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Optimal design for epoxy polymer concrete based on mechanical properties and durability aspects

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Running headline: Optimal design for epoxy polymer concrete based on mechanical properties and durability aspects (83 characters)

by

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Optimal design for epoxy polymer concrete based on mechanical properties and durability aspects

Abstract

Polymer concrete has shown a number of promising applications in building and construction, but its mix design process remains arbitrary due to lack of understanding of how constituent materials influence performance. This paper investigated the effect of resin-to-filler ratio and matrix-to-aggregate ratio on mechanical and durability properties of epoxy-based polymer concrete in order to optimise its mix design. A novel combination of fire-retardant, hollow microsphere and fly ash fillers were used and specimens were prepared using resin-to-filler ratios by volume from 100:0 to 40:60 at 10% increment. Another group of specimens were prepared using matrix-to-aggregate ratios from 1:0 decreasing to 1:0.45, 1:0.90 and 1:1.35 by weight at constant resin-to-filler ratio. The specimens were inspected and tested under compressive, tensile and flexural loading conditions. The epoxy polymer matrix shows excellent durability in air, water, saline solution, and hygrothermal environments. Results show that the resin-to-filler ratio has significant influence on the spatial distribution of aggregates. Severe segregation occurred when the matrix contained less than 40% filler while a uniform aggregate distribution was obtained when the matrix had at least 40% filler. Moreover, the tensile strength, flexural strength and ductility decreased with decrease in matrix-to-aggregate ratio. Empirical models for polymer concrete were proposed based on the experimental results. The optimal resin-to-filler ratio was 70:30 and 60:40 for non-uniform and uniform distribution of aggregates, respectively, while a matrix-to-aggregate ratio of 1:1.35 was optimal in terms of achieving a good balance between performance and cost.

Keywords

Epoxy polymer concrete; resin-to-filler ratio; matrix-to-aggregate ratio; empirical modelling; properties; optimal design.

1. Introduction

Concrete, the second most consumed material in the world after water, is increasingly being used due to the rapid growth of the construction sector particularly in developing countries. Its high compressive strength, excellent elastic modulus and durability, and widespread availability at low cost are the key advantages. However, the use of Portland cement concrete may be limited in applications where high tensile strength, good bond strength or excellent resistance to certain extreme exposure conditions are required. One approach to overcome these limitations is through the use of polymer concrete. The characteristics of high tensile strength, good bond strength, excellent durability, fast curing times, low permeability, and casting flexibility make polymer concrete an interesting alternative construction material [1-5]. The construction sectors are accepting alternative materials beyond the traditional approach [6-8].

Polymer concrete consists of aggregates bonded together by a resin instead of a cement. The most commonly used resins are epoxy [9], polyester [10] and vinyl-ester [11]. Although polyester and vinyl-ester resins are less expensive, epoxy resins are preferable because of their excellent mechanical and thermal properties, superior resistance to humidity, low shrinkage and high elongation that produces durable and flexible polymer matrix [12]. To mitigate the high cost of epoxy resins, a range of fillers can be added to dilute the resin content. Fly ash is the commonly used filler in polymer concrete [13]. This study employed two other fillers named a fire-retardant filler and hollow microsphere to improve fire and shrinkage performances respectively. The main application for polymer concrete is in chemical storage, but this has been recently extended to include bridge decks, concrete crack repairs, railway sleepers, pavement overlays, decorative construction panels, waste-water pipes and other structures in aggressive environmental conditions [1, 11, 14, 15].

While polymer concrete offers superior mechanical performances over Portland cement concrete, the main challenge is their prohibitive cost. Polymer concrete is approximately 5-10 times more expensive than normal concrete and therefore, their application is currently limited to structures where an enhanced performance justifies the higher cost. Despite their use in many building and construction applications, there is limited attempt to establish design procedure for polymer concrete [11]. The current approach of selecting mix proportions is random or based on current experience for Ordinary Portland Cement concrete. The extensive literature review suggest that the only reported studies are [16, 17], which developed design procedure based on a small variation of resin (only 4%) and aggregate sizes. Following experimental and analytical approaches, Muthukumar and Mohan [16] optimised polymer concrete composed of different quantities of furan resin, silica aggregates and microfiller. Their findings suggested that the best mechanical properties (compressive, tensile and flexural) can be obtained when the polymer concrete contains 8.5% resin, 76.5% aggregates and 15% microfiller. Recently, Jafari et al. [17] attempted to optimise polymer concrete with three different polymer ratios (10%, 12%, and 14% by weight) and two different coarse aggregate sizes (4.75–9.5 mm and 9.5–19 mm) tested at temperature levels (−15 °C, +25 °C, and +65 °C). Based on compressive, splitting-tensile, and flexural strengths, they suggested that the optimum mix should contain 14% of polymer and coarse aggregates from 9.5 to 19 mm when tested the concrete at a temperature of −15 °C. However, these studies did not elaborate on how the coarse aggregates were distributed in polymer matrix and how durability aspects such as alkaline and hygrothermal environments affects the polymer properties, which are critical for an optimal mix polymer concrete design. Therefore, an improved understanding of the effects of mix parameters on the performance of polymer concrete and an approach for optimal mix design [3] are deemed necessary.

Several parameters affect the properties of polymer concrete such as the type and content of the resin and filler, curing method, curing temperature, humidity and particularly, resin-to-filler ratio and matrix-to-aggregate ratio [18]. Lokuge and Aravinthan [11] studied polymer concretes made with three different resins (polyester, vinylester and epoxy resin) and observed that epoxy and vinylester resins produced concrete with better mechanical properties compared to polyester. The effect of different fillers (fly ash and silica fume) on the mechanical properties of polymer concrete has been studied by Bărbuţă et al. [19] and they concluded that the addition of these fillers improves the mechanical properties of polymer concrete. Elalaoui et al. [9] studied the mechanical properties of epoxy polymer concrete after exposure to high temperatures and they observed a significant strength loss occurred at temperatures greater than 150^{o}C. The effects of water absorption on the mechanical properties of epoxy resin system has been studied by Nogueira et al. [20] and their study found a gradual reduction in tensile properties with increase in absorbed water. Nevertheless, the effects of resin-to-filler ratio and matrix-to-aggregate ratio remain unclear, yet optimising these parameters may have major performance and cost implications.

To understand the influence of these parameters, the study first prepared and investigated seven polymer matrices with different resin-to-filler ratios and shortlisted four of these for further investigation under elevated temperature. Subsequently, the most suitable matrix for durability study was determined. Polymer concrete specimens were prepared with four different matrix-to-aggregate ratios to investigate its effect on the mechanical properties from which the optimal mix was identified. Finally, empirical models for strength and stiffness of polymer concrete were proposed and compared with the existing models for normal Portland cement concrete. The outcome of this study will help better understand the properties of epoxy polymer concrete and its component material optimisation.

2. Experimental program

2.1. Materials

The epoxy polymer concrete was prepared using a mixture of resin, fillers and coarse aggregate as described below:

2.1.1. Resin

The resin used in this study was a DGEBA (diglycidyl ether of bisphenol-A) type liquid epoxy resin produced from bisphenol A and epichlorohydrin. It has medium viscosity (110 – 150 poise at 25°C) which helps to disperse the filler and provides a good resistance to settling. It also has good mechanical properties and a high level of chemical resistance in the cured state. The resin has a density of 1.068 g/cm^3 and epoxy molar mass of 190 g, i.e. the amount of resin per gram equivalent of epoxide. For curing, the resin was mixed with an amine based liquid hardener. The amine hydrogen equivalent weight of the hardener was 60 g while the measured density was 1.183 g/cm^3. To make the resin mix reactive, one equivalent weight of resin (190 g) was mixed with one equivalent weight of hardener (60 g). When cross-linked and hardened with curing agents, the desired properties can be obtained.

2.1.2. Fillers

A novel combination of three fillers: fire retardant filler (FRF), hollow microspheres (HM) and fly ash (FA) were used in the preparation of polymer concrete. FRF is a non-toxic, non-corrosive and smoke-suppressant material, and effective fire-retardant due to its thermodynamic properties that absorb heat and release water vapour. This filler was used to help address a limitation of polymer concrete that is its inability to withstand high temperatures [21]. HM are lightweight, hollow, spherical, low density, free-flowing, alumino-silicate powder that is added to reduce weight, shrinkage and cracking, and improve flow and workability. Fly ash is added to improve the performance of epoxy concrete by resisting ultraviolet radiation and reducing the permeability of water and aggressive chemicals due to the fact that spherical and smooth surface of fly ash can reduce the average pore size [1, 22, 23]. The absolute density of FRF, HM and FA were 2.411, 0.752 and 2.006 g/cm^3 while their particle size ranged between 75- 95 µm (surface area 3.4 m^2/g), 20-300 µm and 0.1-30 µm (surface area 4 m^2/g), respectively. The combined action of these fillers is expected to produce a highly durable polymer concrete.

2.1.3. Coarse aggregate

Aggregates used were angular limestone obtained from quarry in crushed form, which were then washed and screened for cleanliness and gradation. The angular shape and rough surface texture of the aggregates creates a strong bond with the epoxy matrix and therefore contribute to higher strength development. The aggregates had a nominal particle size of 5 mm, absolute density of 2.929 g/cm^3 and are free from undesirable impurities that might interfere with the setting and hardening of the epoxy resin matrix. Single-sized coarse aggregate was used because preference is given on specific gravity and the spacing between aggregates is such that it can be easily filled with the epoxy matrix and fillers used in this study.

2.2. Specimen preparation

Casting of polymer concrete was done by three steps. Firstly, the fillers were dry mixed at FRF : HM : FA weight ratio of 100 : 10 : 30. This produced a combined filler density of 1.976 g/cm^3. After several trial mixes, this mixing ratio was found to provide a good balanced combination of fillers to the polymer concrete. The required amount of coarse aggregates were also prepared for the mix. Secondly, the resin and hardener were mixed at resin-to-hardener weight ratio of 100 : 32. This produced a combined density of 1.094 g/cm^3. This ratio is based on the requirement of mixing one equivalent weight of resin (190 g) to one equivalent weight of hardener (60 g) to produce a reactive mix that can maintain its fluidity for around 120 minutes before complete polycondensation [1]. Finally, the mixed filler was added to the resin system and stirred until the matrix became homogeneous. Then, the coarse aggregate was added to the matrix and mixed approximately 5 mins to obtain a fresh polymer concrete. All mixing was done by hand since the volume of each mix was small and easy to handle. An earlier study showed that hand mixed polymer concrete does not require vibration for the manufacture of polymer railway sleepers in order to obtain good compaction and consistent properties [14].

2.2.1. Design of optimal resin-to-filler ratio

To determine the optimal resin-to-filler ratio, different resin-to-filler ratios from 100:0 to 40:60 by volume were prepared. The optimal resin-to-filler ratio was determined based on two criteria (a) aggregate particle distribution in polymer matrix and (b) temperature effect on compressive properties of polymer matrix. Seven mixes with different resin-to-filler ratios were prepared at constant aggregate volume fraction of 30% for investigating the aggregate particle distribution in polymer matrix. These samples were not compacted since the purpose was to check the distribution of coarse aggregates and any compaction would affect their natural distribution. Table 1 provides the seven mix proportions for investigating aggregate distribution where the first two rows (resin + hardener and combined fillers) represent the mix proportions for polymer matrix from which four mixes were shortlisted for investigating temperature effects on compressive properties of polymer matrix. The optimal resin-to-filler ratio can be determined at this stage.

Table 1: Mix proportions for investigating aggregate particle distribution

Sample ID	F_0	F_{10}	F_{20}	F_{30}	F_{40}	F_{50}	F_{60}
Resin-to-filler ratio	100:0	90:10	80:20	70:30	60:40	50:50	40:60
Resin + Hardener (gm)	158	142	126	110	95	79	63
Combined fillers (gm)	0	29	57	86	114	143	171

Aggregates (gm)	181	181	181	181	181	181	181
Density (kg/m^3)	1732	1770	1817	1840	1869	1873	1834

Note: Resin-to-filler ratio (by volume) = (Resin + Hardener) : Filler

It can be seen that the optimisation of the resin-to-filler ratio is based on aggregate particle distribution and thermo-mechanical properties, without any considerations for durability aspects. Therefore, the optimal polymer matrix were further exposed to four different environmental conditions and tested over a period of one year to examine their durability properties.

2.2.2. Design of optimal matrix-to-aggregate ratio

The optimal matrix-to-aggregate ratio were determined based on the effect of aggregate volume fraction on mechanical properties of polymer concrete. To investigate the effect of aggregate volume fraction on mechanical properties, cylindrical (50 mm in diameter and 100 mm in height) and beam specimens (25 × 25 × 250 mm) were cast in plastic moulds and plywood formworks, respectively for compressive, splitting tensile and flexural strength tests. The samples were demoulded next day and cured at room temperature (20°C) at 30% relative humidity and tested after 7 days. Unlike conventional Portland cement concrete, epoxy polymer concrete generally achieves approximately 90% of its 28-day strength in 7 days [24].

Four different matrix-to-aggregate ratios of 1:0, 1:0.45, 1:0.90 and 1:1.35 by weight at a constant resin-to-filler ratio (optimal one) were prepared to investigate their effect on mechanical properties. It should be noted that the resin-to-filler ratio is measured by volume while the matrix-to-aggregate ratio is considered by weight. This is because the use of three different fillers having different densities makes the design by weight basis complicated for resin-to-filler mix. Once the resin-to-filler ratio is finalised, coarse aggregate can be easily added to the matrix by traditional weight based mixing. Many trials involving mixes beyond the selected range of the mixing ratio were also prepared but these were not considered in the reported study because of their low workability checked by visual inspection of entrapped air voids formation [1]. The cylindrical polymer concrete specimens were compacted in three equal layers by rodding each layer uniformly for 25 times. The mix proportions of the materials are provided in Table 2.

Table 2: Mix proportions for investigating the effect of matrix-to-aggregate ratio

Matrix-to-aggregate ratio	1:0	1:0.45	1:0.90	1:1.35
Resin-to-filler ratio	60:40	60:40	60:40	60:40
Resin + Hardener (gm)	1189	971	821	711
Combined fillers (gm)	1431	1169	988	856
Aggregates (gm)	0	971	1642	2132
Volume of aggregates (%)	0	18	31	40

Note: Matrix-to-aggregate ratio (by weight) = (Resin + Hardener + Filler) : Aggregate

2.3. Strength and durability testing

The polymer concrete cylinders prepared for aggregates distribution study were sectioned through the longitudinal direction using wet-cutting diamond blades to observe the spatial distribution of coarse aggregates within the polymer matrix (Fig. 1a). A careful observation of the distribution of aggregates in different resin-to-filler ratios and the performance of polymer matrix at different temperature helps to determine the optimal polymer matrix for further testing in the next stage. Four shortlisted polymer matrices were then prepared, cured for 7 days and tested on small cylindrical samples (25 mm in diameter and 25 mm in height) at room temperature (RT, 20^{o}C), 30^{o}C, 40^{o}C, 60^{o}C and 80^{o}C under compressive load (Fig. 1b).

To ensure durability performance of the optimal polymer matrix, the small cylindrical samples were exposed to air, water, saline solution, and hygrothermal environmental conditions (Fig. 1c and 1d). Air exposure with 20^{o}C and 30% humidity was taken as the control environment. Water exposure was carried out by immersing specimens in tap water at room temperature in a glass container with lid (to prevent evaporation). Exposure to saline solution was carried out in the same manner, but using 3.5% sodium chloride (by weight) solution to mimic seawater salinity. To simulate the common hygrothermal environment, specimens were placed in a water bath filled with tap water at constant 40°C temperature. The specimens exposed to air, water and saline solution were tested under compression over a period of one year, specifically at 7-day, 1-month, 2-month, 4-month, 6-month and 1-year while the hygrothermal samples were tested at 1-day, 3-day, 7-day and 1-month due to limited facilities.

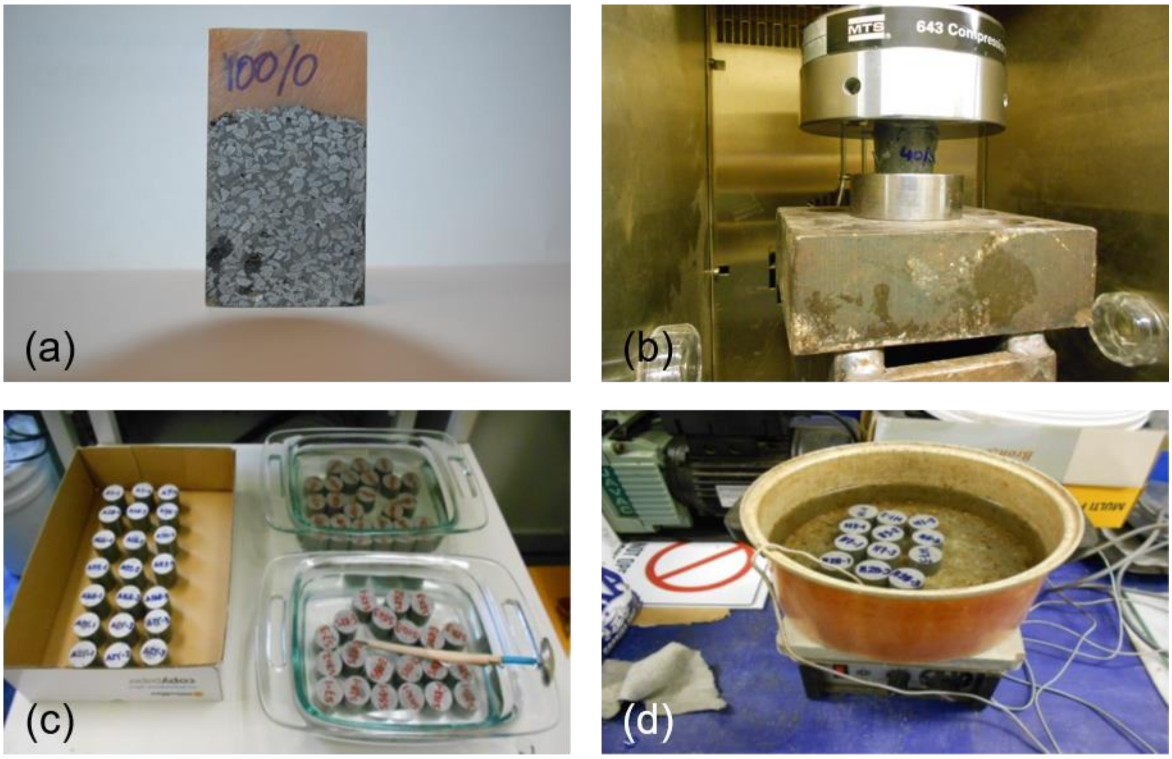


Fig. 1: Methods for determining optimal resin-to-filler ratio and durability study on optimal matrix: (a) aggregates distribution along height, (b) compression testing under elevated temperature, (c) conditioning of optimal matrix in air, water and saline solution, and (d) optimal matrix in hygrothermal condition.

The concrete prepared with optimal polymer matrix were tested under compression (Fig. 2a), splitting tension (Fig. 2b) and flexural (Fig. 2c) loading conditions according to ASTM

C39 [25], ASTM C496 [26] and ASTM C293 [27] standards respectively, to determine the compressive strength and modulus of elasticity, splitting tensile strength, and flexural strength. The nominal dimension of the concrete cylinder was 50 mm diameter and 100 mm in height while the beam specimen was 25 × 25 × 250 mm and tested at 200 mm span. Three replicates for each specimen type and property were tested and averaged. Prior to testing, the height and diameter of each cylinder were measured for strength calculation and confirming the dimensions do not differ by more than 2 % as per requirements of ASTM C39. The load was applied until the load indicator shows a decreasing trend and the specimen displayed a well-defined fracture pattern. The splitting tensile strength and flexural strength were determined by the relationship of $f_{ct} = 2P/\pi dL$ and $f_{cf} = 3PL/2bd^2$, where, P, L, b, d, f_{ct} and f_{cf} are the maximum applied load, cylinder length or span length, width of the beam, diameter of cylinder or depth of the beam, splitting tensile strength and flexural strength, respectively.

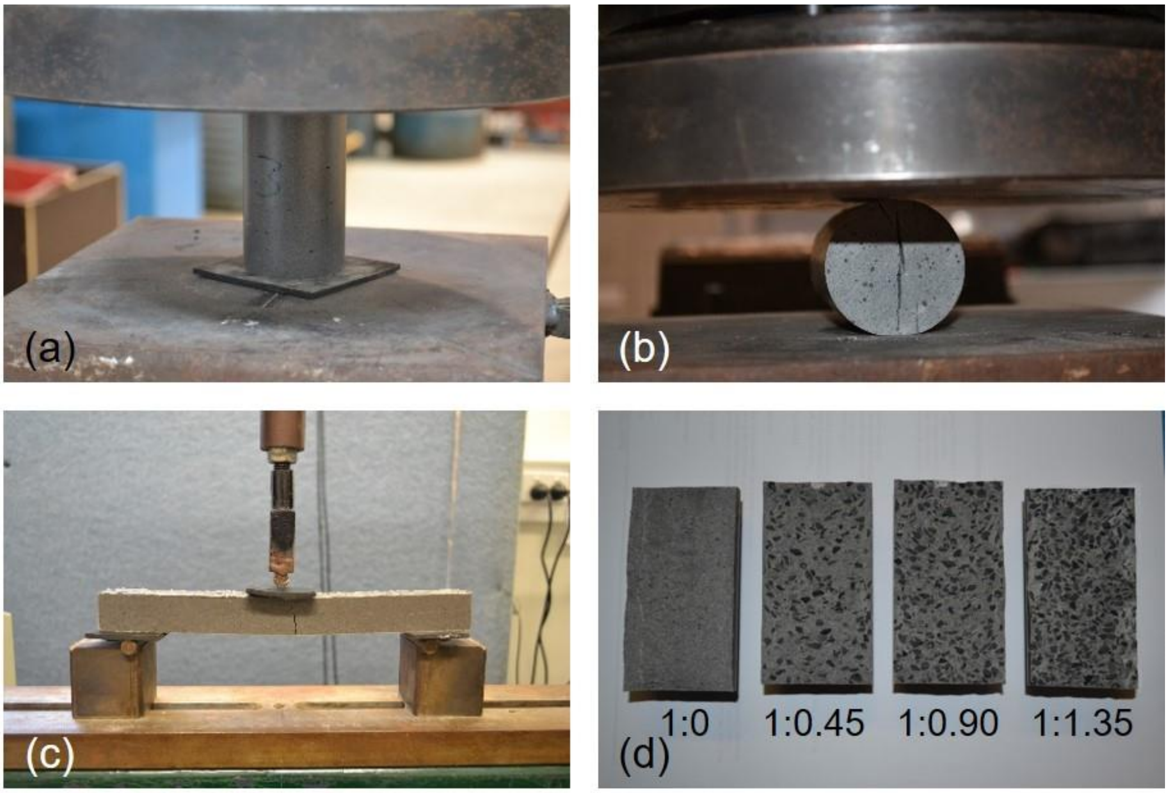
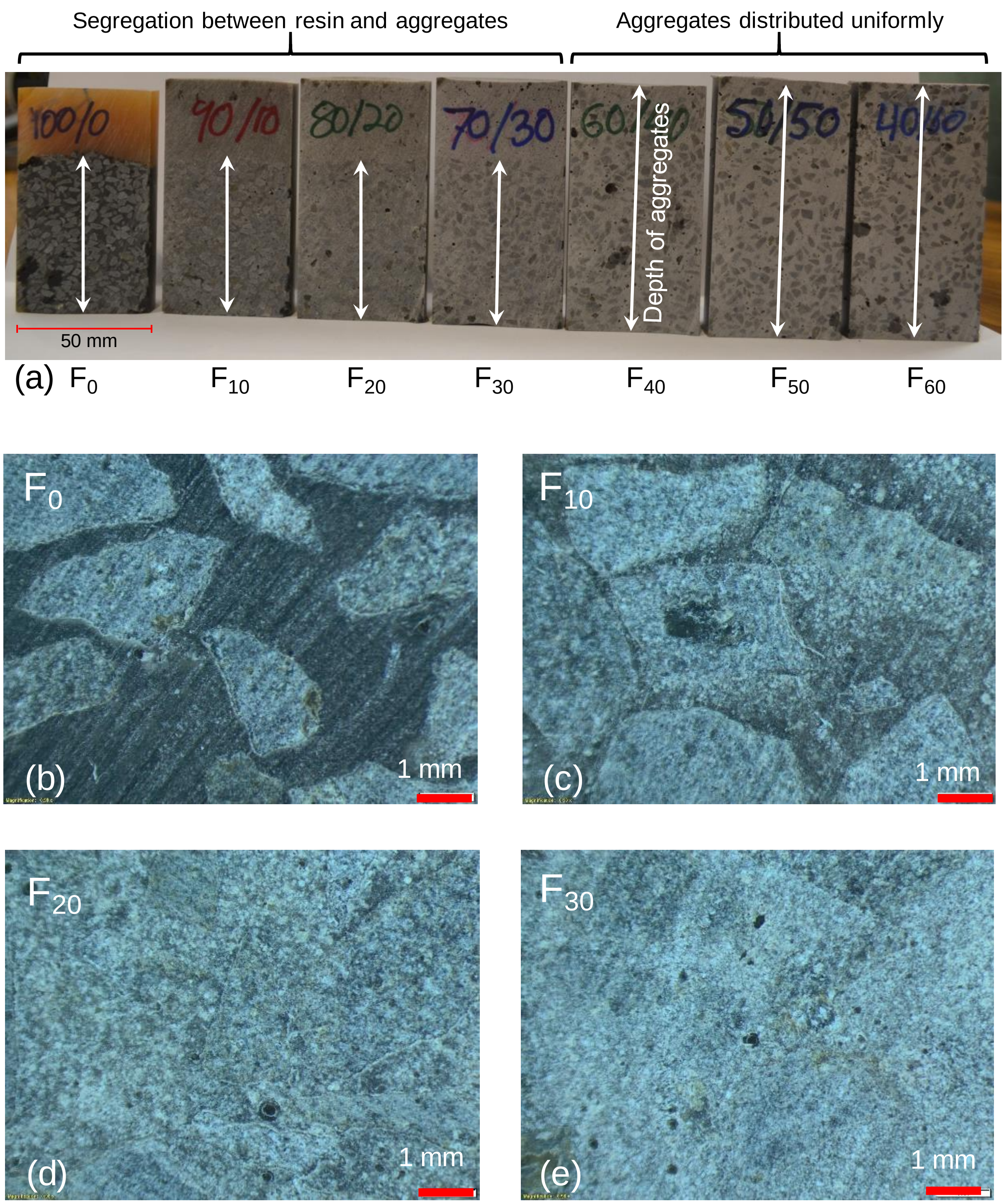


Fig. 2: Strength testing to determine optimal matrix-to-aggregate ratio: (a) compression, (b) splitting tension (c) flexure, and (d) distribution of aggregates in the optimal matrix

3. Results and discussion

3.1. Effect of resin-to-filler ratio on aggregates distribution

Resin binds the aggregates together and gives the polymer concrete its strength. Polymer concrete with low resin content results in a brittle product and is normally very dry and difficult to work with. The flowability of the concrete greatly depends on the resin-to-filler ratio. Fig. 3(a) shows the distribution of aggregates in polymer concrete composed of different resin-to-filler ratios starting from 100:0 decreasing to 90:10, 80:20, 70:30, 60:40, 50:50 and 40:60 denoted by F_0, F_{10}, F_{20}, F_{30}, F_{40}, F_{50} and F_{60}, respectively. In contrast to the traditional concept, this study applied a new approach of selecting aggregates based on their specific gravity (SG) rather than their size. The coarse aggregates are heavier (SG = 2.929) and hollow microspheres are lighter (SG = 0.752) than resin systems (SG = 1.096). The new approach results in mix formulations with excellent flowability. The use of high resin-to-filler ratio (F_0 to F_{30}) produces a light and flowable matrix, which is less capable of keeping the denser aggregates in suspension prior to setting. At filler content of 40% and above, uniform distribution of the coarse aggregates throughout the full depth of the concrete is achieved and no distinct separation between the aggregates and matrix can be observed. At high filler content (F_{40} to F_{60}), the resin matrix was less flowable and settlement of aggregates did not occur. However, the fillers were distributed uniformly in the concrete for all resin-to-filler ratios. This is due to the small particle size and the use of low density HM filler that help to stay in suspension within the resin.


Segregation between resin and aggregates
Aggregates distributed uniformly
100/0
90/10
80/20
70/30
Depth of aggregates
50 mm
(a)
F0
F10
F20
F30
F40
F50
F60
(b)
(c)
(d)
(e)
1 mm

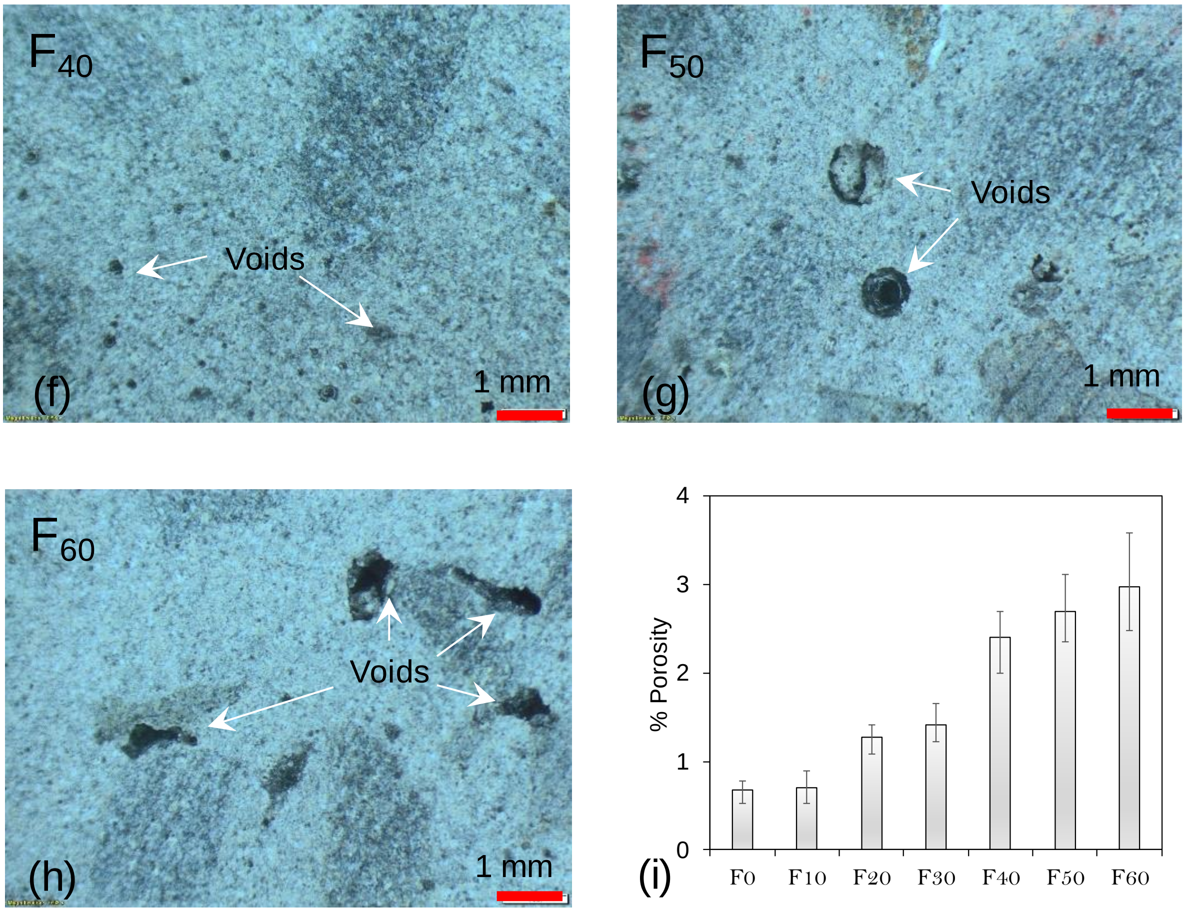


Fig. 3: (a) Polymer concrete with different resin-to-filler ratio showing the distribution of coarse aggregates and epoxy matrix, (b – h) microscopic observation for voids in the samples, and (i) total porosity.

Figs. 3(b) to 3(h) shows the microscopic observation of the specimens from Fig. 3(a). One important aspect noted in Figs. 3(b) to 3(h) is the presence of air voids in the less flowable matrices (i.e., from F_{40} to F_{60}). The number and size of these air voids increased with the decrease in resin-to-filler ratios. These are air bubbles were entrapped during concrete mixing and not completely removed because the samples were prepared without any compaction as explained in Section 2.2.1. However, mixes with high resin-to-filler ratios (i.e., from F_0 to F_{30}) entrapped less air due to their good flowability.

Density increased gradually from 1732 to 1869 kg/m^3 with increase in filler from F_0 to F_{40} and then remained fairly consistent at F_{50} (1873 kg/m^3) and F_{60} (1834 kg/m^3) as shown in

Table 1. The increase in density is due to the higher specific gravity of the combined fillers (1.976) compared to resin (1.096). However, the slight decrease in density at F_{60} is mainly due to the formation of large voids. The average void size and total porosity were analysed using "TBitmap" software on the microscopic images. It was observed that the average diameter of the voids gradually increased from 265 to 560 µm and the porosity (Fig. 3i) increased from 0.7% to 3% with increase in filler from 0% to 60%. Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX) have been carried out in a previous study [1] by the authors and not repeated in the present study as the type of resin and filler used are the same. Moreover, the SEM analysis showed that fracture occurs through the filler [1] indicating a good bond between resin and fillers.

A careful inspection of the specimens in Fig. 3(a) shows that the colour of the matrix changes from orange (F_0 mix) to grey (F_{60} mix) with the decrease in resin-to-filler ratio. This can be attributed to the dark grey colour of fly ash and its increasing content in the matrix with decrease in resin-to-filler ratio. The darkness of the matrix could block ultraviolet radiation and protect the concrete from physical and mechanical deterioration due to photo-oxidative reactions that alter its chemical structure [1]. However, the hardened F_0 mix achieved a smooth exterior surface and surface roughness increased with decrease in resin-to-filler ratio. Surface smoothness is particularly important for decorative works, so there are advantages and disadvantages of decreasing resin-to-filler ratio. As such, four mixes from low to high filler content were shortlisted (F_0, F_{20}, F_{40} and F_{60} mix) for investigating the effect of temperature.

3.2. Effect of temperature and resin-to-filler ratio on stress-strain behaviour

An in-depth understanding of temperature effects on mechanical properties is important for the design of polymer concrete for outdoor applications. The compressive stress-strain behaviour of the four shortlisted matrix are plotted in Figs. 4(a) to 4(d) while the variations of the strength and elastic modulus are illustrated in Figs. 4(e) and 4(f), respectively. The stress-strain plot indicates that the behaviour of specimens heated at 60^{o}C or above are very different to those heated up to 40^{o}C. At 60^{o}C or above, the specimens deformed drastically and showed significant drop in strength and stiffness. At 40^{o}C, the strength and modulus of elasticity retained up to 50% of the values at room temperature while the retention is only approximately 10% at 60^{o}C. This is because of the lost of internal resistance at 60^{o}C which is the glass transition temperature of the polymer matrix as determined by the authors in a previous study [1]. At glass transition temperature, the specimen changes from a hard, rigid or glassy state to a softer, compliant or rubbery state [28].

A general observation is that a lower resin-to-filler ratio achieved a slightly lower strength, but higher elastic modulus at the same temperature. Furthermore, the stress-strain curves show that the ductility of the polymer decreases significantly with decrease in resin-to-filler ratio. This is due to lowering of the bonding capability between resin and filler on which the strength of the matrix is dependant. On the other hand, the higher modulus of filler compared to resin increases the overall stiffness properties with the decrease of resin-to-filler ratio. Moreover, Figs. 4(e) and 4(f) indicate that the reductions in mechanical properties between 20^{o}C and 80^{o}C are less for polymers containing high amount of fillers. This phenomenon can be attributed to the heat absorption capacity of fillers, therefore the higher the fillers content, the higher the heat resistance and lower the negative effect of temperature on strength and elastic modulus.

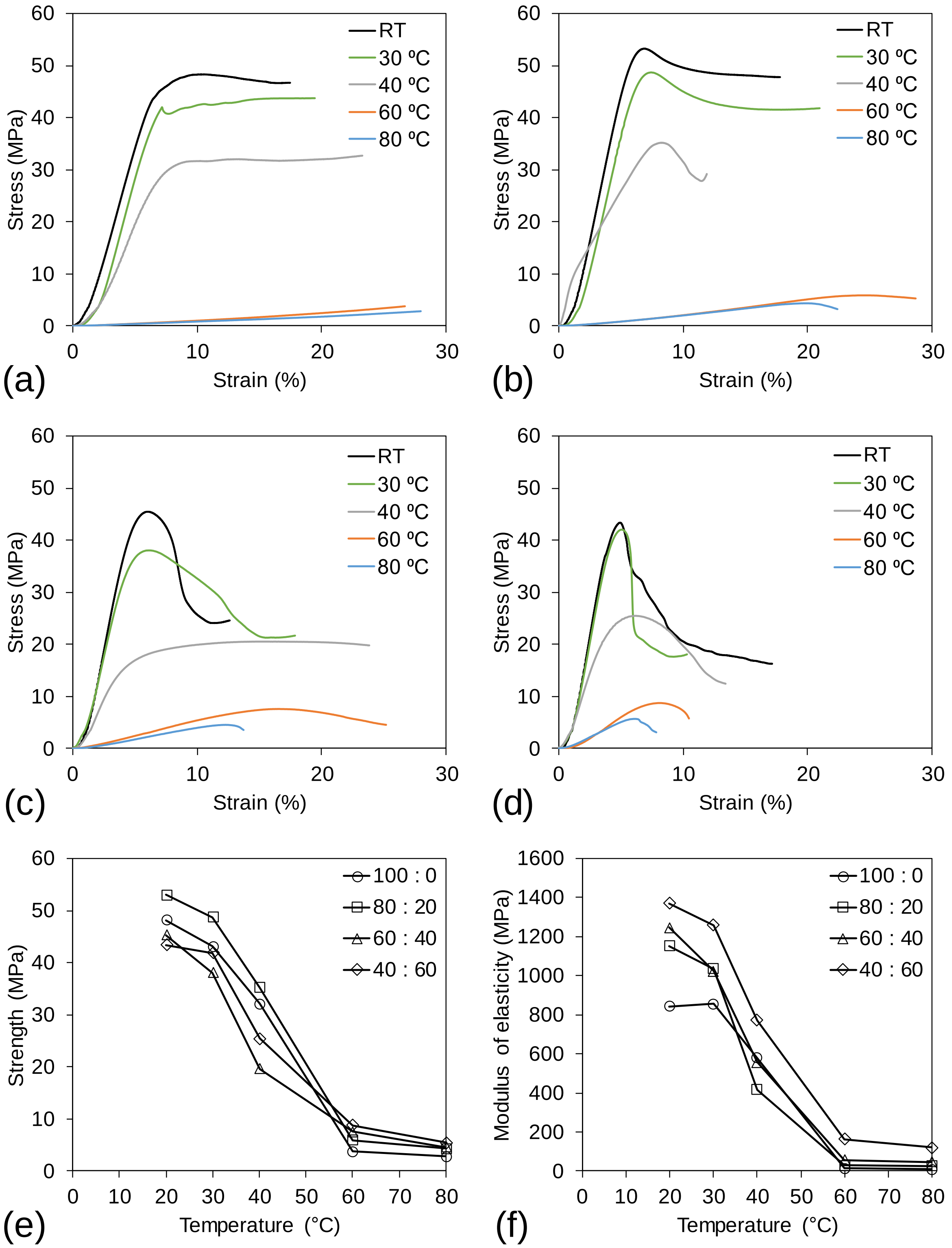


Fig. 4: Effect of temperature on compressive stress-strain behaviour of samples with resin-to-filler ratio (a) 100:0, (b) 80:20, (c) 60:40 and (d) 40:60. Figure (e, f) shows decrease in strength and elastic modulus at elevated temperature.

Previous study by the authors [1] on the properties of polymer matrix (without aggregates) found an optimal resin-to-filler ratio of 70:30 on the basis of mechanical properties. However, the current study shows that this mix is not capable of achieving a uniform distribution of aggregates and thus inappropriate for concrete. Therefore, the 70:30 mix would not be suitable for investigating the effect of matrix-to-aggregate ratio. Moreover, the detrimental effect of temperature on mechanical properties decreases with the increase in fillers content. The results from this study suggests that the 60:40 resin-to-filler ratio is a more appropriate matrix and so this will be used to prepare polymer concretes with different aggregate contents for strength and durability testing.

3.3. Effect of environmental conditions on strength and absorption

The effects of exposure to air, water, saline solution and hygrothermal conditions on the stress-strain behaviour, strength and absorption properties of the optimal polymer matrix (60:40) are shown in Fig. 5. The data were recorded up to 1 year in air, water and saline conditions and up to 1 month in hygrothermal condition after taking the initial reading on 7 days cured (20ºC, 30% relative humidity) specimens. Fig. 5 (a-d) shows that the exposure type and duration induced a small effect on the initial slope of the stress-strain curve and a much more noticeable effect on strength. Unlike Portland cement-based matrix, the polymer matrix showed a significant amount of plasticity beyond peak stress, and therefore a less brittle failure.

Fig. 5 (e) plots the variation in compressive strength for different exposure conditions and times. The increase in strength with time is expected for air exposure. However, it is interesting to see that strength also increased when the specimens were exposed to water, saline solution and hygrothermal environments. Strength increased by up to 33%, 26% and 25% for air, water and saline conditions respectively, during the first 4-month period and then no significant changes were noticed thereafter. The rate of strength increase was slightly higher in hygrothermal condition for the measured period and this suggests that the combination of water and heat curing is beneficial.

Fig. 5 (f) shows the effect of exposure condition on the specimen weight over time. It can be seen that weight increased in all environmental conditions, but at different rates. After 1 year of exposure, the largest increase occurred in water (0.45%), followed by saline solution (0.42%) and air (0.13%). This shows that the polymer matrix can absorb a small amount of water in a wet environment. The slight reduction in weight gain in saline environment is probably due to salt deposition on the surface. Samples in air achieved the lowest weight increase which is expected. In contrast, samples in hygrothermal condition absorbed the most water compared to all other environments (after 1 month exposure) because the absorption process is accelerated at elevated temperature. In any case, the percentage of water absorption for polymer matrix (up to 0.45%) is significantly lower when compare to the absorption capacity of ordinary Portland cement-based grouts which can be up to 30% [29].

The strength development and water absorption results in different environmental conditions suggest that the polymer matrix has excellent durability against these aggressive environments. After selecting the suitable resin-to-filler ratio of 60:40 and assessing the durability aspects of the selected matrix, the next section investigates the effect of matrix-to-aggregate ratio to obtain an optimal mix proportion for polymer concrete.

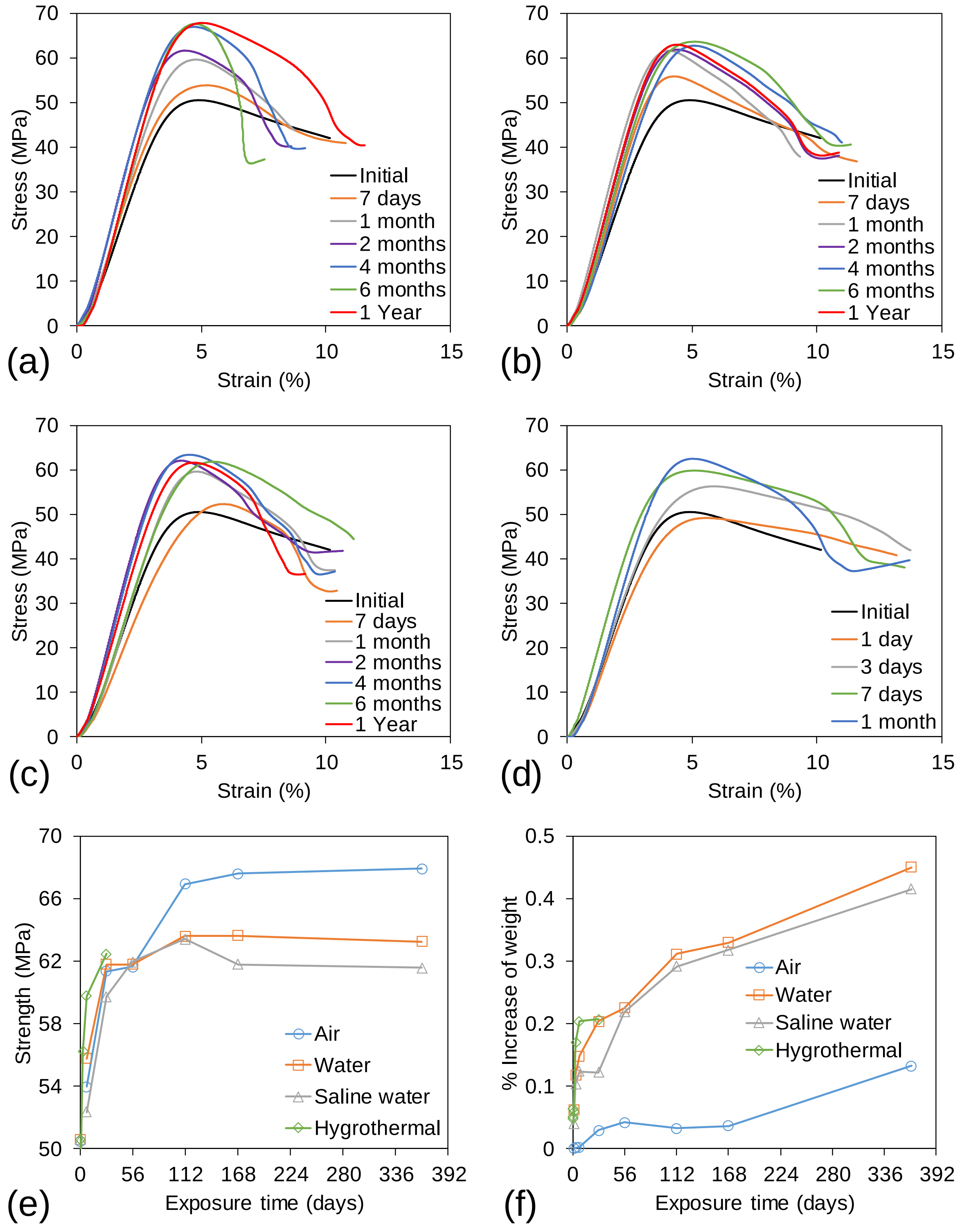


Fig. 5: Compressive stress-strain behaviour of the optimal polymer matrix (60:40 resin-to-filler ratio) after up to 1 year exposure in (a) air, (b) water, (c) saline solution and (d) hygrothermal conditions. Figure (e, f) show the effect of exposure on strength and absorption properties.

3.4. Effect of matrix-to-aggregate ratio on mechanical properties

The effects of matrix-to-aggregate ratio on the mechanical properties of polymer concrete are presented in Fig. 6(a) to Fig. 6(d). The compressive stress-strain behaviour in Fig. 6(a) shows that for the same stress level, strain decreases with increase in aggregate fraction. Therefore, the corresponding decrease in the epoxy matrix fraction makes the concrete stiffer. This effect was also observed in the post peak behaviour. The failure process of the mix without aggregate (1:0 mix) is much more ductile and shows a greater level of plasticity. The peak stress of the 1:0 mix occurred at 0.035 mm/mm strain while the ultimate failure strain was 0.072 mm/mm. In contrast, the post peak behaviour of the mixes with aggregates (i.e., 1:0.45, 1:0.90 and 1:1.35 mixes) is relatively more brittle with peak stress occurring around 0.025 mm/mm strain and ultimate failure strain around 0.035 mm/mm which decreases slightly with the increase of aggregates. Therefore, ductility decreases with the increase in aggregate content.

Fig. 6(b) shows the variation of compressive strength and compressive modulus of elasticity with matrix-to-aggregate ratio. With the exception of the 1:0 mix, compressive strength slightly increases with the decrease in matrix-to-aggregate ratio. The higher strength of 1:0 mix (42.3 MPa) compared to 1:0.45 mix (34.1 MPa) can be attributed to its uniform stress distribution along the depth of cylinder. In contrast, the 1:0.45 mix would experience non-uniform stress distribution due to the presence of stiff aggregates and high stress concentration at the aggregate-matrix interface, which can cause early failure of the specimen. However, when comparison is made between mixes with aggregates (i.e., from 1:0.45 to 1:1.35), the slightly increasing trend of compressive strength (i.e., from 34.1 MPa to 39.9 MPa) is due to the gradual increase of aggregate volume in the mix that has higher crushing strength (30 to100 MPa) than the matrix. The slope of the stress-strain curve represents the modulus of elasticity and this increases from 1.86 GPa to 2.26 GPa with decrease in matrix-to-aggregate ratios from 1:0 to 1:1.35. This is due to the fact that the aggregate has higher elastic modulus (15 to 55 GPa) than the polymer matrix. As such, the elastic modulus of the polymer concrete increases with increase in compressive strength.

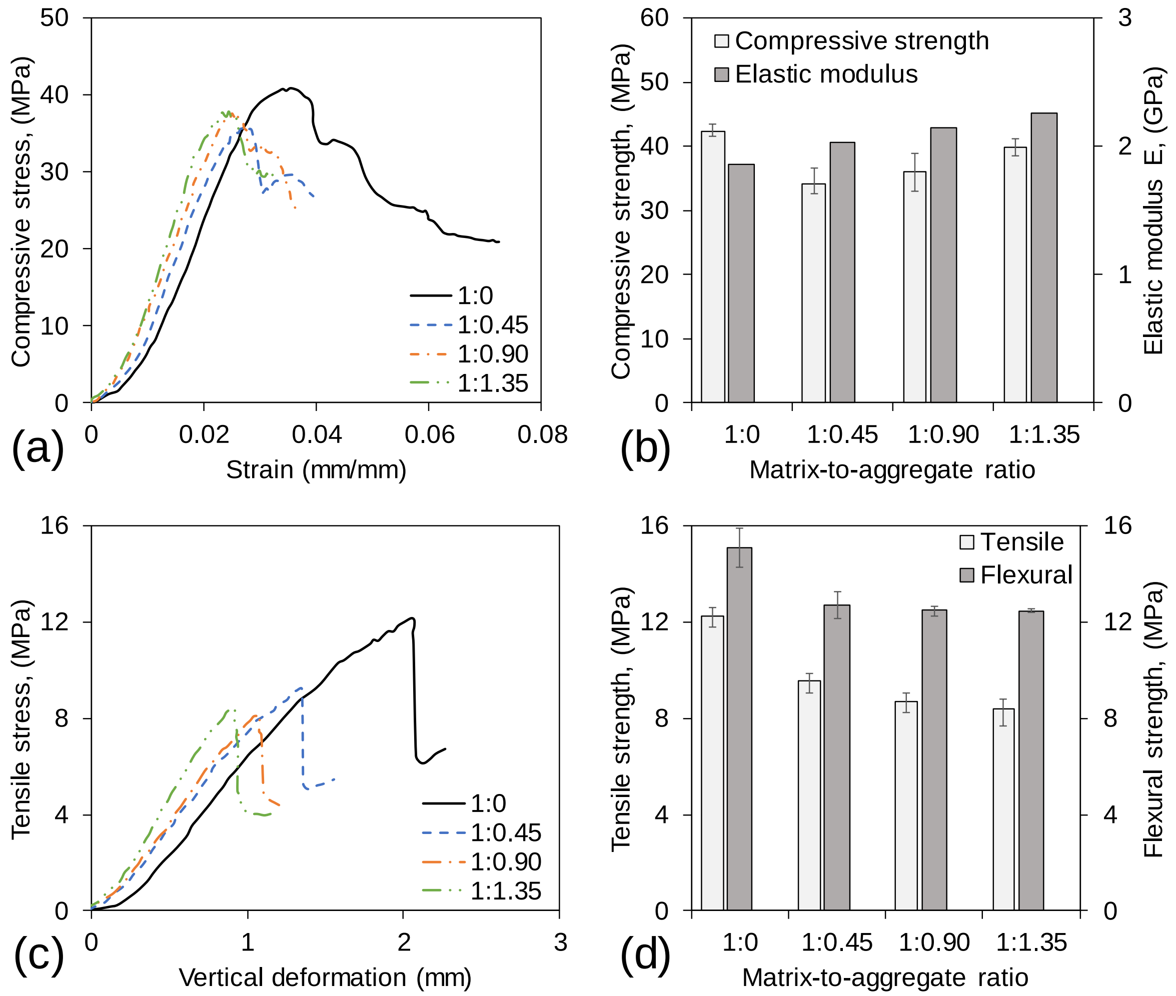


Fig. 6: Effect of matrix-to-aggregate ratio on mechanical properties (a) Compressive stress-strain behaviour, (b) Compressive strength & elastic modulus, (c) Tensile stress-deformation behaviour and (d) Splitting tensile and flexural strength

Fig. 6(c) shows the tensile stress-deformation behaviour of polymer concrete for different matrix-to-aggregate ratios. Similar to the behaviour in compression, deformation at the same load level increases with the increase in matrix-to-aggregate ratio. However, one critical difference is that the mode of failure in tension is much more brittle compared to the failure mode in compression. Beyond the ultimate tensile load, a significant drop in load (~

50%) occurred suddenly followed shortly by ultimate failure as shown in Fig. 6(c). This behaviour was observed in all samples.

Fig. 6(d) shows the splitting tensile and flexural strengths with variation in matrix-to-aggregate ratio. The splitting tensile strength ranged from 8.4 to 12.3 MPa, while the flexural strengths ranged from 12.5 to 15.1 MPa. It can be seen that both the splitting tensile and flexural strengths slightly decreased with decrease in matrix-to-aggregate ratio. This is due to the decrease of resin content that binds the aggregate together and the concrete containing a lower percent of resin resulted in a lower tensile and flexural strength as evident from [1, 11]. It can be noted that the flexural strength is approximately 35% higher than the splitting tensile strength for the same matrix-to-aggregate ratio. This is perhaps due to the assumption of linear elastic behaviour of the flexural specimens until failure (i.e., $f_{cf} = 3PL/2bd^2$) which provides slightly higher flexural strength than the actual. The higher flexural strength may also be attributed to the differences in failure modes of splitting tensile and flexural test. The flexural failure occurs on a small area of the bending section of beam due to the high compressive resistance above neutral axis (Fig. 2c) whereas in the splitting tensile test, the entire longitudinal section of the cylinder is under maximum stress (Fig. 2b). Therefore, it is more likely to find a weak point in the splitting section from which cracking initiates and propagates. This could explain why splitting tensile strength is lower than the flexural strength.

4. Empirical modelling

The lack of information on predicting the behaviour of polymer concrete motivated this study to develop empirical relationships between compressive strength, elastic modulus, tensile strength and flexural strength. To increase the reliability of the proposed model, this study also considered the data from published research. These relationships are compared to those for conventional Portland cement concrete.

4.1. Elastic modulus

Elastic modulus, the slope of the initial linear portion of the stress-strain curve, generally increase with the strength of concrete. The two main parameters that affect elastic modulus of concrete are density and compressive strength [30]. The unit weight of concrete is an important parameter for estimating elastic modulus particularly in case of low density concrete. Theoretically, the density of polymer concrete (γ) can be estimated using the density of its constituent ingredients as provided in Eq. (1). The ratio of w/γ represents the percentage weight of each ingredient in the concrete mix to the density of the corresponding ingredient such as resin, hardener, FRF, HM, FA and aggregate. The effect of density on the stiffness of concrete is high for low strength concrete [30].

$$\gamma = \frac{100}{\sum \frac{w_i}{\gamma_i}} \quad (1)$$

The American Concrete Institute (ACI) recommends an empirical equation to estimate modulus of elasticity for normal strength conventional concrete from its compressive strength. Fig. 7 plots the modulus of elasticity with respect to compressive strength according to the ACI code and experimental results. It can be seen that the elastic modulus of the polymer concrete does not follow ACI code [31], and in fact it is substantially lower than the conventional concrete. The more appropriate model for resin based polymer concrete is expressed in Eq. (2), where both the modulus of elasticity (E) and compressive strength of concrete (f_c) are expressed in MPa.

$$E = 530\sqrt{f_c} \quad (2)$$

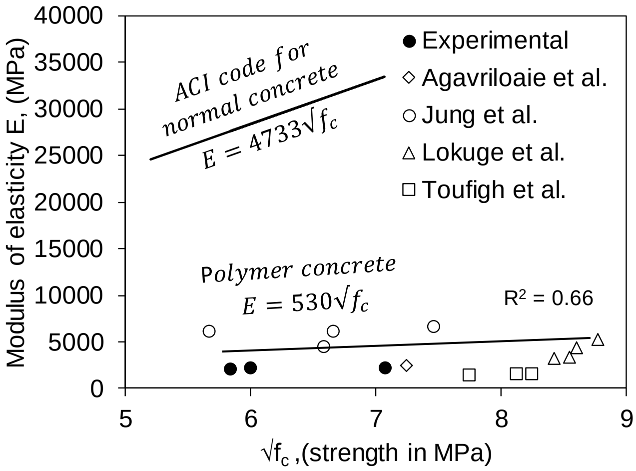


Fig. 7: Relationship between modulus of elasticity and compressive strength for conventional concrete and polymer concrete [3, 5, 11, 32].

At the same compressive strength, the elastic modulus of polymer concrete is significantly lower than the normal concrete. This is probably due to the lack of coarse aggregates, low sand fraction and low elastic modules of the epoxy matrix in polymer concrete relative to cement-based matrix. Nevertheless, a flexible concrete is desirable when it is used for the purpose of binding and coating material for structural load carrying components. For example, the recent development in polymer railway sleeper manufactured from composite sandwich panels (load carrying components) bonded and coated with polymer concrete (provides structural integrity) requires a flexible concrete material to ensure the failure in main structural load carrying components under bending load [14, 33, 34].

4.2. Splitting tensile strength

Splitting tensile strength is an important parameter to evaluate the shear resistance provided by concrete and to determine the development length of reinforcement. The splitting tensile strength is generally greater than direct tensile strength. The Australian standard of concrete structures AS 3600 [35] proposed that the splitting tensile strength is 40% of the square root of compressive strength. Fig. 8 plots the AS 3600 model and the experimental splitting tensile strength, against compressive strength. The tensile strength of polymer concrete increases with the increase of compressive strength. For same compressive strength, it can be seen that tensile strength of the polymer concrete is 2.25 times higher than the conventional Portland cement concrete. This suggests a stronger bond between polymer matrix and aggregates compared with cement-based matrix. This also suggests that there is less inherent flaws within the polymer matrix that would propagate and contribute to failure under tension. The higher tensile strength of polymer concrete makes it a potentially viable material for many civil engineering applications. The relationship between tensile (f_{ct}) and compressive (f_c) strength of the polymer concrete can be expressed by Eq. (3).

$$f_{ct} = 0.9\sqrt{f_c} \quad (3)$$

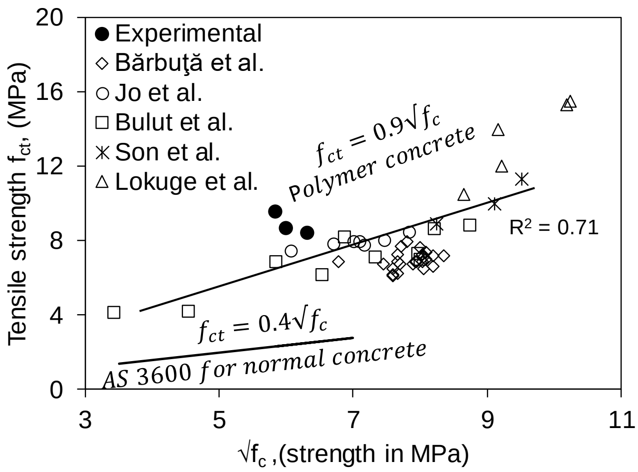


Fig. 8: Tensile and compressive strength relationship [11, 19, 36-38]

4.3. Flexural strength

Flexural strength measures the capacity of concrete to resist failure in bending. The flexural and tensile properties are correlated. Fig. 9 plotted the AS 3600 code for normal Portland cement concrete and the flexural test results from polymer concrete. Similar to tensile strength properties, the flexural strength of polymer concrete increases with increase in compressive strength. The AS 3600 code proposed that flexural strength for conventional concrete is 60%

of the square root of compressive strength, but this does not capture the behaviour of polymer concrete and significantly underestimates its flexural strength. Therefore, a suitable correlation between flexural and compressive strength needs to be developed for polymer concrete. The relationship between flexural strength (f_{cf}) and corresponding compressive (f_c) strength of the polymer concrete can be expressed by Eq. (4). This suggest that the flexural strength of polymer concrete is approximately three times higher than the normal concrete with same compressive strength.

$$f_{cf} = 1.9\sqrt{f_c} \quad (4)$$

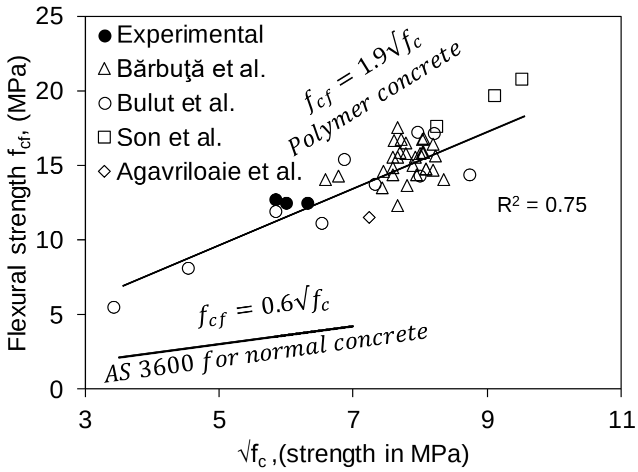


Fig. 9: Flexural and compressive strength relationship [5, 19, 37, 38]

From the analyses presented in this section, it is clear that existing empirical models developed for conventional Portland cement concrete are not suitable for predicting the behaviour of polymer concrete. It is discussed earlier that the polymer concrete undergoes a polycondensation reaction to attain structural strength. The composition of the hardened polymer matrix is not the same as cement based concrete. The regression analysis of the proposed models for modulus, tensile and flexural strength gave R^2 values of 0.66, 0.71 and 0.75, respectively which is shows strong correlation compared with other advance modelling such as artificial neural network approach for normal concrete [39]. However, the simplified empirical relationship to estimate elastic modulus, splitting tensile and flexural strength could be improved further by establishing a material design process such as micromechanical models that bridge microscale to mesoscale. This is beyond the current scope of the study but recommended for future work. It should be noted that the proposed relationships for polymer concrete are based on the results from a normal strength concrete. Therefore, further investigation on high strength concrete with different types of resin and aggregate sizes or gradation need to be conducted to verify the reliability of the proposed models.

5. Optimal design for polymer concrete

The term optimal design refers to the effective use of resin-to-filler ratio and matrix-to-aggregate ratio that achieves the desired physical and mechanical properties of the polymer concrete. This study has formulated two types of polymer concrete: (a) concrete with uniformly distributed aggregates and (b) concrete separated by resin rich and aggregate rich layers as shown in Fig. 3(a). Typically, concrete with uniformly distributed aggregates is preferred for structural elements where the main purpose is to carry loads, while concrete showing high degree of segregation is not desirable. However, recent studies suggest that a composite with resin rich and aggregate rich layers is advantageous for the purpose of bonding and coating structural components. For example, a recently developed polymer railway sleeper was manufactured from sandwich panels where the panels were bonded together using layer-based polymer concrete to achieve higher compressive strength at the top half and greater tensile strength at the bottom half of sleeper [14]. Therefore, it is required to optimise both types of concrete for their effective utilisation in civil construction.

The formation of resin rich and aggregate rich layers in concrete are dependent on the resin-to-filler ratios. The resin-to-filler ratios from 100:0 to 70:30 (from F_0 to F_{30}) can produce a layered concrete while ratios between 60:40 and 40:60 (from F_{40} to F_{60}) produce a more homogeneous material. The results of this study suggest that there are no major differences in flowability and void formation between resin-to-filler ratios from 100:0 to 70:30 (Fig. 3).

However, the increase in filler content produces a less transparent matrix that would be more effective in blocking ultraviolet radiation. In addition, there is also a cost advantage since resin is the most expensive component in polymer concrete as discussed in [1]. Therefore, the optimal resin-to-filler ratio would be 70:30 (i.e., F_{30}) to produce a layered concrete. On the other hand, the concrete with uniformly distributed aggregate (from F_{40} to F_{60} mix) contains voids as a result of their low workability (Fig. 3). The void content increased with the decrease in resin-to-filler ratio. A high percentage of void can create a porous microstructure that may allow unwanted liquids and gases into the concrete. Thus, the optimal resin-to-filler ratio would be 60:40 (i.e., F_{40}) to achieve a well compacted durable polymer concrete with uniform distribution of aggregates.

The matrix-to-aggregate ratio has an influence on the mechanical properties of polymer concrete. The tensile strength decreased by 22%, 29% and 32% and flexural strength decreased by 15.8%, 17% and 17.3% with the increase of coarse aggregate by 1, 2 and 3 times of the resin, respectively. It can be seen that there are no major differences in the variation of strength even when the aggregates are increased by 3 times of the resin (i.e., 1:1.35 mix). However, mixes with much higher aggregate contents were not considered for investigation because of their low workability. The major challenges associated with the use of polymer concrete are their high cost, odour, toxicity and flammability due to the use of resin [40]. It can be expected that decreasing the matrix-to-aggregate ratio could mitigate some of these challenges, e.g. lowering the cost [1], odour and toxicity, and improve fire resistance by reducing resin content per unit volume of concrete. However, further investigation is needed to verify this. Based on the results from this study, the optimal matrix-to-aggregate ratio is 1:1.35 to achieve a good balance between cost, durability and mechanical properties.

6. Conclusions

Epoxy polymer concrete with different resin-to-filler ratios and matrix-to-aggregate ratios were investigated by physical observation, mechanical and durability testing. Empirical models were proposed to predict the behaviour of polymer concrete. The optimal resin-to-filler ratio and matrix-to-aggregate ratio were determined from which the following conclusions are drawn:

- The distribution of aggregates within the concrete is heavily dependent on the resin-to-filler ratio of the mix. Mixes with low filler content (< 40%) show significant segregation and produces a layered polymer concrete with resin rich layer at the top and aggregate rich layer at the bottom. In contrast, a uniform distribution of aggregates was achieved throughout the depth of concrete when the polymer matrix contained at least 40% filler (60% or less resin). This was due to reduction of flowability of the epoxy matrix.
- The higher the fillers in the matrix the lower the negative effect of temperature due to the heat absorption capacity of fillers and consequently the lower the loss of strength. Epoxy-based polymer matrix shows excellent durability against air, water, saline solution and hygrothermal environments.
- The mechanical properties of the polymer concrete are influenced by the matrix-to-aggregate ratio. A decrease in matrix-to-aggregate ratio decreases the tensile strength, flexural strength and ductility. This is because the tensile and flexural properties, and ductility are dependent on resin content in the concrete.
- The tensile strength of polymer concrete is more than 2 times higher than conventional Portland cement concrete because of the better bonding characteristics between the matrix and aggregates. Flexural strength of polymer concrete is about 35% higher than its splitting tensile strength.
- Existing empirical models for elastic modulus, tensile strength and flexural strength that were developed for conventional Portland cement concrete are not applicable to epoxy

polymer concrete. New models are proposed for elastic modulus, tensile strength and flexural strength of polymer concrete.

- The optimal resin-to-filler ratio is 70:30 to achieve a layered composite material and 60:40 to achieve a homogenous material with uniform distribution of aggregates. Furthermore, the optimal matrix-to-aggregate ratio is 1:1.35 to ensure a good balance between performance and cost.

A careful selection of resin-to-filler ratio and matrix-to-aggregate ratio in the mix design can mitigate some of the limitations of epoxy polymer concrete such as cost, odour and toxicity. The unique combination of fire-retardant filler, hollow microsphere and fly ash may able to improve fire resistance, minimise shrinkage, control crack propagation and improve durability. However, further investigations are required on these areas to fully understand their effects and to increase confidence in their usage.

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