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# EFFECTS OF WATER ABSORPTION ON MECHANICAL PROPERTIES OF GLASS MICROBALLOONS/VINYL ESTER SYNTACTIC FOAMS

# ZULZAMRI SALLEH<sup>1,2</sup>, MD MAINUL ISLAM<sup>1</sup>, JAYANTHA ANANDA EPAARACHCHI<sup>1</sup> and MD TAWHIDUL ISLAM KHAN<sup>3</sup>

<sup>1</sup>Centre for Future Materials School of Mechanical and Electrical Engineering Faculty of Health, Engineering and Sciences University of Southern Queensland Toowoomba, Queensland 4350 Australia e-mail: Mainul.Islam@usq.edu.au <sup>2</sup>Universiti Kuala Lumpur Malaysian Institute of Marine Engineering Technology 32200, Lumut, Perak Malaysia <sup>3</sup>Department of Advanced Technology Fusion Faculty of Science and Engineering Saga University Saga 840-8502 Japan

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#### Abstract

In this study, the water absorption of glass microballoon filled vinyl ester syntactic foams at room temperature (25°C) and high temperature (70°C) hygrothermal conditions was investigated. Water absorption rates varied due to the effect of density of syntactic foams because the pore and void containment attributed to higher glass microballoon content. The diffusion rate was varied when the syntactic foams were immersed in water with different conditions, such as salt water (SW) being slower than fresh water (FW) and double distil water (DD) due to the effect of pores. The mechanical properties of syntactic foams when immersed in different types of water at room temperature and under hygrothermal conditions for 30 days and 60 days also varied. It was found that both compressive and tensile modulus decreased when more microballoon content was added and when immersed for a long duration. This phenomenon was also attributed to pores and voids expanding their size compared to dry specimens. Hence, this was further evidence that porosity and void containment in the syntactic foams affected the mechanical and thermal properties.

# 1. Introduction

The multi-functional particulate composites, which are called syntactic foams, have a broad range of applications either in structural engineering as damper, insulator or as flame retardant materials. They are also used as core materials in sandwich panels for marine structures [1]. Light weight material, as a way to save weight, is the main objective that needs to be achieved for all applications. Weight sensitivity is exhibited with higher specific properties by syntactic foams, which are made from cenospheres, glass microballoons or hollow microspheres mixing with a polymeric resin, may be promising in marine applications [2-8]. These are useful for a deep sea environment as buoyancy aid materials [9]. Hence, water absorption is highly recommended to be investigated in order to determine the viability for these applications. Porosity occurs in most of syntactic foams, which contributes to the water or moisture absorption characteristics. A very low moisture absorption coefficient is found, along with high compressive strength for closed porosity [9]. These researchers also found that there is no significant difference in compressive strength for low temperature conditions when compared to dry specimens, while it has detrimental effects in higher temperature conditions. They found that thermodynamic temperatures affects dynamic compressive behaviour when compared to environmental temperatures [10]. The electrical conductivity increases in the process of de-ionised water absorption for temperatures ranging from 20 to 200°C, for the duration of 18 months [11]. In higher humidity, the weight gain increases for different types of water [12]. The strength of phenolic syntactic foams decreases by almost 30% in total when weight gain occurs by 70% within 500 days [13]. Moisture absorption for different types of water reduces flexural properties, particularly modulus of elasticity [14]. Based on the literature mentioned above, most of it focused on the effects of the degradation mechanism and residual mechanical properties of syntactic foams for hygrothermal properties, hydrolytic ageing and moisture absorption at environmental temperatures. Moisture diffusion in polymeric composites has been shown to be governed by three different mechanisms [15-19]. The first mechanism involves diffusion of water molecules inside the micro gaps between polymer chains. The second mechanism involves capillary transport into the gaps and flaws, which are interfaces between fibre and matrix. The third mechanism involves transport of micro-cracks in the matrix arising from the swelling of fibres (particularly in the case of natural fibre composites) [20].

In this study, the focus was on the effect of three types of water, namely, fresh water (FW), double distil water (DD), and salt water (SW), on mechanical properties such as compressive and tensile properties, with respect to water absorption changes. Investigations were also carried out to determine the effects of water absorption on the internal structure of syntactic foam composites with regards to the voids and porosities.

# 2. Materials and Method

Syntactic foams were formed from a mixture of vinyl ester resin and borosilicate glass microballoon filler. Methyl ethyl ketone peroxide was used as a hardener. The mean particle size distribution and bulk density of this microballoon are  $72\mu m$  (5-150 $\mu m$ ) and  $110 \text{kg/m}^3$ , respectively. Five different compositions of microballoon content were used, namely, 2, 4, 6, 8, and 10wt.%.

# 2.1. Water absorption with different types of water

The ASTM 5229 was followed in this study to evaluate the behaviour of water absorption of syntactic foams. The specimens were immersed into three types of different aqueous environments originally taken from dam water, called fresh water (FW), double distil water (DD) which was prepared with two-time condensation process, and red sea salt dissolved with tap water, which was procured from Ocean Company Ltd to produce salt water (SW). The experiment was performed in room temperature (25°C) and all the specimens were immersed in a plastic container, as shown in Figure 1. The duration of the immersion process was longer than 60 days for all specimens.



Figure 1. The immersion process of syntactic foams in plastic container.

The specimen weight was measured prior to determining the density of the foam for two different durations, namely, after being immersed for 30 days and 60 days for both types of compressive and tensile specimens. From the Rule of Mixture (ROM), the density of cavity porosity can be calculated by using the Equation (1),

$$\emptyset_p = 1 - \frac{\rho_{\exp}}{\rho_{\text{theo}}},\tag{1}$$

where  $\rho_{exp}$  and  $\rho_{theo}$  are the experimental density and theoretical density, respectively.

For the water absorption measurements, the specimens were withdrawn from the water, wiped dry to remove the surface moisture, and then weighed using an electronic balance with an accuracy up to  $10^{-4}$ g to monitor the mass during the ageing process. The moisture content, W(t) absorbed by each specimen was calculated from its initial weight,  $W_0$  and weight of the specimens at time t,  $W_t$  as follows:

$$W(t) = \left[\frac{W_t - W_0}{W_0}\right] \times 100\%.$$
 (2)

Alomayri et al. [21] used the Fick's behaviour for water absorption behaviour in their study. The following formula was used [22, 23];

$$\frac{M(t)}{M(\infty)} = 4 \times \left(\frac{D \times t}{\pi \times h^2}\right)^{\frac{1}{2}},\tag{3}$$

where M(t) is the water content at time  $t, M(\infty)$  is the equilibrium water content, D is the diffusion coefficient, and h is the sample thickness. Diffusion coefficient, D is calculated from the slope of moisture content versus the square root of time by:

$$D = \pi \times \left(\frac{h}{4M_{\infty}}\right)^2 \times \left(\frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}}\right)^2.$$
(4)

Assuming that the absorption process is linear at an early stage of immersion, times are taken at the beginning of absorption process, so that the weight change is expected to vary linearly with the square root of time. The actual specimens were measured before starting the water absorption process. The thicknesses of specimens varied for both compression and tensile testing which are difficult to control during the fabrication process. An average value of thickness for: compression is 50 mm and tensile is 10 mm, while an average surface area for: compression is  $600 \text{ mm}^2$  and tensile is  $90 \text{ mm}^2$ .

# 2.2. Investigation on hygrothermal properties for different types of water

The hygrothermal process began with the setup shown in Figure 2. The specimens were soaked in the aluminium bowl placed on the heater plate at a setting temperature of 70-80°C, connected to the thermostat box controller. The thermocouple was also placed in the water to detect the temperature and was connected to the thermostat box controller. All the tensile and compression specimens were immersed in the bowl, as shown in Figure 2. During the hygrothermal process, the water needed to be topped up every two days because of the evaporation of water. The specimens were taken out at same time every day to measure the weight gain percentage. Similar to the water absorption process, all the specimens were wiped and dried before being placed on a digital weighing machine to determine their mass. This step was repeated for a duration of 30 days and 60 days before being changed to another type of water, before proceeding to mechanical testing. The specimens that achieved the saturation weight at the end of the process would proceed to mechanical testing employing an MTS machine.



Figure 2. Setup for hygrothermal experimentation of syntactic foams.

# 3. Results and Discussion

#### 3.1. Density of compressive specimens

The foam densities for compressive specimens with water absorption, as shown in Figure 3, are comparable with theoretical values. Generally, all the specimens had decreased densities when immersed into the FW, DD, and SW for different wt.% of glass microballoons. In the graph, the densities of syntactic foams showed a slight drop when immersed in water. The dry specimens' densities also decreased when more glass microballoon was added, namely, at 8-10wt.%, but their densities were remain almost constant at a lower content of glass microballoons, namely, at 2-6wt.%. Among all the specimens, syntactic foams immersed in SW showed higher density of 1231kgm<sup>-3</sup>, while the dry specimen density was 1118kgm<sup>-3</sup> for the 2wt.% of microballoons. The density for FW showed a consistent decreasing trend with a minimum density of 693kgm<sup>-3</sup>, belonging to specimen with 10wt.% microballoons. The error bars for graphs show the accuracy of the values for each standard deviation. Syntactic foams immersed in the water changed their densities with different wt.% of glass microballoons, due to absorbing water that contributed to different densities. This attributed to voids and pores contained in syntactic foams, since the water could not enter the polymeric resin, and also hydrated in between glass microballoons and resin, or in the glass microballoons themselves. In addition, syntactic foams that had higher glass microballoon content sank in the bottom of container, indicating that density was greater than 1gcm<sup>-3</sup>, while the lower content ones floated on the water surface. This also supports the fact that the effect of having voids, porosity or debris of glass microballoons in syntactic foams contributes to changes in density. Xua and Li [24] also found that difference in moisture absorption is a possibility when containing all these parameters.





**Figure 3.** Densities of compressive specimens of syntactic foams immersed in different water conditions; (a) dry specimen, (b) fresh water (FW), (c) double distil water (DD), and (d) salt water (SW).

#### 3.2. Density of tensile specimens

Figure 4 shows densities of tensile specimens with water absorption that are comparable to theoretical values. It is revealed that for all the specimens, when immersed in FW, DD, and SW for different wt.% of glass microballoon contents, their densities decreased. From the graph, densities of syntactic foams showed a slight drop when immersed in water. Similar to compressive specimens, the dry specimens here also showed their densities decreased when glass microballoon contents were increased, especially for 10wt.% microballoons. Among all the specimens, syntactic foams immersed in DD showed a higher density of 1280 kgm<sup>-3</sup> for 2wt.% microballoons, while the FW specimen density was 1272kgm<sup>-3</sup> for 4wt.% microballoons. The density for FW showed a trend consistently constant with a minimum density of  $693 \text{kgm}^{-3}$  belonging to one with 10wt.% microballoons. The measured values are verified by examining their deviations at each of graphs using error-bars. Generally, Figure 4 shows that the deviations in density are smaller compared to dry specimens. Syntactic foams that were immersed in the water changed their densities with different wt.% of glass microballoons, due to same reason mentioned in previous section. The longer they were immersed in the water, the density increased more tremendously. This occurred when the tensile specimen shape was rectangular and it may have been much easier for the water to enter into the porosity and voids area, compared to compressive specimens with cylindrical shape.





**Figure 4.** Densities of tensile specimens of syntactic foams immersed in different water conditions; (a) dry specimen, (b) fresh water (FW), (c) double distil water (DD), and (d) salt water (SW).

#### 3.3. Room temperature water absorption of syntactic foams

# **3.3.1.** Compressive specimens

The water absorption graph for compression specimens of syntactic foam in room temperature (25°C) is shown in Figure 5. Each curve represents the average data of three specimens. This graph shows that the absorbed water content increased with increasing immersion time. During the immersion process, the presence of water in the foams was due to the diffusion of water from the surface to the internal areas of syntactic foams. In this study, a quantitative measure of the water entrapped in the pores is given as  $W(t_1)$  where the water content associated with the first weight measurement taken at  $t_1$  = Day 1 of water immersion. Thus, the maximum water absorption due only to diffusion in the matrix material  $W^m$  can be calculated as  $W^s - W(t_1)$ , which is also proposed by Tagliavia [25], where  $W^s$  is the weight of specimen when an equilibrium condition was achieved. Table 1(a)-(c) shows all these parameters related to porosity content in the compressive specimens.





**Figure 5.** Water absorption of compression specimens immersed in different water conditions; (a) fresh water (FW), (b) double distil water (DD), and (c) salt water (SW).

**Table 1(a).** Typical results for fresh water (FW) absorption analysis of compressive specimens

Material type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $(\frac{W^m}{\sqrt{t}})$ $(\%/s^{1/2})$
SF2WC-F	0.65918	0.02942	0.62976	2352	0.0002678
SF4WC-F	0.81238	0.04916	0.76322	2388	0.0003196
SF6WC-F	1.17384	0.14394	1.02990	2477	0.0004158
SF8WC-F	1.47386	0.25897	1.21489	2613	0.0004649
SF10WC-F	1.92902	0.41905	1.50997	2726	0.0005539

Material type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $(\frac{W^m}{\sqrt{t}})$ $(\%/s^{1/2})$
SF2WC-D	0.68007	0.20989	0.38117	1972	0.0001933
SF4WC-D	0.60297	0.04529	0.55707	2160	0.0002579
SF6WC-D	1.01042	0.08775	0.92267	2357	0.0003915
SF8WC-D	1.34567	0.16267	1.18300	2380	0.0004954
SF10WC-D	1.56758	0.21987	1.34771	2471	0.0005454

**Table 1(b).** Typical results for double distil water (DD) absorption analysis of compressive specimens

**Table 1(c).** Typical results for salt water (SW) absorption analysis of compressive specimens

Material Type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $\left(\frac{W^m}{\sqrt{t}}\right)$ $\left(\frac{\%}{s^{1/2}}\right)$
SF2WC-S	0.65725	0.07356	0.58369	2163	0.0002699
SF4WC-S	0.76821	0.12356	0.64465	2354	0.0002739
SF6WC-S	0.97110	0.21328	0.75782	2390	0.0003171
SF8WC-S	1.27061	0.22748	1.04313	2480	0.0004206
SF10WC-S	1.38515	0.39453	0.99062	2612	0.0003793

From Table 1(a) to (c), and comparing with the Figure 5, FW and SW required longer immersion times for saturation with the highest times of  $2471s^{1/2}$  and  $2612s^{1/2}$ , respectively, while DD specimens required a shorter time of maximum  $2471s^{1/2}$ . By observing the ratio  $\left(\frac{W^m}{\sqrt{t}}\right)$  for specimens with 2-10wt.% of glass microballoons, it can be noticed that they exhibited a slight increase in their results within the range of 0.0001933 to 0.0004206%, regardless of wall thickness and water environment. Based on the experiment, the contact surface area between the matrix and the water involved in the diffusion was practically the

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same and water did not diffuse in glass microballoon particles. For the specimens with 10wt.% of glass microballoons, DD water absorption had a high ratio when compared among all water conditions. This might have contributed to the higher cavity/matrix porosity content for the first type, which allowed for larger contact surface areas and faster water diffusion, especially for larger glass microballoon content. Tagliavia [25] suggested that specimens with 60% volume fraction of hollow particles had the highest ratio, regardless of density and water conditions, which is important in determining the water up take in the specimens. Table 1(a)-(c) also shows the measured values of maximum  $W^m$ , for FW specimens, and this quantity spans the range of 0.63-1.5% of the initial weight. As the weight percentage increased,  $W^m$  also increased for any water conditions, which led to an increase in the porosity content. This phenomenon allowed the water to be entrapped inside the pore cavities as well. The calculation of  $\mathcal{Q}_p$  is based on the overall weight of syntactic foams, therefore it is difficult to identify their size and location in the foam. This porosity may entrap the water or air as soon as the specimen is dipped into the water without any actual diffusion taking place. Figure 6 shows the estimation of density of porosity content as a percentage in different water conditions. Among all water conditions, DD water had a higher percentage of porosity estimation compared to others, namely from 4-9%.



**Figure 6.** Estimation of density of porosity in different water conditions for dry specimens, fresh water (FW), double distil water (DD), and salt water (SW) specimens.

The alkalisation of DD water allowed more water to intersect internally, and thus made a contribution to the porosity being exposed in an area of the syntactic foams. When compared between dry and FW specimens, the porosity content was not much different. The SW showed the porosity content increased from 2.8 to 4.8% when the glass microballoons in the syntactic foam were increased. In order to check the accuracy for water absorption (%), the error bars for all graphs were calculated, and it was found that on average they were still within 0.25%.

Polymeric syntactic foams also showed plasticisation behaviour, which induced some chemical and physical modification; this could attract the water getting into the microstructure of syntactic foams. This modification changed the physical properties of polymeric composite materials, such as swelling, hydrolysis, lixiviation and others [26]. Lefebvre and Sauvant-Moynot [26] found that water absorbed in the foams was subjected to irreversible ageing, with three mechanisms related to the material parameters and behaviour: (i) matrix resin hydration, (ii) glass microballoon hydration, interface hydration, and (iii) pore cavities filling with water.

# 3.3.2. Tensile specimens

Figure 7 shows the plots for water absorption at room temperature  $(25^{\circ}C)$  for tensile specimens of glass microballoon syntactic foams. Each curve represents the average data of three specimens. This graph shows that the absorbed water content increased with increasing immersion time, until the equilibrium condition was achieved at 60 days duration. The individual error bar for all graphs also showed that the water absorption (%) for all specimens are not much different which are within the range of 0 to 1.5%. Similar to the compressive specimens, it was intention to investigate the mechanism of water intake allowable into syntactic foams with the equation of water absorption,  $W^s - W(t_1)$  which has also been explained in the previous section. Table 2(a)-(c) shows all these parameters related to the porosity content in the tensile specimens.





**Figure 7.** Water absorption for tensile specimens immersed in different water conditions; (a) fresh water (FW), (b) double distil water (DD), and (c) salt water (SW).

Table 2(a). Typical results for FW water absorption analysis of tensile specimens

Material type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ $s^{1/2}$	Ratio $\left(\frac{W^m}{\sqrt{t}}\right)$ $(\%/\mathrm{s}^{1/2})$
SF2WT-F	1.7414	0.16169	1.57971	2180	0.0007246
SF4WT-F	2.3527	0.34392	2.00878	1905	0.0010545
SF6WT-F	3.08634	0.41221	2.67413	1950	0.0013713
SF8WT-F	4.57965	0.56573	4.01392	2180	0.0018412
SF10WT-F	5.08716	0.65009	4.43707	1764	0.0025153

Material type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $(\frac{W^m}{\sqrt{t}})$ $(\%/\mathrm{s}^{1/2})$
SF2WT-D	2.38990	0.22804	2.16186	2240	0.0009651
SF4WT-D	3.79800	0.43132	3.36668	2477	0.0013592
SF6WT-D	4.70429	0.71120	3.99309	2483	0.0016082
SF8WT-D	6.95298	0.75768	6.19530	2450	0.0025287
SF10WT-D	8.01366	1.04421	6.96945	2645	0.0026350

**Table 2(b).** Typical results for DD water absorption analysis of tensile specimens

**Table 2(c).** Typical result for SW water absorption analysis of tensile specimens

Material type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $\left(\frac{W^m}{\sqrt{t}}\right)$ $\left(\%/\mathrm{s}^{1/2}\right)$
SF2WT-S	2.27251	0.26739	2.00512	2424	0.0008272
SF4WT-S	2.65509	0.39199	2.26310	2579	0.0008775
SF6WT-S	2.95429	0.72097	2.23332	2575	0.0008673
SF8WT-S	3.79707	1.06179	2.73528	2520	0.0010854
SF10WT-S	4.58175	1.54066	3.04109	2494	0.0012194

Results from Table 2(a) to (c) comparing with Figure 7, where the specimens were immersed in DD and SW have the highest saturation with time of  $2645s^{1/2}$  and  $2494s^{1/2}$ , respectively. FW specimens required a shorter time for immersion at a maximum of  $1764s^{1/2}$  when the glass microballoon content was increased in syntactic foams. The duration of 100 days showed that the water absorption mechanism produced the equilibrium trend between water gain and  $(\sqrt{t})$  for all syntactic foams. As a general trend, it can be noticed that in syntactic foams weight gains were higher for DD as compared to FW and SW. A larger scattering in the obtained data was revealed for all the tested

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syntactic foams immersed in FW, including the equilibrium being achieved faster than others. In practice, as the weight percentage of glass microballoon increases, the water uptake also increases. Therefore, the syntactic foam density was expected to have a prominent effect on the weight gain trend because the porosity and void contents might have absorbed the water inside of them. The glass did not absorb water, and the effect of weight gained for any water filling in the pores was eliminated by subtracting the first weight measurement. In this case, the specimens with higher content of glass microballoons experienced with the cavity pores occurred due to much glass debris caused the water uptake to increase. This also happened due to the matrix-particle interface area [25]. They also found poor interfacial bonding between matrix and filler during SEM observation. Therefore, the higher microballoon content led to having a higher interface area.

# 3.4. Hygrothermal properties of syntactic foams

# 3.4.1. Compressive specimens

Hygrothermal trends for all compressive syntactic foams in high temperatures (70°C) are shown in Figure 8. The values shown in this figure are average values based on water absorption by a minimum of three specimens for each composition under each type of water condition. This graph shows that the absorbed water content increased with an increase in immersion time until it achieved the equilibrium condition. The figure shows that the equilibrium of water absorption in room temperature can be achieved at  $1500s^{1/2}$ . However, it took a much longer time at 70°C to attain an equilibrium system, such as that for FW at  $2500s^{1/2}$ . Generally, a large difference in the water absorption tendency could be seen with change in temperature. For example, in room temperature condition, DD type water absorption was less than 1.6%. But at a high temperature, this increased to 14% maximum water absorption in the syntactic foams. Gupta and Woldesenbet [9] also found that water absorption increased at a high temperature between 5 and 10% when immersed in DD and SW conditions.





**Figure 8.** Hygrothermal behaviour for compressive specimens immersed in different water conditions; (a) fresh water (FW), (b) double distil water (DD), and (c) salt water (SW).

 Table 3(a). Typical results for FW hygrothermal analysis of compressive specimens

Material type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $(\frac{W^m}{\sqrt{t}})$ $(\%/\mathrm{s}^{1/2})$
SF2WCH-F	2.15378	0.26666	1.88712	1972	0.0009570
SF4WCH-F	8.35421	0.76653	7.58768	2200	0.0034489
SF6WCH-F	8.50674	0.50031	8.00643	2388	0.0033528
SF8WCH-F	14.95858	0.85576	7.65098	2834	0.0026997
SF10WCH-F	14.72946	0.60691	14.12255	1727	0.0081792

Material type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $(\frac{W^m}{\sqrt{t}})$ $(\%/s^{1/2})$
SF2WCH-D	1.13456	0.34733	0.78723	2613	0.0003013
SF4WCH-D	4.0425	0.48518	3.55732	2726	0.0013050
SF6WCH-D	8.03452	1.02652	7.00800	2835	0.0024720
SF8WCH-D	13.21456	1.43501	11.77955	2924	0.0040286
SF10WCH-D	9.02783	0.72755	8.30028	2954	0.0028098

**Table 3(b).** Typical results for DD hygrothermal analysis of compressive specimens

**Table 3(c).** Typical results for SW hygrothermal analysis of compressive specimens

Material type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $\left(\frac{W^m}{\sqrt{t}}\right)$ $\left(\%/\mathrm{s}^{1/2}\right)$
SF2WCH-S	1.73542	0.15234	1.58308	2385	0.0006638
SF4WCH-S	7.44368	0.70502	6.73866	2475	0.0027227
SF6WCH-S	6.45632	0.55346	5.90286	2615	0.0022573
SF8WCH-S	6.82311	0.55408	6.26903	2832	0.0022136
SF10WCH-S	8.81456	0.83121	7.98335	2998	0.0026629

In Table 3(a) to (c), when compared with Figure 8, DD and SW required longer immersion times for saturation with longest times of  $2954s^{1/2}$  and  $2998s^{1/2}$ , respectively. The data also shows that the percentage of water absorption increased almost 7 times to achieve the equilibrium system in hot water conditions, especially for FW specimens. This is evident that the higher number of porosity and voids occurred in the syntactic foams, and was present near the surface of the syntactic foams, after they opened their surface areas. Gupta and Woldesenbet [9] also reported that this phenomenon might be due to the strength of filler with different levels of wall thickness. Therefore, a higher wt.% of glass

microballoons in syntactic foams contributed more to the matrixinterfacial bonding results  $(W^m)$  for all specimens, for example, 10wt.% composition in all water conditions.

#### 3.4.2. Tensile specimens

Similar results have also been reported for tensile hygrothermal specimens as shown in Table 4(a)-(c). All specimens showed an increased percentage of water absorption, almost 5 to 7 times, compared to room temperature conditions shown in Figure 9. This was attributed to their physical and mechanical properties as well. The immersion time to achieve the equilibrium took longer as  $2910s^{1/2}$  for all specimens with 10wt.% of glass microballoon content. Similar to compression specimens, the span for error bars was also found longer for tensile specimens.





**Figure 9.** Hygrothermal behaviour for tensile specimens immersed in different water conditions; (a) fresh water (FW), (b) double distil water (DD), and (c) salt water (SW).

Table 4(a). Typical results for FW hygrothermal analysis of tensile specimens

Material Type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $\left(\frac{W^m}{\sqrt{t}}\right)$ $(\%/s^{1/2})$
SF2WTH-F	9.02611	1.54673	7.47938	2511	0.0029786
SF4WTH-F	14.67463	2.23415	12.44048	2596	0.0047922
SF6WTH-F	25.2426	3.54672	21.69588	2678	0.0081015
SF8WTH-F	32.35706	3.43520	28.92186	2757	0.0104903
SF10WTH-F	37.15931	3.87659	33.28272	2910	0.0114374

Material Type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $(\frac{W^m}{\sqrt{t}})$ $(\%/\mathrm{s}^{1/2})$
SF2WTH-D	4.67843	0.88797	3.79046	2612	0.0014512
SF4WTH-D	17.11231	3.44563	13.66680	2666	0.0051263
SF6WTH-D	22.22341	5.33241	16.89100	2731	0.0061849
SF8WTH-D	21.00112	1.00342	19.96692	2835	0.0070430
SF10WTH-D	36.88796	6.99806	29.88980	2741	0.0109047

Table 4(b). Typical results for DD hygrothermal analysis of tensile specimens

**Table 4(c).** Typical results for SW hygrothermal analysis of tensile specimens

Material type	Max. W <sup>s</sup> %	W(t <sub>1</sub> ) %	Max. W <sup>m</sup> %	Immersion time $(\sqrt{t})$ s <sup>1/2</sup>	Ratio $\left(\frac{W^m}{\sqrt{t}}\right)$ $(\%/s^{1/2})$
SF2WTH-S	7.59803	1.54321	6.05482	2369	0.0025559
SF4WTH-S	16.13456	2.54362	13.59094	2460	0.0055248
SF6WTH-S	22.22345	4.23452	17.98893	2562	0.0070214
SF8WTH-S	24.25463	3.89765	20.35698	2710	0.0075118
SF10WTH-S	30.11257	4.89765	25.21492	2820	0.0089415

# 3.5. Coefficient of diffusion of syntactic foams

In this study, the slope in Equation (4) was calculated from the firsttime equilibrium achieved, and the diffusion coefficient D calculated from experiment data, is tabulated in Table 5(a). Water diffusivity in the foams is generally higher in FW when compared to other water conditions. This is comparable with increasing the glass microballoon content in syntactic foams as well, where the diffusivity value increased for composites containing higher porosity content. Specimens SF8WT (with 8wt.% of microballoons) and SF10WT (with 10wt.% of microballoons) had large gaps with 3 times higher D values compared to the lower glass content in FW condition. However, D values for all compositions were slightly smaller in SW condition, even though the glass microballoon content was increased. The reason for such a large discrepancy could be attributed to the high matrix porosity content in SF8WT and SF10WT (see Tables 1 and 2 for both compressive and tensile specimens). This is the cause of the large value for  $W^m$  in FW and DD conditions. Similar results were detected when the specimens were immersed at high temperature (70°C) for the hygrothermal conditions. Generally, the water diffusion coefficient, D increased for all water systems in high temperature condition. It was revealed that D had a higher value for specimens SF8WT and SF10WT when immersed in both of FW and DD conditions (see Table 4(b)).

**Table 5(a).** Typical results for diffusion coefficient, D for water absorption in room temperature

	SF2WT		SF4WT		SF6WT		SF8WT		SF10WT	
Diffusion property	(×10 <sup>-9</sup>	mm²/s)	$(\times 10^{-9}\mathrm{mm^2/s})$		$(\times 10^{-9}\mathrm{mm^{2}/s})$		$(\times 10^{-9}\mathrm{mm^2/s})$		$(\times 10^{-9}\mathrm{mm^2/s})$	
	D	CoV	D	CoV	D	CoV	D	CoV	D	CoV
FW	6.94	0.31	17.37	0.25	17.23	0.46	24.08	0.02	58.03	0.14
DD	5.70	0.74	6.40	0.86	2.11	0.43	36.83	0.28	46.80	0.09
SW	4.25	0.06	3.83	0.24	1.31	1.04	1.13	0.58	14.86	1.23

\*CoV: Coefficient of variance

**Table 5(b).** Typical results for diffusion coefficient, D for hygrothermal water absorption temperature

	SF2WT		SF4WT		SF6WT		SF8WT		SF10WT	
Diffusion property	$(\times 10^{-9}  \text{mm}^2/\text{s})$		$(\times 10^{-9}\mathrm{mm}^2\mathrm{/s})$		$(\times 10^{-9}\mathrm{mm^2/s})$		$(\times 10^{-9}\mathrm{mm}^{2}\mathrm{/s})$		$(\times 10^{-9} \mathrm{mm}^2/\mathrm{s})$	
	D	CoV	D	CoV	D	CoV	D	CoV	D	CoV
FW	7.85	0.41	13.57	0.21	15.21	0.05	35.71	0.01	67.63	0.22
DD	6.45	0.23	5.95	0.75	5.88	0.51	57.36	0.54	61.52	0.10
SW	3.22	0.14	2.11	0.31	7.42	0.01	4.34	0.51	21.73	0.66

Tables 5(a) and (b) show a higher diffusion coefficient, D and a maximum of water absorption,  $W^m$ , when immersed into FW, followed by DD and SW conditions. The reduction of these values, particularly in SW condition, were due to changes in the main physicochemical sources in addition to the specimen surface hydration. A similar reduction of D values in SW was also reported by Tagliavia et al. [25] when a different compositions of glass microballoon content were used in syntactic foams. This contributes to the global amount of water absorption and the majority of each mechanism depends on ageing conditions, and the durability of each component. In details, for foams immersed in SW condition, because the ionic species of salt are larger in size than those of normal water, the presence of salt ions in the water interferes with the diffusion of water in the foams, and ionic species of the salt have a much slower diffusion rate compared to that of water. The occupancy of pores in the foams may further reduce the diffusion rate particularly for the higher glass microballoon content. Xua and Li [24] also reported that the diffusion rate is slower in salt water condition when compared to rain water, with a difference of 0.34% in water absorption of glass microballoon/SMP resin. A similar finding was also revealed by Gupta and Woldesenbet [9], who noted that the deposition of salt may give allowance for the pores to be occupied in the foam and contribute to slower D values. However, for syntactic foams which were immersed in FW and DD conditions, the water uptake was more than SW condition. This was because as per the chemistry, both waters were enriched with a lot of nutrients in organic ions such as  $NO_3-$ ,  $SO_4^{2-}$ ,  $F^-$ ,  $NH_4^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Na^+$ ,  $K^+$  and organic species such as  $CH_3COO^-$ ,  $HCOO^-$ ,  $CH_2(COO)_2^{2-}$ ,  $C_2O_4^{2-}$ , as reported by Song and Gao [27]. The water resources from rain contains more aggressive ions that may easily hydrate the polymer matrix in the syntactic foams [27]. Hence, a larger amount of organic materials was deposited in the open pores, causing a greater weight gain than that of smaller inorganic ions in SW.

#### 3.6. Effect of water absorption on mechanical properties

## 3.6.1. Compression testing

Compression test results after being immersed for 30 days in all water conditions at room temperature are shown in Figure 10(b)-(d). Dry specimens (Figure 10(a)) and the specimens immersed in all water conditions at room temperature were compressed to strain as high as 0.20% for compressive properties. From Figure 10(a), it is observed that the stress-strain curves for the dry specimens behaved similarly to the specimens immersed in FW, DD, and SW conditions. These curves showed elastic characteristics at a low strain level. After the elastic region, the stress became nearly constant at considerable strain during further compression, which is referred as the densification stage. At the end of the densification stage, the stress started increasing again. This behaviour was revealed to be more likely for lower glass microballoon content, which took longer to fracture, compared to higher microballoon content that was much lower in rigidity and allowed more intact microballoons to be crushed. It was revealed that the variations of compressive strength after being exposed to FW, DD, and SW were comparable to dry specimens. Among all specimens, the highest strength belonged to SF2WC (with 2wt.% of microballoons), except in the DD condition belonging to the SF4WC (with 4wt.% of microballoons) specimen. On the other hand, similar trends were found in the corresponding stress-strain curves for each condition after immersion for 60 days. Each value represented the average data of three specimens. This indicated a decreasing trend in compressive strength and modulus with an increasing immersion time; however, there was a trend of an increasing maximum compressive strain as immersion time increased after being exposed to aqueous environment. In Figure 11(c), the compressive strength for SF2WC and SF4WC had a closer ultimate strength at 80MPa. The influence of DD through internal surface of syntactic foams after 60 days might have occurred. Generally, all the specimens had a higher ultimate compressive strength after being immersed in all water conditions for 60 days.





**Figure 10.** Typical stress-strain curves from compression tests after water absorption at room temperature (25°C) for (a) dry, (b) FW, (c) DD, and (d) SW specimens.

These compression test results also showed a similar trend in terms of the details displayed for all mechanical properties as shown in Figure 11(a)-(d) for 30 days and 60 days of immersion in all water conditions. The peak strength in Figure 14(a) shows that its decrease in specimens immersed in all water conditions was comparable to dry specimens after being immersed for both 30 and 60 days. However, comparing among all water conditions, specimens immersed in FW and SW showed a higher strength for both 30 and 60 days. A comparison for the compressive modulus is shown in Figure 11(b). The trends showed an increase of at least one-third after 60 days, particularly when immersed in DD and SW conditions, but it was still lower than dry specimens. Specific compressive strength showed an incremental trend when immersed in DD and SW but FW had a decreasing trend, particularly for specimens with higher glass microballoon content for the duration of 60 days. This was due to the higher density of syntactic foams. Not much difference was observed for specific compressive modulus results, as shown in Figure 11(d).





Figure 11. Typical results from compression testing after specimens being immersed at room temperature.

# 3.6.2. Tensile testing

The tensile test results are shown in Figure 12. The graph exhibits the tensile stress-strain curves of the foams immersed in all water at room temperature and shows a decrease in tensile strength when more glass microballoons were added, compared to dry specimens. The specimens SF2WT (with 2wt.% of microballoons) had higher strength in all water conditions for the duration of 30 days, especially for FW with value of 30MPa. Moreover, specimens with higher glass microballoon content, such as SF8WT (with 8wt.% of microballoons) and SF10WT (with 10wt.% of microballoons) immersed in SW, showed the lower strength values of 8-10MPa. This indicated that the immersed foams in SW had more ductility than in other water due to the moisture content in the foams, which had more salinity in chemical reaction. This might have caused more severe plasticization of syntactic foams but lower strength.





**Figure 12.** Typical stress-strain curves from tensile tests after water absorption at room temperature (25°C) for (a) dry, (b) FW, (c) DD, and (d) SW specimens.

While specimens were immersed in all water conditions for 60 days duration, they exhibited very large decrease in tensile strength with the highest value of 20MPa only. This discrepancy might have occurred due to hydrolytic ageing of glass microballoons, especially when immersed in FW and DD, which had more  $OH^-$  content compared to SW. In addition, it also caused more weakening between the matrix and glass microballoon interface when immersed for a long duration, regardless of the water condition. Sauvant-Moynot et al. [11] had the same finding and reason for the decrease in stiffness, namely, due to an interface problem.

A comparison of mechanical properties for all specimens immersed in FW, DD, and SW can also be observed in Figure 16. The tensile strength increased when immersed for 30 days in FW and DD conditions, led by specimen SF2WT. Generally, tensile strength showed a decrease when more glass microballoons were added in all water conditions. It was also revealed that the tensile modulus showed a decrease for all specimens and had a similar trend for dry specimens. This might have been due to a debonding problem that occurred between matrix and resin, and as a result the connectivity was loose, which was detected during the tensile

testing of the specimens. Moreover, specific tensile strength showed an increasing trend when immersed in FW and DD, especially for SF2WT specimens as 0.04MPa/kgm<sup>-3</sup>, but decreased in SW condition. Even though specimen SF2WT had the highest specific tensile modulus of  $16 MPa/kgm^{-3}$  when immersed in DD, other specimens still showed a trend of decrement. The porosity could have contributed to these results because the lighter materials, such as higher the microballoon content, higher chance of having more porosities, compared to the higher resin specimens like SF2WT. Similar results were found for the specimens immersed for the duration of 60 days in all water conditions, as shown in Figure 13(a). Generally, the tensile strength had a lower value when compared to dry specimens. When immersed in DD, the specimen SF6WT (with 6wt.% of microballoons) had the highest values while SF10WT had the lowest value as 22MPa and 5MPa, respectively. The tensile modulus showed a decreasing trend for both immersed in FW and SW conditions, but DD still led to this behaviour. Specimen SF6WT had an increase by 20% when compared to dry and FW conditions, while it increased by 50% when compared with SW. Similar to tensile strength, the specific tensile strength also showed an increase of 2-8% when immersed in both of FW and DD. While specimen SF10WT showed not much difference when compared to DD and SW conditions as 0.008MPa/kgm<sup>-3</sup>, this was related to the higher possibility of porosity content in the syntactic foams. The specific tensile modulus in Figure 13(d) showed a slight increase for specimen SF2WT and the highest value belonged to SF4WT as 12MPa/kgm<sup>-3</sup>, which was closer to a dry specimen while SF10WT slightly decreased in all water conditions.





SW

Dry

(a)

Dry

FW

DD





**Figure 13.** Typical results from tensile testing after specimens being immersed at room temperature.

### 3.7. Effect of hygrothermal on mechanical properties

#### 3.7.1. Compression testing

Compression test results for syntactic foams immersed in FW, DD, and SW at 70°C are shown in Figure 14 for both 30 and 60 days. From Figure 14, it is observed that the stress-strain behaviour for the 30-day specimens was found to be similar to those immersed for 60 days in FW, DD, and SW. These curves showed elastic characteristics at a low strain level. After the elastic region, the stress became nearly constant during further compression, which is referred as the densification stage. At the end of the densification stage the stress started increasing again. This behaviour was common for all syntactic foams with having much lower rigidity and more intact microspheres to be crushed. Meanwhile, foams immersed in FW and SW for 60 days exhibited a decrease in yield compressive strength, which was ascribed to water absorption and debonding and damage at interfaces [21]. However, the excepted specimen SF2WT kept its strength at 80MPa in all water conditions with long duration. Also in this figure, a comparison of the yield compressive strength for both hydrolytic and salinity foams is shown clearly. This

revealed that, for hydrolytic specimens, a decrease in the yield strength was seen as compared to the salty specimens. Furthermore, foams immersed in FW for 60 days showed a further decrease in yield strength by 25% and 15-20% for DD specimens, as compared to those immersed in SW.



**Figure 14.** Typical hygrothermal compressive graphs for (a) FW, (b) DD, and (c) SW for 30 and 60 days.

The mechanical properties of syntactic foams can be explained in detail in Figure 15. As shown in the graphs, the majority of the specimens had a decrease in their compressive strength after being immersed for 30 and 60 days in all water conditions when compared with dry specimens. However, specimens SF2WT, SF6WT, and SF8WT showed an increase in 30 days while SF2WT, SF4WT, SF8WT, and SF10WT also showed an increase in their strength in 60 days. Additionally, the compressive strains of hydrolytic foams at the yield compressive strength increased. This indicates that the stiffness of all types of foams decreased due to the presence of moisture in the specimens. FW and DD made the foam softer and brittle than SW did. This caused a considerable decrease in modulus as shown in Figure 15(b). These results were attributed to the moisture content in the foams and the possibility of material property degradation. The foams in water might undergo a faster degradation than in air [24]. Consequently, syntactic foam specimens could be compressed to a higher degree of strain without generation of cracks. Additionally, another reason for decrease in stiffness was hydrolytic ageing of glass, which was not only a direct factor in glass microballoon breakage, but also an indirect factor as a promoter for weakening the matrix-glass microballoon interface [11].





Figure 15. Typical results for hygrothermal compressive tests after being immersed at the temperature of  $70^{\circ}$ C.

# 3.7.2. Tensile testing

Results from tensile testing are shown in Figure 16. The tensile stress-strain curves of the foams immersed in water exhibited a decrease in strength for both 30 and 60 days when glass microballoon content was added. Moreover, foams immersed in SW showed a larger decrease in strength than those in FW and DD for both durations. This indicates that the immersed foams had more ductility than dry foams due to the presence of moisture in the foams, which might cause plasticisation of the resin. Moreover, SW made the foam more ductile than DD and FW conditions. The reason for the decrease in ductility was same as that in compression tests. In addition, the presence of porosity and voids in syntactic foams also contributed to the ductility regardless of the water condition.



**Figure 16.** Typical hygrothermal tensile graphs for (a) FW, (b) DD, and (c) SW for both 30 and 60 days.

The tensile behaviour of syntactic foams can be elaborated in detail in Figure 17. A comparison of tensile strength, modulus, specific strength, and specific modulus are demonstrated in these graphs. Tensile strength of SF2WT was a little bit higher as 32MPa when immersed in SW compared to dry specimens, but both FW and DD ones were still below this result after 30 days. However, while SF4WT showed a higher result as 33MPa when immersed in DD, the rest were still lower than this after 60 days.





**Figure 17.** Typical results for hygrothermal tensile tests after being immersed at temperature of 70°C.

In comparing modulus values, it can be seen that all types of syntactic foams were affected due to the presence of moisture in the specimens after being immersed in high temperature conditions. Specimens SF4WT, SF6WT, SF8WT, and SF10WT showed a reduced tensile modulus for all water conditions in 30 days, namely, 5-15% reduction. However, SF2WT still had a higher value of 9531MPa, which was still below the dry specimen value. When immersed in high temperature conditions for 60 days, it was revealed that tensile modulus decreased, especially for specimens SF2WT, SF4WT, SF8WT, and SF10WT. These behaviours could be attributed to two factors: the moisture content entrapped in the porosity regime in the specimens, and the possibility of material property degradation. According to Figure 15, the water absorption for compressive specimens at high temperature was much higher than room temperature at around 16% maximum. Conversely, for tensile specimens, according to the Figure 17, the water absorption was around 35% maximum when compared to the room temperature conditions.

A considerable decrease in modulus revealed that water was infused in the specimens, allowing for cavity and matrix porosity to contain the water inside. Due to being brittle and easily cracked, the strength as well as modulus were reduced in the high temperature specimens, which indicated an occurrence of some additional events in the material. It should be noted that the thermal and water absorption into the porosity area induced strains that generated the syntactic foams to disintegrate the glass microballoons, which could then lead to fracture. Due to the fracture of the glass microballoons, it could develop cavity or matrix porosity, and as a result not only the strength and modulus of syntactic foams would go down, but further water absorption would also increase. In addition to this, it should also be noted that when more glass microballoons were added, the potential for them to be broken was higher and it generated more porosity. Therefore, the hygrothermal strain could cause rupture of the matrix resin in some places near the glass microballoons, which would reduce the strength of syntactic foam specimens. Finally, contributions from the hygrothermal conditions could also expand air being trapped in the porosity and voids which would then slightly reduce the strength and modulus of the specimens.

# 3.8. Fractographic examination of the tested compressive specimens

Fractographic examination of the tested compressive specimens (see Figure 18) shows the various toughening mechanisms in syntactic foams after being immersed in FW, DD, and SW conditions. Fractured, crushed and de-bonded glass microballoons, and matrix deformation were observed. The de-bonded glass microballoons were clearly revealed in the

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specimens in SW where the matrix gap was larger than other specimens immersed in FW and DD, as shown in Figure 18(a). Generally, porosity and voids occurred in all the specimens. The effect of the hygrothermal behaviour on the specimens was obvious when being immersed in SW at high temperature. Ray [28] reported that this thermal stress or hydrothermally generated porous and weaker interface could allow capillary flow of absorbed water in the composites at higher conditioning temperatures. He also reported the occurrence of porosity entrapped with water inside the glass microballoons, or voids could also reasonably cause matrix deformation cracking, resin de-bonding and microballoon de-bonding [28]. As can be seen in Figure 18(b)-(c), de-bonding failure modes occurred clearly indicated that the water absorption was higher in the hygrothermal process, which might be due to degradation phenomena (Subsection 3.4). The probable reasons for such degradation may be related to the weakening of higher thermal and moisture induced swelling stresses at the interface and/or in the matrix resin. It may also be hypothesized that this conditioning environment could result in either breakdown of chemical bonds or secondary forces of attraction at the interface.

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(b)



(c)

**Figure 18.** SEM images of the fracture surfaces of the specimens after being immersed in (a) SW, (b) FW, and (c) DD.

# 4. Conclusions

The behaviour of glass microballoons/vinyl ester syntactic foams, a type of particulate composites, was investigated for their potentiality in marine applications, such as water absorption in room temperature and high temperature hygrothermal conditions. The density of syntactic foams was investigated after immersing the specimens in three different types of water such as fresh water (FW), double distil water (DD), and salt water (SW) at room temperature ( $25^{\circ}$ C) and high temperature ( $70^{\circ}$ C) for the durations of 30 days and 60 days. The density of syntactic foams varied when glass microballoons were added from 2 to 10wt.% for all water conditions. The density of compressive specimens for a duration of 30 days showed an average value below 1000kgm<sup>-3</sup>, while tensile

specimens showed an average value below 1100kgm<sup>-3</sup>. It was revealed that density of compressive specimens was higher in FW and SW for a duration of 60 days while that of tensile specimens was higher in FW and DD. This was attributed to voids and pores contained in syntactic foams, since the water could not enter the polymeric resin nor hydrate in between glass microballoons and resin, or in the glass microballoons themselves. In addition, SEM photos also revealed that some specimens had cavity porosity, which was filled up with small glass microballoons, and debris from broken microballoons that could discard the water from spreading in syntactic foams.

Water uptake by compressive specimens at room temperature showed an increase in their maximum weight  $(W^s)$  when glass microballoon content was added until the equilibrium condition was achieved. Syntactic foam had the highest maximum weight in FW with 1.92902% for 10wt.% of glass microballoons, when compared to other conditions. The maximum diffusion rate  $(W^m)$  was also highest in FW with a value of 1.50997% and lowest rate was in SW with a value of 0.99062%. Tensile specimens showed the water uptake was achieved at the highest maximum weight in DD with 8.01366% for 10wt.% of glass microballoons, when compared to other conditions. The maximum diffusion rate and weight gain was also higher in DD with 6.96945 and 8% for 10wt.% of glass microballoons, respectively. The lower water gain revealed in SW, namely, a maximum of around 5% only. The alkalisation properties of DD might have contributed to this result. Other factors considered were poor interfacial bonding between matrix and microballoons and plasticisation behaviour of polymeric syntactic foams.

Results from hygrothermal compressive specimens showed that the maximum weight increased to average of around 7 times higher than the room temperature ones. Moreover, the equilibrium condition that could be achieved from the water uptake also showed an increase, especially when immersed in FW and DD. This might be attributed to the presence of porosity and voids near the surface of the specimens, which opened the surface area in hot conditions. Hygrothermal tensile specimens had a higher result, almost 7 times as high, when compared to compressive specimens for the maximum weight and maximum diffusion rate. In physical properties comparison with different shapes of specimens, rectangular ones for tensile specimens, allowed the surface to absorb more water, which included an increase in porosity in the foams. The highest  $W^s$  was detected in FW with a value of 37.15931% for specimens with 10wt.% of glass microballoons. The highest diffusion rate percentage belonged to the same group of specimens.

With regards to rate of water diffusivity (D), D was generally higher in FW when compared to other conditions. However, D values for all compositions were slightly smaller in the SW, even though the glass microballoon content was increased. The reason for such a large discrepancy could be attributed to the high matrix porosity in the foams. Similar results were detected when the specimens were immersed at high temperature for hygrothermal testing. The reduction of D in SW was related to enrichment of organic ions that made glass microballoons much closer to each other. In addition, the plasticisation of matrix resin was more severe in hygrothermal conditions, especially when de-bonding occurred, and the gap would be closed and reduction of water entrapment in the porosity area as well.

Variations of compression strength in room temperature conditions were revealed after being exposed to FW, DD, and SW, which were compared to dry specimens. The compressive behaviour was revealed to be more likely for lower glass microballoon content, which took longer to fracture, compared to higher microballoon content that was much lower in rigidity and allowed more intact microballoons to be crushed. This indicated a decreasing trend in compressive strength and modulus with an increasing immersion time. However, there was a trend of an increasing maximum compressive strain as immersion time increased after being exposed to aqueous environments. It was also revealed that the tensile modulus showed a decrease for all specimens and had a similar trend for dry specimens. This might be due to de-bonding problem that occurred between matrix and resin, and as a result the connectivity was loose, which was detected during the tensile testing of the specimens.

The hydrolytic compressive specimens in hygrothermal conditions showed a decrease in strength as compared to the salty specimens. Furthermore, the mechanical properties of the foams immersed in FW for 60 days showed a further decrease in strength by 25% and 15-20% in DD specimens, as compared to those immersed in SW. Additionally, the compressive strain of hydrolytic foams at ultimate compressive strength increased. This indicated that the stiffness of all types of foams was lowered due to the presence of moisture in the specimens. FW and DD made the foam softer and more brittle than the SW did. The tensile stress-strain curves of the foams immersed in hygrothermal conditions exhibited a decrease in tensile strength when glass microballoon content was added for the durations of 30 and 60 days. Moreover, foams immersed in SW showed a larger decrease in tensile strength than those in FW and DD for both durations. This indicates that the immersed foams had more ductility than dry foams due to the presence of moisture in the foams due to probable plasticisation of the matrix resin. Moreover, SW made the foams more ductile than DD and FW.

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