Polymer Concrete and Polymer Mortar Using Resins Based on Recycled Poly(ethylene Terephthalate)

K. S. REBEIZ,¹ D. W. FOWLER,¹ and D. R. PAUL²

¹Department of Civil Engineering, and ²Department of Chemical Engineering, The University of Texas, Austin, Texas 78712

SYNOPSIS

Fundamental properties of polymer concrete (PC) and polymer mortar (PM) made from unsaturated polyester resins based on recycled poly(ethylene terephthalate), PET, are described. Mechanical properties investigated include strength, modulus of elasticity, ductility index, Poisson's ratio, coefficient of thermal expansion, shrinkage, and exotherm. Durability properties include chemical resistance, water absorption, sand-blast resistance, and thermal cycles of thin overlays on portland cement concrete slabs. Resins based on recycled PET offer the possibility of a lower source cost for forming useful PC and PM (PC/PM) based products. The recycling of PET in PC and PM would also help alleviate an environmental problem and save energy.

INTRODUCTION

Polymer concrete (PC) and polymer mortar (PM) are composite materials made of a resin and inorganic fillers. PC uses coarse and fine aggregates while PM only uses fine aggregates. The coarse and fine aggregates have to be dry to ensure good bond between the polymer matrix and the inorganic aggregates. The liquid resin is mixed with the inorganic aggregates and allowed to cure. Initiators and promoters are added to the resin just prior to its mixing with the inorganic aggregates to initiate the polymerization reaction. The curing or working time of PC and PM is controlled by the quantity of promoters added to the resin. Compared to cementbased materials, PC and PM are very strong and durable materials. The fast curing time of these products is another important advantage in many construction applications (PC and PM cure in a few hours while portland cement concrete cures in a few days or weeks). However, the high cost of resins used in the production of PC and PM makes it expensive relative to cement-based materials and is the main disadvantage of the material.

Recently, some work has been done on the production of unsaturated polyester resins based on recycled poly (ethylene terephthalate), PET.¹ The PET waste, typically found in beverage bottles that are collected and recycled in many areas of the United States, is depolymerized using ethylene, propylene, or neopentyl glycol. The resulting glycolized products consist of oligomers and monomers. These oligomers are then reacted with unsaturated dibasic acids or anhydrides to form unsaturated polyester resins. A variety of proprietary ingredients may be added to give the resin specific properties like flexibility or rigidity. Unsaturated polyester resins made from virgin materials usually use phthalic anhydride or isophthalic acid instead of terephthalate acid.

If unsaturated polyester resins based on recycled PET could be used to produce good quality PC or PM at a lower price, their use would be important to the success of the PC and PM industry. This paper will therefore present the mechanical and durability properties of PC and PM made from two resins based on recycled PET that appear to have potential applications.

MATERIALS

Two different unsaturated polyester resins, resin 1 and resin 2, supplied from commercial sources, were used in this study. These resins were diluted with styrene. The two resins vary in their formulations,

Journal of Applied Polymer Science, Vol. 44, 1649–1655 (1992) © 1992 John Wiley & Sons, Inc. CCC 0021-8995/92/091649-07\$04.00

physical properties, and potential applications. Resin 1 and resin 2 used 40% and 30% by weight, respectively, of recycled PET in the alkyd portion of their formulations, i.e., before the addition of styrene. The recycled PET used in the production of resin 1 was not purified to the same extent as that used in the production of resin 2. In most PC or PM applications, the purity of the recycled PET is not very important, which should minimize the cost of resins based on recycled PET. Resin 1 had low viscosity and excellent wetting properties. It is therefore suitable for PC (high aggregate-to-resin ratio) as well as for PM (low aggregate-to-resin ratio) applications. Resin 2 had relatively high viscosity and relatively poor wetting properties. It is therefore only suitable for PM applications. The physical descriptions of resins 1 and 2 are shown in Table I.

The mix design for the clear cast resin was 1% (by weight of resin) of methyl ethyl ketone peroxide (MEKP) initiators and 0.1% (by weight of resin) of 12% solution cobalt naphthenate (CoNp) promoters, added to the resin immediately prior to casting. The resins were cast in 120 mm \times 160 mm rectangular molds and allowed to cure at room temperature. After demolding, the clear cast resins were shaped to conform to type I specimens of ASTM D638.

The mix design for PC and PM specimens was optimized for workability, strength, and economy. PC specimens using resin 1 as a binder are referred to as PC1. PC1 mix design was 10% resin, 45% 10mm oven-dried pea gravel, 32% oven-dried sand, and

Table IPhysical Properties of Descriptionsof Unsaturated Polyester Resins

Properties and		
Descriptions	Resin 1	Resin 2
Percentage by Weight of	40	30
Recycled PET in		
Resin Alkyd Portion		
Percentage by Weight of	40	30
Styrene Monomer in		
Resin		
Viscosity at 25°C (cps)	110	700
Weight Average Molecular Weight	3340	8000
Number Average Molecular Weight	1000	1500
Acid Number	8	20
Specific Gravity	1.09	1.16
Appearance	Dark	Clear
	green	amber

13% fly ash. PM specimens using resins 1 and 2 as binders are referred to as PM1 and PM2, respectively. PM1 and PM2 mix consisted of 20% resin, 60% oven-dried sand, and 20% fly ash. One percent (by weight of resin) of MEKP initiators and 0.1% (by weight of resin) of 12% solution CoNp promoters were added to the resin immediately prior to mixing. Mixing was done using conventional concrete mixers for a period of about 3 min. Specimens were then cast in molds and allowed to cure at room temperature. The age at testing of the specimens was 3 days unless otherwise specified.

TESTING

The clear cast resins (neat resins without the use of aggregates) were evaluated for their tensile properties according to ASTM D638. The cross-head speed was 5 mm/min and the elongation was measured over a 50-mm gage length using an electronic extensometer. Tensile strength, Young's modulus, and elongation at break were obtained from this test.

There are no standard tests that are directly applicable to PC or PM specimens. However, in the evaluation of mechanical and durability properties of PC and PM materials, ASTM standards applicable to cement-based materials were used as guidelines.

Compression tests were performed on 76 mm $\times 152$ mm cylinders. A constant loading rate of 44,500N/min was applied. Electrical strain gages, bonded to the specimens and connected to a data acquisition system, were used to read strains. Compressive strength and Young's modulus were obtained from this test.

Flexural specimens used 50 mm \times 50 mm \times 305 mm beams. The beams were loaded in third-point loading at a uniform rate of 2,225N/min. A dial gage was used to read the midspan deflection of the beam. Flexural strength, flexural Young's modulus, and ductility index (which is the ratio of deflection at ultimate load to deflection at the proportional limit) were obtained from this test.

Bond strength specimens were thin overlays, about 12 mm thick, cast directly (without the use of a primer) on sandblasted portland cement concrete slabs. Circular grooves (100 mm diameter) were cored through the overlays and into the portland cement concrete substrate. Circular steel disks were then bonded to the sandblasted overlay at the cored locations using a strong epoxy. The disks were then pulled out in direct tension to determine the type and magnitude of the bond failure. The pull-



Figure 1 PC and PM shrinkage measuring device.

out test method is fully described in an American Concrete Institute Committee 503 Report.²

The thermal expansion test used 76 mm \times 152 mm cylinders. Longitudinal electrical strain gages were bonded to the specimens using a special epoxy system insensitive to high temperatures (other epoxy systems showed improper behavior above 55°C). The strain gages were then connected to a switch and balance unit in a full-bridge configuration. An aluminum specimen, with a known coefficient of thermal expansion, was also tested to measure the validity of the test data. The specimens were subjected to thermal cycles beginning at room temperature. The temperature was increased to 70°C, decreased to -25°C, and then returned to room temperature. Strain and temperature readings were taken in increments of six degrees. For each of the six degree increments, the specimens were left at a constant temperature for a minimum of 8 h to ensure thermal stabilization before the strain and temperature readings were taken. Thermocouples, attached to the surface of the specimens and connected to a digital temperature indicator, were used to monitor specimen temperatures. The coefficient of thermal expansion was obtained from this test.

Shrinkage testing did not follow ASTM guidelines applicable to cement-based materials because the shrinkage mechanism in PC and PM is different than in cement-based materials. PC and PM only experience short-term shrinkage (plastic shrinkage) due to resin polymerization, whereas cement-based materials experience both short-term shrinkage (plastic shrinkage) and long-term shrinkage (drying shrinkage) due to water evaporation from the cement paste. A special method was therefore used to measure shrinkage strains. Shrinkage specimens consisted of 76 mm \times 76 mm \times 305 mm beams cast inside Teflon-lined molds. The molds were wrapped in a plastic sheet to reduce the effect of ambient temperature changes on the plastic shrinkage readings. Immediately after mixing and placing the materials in the molds, a shrinkage measuring device, connected to a data acquisition system, was carefully inserted into the fresh mix. The device, shown in Figure 1, consists of a horizontal rod to which two removable angles were attached. One angle was fixed while the other was free to move on roller bearings. A direct current differential transformer (DCDT), attached to the rod, was used to record the longitudinal displacement induced by shrinkage.

The peak exotherm was measured by inserting thermocouples inside the shrinkage specimens and connecting them to a digital temperature indicator.

The water absorption test used 50-mm cubes cut from the flexural specimens previously tested. The cubes were oven dried, weighed, and submerged in tap water. At 1, 4, 7, 19, 28, and 54 days, cubes were removed from water, their surfaces were dried, and their weights taken again to compute the water absorption percentages.

The chemical resistance specimens were also 50mm cubes cut from the flexural specimens. The specimens were cleaned, weighed, and submerged in acetone and 10% solutions of sulfuric acid, hydrochloric acid, and sodium hydroperoxide. Cubes removed at 1, 4, 7, 19, 28, and 54 days were surface dried and weighed. The chemical resistance was studied by analyzing changes in weight of the specimens and visually inspecting any physical defects.

The sandblast abrasion test was performed on 150 mm \times 150 mm \times 15 mm specimens. Portland cement concrete slabs tested in the same conditions served as control specimens. The test was conducted in a sandblasting cabinet using No. 20 to 30 Ottawa sand and an air pressure of 415 kPa. Each of the specimens was sandblasted for a period of 1 min on a 25-mm diameter area at eight different locations. The weights of the specimens were recorded before and after testing to determine the amount of material loss due to the sandblasting.

Table IITensile Strength Propertiesof Clear Cast Resins

Tensile Strength Properties	Resin 1	Resin 2
Tensile Strength (MPa)	21.1	13.3
Young's Modulus (GPa)	1.24	0.30
Elongation at Break (%)	2.4	53.5

Properties	PC1	PM 1	PM2
Compressive Strength (MPa)	91.8	90.2	72.2
Compressive Modulus (GPa)	28.6	16.7	6.7
Flexural Strength (MPa)	18.7	18.0	14.6
Flexural Modulus (GPa)	26.6	10.6	4.4
Tensile Bond Strength to Portland Cement			
Concrete (MPa)	1.9	2.7	2.6
Ductility Index ^a	2.8	3.1	4.5
Coefficient of Thermal Expansion for 20°C			
$\leq T \leq 70^{\circ}$ C (10^{-6} mm/mm/°C)	14.9	38.8	41.7
Poisson's Ratio	0.27	0.28	0.32
Shrinkage (10^{-3} mm/mm) at 24 h	2.1	7.5	4.9
Peak Exotherm (°C) at 2 h	31	42	40

 Table III
 Average Mechanical Properties of PC and PM Specimens

* Ratio of beam deflections at ultimate to deflections at proportional limit.

Thermal cycling test was done on 6-mm and 12mm thick overlays cast directly on portland cement concrete slabs. The specimens were put in an environmental chamber and subjected to thermal cycles. In each cycle, the temperature varied from -25° C to 70°C over a 24-h period. Thermocouples, inserted inside the overlays and at different depths, were used to ensure that all specimens had the same temperature. After 0, 25, and 80 thermal cycles, specimens were removed and the overlay was tested for its tensile bond strength to portland cement concrete using the pull-out test method.

RESULTS AND DISCUSSION

The tensile properties of the clear cast resins based on recycled PET, shown in Table II, are very important in the potential applications of PC or PM materials. Resin 1, being a rigid resin with high modulus and low elongation at break, is suitable for precast applications. Resin 2, being a more flexible resin with lower modulus and higher elongation at break, is suitable for overlay applications.

A summary of the mechanical properties of PC1, PM1, and PM2 is shown in Table III. These properties are comparable to PC and PM using virgin resins and tested for the same conditions at the University of Texas. Thus, they may be used for the same applications.

PC is typically used in precast applications. The greatest needs for precast products are, in order of importance: utility components (vaults, junction boxes, trench lines, floor drains, etc.); building panels; transportation-related components (median barriers, bridge panels, railroad ties, tunnels liners,

etc.); high voltage insulators; and flooring.³ PC is an excellent material for precast components because the fast cure time enables the fast and efficient use of forms and other production facilities. The excellent strength and durability properties permit the use of thinner sections, thus reducing dead loads in structures and minimizing transportation and erection costs. The ability to use colors and exposed aggregate finishes gives PC attractive appearances. Rigid resins should be used in precast applications because they produce high modulus PC that deflect and creep less under the action of loads. Low shrinkage is important because excessive shrinkage strains may significantly affect the dimension of precast structures, thus making their erection, assembly, or use more difficult. The coefficient of thermal expansion is important when PC is used in conjunction with other materials such as steel or portland cement concrete. The reason is that the coefficient of thermal expansion for PC is about two times higher than for steel or cement concrete. Hence, when PC is used with portland cement concrete or steel in a composite structure, changes in temperature will create stresses at the interface between the two materials that may eventually deteriorate the structure.

PM is typically used in the repair and overlay of pavements and bridges. The excellent mechanical and durability properties of the material allow the use of less materials (meaning less dead loads on the structures), eliminate the need to reconstruct approach slabs for bridges, and reduce maintenance and repair activities. The very fast cure time of the material results in minimum traffic disruption, shorter delays, and greater safety, and eliminates the need to construct expensive detours. Low mod-



Figure 2 Typical compressive stress versus strain for PC and PM.

ulus and good bond strength to the substrate are important properties when PM is used in overlays and repairs. Relatively low shrinkage is desirable because studies have reported that excessive shrinkage strains may cause delamination between the overlay and the substrate.⁴ Flexible resins should be used to produce low modulus PM (capable of stretching) in applications where large thermal or mechanical movements are to be expected.

The properties of PC1 are very different from PM1 and PM2 because PC1 used a weight aggregateto-resin ratio of 9-to-1, while PM1 and PM2 used a weight aggregate-to-resin ratio of 4-to-1. PC1 has a higher Young's modulus and lower Poisson's ratio than PM1 and PM2, as shown in Figure 2, because inorganic aggregates have a higher Young's modulus and lower Poisson's ratio than the resin binder. It is also observed that PM1 has a higher Young's modulus than PM2 because resin 1 is rigid while resin 2 is flexible.



Figure 3 Typical thermal expansion for PC and PM.



Figure 4 Typical shrinkage for PC and PM.

The amount of filler influences the thermal expansion behavior of polymer composites.⁵ Inorganic fillers or aggregates are considerably stiffer and have a lower coefficient of thermal expansion than the polymer binder. Therefore, as the aggregate-to-resin ratio in the polymer composite increases, the coefficient of thermal expansion decreases.⁶ The same phenomenon occurs with polymer composites using resins based on recycled PET. Figure 3 shows the thermal expansion of PC1, PM1, and PM2 polymer composites. PM1 and PM2, made with weight aggregate-to-resin ratio of 4-to-1, have a higher coefficient of thermal expansion at temperatures greater than 22°C than PC1 which is made with a weight aggregate-to-resin ratio of 9-to-1. Both polymer mortars show significant changes in thermal expan-



Figure 5 Typical exotherm for PC and PM.

sion characteristics in the temperature range 0°C to 20°C. Others have noted similar behavior with other materials.⁷

PC1 has a lower ultimate shrinkage and peak exotherm than PM1 and PM2, as shown in Figures 4 and 5, since less resin is available for polymerization in PC1 than in PM1 and PM2. Most of the shrinkage strains in PC1, PM1, and PM2 took place within the first 8 h after mixing and stopped after 24 h. The peak exotherm for PC1, PM1, and PM2 occurred about 2 h after mixing.

The durability properties of PM are shown in Table IV. The PM tested had low water absorption, good abrasion resistance, and good chemical resistance with the exception of acetone. These properties are important in various applications. A low water absorption is important when thin PM overlays are used in pavements and bridges because they prevent the intrusion of water and salts that could cause corrosion of the reinforcing steel in the portland cement concrete substrate. Good chemical resistance is important when PM overlays are used to protect industrial floors, tanks, or pipes from aggressive liquids. The abrasion resistance allows PM overlays to resist the action of traffic in pavements, floors, and bridges, and the erosion of water in dams and other hydraulic structures.

Figure 6 shows the tensile bond strength of PM1 and PM2 overlays on portland cement concrete slabs as a function of number of thermal cycles. This thermal cycle test is an accelerated procedure designed to simulate the effect of large variations in temperature that may occur in structures such as pavements and bridges. Variations in temperature in overlays can result in high stresses at the interface between the overlay and the portland cement concrete substrate because the coefficient of thermal expansion of unsaturated polyester concrete is high compared to portland cement concrete. The repeated changes in temperature, or thermal cycles, may

Table IVAverage Durability Propertiesof PM Specimens

Properties	PM 1	PM2
Water Absorption	< 1%	< 1%
Chemical Resistance		
Sulfuric Acid	Excellent	Excellent
Hydrochloric Acid	Excellent	Excellent
Sodium Hydroperoxide	Excellent	Excellent
Acetone	Poor	Poor
Sand Blast Abrasion	$\mathbf{Excellent}$	Excellent



Figure 6 Thermal cycling effect on bond strength of PM overlays on portland cement concrete slabs.

eventually reduce the bond strength between the overlay and the portland cement concrete substrate which may cause delamination and increase the permeability to water and chloride ions. Before thermal cycling, the bond strength of PM1 and PM2 overlays to the portland cement concrete substrate was good. After thermal cycling, a reduction in bond strength occurred between PM1 and PM2 overlays and the portland cement concrete substrate. The loss in bond was very substantial for PM1 and the overlays delaminated completely after 80 thermal cycles. Conversely, the loss in bond was not very substantial for PM2 and the bond strength of the overlay was still good after 80 thermal cycles. Therefore, the use of flexible resin binders with low modulus and high elongation at break is important to prevent thermal cycling failure of overlays. Low modulus resin binders are desirable because they produce low modulus PM overlays, offsetting the effect of the higher coefficient of thermal expansion. Resins with high elongation at break, capable of stretching during thermal movements, are desirable. Polyester resins which have an elongation at break of less than 10% do not perform as well as polyesters with an elongation of 20 to 50%. The same kind of behavior was observed in PC or PM overlays made with virgin unsaturated polyester resins.⁸

CONCLUSION

Unsaturated polyester resins based on recycled PET rather than all virgin materials can be used to produce good quality PC and PM products at a lower cost. In most PC or PM applications, the recycled PET does not have to be purified, e.g., removal of colorants, to the extent other PET recycling applications might require, which should minimize the cost of resins based on recycled PET.

In precast components, PC should be based on rigid resins. Resins with low viscosity and good wetting properties are important since they produce workable PC with high aggregate-to-resin ratio. A high aggregate-to-resin ratio is not only desirable for economy (since less resin, the expensive component, is used), but it also improves the properties of the PC by increasing the modulus, decreasing thermal expansion and shrinkage, and providing better heat and flame resistance.

In overlay or repair applications, PM should be based on flexible resins capable of stretching when subjected to large thermal or mechanical movements. The absence of coarse aggregates in PM permits the application of very thin sections and the effective use of shotcrete equipments in horizontal, vertical, or irregular surfaces. The low aggregate-toresin ratio provides PM with low Young's modulus, an important consideration in overlays and repairs.

The excellent strength and durability properties, and the fast curing time make PC and PM products cost efficient. Their long life will also provide for a long term disposal of the PET waste. PC and PM products could be crushed at the end of their life and hence transformed into excellent sources of aggregates for various concrete, highway, or subbase applications.

The authors acknowledge the support for this research from the Advanced Research Program of the Texas Higher Education Coordinating Board.

REFERENCES

- U. R. Vaidya and V. M. Nadkarni, "Unsaturated Polyester Resins from Poly (ethylene Terephthalate) Waste," in *Industrial & Engineering Chemistry Re*search, Vol. 26, 1987, pp. 194–198.
- American Concrete Institute Committee 503 Report, Appendix A, American Concrete Institute, Detroit, MI, 1979.
- D. W. Fowler, Future Trends in Polymer Concrete, American Concrete Institute Special Publication 116– 8, 1989, pp. 129–143.
- A. Al-Negheimish, Bond Strength, Long Term Performance and Temperature Induced Stresses in Polymer Concrete-Portland Cement Concrete Composite Members, Ph.D. dissertation, The University of Texas, Austin, Texas, 1988.
- V. I. Solomatov, Polymer-Cement Concretes and Polymer-Concretes, United States Atomic Energy Commission, Division of Technical Information, Translation Series 7147, 1970.
- J. A. Manson and L. H. Sperling, *Polymer Blends and* Composites, Plenum Press, New York and London, 1976.
- R. Letsch, Zum Verformungsverhalten von Epoxidharzmoerteln bei stationaeren und instationaeren Temperaturen, Polymers in Concrete, Fourth International Congress, Darmstadt, Germany, 1984.
- 8. M. M. Sprinkel, Polymer Concrete Overlay on the Big Swan Creek Bridge, Virginia Highway and Transportation Research Council, Report 37, Charlottesville, Virginia, 1986.

Received February 8, 1991 Accepted May 14, 1991