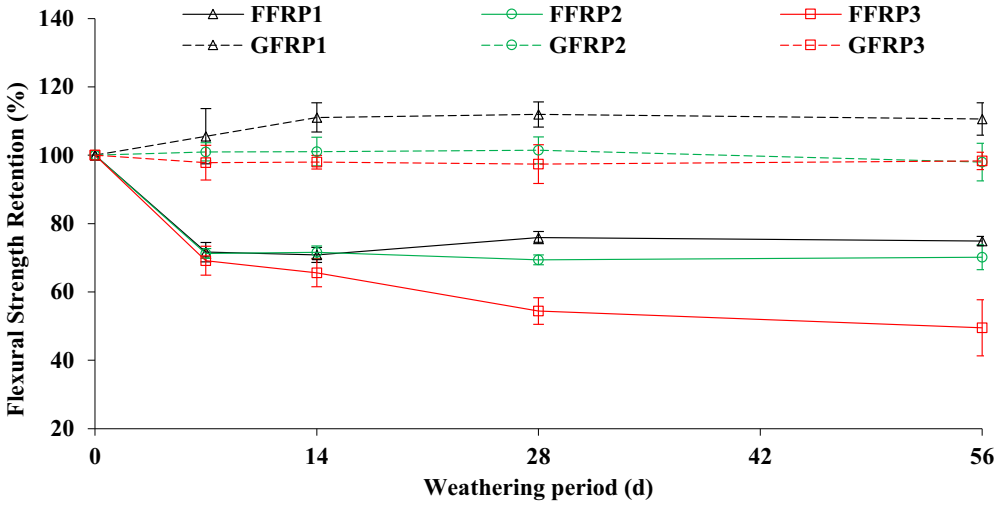


Fig. 10. Flexural strength retention as a function of weathering period for flax and glass fibre reinforced polymer composites under accelerated weathering.

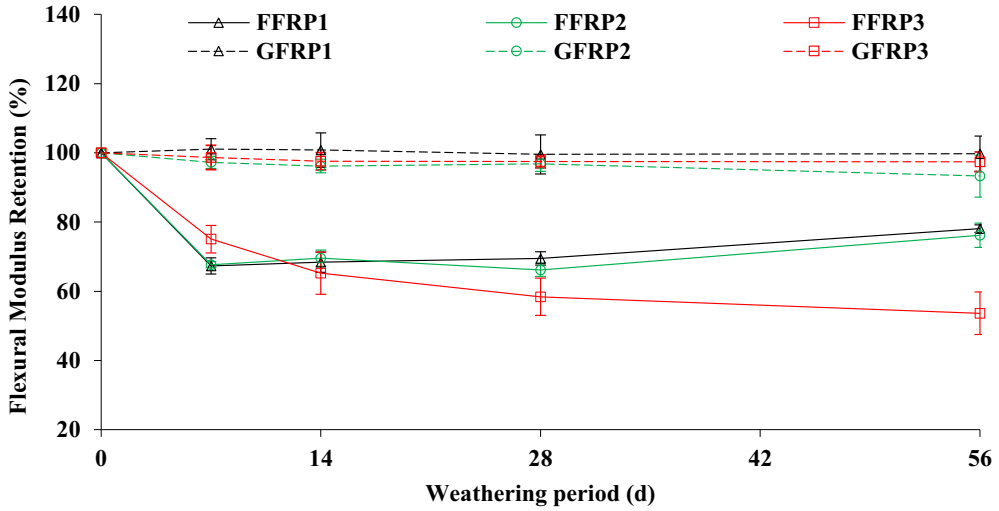


Fig. 11. Flexural modulus retention as a function of weathering period for flax and glass fibre reinforced polymer composites under accelerated weathering.

respectively, which corresponds to a drop to approximately 25, 30, and 50%, as presented in Fig. 10. The flexural modulus of unaged FFRP1, FFRP2, and FFRP3 was found to be 26.65, 26.45, and 24.89 GPa, respectively, as illustrated in Table 2. After 56 d weathering, the modulus decreased in a similar manner to flexural strength, and was found to be 20.82, 20.16, and 13.36 GPa, respectively. Thus, the flexural modulus has approximately lowered to 22, 24, and 46%, respectively, after 56 d of weathering, as illustrated in Fig. 11.

Comparing the results of FFRP1 and FFRP2, in Figs. 10 and 11, flax/petro-epoxy composites (FFRP1) revealed slightly higher resistance to ageing, compared to flax/bio-based epoxy composites (FFRP2), which may be likely to the bio-based content in the polymers of FFRP2 case. Notably, the flax/acrylic composites (FFRP3) exhibited the lowest resistance to ageing, amongst all cases, likely due to the poor interfacial adhesion between flax fibre and the acrylic thermoplastic resin. In general, flax fibres tend to swell, due to moisture intake, leading to disruption of the fibre/matrix interface. Unexpectedly, in the case of FFRP3, revealed the highest amount of moisture uptake, after 56 d of ageing (approximately 5 wt.%, *see* Table 2), which likely causes more pronounced damage within the composite structure. This was unexpected, due to the thermoplastic nature of the polymer, which is expected to be more hydrophobic than thermosets.

Yan et al. [33] reported a drop of approximately 10% of flexural strength and modulus due to weathering up to 1500 h (≈ 63 d) for woven flax fabric reinforced petro-epoxy composites. On the other hand, Wang and Petru [55] reported a drop of 11, 15, 16% flexural strength and 21, 32, 36% flexural modulus after 60, 120, 180 d, respectively, of natural ageing in outdoor environment for UD flax/petro-epoxy composites. The degradation of flexural properties for the flax/petro-epoxy case, is more pronounced in the current study (25% drop of flexural strength and 22% drop of flexural modulus), than in previous studies [33,55], likely due to the harsher ageing environment employed here. However, the rate of degradation increased as a function of extended ageing period as revealed in Ref. [55].

In case of bio-epoxy based flax composites (FFRP2), the revealed degradation in flexural strength (30%) and flexural modulus (24%), coincides to the findings reported in Taylor et al. [34], after subjecting untreated flax/bio-based polymer composites to 1000 h (≈ 42 d) of weathering. That said, the amount of bio-based content (23% in this work compared to 85% in Taylor et al. [34]) did not showcase a significant difference in degradation due to weathering. In the case of flax/acrylic composites (FFRP3), the current study presents a significant drop of flexural strength and modulus, of approximately 50 and 46%, respectively, which is in close agreement to what has been reported earlier and is claimed to be caused by poor interfacial adhesion between flax fibres and acrylic thermoplastic resin [31,56].

Contrary to flax composites, the flexural properties of glass composites were significantly less affected by weathering as illustrated in Table 2. For unaged GFRP1, GFRP2, and GFRP3 composites, the flexural strength was found to be 876.92, 989.89, and 1016.49 MPa, respectively, while the flexural modulus was calculated to be 44.07, 46.02, and 46.17 GPa, respectively. After 56 d weathering, GFRP1, GFRP2, and GFRP3 composites, flexural strength changed to 111, 98, and 98%, respectively, while the equivalent change for flexural modulus was calculated to be 100 (no change), 93, and 97%, respectively. As such, the three GFRP cases, revealed very small differences in long-term flexural performance. Petro-epoxy composites (GFRP1) exhibited the highest performance in flexure, while bio-based epoxy composites (GFRP2) performed marginally lower than GFRP1, mainly due to bio-based content in the resin system. Glass/acrylic composites (GFRP3) exhibited flexural properties slightly lower than that of GFRP1 and GFRP2. In all cases, it could be postulated that, in comparison with flax composites, the studied glass composites were not negatively affected by the imposed weathering cycle, rather in the case of GFRP1 flexure strength improved, most likely due to possible post-curing or matrix hardening [29,48,57].

Sousa et al. [37] has reported 78 and 87% flexural strength retention for glass/vinyl ester composites after 42 months of natural weathering and 3000 h (~ 125 d) of artificial weathering, respectively. In the same study, flexural strength retention of glass/polyester composites, was reported to be 95 and 112% after 42 months of natural weathering and 3000 h (~ 125 d) of artificial weathering, respectively. These two studies of petro-based thermoset polymer (vinyl ester and unsaturated polyester)/glass composites were performed for a significantly longer period than the current study (56 d). Interestingly, the retention of flexural strength was 95–112% for glass/unsaturated polyester composites, in spite having a prolong ageing period, which is coherent with the degradation values of petro-epoxy based glass thermoset composites (GFRP1). This may be due to the effect of possible cross-linking, post--curing, or matrix hardening phenomena [29,48,57]. However, the glass/vinyl ester composites showed degradation of their flexural strength, which is expected after a prolong ageing period, and may be caused due to the degradation of polymer matrices and interface weakening by the action of weathering [37]. On the other hand, the bio-based epoxy/glass composites (GFRP2) exhibited slightly lower retention of flexural strength and modulus than the GFRP1 which may be due to the presence of bio-based content in the bio-based epoxy resin system.

In the case of glass/acrylic thermoplastic (GFRP3), the degradation behaviour of flexural properties is similar with the degradation of glass/polypropylene (PP) thermoplastic composites under outdoor weathering environment for the first 2 months of ageing period [38]. No significant damage on the flexural properties was reported for both thermoplastic composite materials. This may be due to the increased interfacial adhesion of glass fibres with the thermoplastic matrix [38].

Viscoelastic properties

As explained in the Section 2.2.3.2, viscoelastic behaviour of the developed laminates, was studied using Dynamic Mechanical Analysis (DMA). From DMA testing, Storage Modulus (E') and Glass transition temperature (T_g) were calculated and recorded over the course of ageing duration, in order to study the viscoelastic performance of the tested composites. The viscoelastic properties of unaged and aged flax composites and glass composites are presented in Table 3. Figs. 12 and 13 depict the E' and tan δ curves as a function of temperature and weathering period for flax fibre composites (FFRP1, FFRP2, FFRP3) and for glass fibre composites (GFRP1, GFRP2, GFRP3), respectively.

From the literature, it is known that a decrease of E' values due to weathering, corresponds to photo-degradation, matrix cracking, swelling, etc. [58], while improvement of E' values are mainly due to residual post-curing (additional crosslinking), or a result of increased crystallinity in the polymer system [59–61].

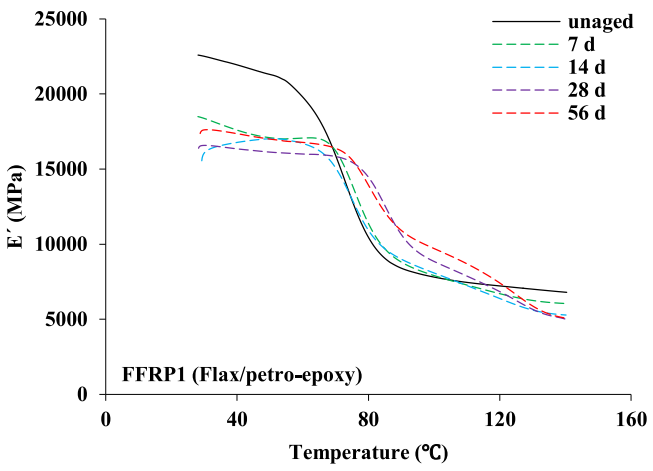
Table 3
Viscoelastic properties of unaged and weathered (aged) composite samples.

Composite laminates	Time (d)	E' (GPa) at 30 °C	Peak height of tan δ curve	T_g from tan δ_{max} (°C)
FFRP1	0	21.99±2.19	0.187±0.01	75.53±1.20
	7	17.24±0.83	0.154±0.01	79.05±1.28
	14	15.79±1.41	0.149±0.01	78.21±1.99
	28	14.64±3.61	0.124±0.01	85.67±3.44
	56	17.20±1.84	0.112±0.01	83.88±2.12
FFRP2	0	20.96±0.52	0.200±0.01	90.19±1.33
	7	17.08±0.64	0.216±0.01	89.30±1.92
	14	16.29±1.83	0.194±0.03	86.42±4.29
	28	15.89±2.89	0.184±0.01	90.10±2.75
	56	13.90±2.66	0.187±0.01	89.08±1.39
FFRP3	0	19.24±1.55	0.350±0.05	101.12±0.43
	7	15.11±3.03	0.447±0.02	107.03±0.53
	14	10.14±1.94	0.475±0.03	111.20±0.92
	28	7.04±0.72	0.556±0.03	120.22±4.53
	56	7.29±2.08	0.566±0.04	116.54±1.02
GFRP1	0	37.84±4.55	0.324±0.08	69.94±0.49
	7	29.26±1.84	0.343±0.01	71.69±1.12
	14	31.08±2.03	0.326±0.01	72.66±1.12
	28	31.01±1.02	0.324±0.01	73.11±1.15
	56	29.66±3.26	0.297±0.01	69.84±0.72
GFRP2	0	34.02±1.72	0.287±0.01	91.18±1.05
	7	33.79±2.32	0.311±0.01	94.99±1.45
	14	32.43±3.71	0.308±0.01	94.54±0.76
	28	32.56±3.82	0.321±0.01	92.80±0.76
	56	31.20±1.49	0.330±0.01	90.39±0.22
GFRP3	0	33.44±5.61	0.742±0.01	95.08±0.84
	7	33.48±3.15	0.685±0.03	93.83±0.95
	14	38.43±1.18	0.684±0.02	96.11±1.00
	28	36.47±0.98	0.671±0.03	94.64±0.55
	56	32.38±3.37	0.648±0.02	96.61±0.39

The Storage Modulus, E', of the unaged FFRP1, FFRP2, and FFRP3 composite laminate coupons, were found to be 21.99, 20.96, and 19.24 GPa, respectively, as tabulated in Table 3. After 56 d of weathering, E' values were recorded to be 17.20, 13.90, and 7.29 GPa, respectively. Fig. 14 depicts the retention values of E', as a function of weathering period for all the studied composites. As can be seen, the E' values of all the flax fibre composites, reduced after ageing, in comparison to the unaged, due to the synergistic effects of UV radiation, heat and moisture, as described in the previous sections and well evident from the SEM images (Section 3.1).

The retention of E' of FFRP1 coupons, was found to change to 78, 72, 67, and 78% after 7, 14, 28, and 56 d of weathering while FFRP2 coupons, revealed an almost similar behaviour due to ageing, although, after 56 d, FFRP2 revealed marginally lower E' values than FFRP1, but within the experimental scatter, which could be attributed to the bio-based content of the bio-based resin. In the case of FFRP3, E' values declined significantly, compared to FFRP1 and FFRP2, which was aligned with the flexure performance results. After 7 d of weathering, the retention of E' values of FFRP3 was to be almost similar to that of FFRP1 and FFRP2, however, after that, E' values dropped significantly. As stated earlier, this may be due to the poorer fibre/matrix interfacial adhesion, caused due to the increased moisture content.

For the case of GFRPs, E' values for the unaged GFRP1, GFRP2, and GFRP3 composites, was calculated to be 37.84, 34.02, and 33.44 GPa, respectively, while after 56 d of ageing, E' values dropped to 29.66, 31.20, and 32.38 GPa, respectively (Table 3). From Fig. 14, it can be seen that the percentage change of E' values was found to be 77, 82, 82, and 78% for GFRP1, 99, 95, 96, and 92% for GFRP2, and 100, 115, 109, and 97% for GFRP3, after 7, 14, 28, and 56 d weathering, respectively. In specific, GFRP1 revealed a drop in E' values after 7 d of ageing. After that, E' values increased slightly (14 d) and later remained unaffected, until the end of ageing period. On the other hand, to GFRP1, in the case of GFRP2, E' values increased slightly after 7 d of ageing, and later E' values remained practically unaffected until the end of ageing period.



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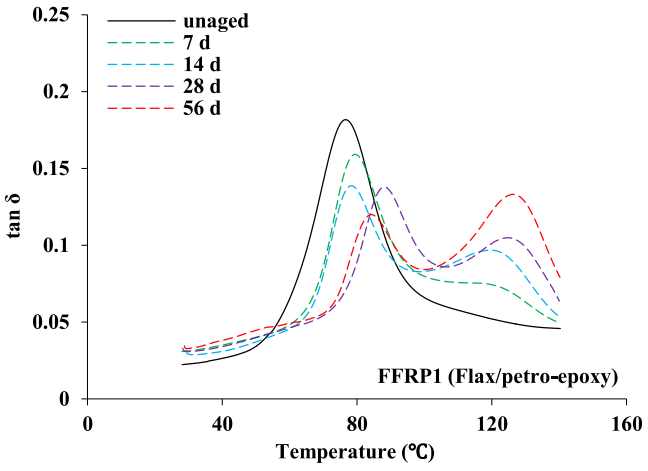

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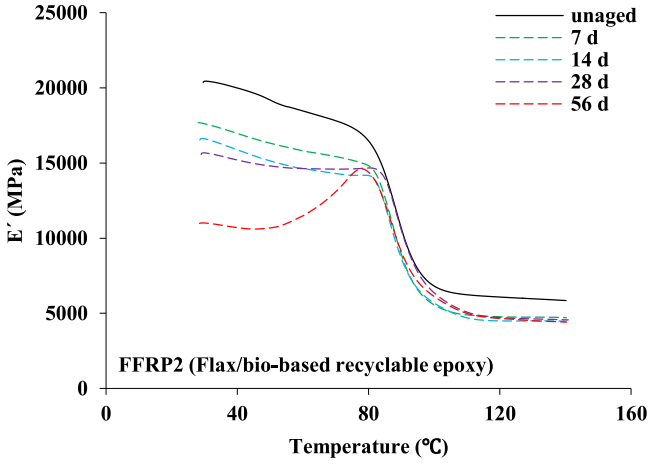

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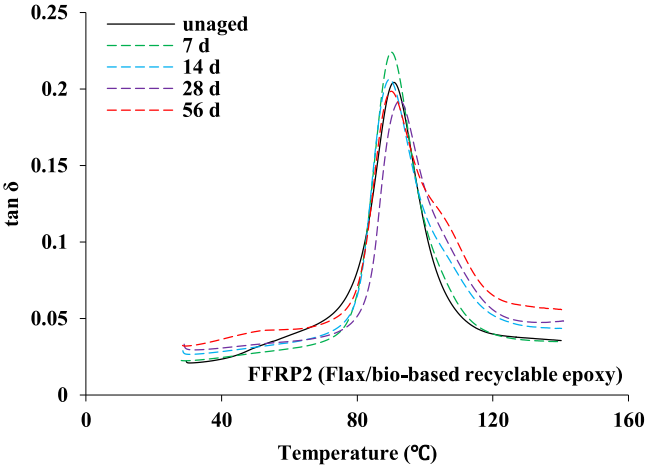

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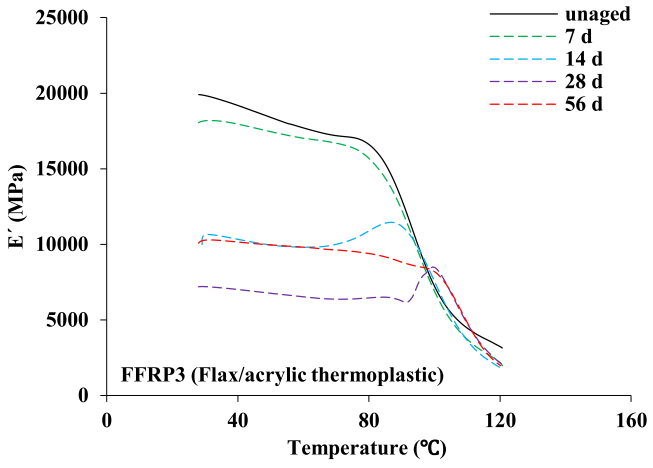

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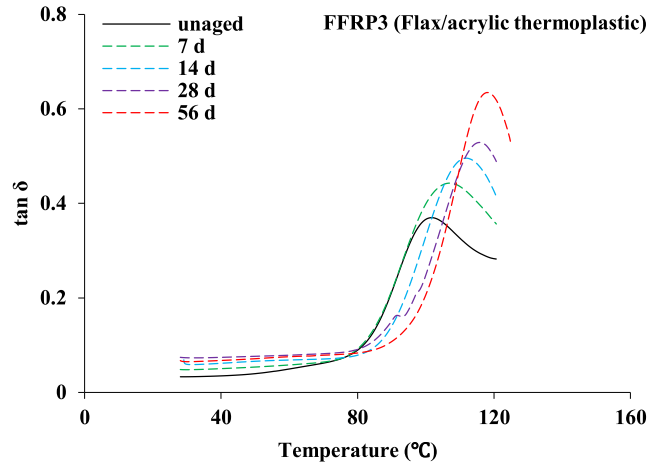

(f)

Fig. 12. The E' and tan δ curves as a function of temperature and weathering period for flax fibre composites: FFRP1 (a, b), FFRP2 (c, d), and FFRP3 (e, f).

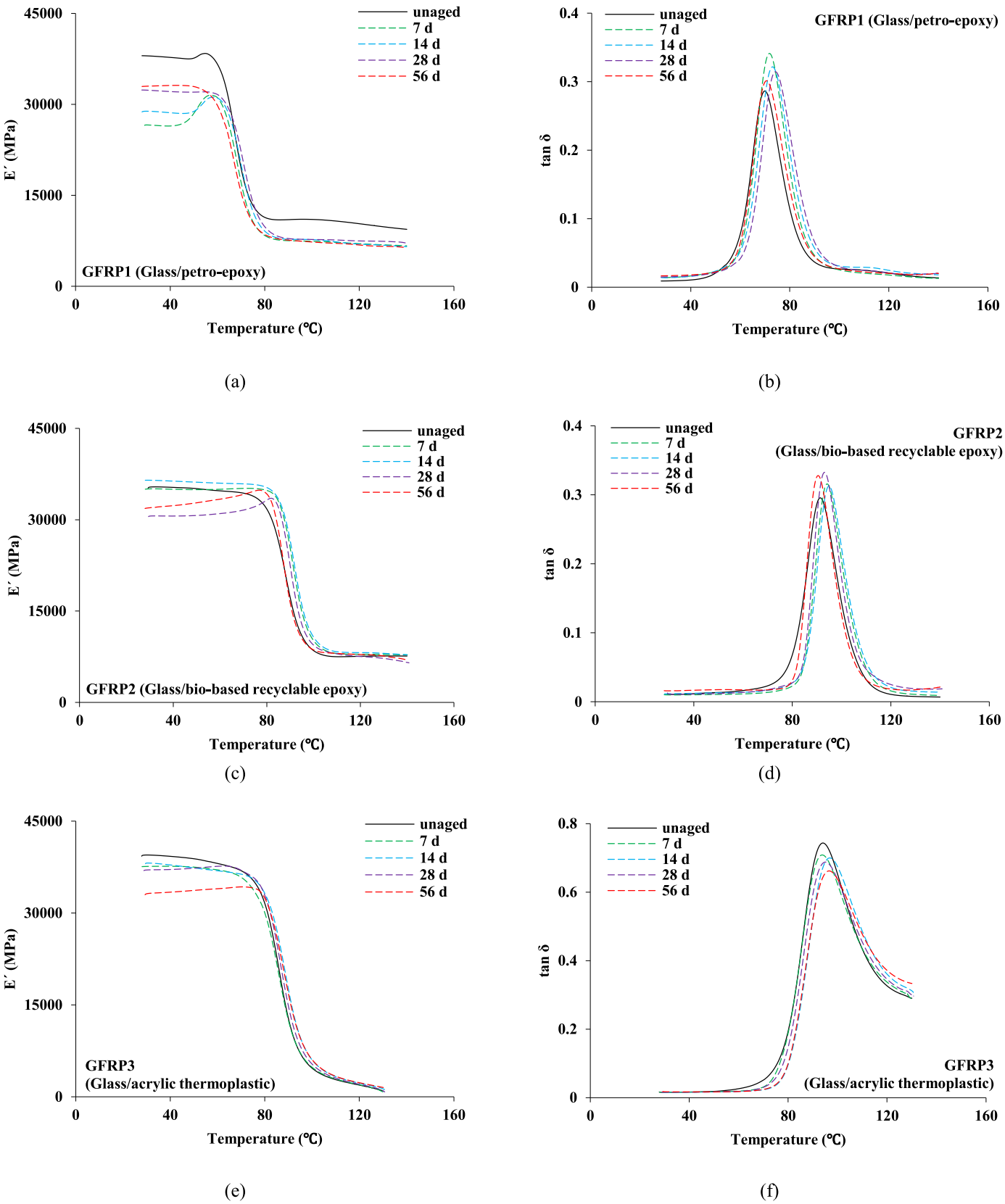

Fig. 13. The E' and tan δ curves as a function of temperature and weathering period for glass fibre composites: GFRP1 (a, b), GFRP2 (c, d), and GFRP3 (e, f).

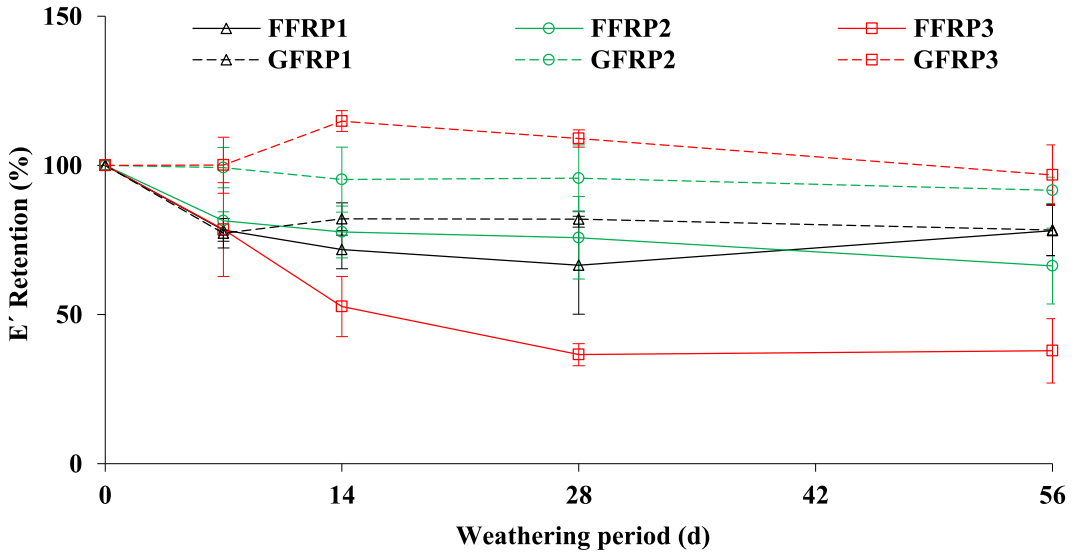

Fig. 14. Retention (%) of E' as a function of weathering period for flax and glass fibre reinforced polymer composites under accelerated weathering.

For the case of GFRP3, E' values increased for 14 d of ageing, while then E' values declined for 28 d and 56 d, respectively, reaching similar E' value retention to GFRP2, at the end of ageing. As such, contrary to long-term flexure performance, it may be postulated that glass/bio-based epoxy and glass/acrylic thermoplastic composites (GFRP2 and GFRP3) performed better than the case of glass/petro-epoxy composites (GFRP1).

Overall, the degradation of E' was more pronounced for flax composites than that of the glass composites, due to the increased moisture sensitivity [13,14]. Since DMA testing was conducted on a 3-point bending mode, the recorded values are complementary to flexural properties, and as such, E' value could be indicative of flexural stiffness. It is also evident, that for the same polymer matrix, the degradation of E' is more severe for flax composites than the glass composites, denoting that, storage modulus (E) is mainly dependant on the properties of reinforcing fibres, as well as their effects to the final composite.

The tan δ (damping) curves for flax fibre composites (FFRP1, FFRP2, and FFRP3) and glass fibre composites (GFRP1, GFRP2, and GFRP3) are presented in Fig. 12 (b, d, f), and Fig. 13 (b, d, f), respectively. As can be seen, damping properties of FFRP1 and FFRP3 composites, were significantly more affected by accelerated weathering, compared to the cases of FFRP2, GFRP1, GFRP2, and GFRP3. Fig. 15 depicts the change of T_g as a function of weathering time for all the studied composites. The T_g values for unaged FFRP1, FFRP2, and FFRP3 composites were recorded to be 75.53, 90.19, and 101.12 °C, respectively, as tabulated in Table 3, which eventually changed to 83.88, 89.08, and 116.54 °C, respectively, after 56 d of weathering. Hence, an increase of T_g revealed for FFRP1 (11%) and FFRP3 (15%) during the entire ageing period while FFRP2 demonstrated almost similar or a slight decrease in T_g values.

In the case of FFRP1 (Fig. 12b), a sharp peak of tan δ curve was recorded, for the unaged coupon case, whilst for the case of aged coupons, tan δ curves revealed shifted peaks at lower tan δ values, as well as one additional peak, that appeared in the rubbery plateau region, after 14, 28, and 56 d of ageing. The value of T_g increased, which may be indicative of post-curing or an effect of polymer hardening. The first peak in the transition region corresponds to the T_g [32,62] of the material, while emergence of 2nd peak in the rubbery plateau region, may be related to the absorption of moisture within the composite structure [32,59], or the presence of immobilized layers of epoxy resin polymer between reinforcing fibers and polymer matrix (interface), as a

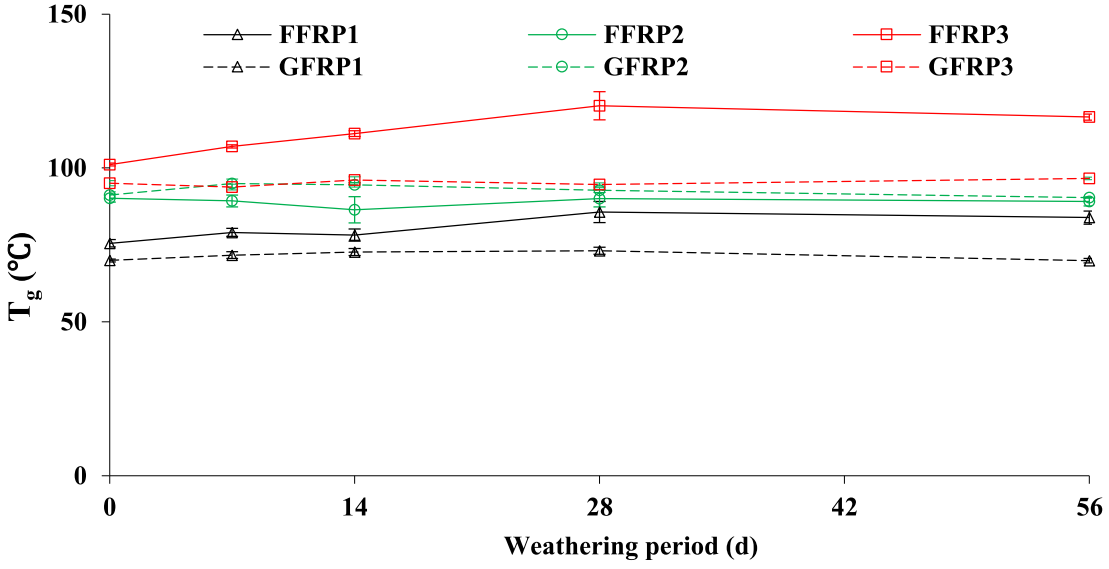

Fig. 15. Change of T_g (°C) values as a function of weathering period for flax and glass fibre reinforced polymer composites under accelerated weathering.

phenomenon of micro-mechanical transition [32,62].

For the case of FFRP2 (Fig. 12d), the tan δ curves of the unaged and aged composites were found to be almost identical. In specific, the tan δ peak after 7 d of ageing, increased to slightly higher values (peak value of 8% higher than unaged ones), while then diminished to lower values, after 28 d of ageing. However, the T_g remained almost similar to the unaged ones after 28 d as a function of weathering (only 1% decrease occurred), which is indicative of appropriate interfacial adhesion and post-curing or matrix hardening phenomena. For the case of FFRP3, as can be denoted from Fig. 12f, the tan δ peak values (hence T_g) of the conditioned composites has risen significantly (in a range of 6–19% of T_g values) in comparison to reference case. This is attributed to the effect of moisture to both the flax fibre reinforcement and acrylic matrix (given the increased moisture content, *see* Table 2), as well as fibre/matrix interface damage [59,62,63]. Thus, it could be postulated that the flax/bio-based epoxy composite (FFRP2) performed best in terms of damping performance, amongst the three FFRP composite cases.

The tan δ curves of glass fibre composites (GFRP1, GFRP2, and GFRP3) are illustrated in Fig. 13 (b, d, f), respectively. As can be seen from Fig. 13 (b, d, f), overall damping performance of GFRPs, has not been significantly affected, by accelerated weathering. The T_g of unaged GFRP1, GFRP2, and GFRP3 composites was calculated to be 69.94, 91.18 and 95.08 °C, respectively, as tabulated in Table 3. After 56 d of weathering, the T_g values changed to 69.84, 90.39, and 96.61 °C, respectively, demonstrating only a slight change of T_g.

Higher and sharp tan δ peaks in the glass transition region were observed for GFRP1 and GFRP2, and a slightly lower tan δ peaks exhibited for GFRP3 after weathering, hence no noticeable change of T_g observed which is also evident from the Table 3 and Fig. 15. In the case of GFRP1, the T_g values increased 3–5% from 7 to 28 d of ageing, however, then after 56 d it returned to the values near unaged composite sample. A similar change of T_g reported for the GFRP2 composite case while GFRP3 reported very minor change of T_g (- 1 to 2%, from 7 to 56 d ageing period). So, glass/acrylic thermoplastic composites (GFRP3) showed slightly good performance in terms of damping and retention of T_g due to weathering.

There may be several reasons for the stability of the damping or T_g values for glass fibre composites during the accelerated weathering period such as hydrophobicity of the glass fibres which were not affected by moisture, heat or UV ray, and excellent interface between glass fibres with these polymer matrices which was not damaged significantly due to the weathering actions, moreover, there may be the phenomenon of extra post-curing, cross-linking, or enhanced crystallinity in the polymer systems which yielding in improved T_g [37,59,64].

Flax composites vs. glass composites

Fig. 16 illustrates a comparative chart, of flexural and viscoelastic properties, of the retention (%) values after 56 d of ageing, for all the studied types of composites. As can be seen from Fig. 16, GFRP composites exhibited significantly higher values of retention, for flexural strength, flexural stiffness, storage modulus (E), and glass transition temperature (T_g), than their FFRP counterparts. This reveals that the reinforcing fibres dominate the long-term performance of the composites, while the type of polymer matrix has less influence.

In the case of flax fibre composites, petro- and bio-based epoxy composites (FFRP1 and FFRP2) performed better than the flax/acrylic thermoplastic composites (FFRP3), against accelerated weathering. However, glass/acrylic thermoplastic composites (GFRP3) exhibited excellent retention of mechanical and thermomechanical properties after ageing. amongst all GFRP composites, flexural strength, flexural modulus as well as T_g values, were not affected. However, the drop of E'

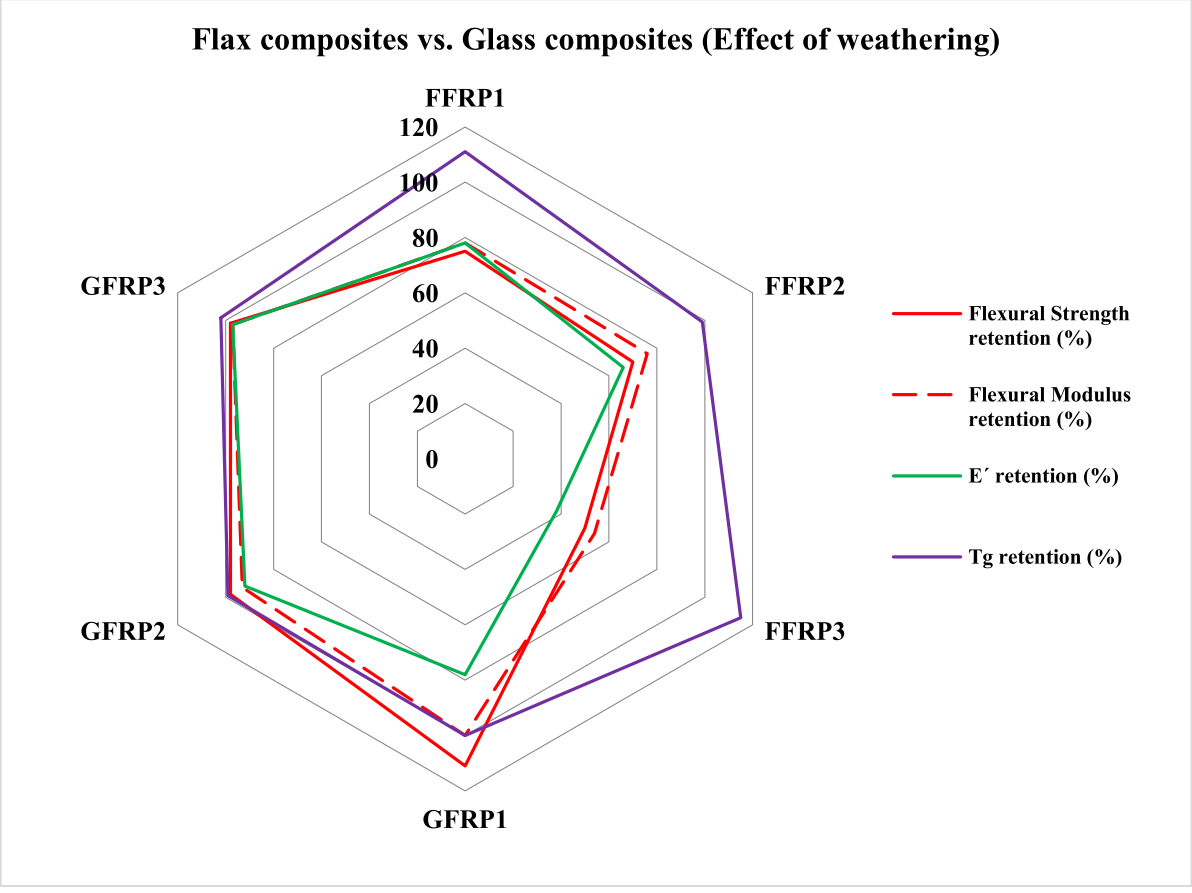

Fig. 16. Comparative properties of flax fibre composites and glass fibre composites after 56 d weathering, based on their retention values.

was comparatively higher for GFRP1 (78% retention) than GFRP2 (92% retention) and GFRP3 (97% retention). Overall, acrylic thermoplastic polymer-based glass composites (GFRP3) performed comparatively well than both epoxy-based glass composites, which can be written as GFRP3>GFRP2>GFRP1.

Conclusion

In this research, flax and glass fibre composites were fabricated with three polymer matrices (petro-epoxy, bio-based recyclable epoxy, and acrylic thermoplastic liquid resin) to investigate their performance under weathering for a duration of 56 days. The long-term performance of the composites was assessed by visual analysis, flexural and DMA characterization as well as SEM analysis. The following conclusions can be highlighted based on the outcome of this work:

- The accelerated weathering has a significant effect on the physico-mechanical and viscoelastic properties of flax fibre composites due to the weathering sensitivity of flax fibres and mainly the increased hydrophilicity. Degradation was also confirmed by discoloration and SEM. As expected, glass fibre composites, performed best amongst others against weathering, exhibiting marginal deterioration, for all polymer matrix cases. As such, it may be postulated, that the type of reinforcing fibres dominates composite`s behaviour to ageing while the type of polymer matrix has less influence.
- After 56 days of weathering, flexural and viscoelastic properties degradation was more pronounced for flax/acrylic thermoplastic composites (FFRP3) compared to the other two flax fibre composites cases (FFRP1 and FFRP2), which could be attributed to the poor fibre/matrix interfacial adhesion. However, glass/acrylic thermoplastic composites (GFRP3) revealed overall adequate performance (almost 100% retention of properties) in the entire ageing period due to the improved fibre/matrix interfacial adhesion.
- Overall, the bio-based recyclable epoxy matrix revealed almost similar behaviour (flexural and viscoelastic) to the studied petro-based epoxy matrix, after ageing, in both flax and glass fibre reinforced composite case, which is a promising outcome when it comes to future sustainable composites. Moreover, better retention of T_g and damping performance was revealed for flax/bio-based recyclable epoxy composites (FFRP2) than their petro-based counterpart (FFRP1) and flax/acrylic thermoplastic composites (FFRP3). On the other hand, the change of T_g for all the glass fibre composites was insignificant during the entire ageing period. It is also worth noting that, all the composite specimens retained the threshold value of T_g (60 °C) which is essential for civil engineering structural materials.

In summary, it was revealed that reinforcing fibres play a dominant role in performance, after accelerated weathering, with glass fibre composites exhibiting superior performance over flax composites, indicating the need for long-term performance improvement of flax fibre reinforcement, via enhancing fibre/ matrix interaction, fibre protection, etc., to ensure the adoption of circular natural fibre composites in engineering applications.

CRediT authorship contribution statement

S.C. Das: Conceptualization, Methodology, Experimental work, Data curation, Formal analysis, Visualization, Writing – original draft, review & editing. **A.D. La Rosa:** Co-supervision, review final document. **S. Goutianos:** Co-supervision, Methodology, review final document. **S.A. Grammatikos:** Conceptualization, Methodology, Project administration, Funding acquisition, Resources, Supervision, Validation, Writing – review & editing.

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Declaration of Competing Interest

None.

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