# EFFECT OF MONTMORILLONITE ON STRENGTH, EXPANSIVE COEFFICIENT AND SETTING SHRINKAGE OF RECYCLED UNSATURATED POLYESTER CONCRETE

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## Abstract

The effects of resin (binder), filler content, and montmorillonite contents on the strength, expansive coefficient, and linear setting shrinkage of a recycled unsaturated polyester resin-based polymer concrete are reported. In this study, heavy calcium carbonate was used as the filler. An aggregate grading, which resulted in the least void content, was used. Also, montmorillonite was used to achieve low shrinkage. The resin content was varied between 10 and 15 percent, and the filler content between 3 and 11 percent. Montmorillonite content was varied between 0 and 8 percent. The strength of polymer concrete increased with resin content initially, but decreased beyond a particular level of resin content. A fairly one-to-one correspondence was observed between the compressive strength and coefficient of thermal expansion of the PC. The shrinkage also increased linearly with resin content. It is also increased with filler and montmorillonite on the shrinkage is discussed. The practical significance of PC with and without montmorillonite was identified.

## 1. Introduction

Time-dependent behavior of polyester-based materials such as polymer mortar or polymer concrete is recognized as an important aspect of mix design. Typically, creep and shrinkage of polyester-based materials exhibit complex time-dependent behavior. Thus, such time-dependent behavior is considered an essential design factor of the safety and serviceability of precast structures, especially, during construction and at early age of curing. In the construction industry, the early age properties of polyester-based materials have become increasingly important because, the use of rapidly cured materials can accelerate a construction process or shorten the production cycle of precast members [8].

The early age material properties of polyester-based materials, as influenced by setting shrinkage, have been investigated theoretically and experimentally. In some cases, the extent of setting shrinkage can significantly change the early age material properties and their development over time [6]. Recently, many polymer-based materials have been developed for potential civil engineering applications. Polymer concrete is one of the most promising materials. Polymer concrete has various advantages over conventional, Portland cement concrete. They have higher compressive, tensile, and flexural strengths. They also have fast curing time, an important advantage in many construction applications; polymer concrete materials cure in several hours or less, whereas Portland cement based materials take 2~4 weeks [1, 7]. Also, they have excellent resistance to impact, abrasion, weathering, chemicals, water, and salt sprays. However, the early age properties of these polymer concrete with unsaturated polyester resin have not been carefully investigated [3, 11].

Unsaturated polyester resins are used extensively as matrix materials in resin mortar systems. Conventional unsaturated polyester resins characteristically shrink about 5 to 10% by volume during curing process [10]. This shrinkage is unfavorable for molding properties despite the many excellent characteristics of the resins. Because, the resin

mortars have high elastic modulus and their relatively low tensile strength owing to the high content of aggregates, the curing shrinkage is liable to induce cracks in molded parts [2, 12]. It is well known that low shrinkage resin compounds consist of unsaturated polyester resins mixed with certain thermoplastics such as polystyrene, acrylic polymer, polyvinyl acetate, etc. [5]. However, because low shrinkage behavior is complicated because it depends on many factors, such as components and curing conditions, several different hypotheses have been postulated.

In this study, the low shrinkage behaviors of recycled unsaturated polymer resin mortar were observed at the ambient curing temperature, and the low shrinkage mechanisms in recycled unsaturated polymer resin mortar were elucidate.

## 2. Experimental Programs

### 2.1. Materials

The relevant details of the coarse and fine aggregates used are given in Table 1. The sieve analysis of the fine aggregate is presented in Table 2. The nature and properties of fillers are given in Table 3. The aggregate and fillers were dried in an oven at 110°C  (230F) for 24 hours prior to their use. The properties of montmorillonite are given in Table 4. Two types of binder system–unsaturated polyester styrene type USP-1 (recycled) and USP-2 (verjin) were used, and their technical details are summarized in Table 5.

**Table 1.** Aggregate type and properties

| Aggregate type | Maximum size of aggregate, mm | Fineness modulus | Specific gravity |
|:---:|:---:|:---:|:---:|
| Course | 10 | 6.00 | 2.65 |
| Fine | 4.75 | 2.40 | 2.65 |

**Note.** 1mm = 0.039in.

**Table 2.** Particle size analysis of fine aggregate

| Sieve size, mm | 4.75 | 2.36 | 1.18 | 0.60 | 0.030 | 0.15 |
|---|---|---|---|---|---|---|
| Percent passed | 100 | 99.3 | 89.5 | 63.9 | 8.3 | 0.00 |

**Note.** 1mm = 0.039in.

**Table 3.** Detail of filler

| Filler | Average particle diameter microns | Specific gravity |
|---|---|---|
| Heavy calcium carbonate | 5.55 | 2.20 |
| Fly ash | 1.50 | 2.70 |

**Table 4.** Details of montmorillonite

| Specific gravity | Viscosity (sec.) | Strength (N/mm²) | Filtrate (ml) | PH |
|---|---|---|---|---|
| 1.22 | 43.2 | 2.16 | 18.4 | 10.4 |

**Table 5.** Relevant details of binders

| Binder | Styrene content | Hardener | Accelerator | Working life at 30C, mim. | Viscosity at 30C, cps |
|---|---|---|---|---|---|
| USP-1 | 36 | 5 | 2 | 150 | $312^{\xi}$ |
| USP-2 | 44 | 1 | 2 | 180 | $97^{\xi}$ |

**Note.** $\xi$ of resin only (Brookefield viscometer; spindle no. = 2).

## 2.2. Shrinkage measurement apparatus

A shrinkage measurement apparatus was specifically fabricated by modifying a commercially available length comparator (shrinkage measurement device conforming to BIS: 4031–1968), primarily used to measure the shrinkage of cement concrete and shrinkage measurement devices were also devised earlier by Ohama, Stanley et al., and Dupont Company [12]. However, the modified length comparator was found to be more adaptable to the local laboratory conditions. A schematic diagram of the apparatus is shown in Figure 1. It consists of two threaded circular rods, each of 25.4mm (1in.) in diameter and 550mm (21.65in.) in length. The threading was provided at both ends of each rod up to a length of 130mm (5.12in.). These rods were fixed to a channeled metallic frame (thickness 5.28mm (0.21in.)) through bolts, and this frame was firmly fixed to a heavy table. Two cross plates, A and B, each of 0.002mm (78.7 micro in.) in sensitivity and 10mm (6.39in.) in travel, were fixed at the middle of the two cross plates, A and B, respectively, such that the plungers of the dial gauges faced each other. A hole was provided at an appropriate place in the channeled frame, so that the plunger of dial gauge B could protrude to come in contact with the test specimen. It was ensured that, there was no hindrance of any kind to the movement of the plunger of dial gauge B, when it passed through the hole in the frame. All the metallic parts of the device were made of mild steel. A preferred position could be to fix both cross plates on one side of the channeled frame, in which case no hole was needed in the frame. But, the plates in this instance were placed on either side of the frame due to the limitations of rod lengths.
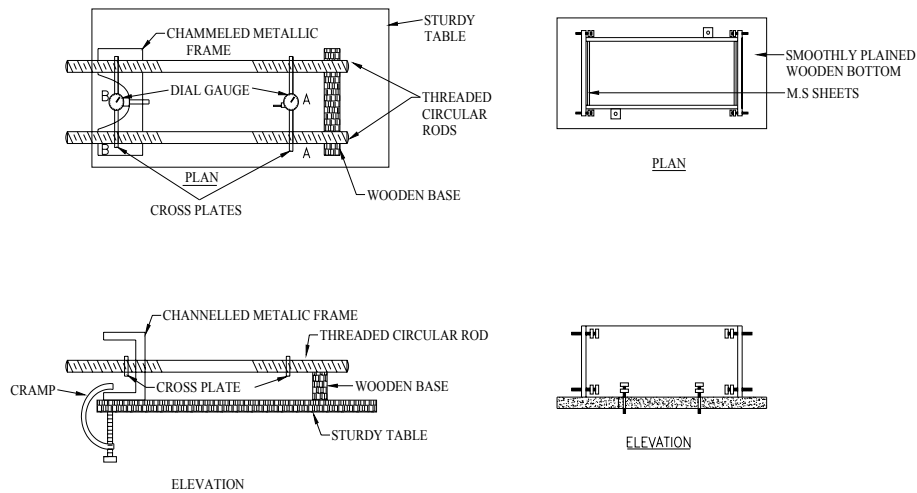
**Figure 1.** Shrinkage measuring device.

## 2.3. Shrinkage specimens

Polymer concrete specimens in this experiment were divided into three types. The first type consisted of 100mm square by 400mm bars for initial setting shrinkage, shown in Figure 2. The second type consisted of 25mm square by 285mm bars, whose mold is shown in Figure 3, tests setting shrinkage, coefficient of thermal expansion, and strain by using the strain gauge inserted into above bars according to ASTM C531. And the last type is a $\varphi 75 \times 150$mm cylinder, which was used to test the compressive strength of polymer concrete according to the content of montmorillonite. The proportions of binder and filler contents were varied, as shown in Table 6. The filler was mixed with the aggregate mixture and thoroughly dispersed. The binder, containing appropriate amounts of hardener and accelerator, was then added to the aggregate filler mixture, and a homogeneous polymer concrete mixture was prepared. The dry coarse and fine aggregates were first added to the filler, and montmorillonite in accordance with mix proportions and mixed for at least 2 minutes before adding unsaturated polyester resin. After

mixing, methyl ethyl kepton peroxide was slowly added in unsaturated polyester and was mixed for sufficiently long time. The mixture was poured slowly into the mixing equipment, which already contained the former mixture of aggregates, filler, and montmorillonite. And final mixture was poured into respective appropriate molds to obtain a good mixture. The molds were then vibrated for 2 minutes to eliminate air voids in the mixture. The specimens were placed at room temperature for a day, before demolding. The specimens demolded were placed at various temperatures and for various durations according to the experimental and material variables.
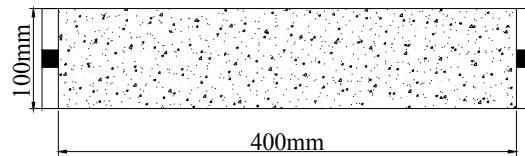
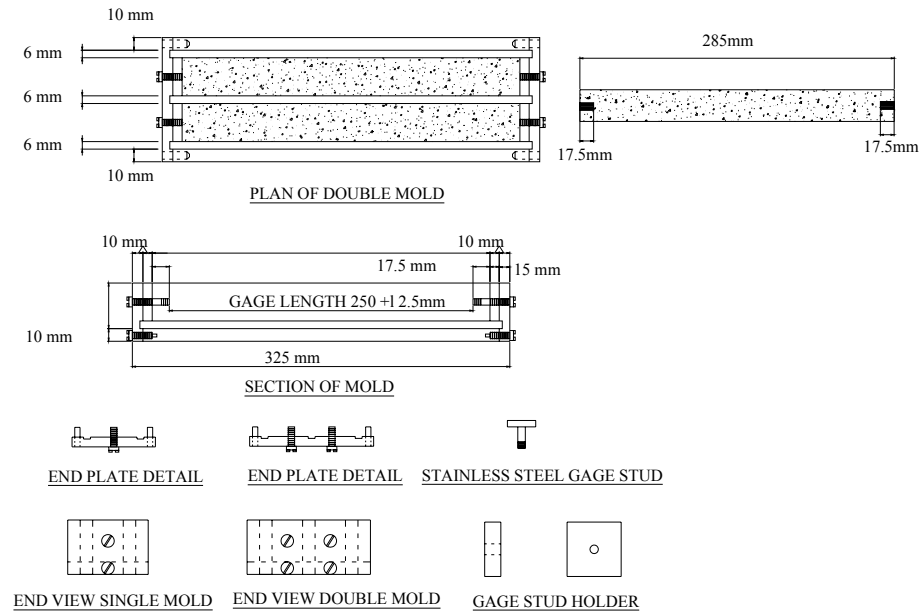**Figure 2.** Sketch of mold of initial setting shrinkage.

**Figure 3.** Sketch of mold of coefficient of thermal expansion.

**Table 6.** Proportions of binders and filler used

| Property investigated | Binder used | Binder content varied percent | Filler content varied percent | Mont. content varied percent | No. of specimen per each variation |
|---|---|---|---|---|---|
| Comp. strength | USP-1 | 10, 12, 15 | 3 ~ 11 | 0 ~ 8 | 3 |
| | USP-2 | 10, 15 | 5 ~ 11 | 0 ~ 8 | 3 |
| Setting shrinkage | USP-1 | 10, 12, 15 | 3 ~ 11 | 0 ~ 8 | 3 |

### 2.4. Curing method

30% methyl ethyl ketone peroxide (MEKPO) solution and 8% cobalt octoate (CoOc) solution were used as an initiator and an accelerator, respectively. The initiator and the accelerator were added in concentrations of 3.0 and 1.5phr, respectively. N, N-dimethyl aniline (DMA) or acethyl acetone (AcAc) was employed to reduce curing time. These co-accelerator concentrations influenced the low shrinkage property of polyester resin. Furthermore, *p*-tertiary buthyl catechol (PTBC) used as an inhibitor was kept at $20\pm1°C$, the standard temperature, before the addition of the initiator [1, 9].

### 2.5. Measurements

The curing process of the resin mortar was continuously monitored by the strain gauge. The thermocouple embedded at the center of the test piece was removed, so that the test piece could move freely. The curing process was monitored for over 24 hours. Measured values by the strain gauge were compared with the external dimension variations, as shown in Figure 4. The exact relation between them was confirmed, though the measured values were 0.1% larger than the external dimension variations in either the expanded or shrunken state.

**Figure 4.** Strain gauge setup for length change.

The sides of the mold were opened immediately after the casing was completed. The specimen with the thin sheets at its ends was placed (along with wooden base) between the plungers of the dial gauges of the shrinkage-measurement device, and the initial readings were recorded. The entire casting operation took about 5 minutes.

The first readings in the dial gauges were treated as reference to calculate the shrinkage of PC. Subsequent readings on the dial gauges were taken after 20min, 30min, 1hr, 2hr, 3hr, 4hr, 5hr, 10hr, 15hr, 20hr, and 24hr of commencement of casting. During the period of the experiment, the laboratory temperature varied between 28 to 32°C (82.4 to 89.6F).

The change in length at one end of the specimen was calculated by multiplying the difference in dial gauge readings with the sensitivity of the dial gauge, i.e., 0.002mm. The total change in the length of the specimen would be the sum of the individual changes in length at both ends, calculated separately. The shrinkage of the specimen, in micro strains, was obtained by dividing the change in length of the specimen by its initial length [4].

## 3. Results and Discussion

To confirm the effectiveness of reinforcement, all results obtained from the specimens were compared with those from the control specimen. The following results are discussed: compressive and flexural strengths, thermal expansive coefficient, and setting shrinkage for aging treatment.

### 3.1. Compressive and flexural strengths

The effect of age on the compressive and flexural strength of recycled unsaturated polyester polymer concrete is shown in Figures 5(a) and 5(b). Recycled unsaturated polyester resin-based polymer concrete achieves more than 80% of its 28-day strength in seven days. The high compressive strength of recycled unsaturated polyester resin-based polymer concrete allows the use of thinner sections in precast components, thus reducing dead loads in structures and minimizing transportation and erection costs. As a result, an increase in montmorillonite content from 1% to 5% increased, the recycled unsaturated polyester polymer concrete compressive strength by about 12%. However, an increase in the montmorillonite content 8%, decreased the strength of specimens.
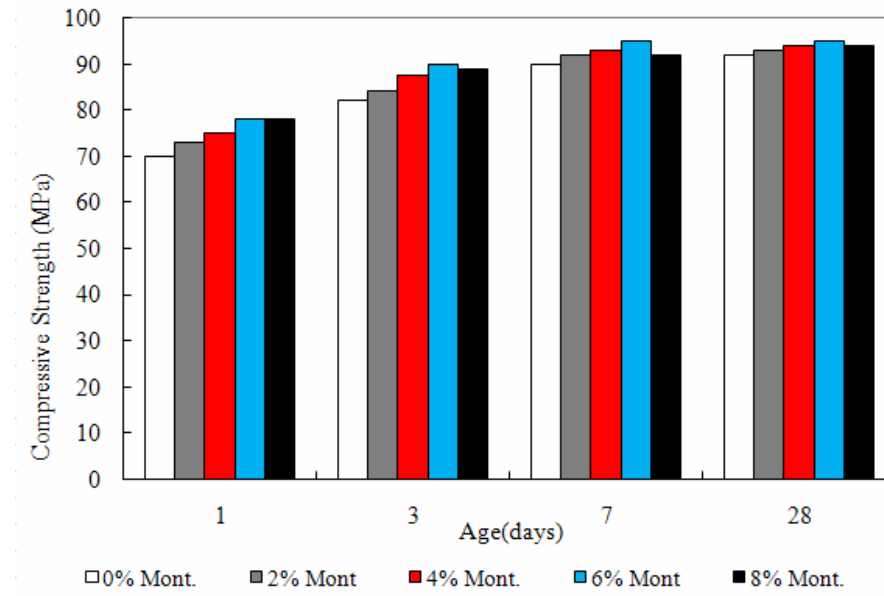
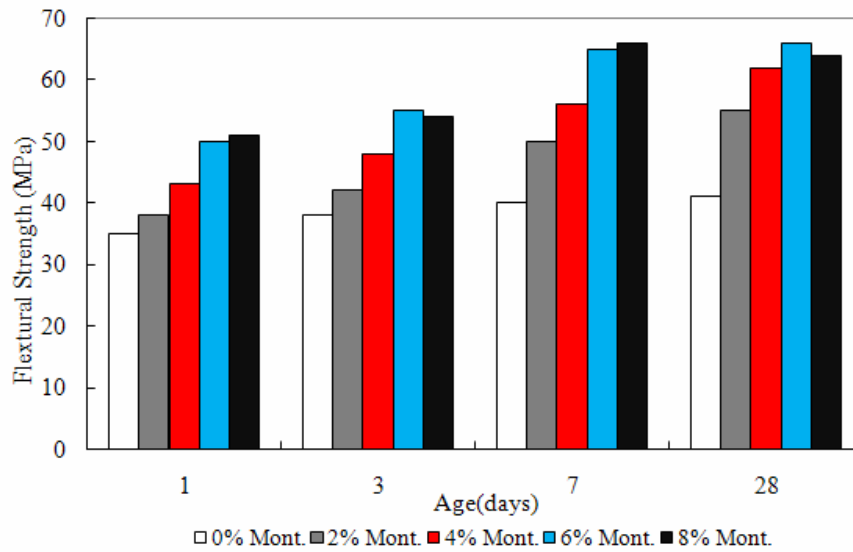**Figure 5(a).** Age effect on compressive strength.



**Figure 5(b).** Age effect on flexural strength.

The effect of temperature on the compressive and flexural strength of recycled unsaturated polyester polymer concrete is shown in Figure 6. Fillers (fly ash, CaCO$_3$) were placed in an environmental chamber at the specified temperature 48hr prior to mixing. After mixing, the specimens were again put in an environmental chamber at the designated temperature for a period of 28 days prior to testing. The selected temperatures were 25°C, 60°C, and 90°C. Actual testing, performed at room temperature, was conducted immediately after removing the specimens from the environmental chamber. When the temperature increased, recycled unsaturated polyester polymer concrete lost strength because of the resulting loss in strength of the resin binder, and the resulting decrease in bond strength between the inorganic aggregates and the resin binder. For example, recycled unsaturated polyester resin in temperature from 25°C to 90°C is more temperature sensitive than the inorganic cement binder used in producing normal Portland cement concrete. However, despite this loss in strength at high temperatures, recycled unsaturated polyester polymer concrete remains at least twice as strong in compression as regular Portland cement concrete.
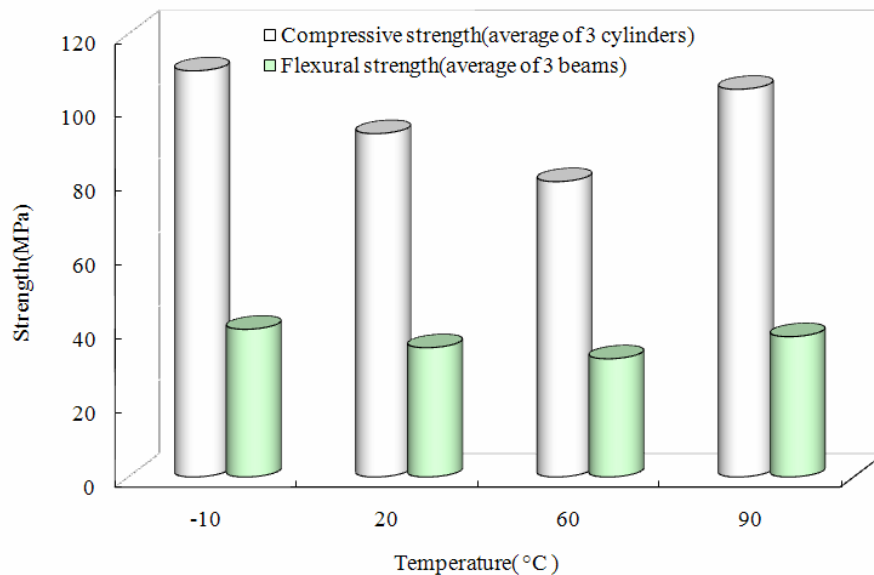


**Figure 6.** Temperature effect on compressive and flexural strength.

### 3.2. Expansive coefficient

The thermal reaction of montmorillonite as the shrinkage reducing agent, and fly ash and calcium carbonate as the filler is represented in Figure 7.
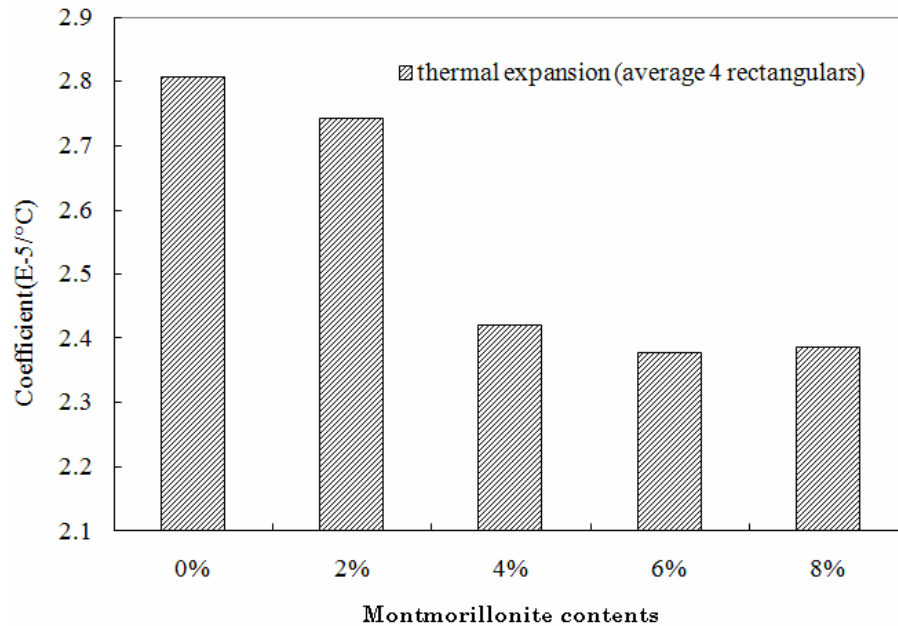


**Figure 7.** Coefficient of thermal expansion on montmorillonite contents.

The coefficient of thermal expansion decreases noticeably according to montmorillonite content. Moreover, specimens that used fly ash as the filler were less affected by temperature than those that used calcium carbonate among the specimens that contained 5% of montmorillonite evenly. The fact that fly ash is little sensitive to heat is useful, when using polymer concrete made by recycled materials in construction materials. The coefficients of thermal expansion of polymer concrete produced by recycled PET maintained a good level than those of general polymer concrete $(2.5 \sim 3.5\text{m} \times 10^{-5}/°\text{C})$, but showed a great difference from the coefficients of thermal expansion of general cement concrete $(0.7 \sim 1.2\text{m} \times 10^{-5}/°\text{C})$.

The effect of curing temperature on specimens of montmorillonite 5% is shown in Figure 8. The coefficient of thermal expansion decreased according to the increase of curing temperature, because of the close up between polymer molecules and the complete hardening by high temperature during the initial curing process. Especially, the values from the specimens cured in water were relatively higher than those cured at high temperature.
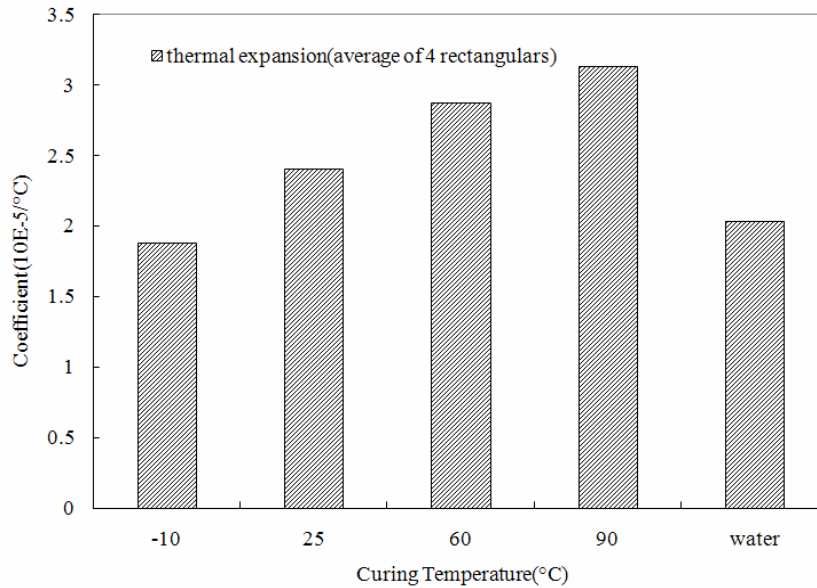


**Figure 8.** Coefficient of thermal expansion on curing temperature.

### 3.3. Shrinkage behavior

The linear setting shrinkage values of different polymer concrete mixes investigated are presented in Table 7. The shrinkage of recycled unsaturated polyester polymer concrete was generally high, varying between micro strains, as compared to that of Portland cement concrete, whose (drying) shrinkage varied usually between 200 and 500 micro strains. However, the measured shrinkage values were in the range of 0.1 ~ 0.3 percent of the reported values were in the range of 0.2 ~ 0.3

percent. The reduced shrinkage values exhibited by the mixes in this study could be due to the use of shrinkage reducer, such as montmorillonite.

**Table 7.** Shrinkage values of polymer concrete mixes (micro strains)

| Montmorillonite content percent | Resin content percent | | |
|:---:|:---:|:---:|:---:|
| | 10 | 12 | 15 |
| 0 | 2401 | 2579 | 3117 |
| 2 | 2240 | 2305 | 2895 |
| 4 | 1938 | 2014 | 2405 |
| 6 | 1540 | 1655 | 2010 |
| 8 | 1220 | 1255 | 1520 |

The development of shrinkage strains with time for recycled unsaturated polyester polymer concrete mixes containing different filler contents and montmorillonite contents are plotted in Figures 9 and 10, for mixes made with resin contents 10, 12, and 15 percent, respectively. Shrinkage developed at a rapid rate in the first few hours of casting (4 to 15hr), then slow down and finally seemed to attain a constant value, with no further or very little increase thereafter, suggesting the near completion of the shrinkage process.
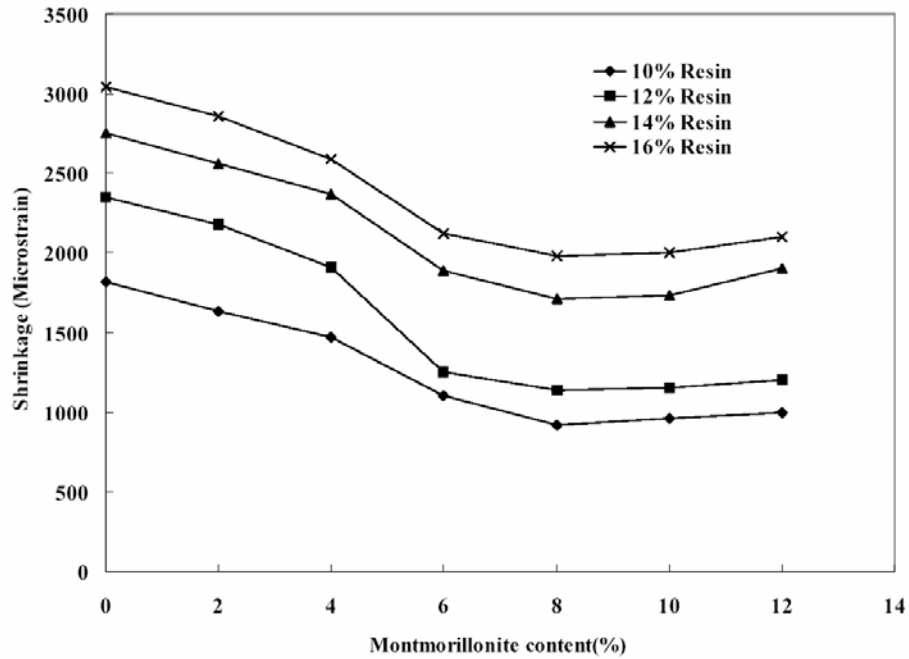
**Figure 9.** Shrinkage variation of recycled unsaturated polyester polymer concrete with montmorillonite content.
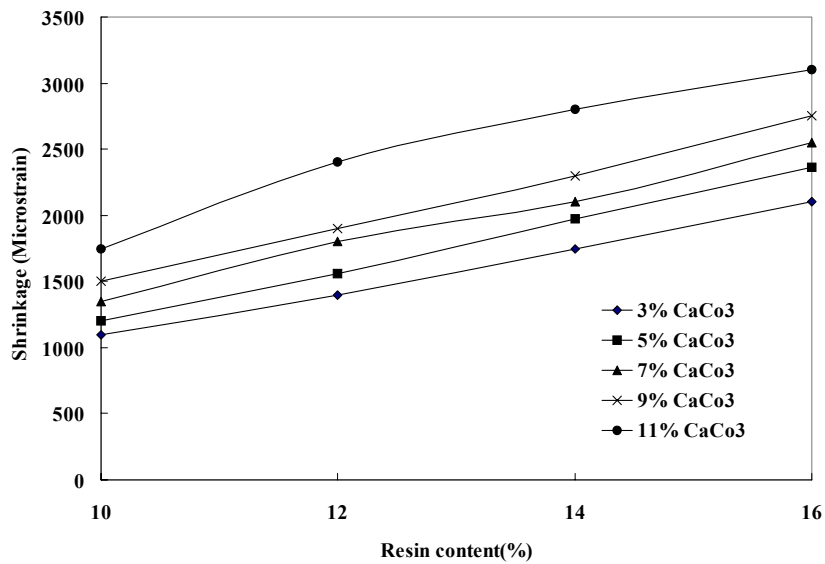


**Figure 10.** Shrinkage variation of RUSPC with resin content.

The variation of shrinkage (at 24hr) with resin and filler contents is plotted in Figures 11 and 12, respectively. The shrinkage increases linearly with resin content for all proportions of the filler content because, the shrinkage of polymer concrete is principally due to its binder content and no shrinkage of the aggregate.
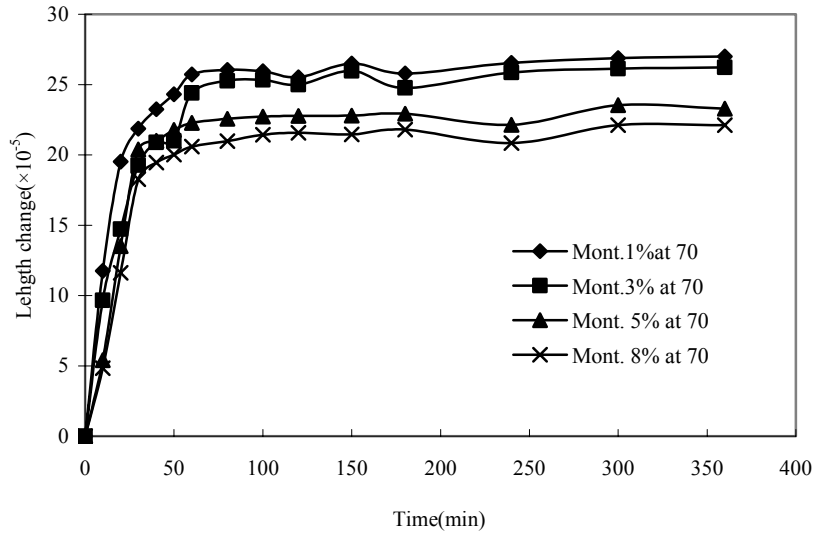


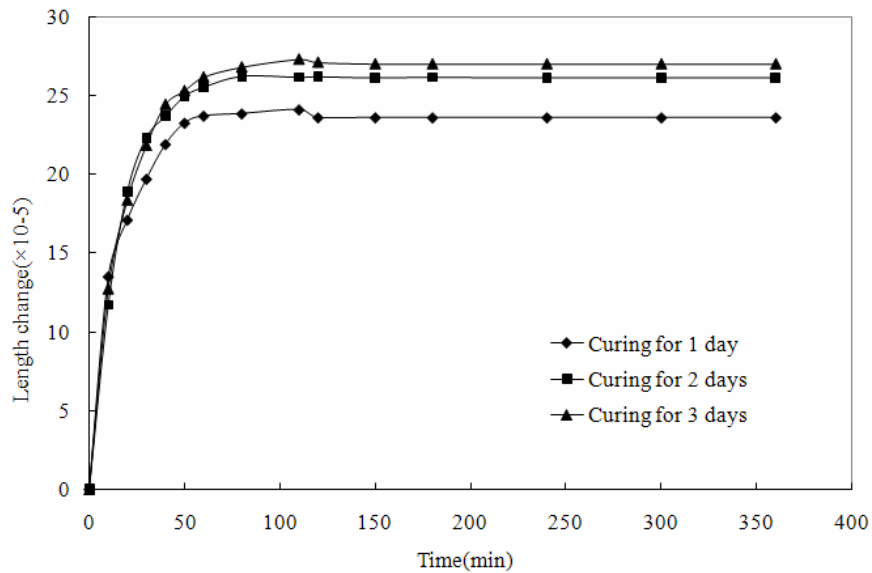**Figure11.** Shrinkage variation of RUSPC with time.

**Figure 12.** Length change of RUSPC with curing days.

The shrinkage of polymer concrete increases with the filler content also, but the rate of increase seems to decrease at the higher filler content. A possible reason for these increases in shrinkage with filler content could be as below.

As the polymerizing resin shrinks, the aggregate offers a restraint to the shrinkage. Such a restraint is likely to be due to two mechanisms : a frictional component at the aggregate surface and the relative low compressibility of the aggregate. The filler, which is very fine and soft, when added, is likely to coat the surfaces of the aggregate particles and cause a decrease in the frictional component of the restraint. In such a case, restraint to shrinkage would decrease as the filler is added. However, when the filler is high (such as 5 percent), the influence of the filler content on polymer concrete is not proportional. This is possible since only a small amount of filler needed to coat the aggregate particles, and the filler content beyond this amount is unlikely to cause a further decrease of the frictional restraint. The low shrinkage of polymer concrete with low resin content and zero filler content has a practical significance in the repair, and rehabilitation of deteriorated concrete structures.

## 4. Conclusion

Based on the results obtained in this investigation on setting shrinkage and compressive strength, and expansion coefficient of polymer concrete, the following conclusions can be drawn:

1. Strength test results revealed that, the material could achieve more than 85% of its final strength in one day. This result is an important advantage in many construction and structural applications. The material experiences a loss in strength at high temperatures; this result may be important, if the polymer concrete is used in precast box culvert, for example. However, despite this loss in strength at high temperatures, the material remains strong compared to Portland cement concrete.

2. Montmorillonite considerably affects various properties of polymer concrete. Also, the more the montmorillonite content increases, the more both the setting shrinkage and sensitivity to heat decrease. This result shows that, montmorillonite without a special coupling agent could be used as one of the additives to enhance various properties of polyester resin.

3. The coefficient of thermal expansion of fly ash was relatively lower than that of calcium carbonate because similar chemical composition led to active reaction with montmorillonite.

4. With the addition of montmorillonite 5%, the compressive strength was found to increase by 12%. Improvements in the properties of recycled unsaturated polyester polymer concrete were true for the optimum values of 5% of montmorillonite content. If the percentage of the montmorillonite exceeds 5%, recycled unsaturated polyester polymer concrete properties either remain constant or change in a negative way.

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