Publication Date : 30 April, 2015

Repercussion of mechanical behaviour in mortars with recycled aggregates by an overdose of PET additive

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***Abstract*— At present generation of domestic and industrial waste is a serious problem that must be controlled, so it is necessary to conduct further research of materials that minimize this problem by incorporating domestic and industrial waste in them; these materials must provide suitable properties, and perform the same function as the traditional material complies with virgin materials.**

This research focused on explaining the mechanical effects caused by overdose of unsaturated polyester resin made from post-consumer bottles made with polyethylene terephthalate (PET) (R-PET), cement-R- PET pastas and polymer modified mortars (PMM) with total or partial substitution of the recycled fine aggregate (AR) and additions of R-PET. The TGA and XRD results show that there is less development of the hydration products to a higher content of R-PET, causing reduction in the compressive strength of the PMM.

Keywords— polymer modified mortar, unsaturated polyester, polyethylene terephthalate, overdosage of additives in mortars**.**

I. Introduction

The construction industry requires significant demand for natural resources that cause waste generation; currently the above is a serious social and environmental problem that needs to be managed because the landfills are limited and resources are not unlimited. As feasible sustainable alternative to this problem, the recycling of construction waste and demolition (RCD) allows their use as recycled aggregates (RA) capable of being incorporated in full or partial replacement of natural aggregate (NA) in new elements (mortars recycled, MR); the above in addition to being a sustainable use would reduce the costs of processing and energy consumption used to obtaining [1] [2]. In contrast, this alternative could have physical impairments (porosity and density) in new recycled materials, with the implications of mechanical loss of resistance and of durability.

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On the other hand the plastic materials are also omnipresent in today's industry, of which the polyethylene terephthalate (PET) is the most representative, so its use is precursor also of a lot of waste. Despite being a non-toxic material to humans, it has high resistance to be degraded; however it can be recycled by mechanical and chemical processes; In the first process used (mechanical), is the raw material necessary for the second process (chemical); and the latter used is the called glycolysis, which uses large amounts of any of the glycols following: ethylene glycol, diethylene glycol, etc., yielding as final product a terephthalate monomer bis (2-hydroxyethyl) (BHET), that can be used to synthesize the PET or unsaturated polyester resin for use as densifier mortar additive (R-PET) [3] [4]. Therefore, the study of different dosages of R-PET in the search for improvements mechanical of the MR allow to set a new polymer modified mortar recycled R-PET (PMM); and what dosages are used of the R-PET in these PMMs, may become the cause of undesirable and counterproductive effects (an opposite effect from the intended), so the investigation of the effect of overdosage should be made, settle down and collated with their mechanical properties.

II. Methodology

A. *Preparation of recycled aggregate*

RCD is used from the demolition of a concrete pavement was processed with a jackhammer to reduce its size (<20 cm) in order to introduce in a jaw crusher and get aggregates with a size ≤ 2 cm; This material is then separated into two fractions using sieve # 4: fine and coarse, and of which the granulometric profiles were obtained (Figure 1) (ASTM C33) that allow verifying their suitability and to obtain parameters mix design (ASTM C128 and ASTM C136): fineness modulus, density and absorption (Table 1).

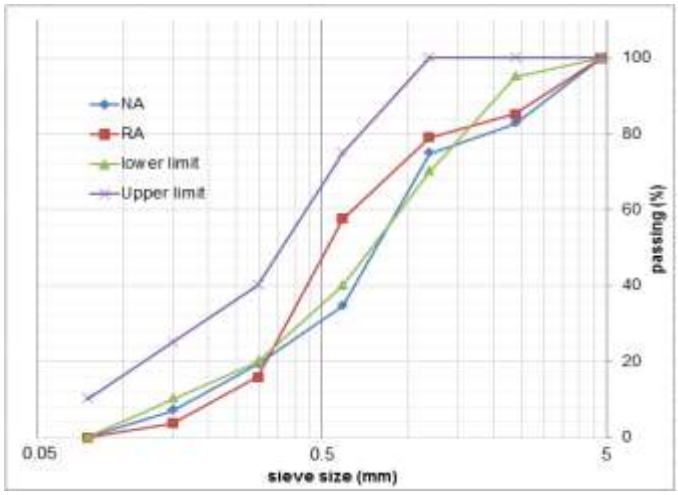

Figure 1 Aggregates granulometry

B. Getting recycled PET flakes

Of the Bottles post-consumer PET used was necessary to eliminate their labels, caps and rings. Then was made a with water wash which allowed cleaning. Finally, they were passed through a mill of 5HP non-commercial, giving as final product, flakes with sizes <1.00 cm, which were washed with water and caustic soda at 50% in order to remove contaminants.

C. Glycolysis of flakes

For depolymerisation of PET flakes, was used propylene glycol (PG) in a relation 2:1, and zinc acetate (AZ) as catalyst. The reaction occurred in a type reactor Vessel brand Syrris with digital control, which is constituted by a glass dual layer through which fluid circulates at a constant temperature (of -40 °C to 200 °C), and a stirrer, keeping the reaction moving, the process begins with increments warming 25 °C every 20 minutes to reach 197 °C, remaining in the latter for 3 hrs. Finally the resulting liquid material, which present a semi-solid consistency, removed from the reactor and at room temperature the colored of this was gray [5] [6] [7].

D. Synthesis of the resin

Maleic anhydride (MA) and adipic acid (AA) were used in a molar ratio of 1.1: 0.5: 0.5 both were weighed and introduced reactor into the Vessel, the reactor was programmed with equal parameters to those established in the reaction prior glycolysis. Then the material was removed from the reactor and allowed to cool to room temperature. Finally, was conducted the curing reaction by dissolving and reducing the viscosity of the resin (60%) with styrene (40%); for use in mixtures, was added to resin methyl ethyl ketone peroxide (MEKP) and cobalt naphthenate (NC) as initiator and catalyst respectively.

Dosage of PMM mixtures and use of R-PET

The study samples were determined it by varying amounts of RA in place of NA (with recommended percentage of content of [1]), and also using various percentages of resin addition by weight of cement (was selected percentages above of previous research [8]). In Table 1 are presented dosages.

Table 1 Dosage of mortar mixtures

Components	Quantities used				
RA	0	25	50	75	100
R-PET	0,5,10,15	0,5,10,15	0,5,10,15	0,5,10,15	0,5,10,15

NOTE: RA is expressed in % of aggregate used. R-PET is expressed in % by weight of cement.

E. Aggregates characteristics

ASTM C128 and ASTM C136 were used for the characterization of the aggregates, thereby obtaining its density and absorption. The behaviour of these is summarized as follows: increased absorption and lower density of AR when compared with AN (see Table 2).

Table 2 Properties of aggregates

Aggregates	Density (gr/cm^3)	Absorption (%)	Fineness modulus
AN	2.58	2.1	2.58
AR	2.24	12.8	3.07

F. Mixing and simple compression test

The PMM was mixed and tested in accordance with ASTM C109 using the amounts in Table 1, to avoid affectation relationship of water/cement (w/c) that could produce the AR (high absorption), these were pre-saturated with water for 10 min. before incorporation. Then the cement was added and finally the R-PET (activated with MEKP and NC to cause polymerization). Three cubes for each age of the curing and mixture study were made in cubic molds 5x5x5cm, then were covered with a plastic film to prevent moisture loss, and after 24 hrs, they were de-moulded and cured in immersion until assayed (Figure 1).

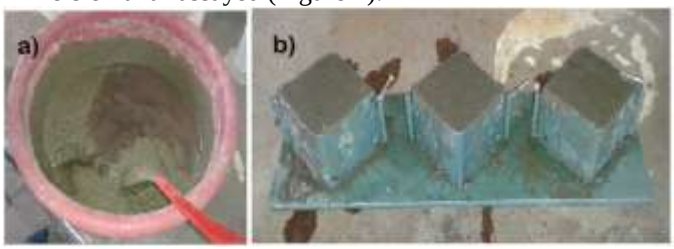

Figure 2 a) Mixing of mortar, b) Cast of molds

The ages of testing the compressive strength study were 7, 14 and 28 days, and the equipment used was a brand press INSTRON 600DXR2081 (Figure 3a).

G. X-ray diffraction (XRD)

Cement pastes with R-PET were characterized to age of 28 days (usual age of characterization) to prevent crystalline phases of the aggregates interfere with XRD analysis signals;

the tests were performed on a diffractometer equipped with a source radiation X-ray PRO PANalyticalX`Pert CuKa , and the test range utilized 5º to 70º of 2θ with steps of scanning 0.05 seg. The ratio a/c used was 0.5 with R-PET content of 0% (control test), 5, 10, and 15%. Before the test, the samples were ground in an agate mortar to a powder similar to fineness the talc, later to be arranged on the sample of holder and introduced on the computer for analysis (Figure 3b).

H. *Thermogravimetric Analysis (TGA)*

Same variables and sample preparation that in the XRD were characterized by TGA, equipment used was a TA Instruments SDT 2960 (Figure 3c), which consisting of an oven with an electronic balance inside that was coupled to a microprocessor control and data analyser. The samples were introduced and were calcined at 1100 ° C with temperature ramps 10ºC/ minute.

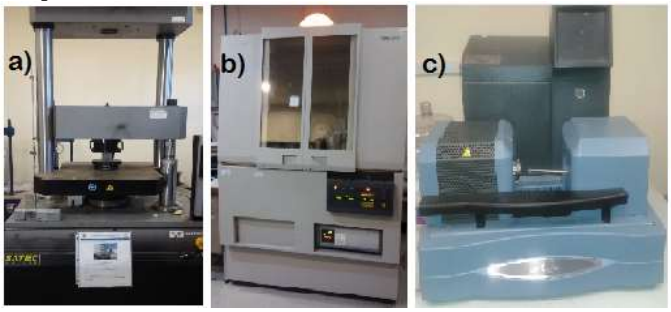

Figure 3 a) Compression test configuration, b) XRD test equipment, c) Test equipment TGA

III. Results

The results try correlate and determine the effect of overdose of R-PET in mechanical resistance of PMM, to explain this behaviour by developing its hydration products.

A. *Simple compressive strength*

The compressive strength of PMM is reduced by affectation in increasing both variables proposed study: a) content of R-PET, and b) RA contents. For the first variable, the increased from 0% to 5% of R-PET content causes loss in strength at an accelerated rate; but could be conceived as a tolerable dosage for matrix PMM since while reducing its strength in this dosage range is better; these materials indicate few losses in the hydration products with reference to the high dosages. As regards the increase to higher dosages (overdose of R-PET), although the rate of loss of resistance is reduced, the matrix does not evolve with the proper training of the hydration products. With respect to the second of the variables, this undesirable effect could be explained by the high porosity of the RA as compared to NA (Table 2). In Figure 4 the results of 28 day PMM are shown [9] [10].

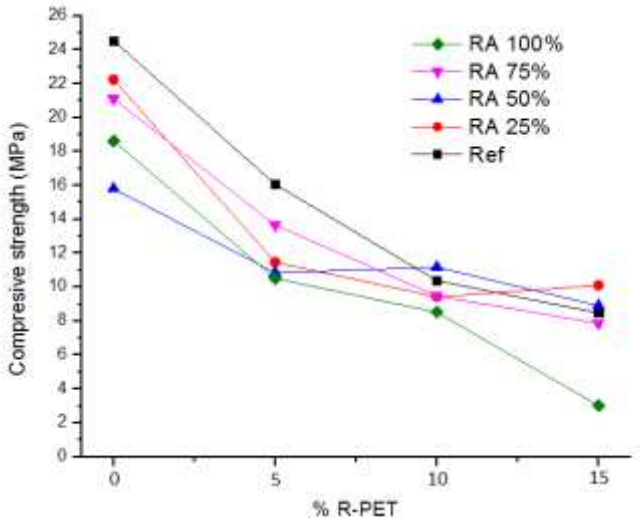

Figure 4 PMM Compressive strength at 28 days

B. *XRD*

XRD was used to assess the degree of hydration of pastes cement-R-PET (C / R-PET) and know the crystalline phases of the hydration products of cement pastes, among these calcium hydroxide ($Ca(OH)_2$) is one of the products more related to this process, at higher $Ca(OH)_2$ higher degree of hydration will have. Samples with higher content of R-PET indicate a decrease in the signal corresponding to $Ca(OH)_2$ which is close to 18 ° of 2θ (see Figure 5); however the above is not conclusive, since the degree of hydration may also be attributed to such factors as obstruction of the R-PET to the development of the hydration products or a reaction between the cement paste and the additive [11] [12]. Therefore, when the peaks of $Ca(OH)_2$ are reduced this directly related to increased R-PET, can be considered as evidence of adverse effect on PMM matrix, but is not conclusive.

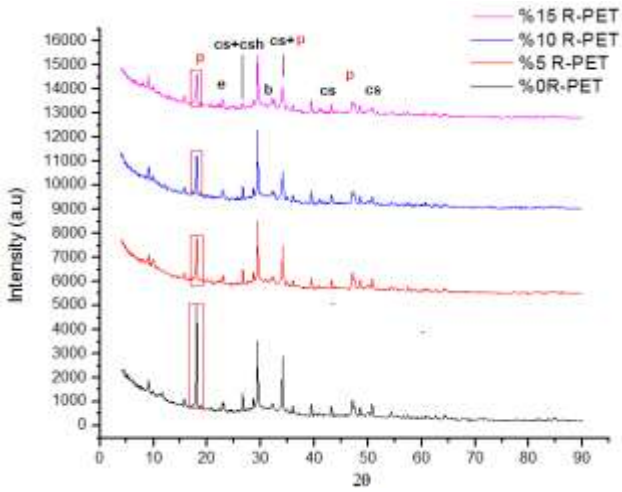

Figure 5 XRD for samples (csh- hydrated calcium silicate, cs-alite, p-portlandite, e- ettringite, b-belite).

C. *TGA*

As a complementary technique to XRD, TGA was used to assess the degree of hydration of cement. This technique establishes the relationship between the losses of non-evaporable water (chemically bound) at different stages of maturity of the cementitious matrix (curing age) with existing chemical compounds. Moreover, one of the principal compounds of interest in this research is the formation of $Ca(OH)_2$ because a high content of it correlates with adequate hydration of the matrix; so the use of R-PET can difficult the interpretation of the test [13].

In Figure 5, it can be seen that the temperature range for pastes ranging from 25 °C to 380 °C, corresponds to the physically absorbed water and chemically bound, the which ranges from 430 °C to 500 °C is attributable to the loss of $Ca(OH)_2$, and finally 500 ºC to 750 ºC to decarbonation of $CaCO_3$.

In the study variables, the significant range is was located between 380 ºC to 500 ºC, where significant drops weight of them were located. The percentage weight loss of the ends in the range the specified temperature, was determined; it can be seen that the compound $Ca(OH)_2$ has an inverse relationship to the content of R-PET. Less amount of compound is produced with increasing R-PET, which suggests affectation by the additive in the development of cement hydration [8].

Between 500 °C and 650 °C, range in which it can be assumed that the loss of R-PET originates, differences from pastes containing 5% and 10% are not significant, maintaining a trend similar to the variable (0% R-PET), but less loss attributed to the Ca $(OH)_2$. However, in the case of pasta with 15% contents of R-PET this trend not continues, this may be produced by a superposition effect of the resin and loss of $Ca(OH)_2$, which can add in this temperature range.

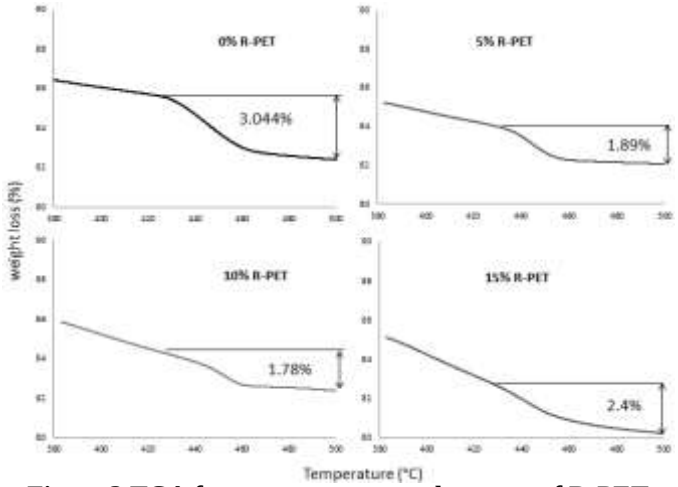

Figure 3 TGA for cement pastes and content of R-PET

IV. Discussion

In accordance with the test results obtained, the incorporation of R-PET in PMM affect the hydration process of the cement paste causing deficiency in his developmental, loss of the cementitious matrix densification, and products reduction that provide mechanical strength thereof.

This is ratified in two irrefutable evidence: First, XRD analysis performed at C/ R-PET has a lower intensity (increase C/R-PET) in the peaks corresponding to $Ca(OH)_2$ (located at 18º 2θ), evidence that hints of the development of the hydration products. Moreover, also are verified, causing affectation of the additive generally all the PMM.

Finally, the variable included in the study incorporation the RCD entails in PMM as the incorporation of R-PET, negative effects such as loss of mechanical strength. These effects may be attributed to the physical characteristics of RA used (lower density with respect to the NA, a high water absorption, and high porosity).

V. Conclusions

Below lists the conclusions of this study:

1. The Use R-PET in modifying cement mortars in large quantities, produces affectations in its resistance to compression.
2. In all cases that the additive was used and according to TGA and XRD analysis, there was a decrease in the formation of Ca $(OH)_2$, which is generated in the cement hydration process, and can be used to evaluate the degree of hydration of pasta.
3. Increasing the relationship C/R results in a reduction in cement hydration and hence a loss in compressive strength..
4. The Use RCD as RA produces lower compression strength, both in mortars containing R-PET as those not (R0%).

VI. References

[1]. Lee, S. T. (2009). Influence of recycled fine aggregates on the resistance of mortars to magnesium sulfate attack. Waste management, 29(8), 2385-2391.

[2]. Braga, M., de Brito, J., & Veiga, R. (2012). Incorporation of fine concrete aggregates in mortars. Construction and Building Materials, 36, 960-968.

[3]. Sánchez A. A. O. (2010). Obtención de oligómeros a partir de la degradación de PET para la formación de resinas poliéster insaturadas, (Obtaining oligomers from PET degradation to form unsaturated polyester resins), (Master thesis) Universidad Autónoma de Nuevo León, México.

[4]. Mahdi, F., A. A. Khan, and H. A. (2007). 3 Physiochemical properties of polymer mortar composites using resins derived from post-consumer PET bottles. Cement Concr. Compos. 29, 241–248.

[5]. Reséndiz, J.R.H. and A.E.M. (2012). Depolimerización de botellas de poli (tereftalato de etileno) (PET) post–consumo mediante glicólisis. I. Efecto del catalizador y del typo de glycol. (Bottles depolymerization of poly (ethylene terephthalate) (PET) by post-consumer glycolysis. I. Catalyst effect of glycol typo). Rev. Iberoam. Polímeros. 13, 3.

[6]. Xi, G., Lu, M., & Sun, C. (2005). Study on depolymerization of waste polyethylene terephthalate into monomer of bis (2-hydroxyethyl terephthalate). Polymer degradation and stability, 87(1), 117-120.

[7]. Elgegren Lituma, M. (2012). Poliésteres insaturados elaborados a partir del reciclaje químico de Poli (Tereftalato de Etileno), PET, y su aplicación como matriz de encapsulamiento de sales, (Unsaturated polyesters made from chemical recycling of poly (ethylene terephthalate), PET, and its use as encapsulation matrix salts). Rev Soc Quím Perú. 75 (1), 26–32.

[8]. Escalante, J. M. M. (2012) Síntesis y caracterización de resina de poliéster insaturado a partir de tereftalato de polietileno (PET) y evaluación de su influencia en la porosidad del concreto con agregado reciclado, (Synthesis and characterization of unsaturated polyester resin from polyethylene terephthalate (PET) and evaluation of its influence on the porosity of recycled concrete aggregate), (Master thesis) Universidad Autónoma de Sinaloa, Facultad de Ingenieria Mochis, México.

[9]. Wu, K. R., Zhang, D., & Song, J. M. (2002). Properties of polymer-modified cement mortar using pre-enveloping method. Cement and Concrete research, 32(3), 425-429.

[10]. Hwang, E. H., Ko, Y. S., & Jeon, J. K. (2008). Effect of polymer cement modifiers on mechanical and physical properties of polymer-modified mortar using recycled artificial marble waste fine aggregate. Journal of Industrial and Engineering Chemistry, 14(2), 265-271.

[11]. Li3, H.M.Y.T. and Z. (2011). Organic and Inorganic Phases in PA- and PU/PA-Modified-Cement-Based Materials. J. Mater. Civ. Eng. 23, 1412–1421.

[12]. Elena, J. and M.D.L. (2012). Application of x ray diffraction (XRD) and scanning electron microscopy (SEM) methods to the Portland cement hydration processes, journal of applied engieeniering science, 2(15), 35-42.

[13]. Vedalakshmi, R., Raj, a. S., Srinivasan, S., Babu, K.G. (2003). Quantification of hydrated cement products of blended cements in low and medium strength concrete using TG and DTA technique. Thermochim. Acta. 407, 49–60.

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