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# **Environmental Enhancement Through Use of Recycled Aggregate Concrete in a Two-Stage Mixing Approach**

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**Running Head:** Recycled Aggregate Concrete for Environmental Enhancement

## **ABSTRACT**

As a high priority of waste management and recycling by the Hong Kong government, Recycled Aggregate (RA) has been used in various construction applications, mainly as sub-grade, roadwork, and unbound materials. However, higher-grade applications are rare. The major barrier encountered is the variation of quality within RA, which causes lower strength, resulted from crystallization of Recycled Aggregate Concrete (RAC). Therefore, the objective of this study is to examine the crystallization of RAC in a Two-Stage Mixing Approach. Following are the five areas of interest: i) investigate the waste problems in construction activities; ii) examine the crystal development on the hydration of cement paste; iii) develop a two-stage mixing approach (TSMA) for improving the performance of RAC; iv) explore the crystallization of TSMA in comparison with the Normal Mixing Approach (NMA) through use of Differential Scanning Calorimetry (DSC); and v) verify the results obtained from DSC analysis with those obtained from compressive strength testing. This study adopted 0, 20, and 100% RA substitution in virgin aggregate and measured by DSC and compressive strength on both TSMA and NMA. TSMA uses only half the water for mixing, forming a thin layer of cement slurry on the surface of RA that will permeate into the porous old cement mortar and fill old cracks and voids in the pre-mix process. The results from DSC analysis clearly demonstrated that TSMA can give a better crystallization of  $\text{CaO} \cdot \text{SiO}_2 \cdot \text{H}_2\text{O}$  [CSH] and  $\text{Ca}(\text{OH})_2$  [CH]. The optimal situation occurs on 20% RA substitution in virgin aggregate, balancing the advantages of each, a finding supported by the results from compressive strength testing. Therefore, TSMA is a superior methodology and open a wider application for the use of RAC.

**Key Words:** crystallization, differential scanning calorimetry, recycled aggregate, concrete, construction.

## **INTRODUCTION**

Construction wastes are considered as having major impacts on the environment. Waste is defined as the by-product generated and removed from construction, renovation, and demolition of workplaces or sites of building and civil engineering structures (Cheung 1993). With the demands to implement major infrastructure projects in Hong Kong, together with many commercial building and housing redevelopment programmes, a large amount of construction waste is being produced. Extra construction materials are usually planned due to the lack of consideration given to waste reduction during the planning and design stages in order to minimize the generation of wastes. The excessive wastage of raw materials, improper waste management, and low awareness of the need for waste reduction are common in the local construction industry.

Although the reuse of concrete waste as Recycled Aggregate (RA) can reduce the consumption of limited resources and thereby save construction costs, the use of RA also has shortcomings, *e.g.*, weaker interfacial behaviour between aggregate and cement paste, higher portions of cement mortar attached and lower quality. (Alexander 1996; Covertry 1999; Farran 1956; Jia *et al.* 1986; Kawano 2000; Keru and Jianhua 1988; Li *et al.* 2001; Popovics 1987; Tomosawa and Noguchi 2000; Xueqan *et al.* 1988). In order to facilitate waste management through the use of RA, it is vital to investigate ways in which its shortcomings can be mitigated.

## **OBJECTIVE**

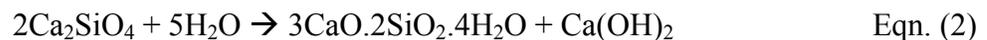
The research objective is to examine the crystallization of Recycled Aggregate Concrete (RAC) for use in a two-stage mixing approach (TSMA). The following are focal issues:

- i) Investigate the waste problems in construction activities,
- ii) Examine the crystal development on the hydration of cement paste,
- iii) Develop a TSMA for improving the performance of RAC,
- iv) Explore the crystallization for TSMA in comparison with that in the Normal Mixing Approach (NMA) through application of Differential Scanning Calorimetry (DSC), and
- v) Verify the results obtained from DSC analysis with those from compressive strength testing.

## CRYSTALLIZATION

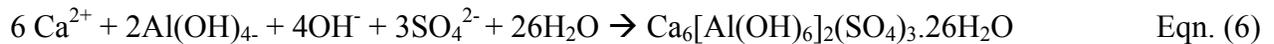
At the micro-structural level, Hardened Cement Paste (HCP) is an intimate but inhomogeneous mixture of a variety of crystalline and quasi-crystalline phases and pores of different sizes and shapes (see Figure 1). Four major compounds: tricalcium silicate [ $3\text{CaO}\cdot\text{SiO}_2$  or  $\text{C}_3\text{S}$ ], dicalcium silicate [ $2\text{CaO}\cdot\text{SiO}_2$  or  $\text{C}_2\text{S}$ ], tricalcium aluminate [ $3\text{CaO}\cdot\text{Al}_2\text{O}_3$  or  $\text{C}_3\text{A}$ ] and tetralcium aluminoferrite [ $4\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot\text{Fe}_2\text{O}_3$  or  $\text{C}_4\text{AF}$ ] occur in ordinary Portland cement (Sha *et al.* 1999).

In concrete, most of the volume is calcium silicate hydrate [ $\text{CaO}\cdot\text{SiO}_2\cdot\text{H}_2\text{O}$ , abbreviated as CSH], the principal hydration product, which is a highly disordered crystalline material of variable composition. Upon the addition of water,  $\text{C}_3\text{S}$  rapidly reacts to release calcium ions, hydroxide ions, and a large amount of heat. The pH quickly rises to greater than 12 because of the release of alkaline hydroxide ( $\text{OH}^-$ ) ions. This initial hydrolysis slows quickly after it starts, resulting in a decrease in heat evolved. The reaction continues slowly and produces calcium and hydroxide ions until the system becomes saturated. Once this occurs, the calcium hydroxide [ $\text{Ca}(\text{OH})_2$ , abbreviated as CH] starts to crystallize. Simultaneously, CSH begins to form. Ions precipitate out of solution, which accelerates the reaction of  $\text{C}_3\text{S}$  to calcium and hydroxide ions. The evolution of heat is then dramatically increased. The formations of CH and CSH crystals provide ‘seeds’ upon which more CSH can form. The CSH crystals grow thicker making it more difficult for water molecules to reach the unhydrated  $\text{C}_3\text{S}$ . The speed of the reaction is now controlled by the rate at which water molecules diffuse through the CSH coating. This coating thickens over time, causing the production of CSH to become slower and slower. The molecular structure of CSH is layered and provides for a very high internal porosity. The process of the hydration of  $\text{C}_3\text{S}$  is given as Equation (1) (MAST 2004).  $\text{C}_2\text{S}$  also affects the strength of concrete through its hydration.  $\text{C}_2\text{S}$  reacts with water in a similar manner compared to  $\text{C}_3\text{S}$ , but much more slowly. The heat released is less than that by the hydration of  $\text{C}_3\text{S}$  because the  $\text{C}_2\text{S}$  is much less reactive. The process of the hydration of  $\text{C}_2\text{S}$  is given as Equation (2) (MAST 2004):



In contrast to CSH,  $\text{Ca}(\text{OH})_2$  [CH], also known as Portlandite, is highly crystalline and has a fixed composition, which comprises more than 20% of the hydration products in a fully cured cement paste (Bhatty 1991). CH appears as thin hexagonal platelets, often layered; typically tens

of  $\mu\text{m}$  across (see Figure 2). With continued hydration, they grow massive, lose their hexagonal outline, and encapsulate other regions of the paste (Ahmed and Struble 1995). Massive blocks of CH crystals can be easily identified in mature pastes. Unhydrated residues of clinker grains, which are crystalline in nature, are present even in well-hydrated systems. They are produced by the hydration of the silicate phases,  $\text{C}_3\text{S}$ ,  $\text{C}_2\text{S}$ , and free lime  $[\text{CaO}]$  as shown in Equations (3) to (5) (Bhatty 1991; John *et al.* 1998; Lea and Desch 1935). It can be seen from Equations (3) and (4) that weight for the hydration of  $\text{C}_3\text{S}$  [Equation (3)] produces three times the amount of CH as does the hydration of  $\text{C}_2\text{S}$  [Equation (4)]. Thus the ratio of these two phases will affect the amount of CH formed.



Another type of crystal commonly found in HCP is ettringite  $[\text{Ca}_6[\text{Al}(\text{OH})_6]_2(\text{SO}_4)_3.26\text{H}_2\text{O}]$ , as reacted in Equation (6) (see Figure 3). Where ettringite is observed in cracks and voids, it appears as needles that have a distinctive low birefringence and low refractive index; it is often found in conjunction with CH (John *et al.* 1998). Situations such as movement of water through concrete or the formation of alkali-silica gel appear capable of causing conditions that allow it to reform. In old concrete, these deposits can be massive. However, as the ettringite is reformed in void spaces, its expansion can be accommodated (John *et al.* 1998). Ettringite is produced by a reaction that requires an excess of the sulphate ion  $\text{SO}_4^{2-}$  over the aluminate phase in the pore solution (John *et al.* 1998). As hardening proceeds, more of the aluminate phase moves into the pore solution, which converts the ettringite to mono-calcium sulfo-aluminate  $[\text{CaSO}_4.2\text{CaO}.\text{Al}_2\text{O}_3.12\text{H}_2\text{O}]$ . This conversion results in a small decrease in volume and thus may occur in the hardened paste without damage. Both ettringite and monocalcium sulfo-aluminate hydrate formed during setting and hardening are usually submicroscopic and require the use of Scanning Electron Microscopy (SEM) observations. Only where ettringite is secondary and reformed from reactions of the  $\text{SO}_4^{2-}$  ion with monocalcium sulfo-aluminate do its crystals become large enough to be observed in thin-section. Localized concentrations of ettringite are controlled by both the amount of water and space at particle surfaces, with

crystallization modified by the surface texture and porosity of the aggregate, which greatly improves the strength (John *et al.* 1998).

## **MATERIALS AND METHODS**

The weaknesses of RA, which include high porosity, high amounts of cracks, high levels of sulphate and chloride content, high levels of impurity, and high cement mortar remains, will affect the mechanical performance of RAC (Ryu 2002). The prerequisite in applying RA to high grade concrete is to overcome these weaknesses.

A new mixing approach, the Two-Stage Mixing Approach (TSMA), is proposed as a means to mitigate the weakness of RA. In the normal mixing approach (NMA), the mixer is first charged with about one-half coarse aggregate, then mixed with fine aggregate, then with cement, and finally with the remaining coarse aggregate. Water is then added immediately before the rotation of the drum or starting the pan (Hong Kong Government 1990). TSMA divides the mixing process into two parts and proportionally splits the required water into two streams, which are added at different times. The TSMA mixing procedure is illustrated in Figure 4, while Table 1 shows the symbols used in the figure.

In order to study the crystallization of RAC, experimental work was undertaken by use of Differential Scanning Calorimetry (DSC) [Seiko Instruments (SII) SSC 5200 TG/DTA 220] (Bhatty 1991). The development of crystallization during the TSMA with RA replacement in virgin aggregate of 0, 20, and 100% was studied. The RA was collected from the Tuen Mun Area 38 centralized recycling plant. Before collection of samples from the cube, the first 4 to 5 mm of the surface was removed by either grinding or filing. These samples were placed in a small and flat container in shallow pans, with the aim of making a good thermal contact between sample, pan, and heat flux plate (Anasys 2004). The samples weighted approximately 10 mg and were collected from cubes stored under water at about 20°C for 28-day or 56-day curing. The samples were heated to 600°C at a constant heating rate of 20°C per minute in a dynamic helium atmosphere.

This experiment was also measured and compared with results from compressive strength testing. The average of three results of measurement with 100mm sized cubes was derived according to BS 1881: Part 116 (1983).

## **RESULTS**

Listed in Table 2 are the results of DSC analysis, which show the major endothermic peaks and their quantification after 28-day and 56-day curing, and the results of the compressive strength testing are tabulated in Table 3.

DSC analysis showed there are two major peaks, peaks 1 and 2, at around 90°C and 450°C, respectively (Poon and Wong 2004; Sha *et al.* 1999). The first endothermic peak, peak 1, is the result of dehydration reactions due to the loss of water from CSH, which reduce cohesive forces between CSH layers and where coarsening of pores starts. The second major peak, peak 2, corresponds to the dehydroxylation of CH, in which the decomposition of CH starts from 350°C to 500°C. A smaller peak is also found in the DSC's curves at around 140°C to 215°C. This endothermic peak is attributed as ettringite, which corresponds to the hydration products of C<sub>3</sub>A (Sha *et al.* 1999). A typical DSC curve is shown in Figure 5.

## **DISCUSSION AND VERIFICATION**

In the general sense, it was found that the more RA substituted in the concrete, the poorer the quality of the concrete. This is because the cohesive force between aggregate and cement paste is weaker in RA than that in virgin aggregate (John *et al.* 1998). This situation is also reflected from the DSC's results; both the quantities of CSH and CH recorded are lowered with increased in amounts of RA substitution for NMA. For example, the peak area of CSH with 0% RA substitution after 28-day curing is 21.85  $\mu\text{V}^\circ\text{C}$ ; while that of 20 and 100% RA substitution are 18.38  $\mu\text{V}^\circ\text{C}$  and 12.97  $\mu\text{V}^\circ\text{C}$ , respectively (see Table 2). Similar situations are also recorded on the peak area of CH and in the curing conditions of 56-day. It is also proved from the results obtained from compressive strength testing after 28-day curing at 55.72 MPa, 53.68 Mpa, and 51.72 MPa for 0, 20, and 100% RA substitution, respectively (see Table 3).

Although the pessimistic situations happened when substituting RA with virgin aggregate, TSMA helps solve the current problem. From the results of DSC analysis, the crystallization of CSH and CH are enhanced when mixed by TSMA compared with that for NMA. After 28-day curing, dehydroxylation of CH at 27.35  $\mu\text{V}^\circ\text{C}$ , 48.31  $\mu\text{V}^\circ\text{C}$ , and 28.61  $\mu\text{V}^\circ\text{C}$  and compressive strength at 56.00 MPa, 65.05 Mpa, and 55.56 MPa were found with 0, 20, and 100% RA substitution, respectively, for TSMA. TSMA therefore provides improvement in comparison with NMA on dehydroxylation of CH at 25.35  $\mu\text{V}^\circ\text{C}$ , 25.30  $\mu\text{V}^\circ\text{C}$ , and 21.23  $\mu\text{V}^\circ\text{C}$  and compressive strength at 55.72 MPa, 53.68 Mpa, and 51.72 MPa, respectively. Figure 6 shows the benefits gained from the use of TSMA.

It is recorded at 20.45  $\mu\text{V}^\circ\text{C}$  and 28.61  $\mu\text{V}^\circ\text{C}$  on the dehydration of CSH and the dehydroxylation of CH, respectively, for TSMA and strength at 55.56 MPa with 100% RA substitution, which achieved better performance than that in the same RA substitution for NMA. It is encouragingly found that the results with 100% RA replacement for TSMA are as competitive as the traditional approach with 0% RA substitution at 21.85  $\mu\text{V}^\circ\text{C}$  and 25.35  $\mu\text{V}^\circ\text{C}$  and 55.72 MPa for the dehydration of CSH, the dehydroxylation of CH and the compressive strength at 28 days, respectively.

The optimal situation occurs at 20% RA substitution in virgin aggregate, in balancing the benefits earned from RA and virgin aggregate, which gained 34.21  $\mu\text{V}^\circ\text{C}$  and 48.31  $\mu\text{V}^\circ\text{C}$  for the dehydration of CSH and the dehydroxylation of CH, respectively, after 28-day curing, and 25.44  $\mu\text{V}^\circ\text{C}$  and 44.59  $\mu\text{V}^\circ\text{C}$ , respectively, after 56-day curing. The results from compressive strength testing correspond with the similar conclusions at 65.05 MPa and 72.04 MPa after 28-day and 56-day curing, respectively, with 20% RA replacement ratio. Furthermore, the crystallization of 20% RA substitution for TSMA is also proved from the examination of Scanning Electron Microscopy (SEM) in Figures 7 and 8. Hence, most studies recommended a limit of 30% RA replacement in concrete mixing (Kikuchi *et al.* 1988).

## **CONCLUSIONS**

To alleviate waste problems from construction and demolition activities, recycling concrete waste as RA provides the greatest benefit to the environment. Due to the variation of quality of RA, higher-grade applications are restricted. The two-stage mixing approach (TSMA) helps solve the current pessimistic situations. The TSMA uses only half of the water for mixing to form a thin layer of cement slurry on the surface of RA, which will permeate into the porous old cement mortar and fill the old cracks and voids in the pre-mix process. Thus, TSMA gained a better crystallization of CSH and CH. From the results of DSC analysis, it was encouragingly found that the 100% RA substitution in TSMA can be as competitive as the traditional mixing approach with no substitution of RA. Furthermore, the optimal situation occurs with 20% RA substitution. These results are in agreement with those measured by compressive strength testing. Therefore, TSMA is a superior methodology and opens up a wider application for recycled aggregate concrete.

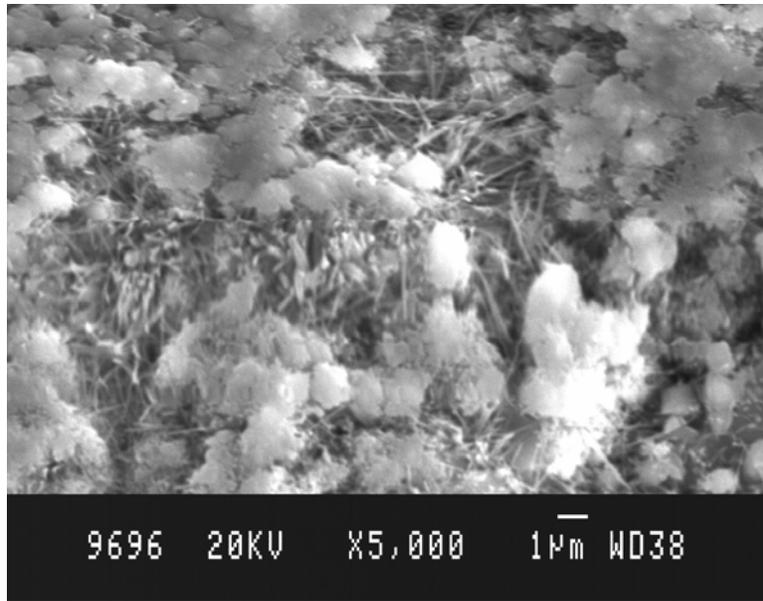
## **ACKNOWLEDGMENTS**

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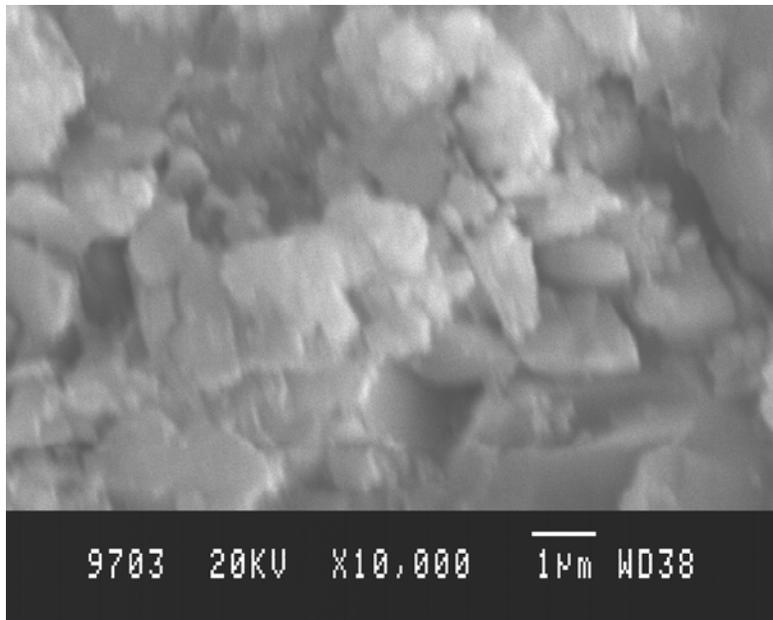
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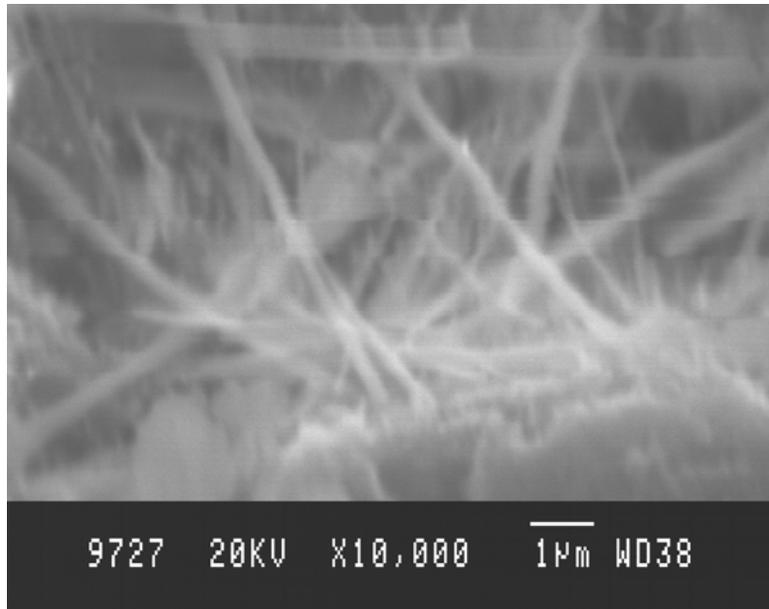
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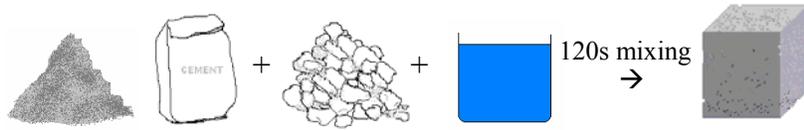
**Figure 1.** SEM micrograph of the hardened cement paste.



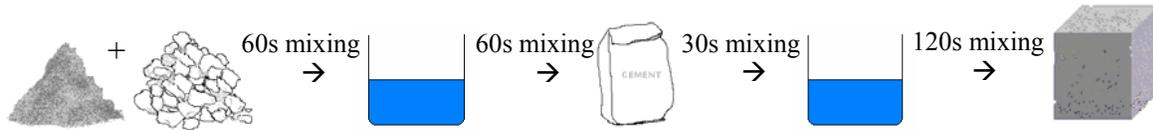
**Figure 2.** SEM micrograph of calcium hydroxide.



**Figure 3.** SEM micrograph of ettringite.

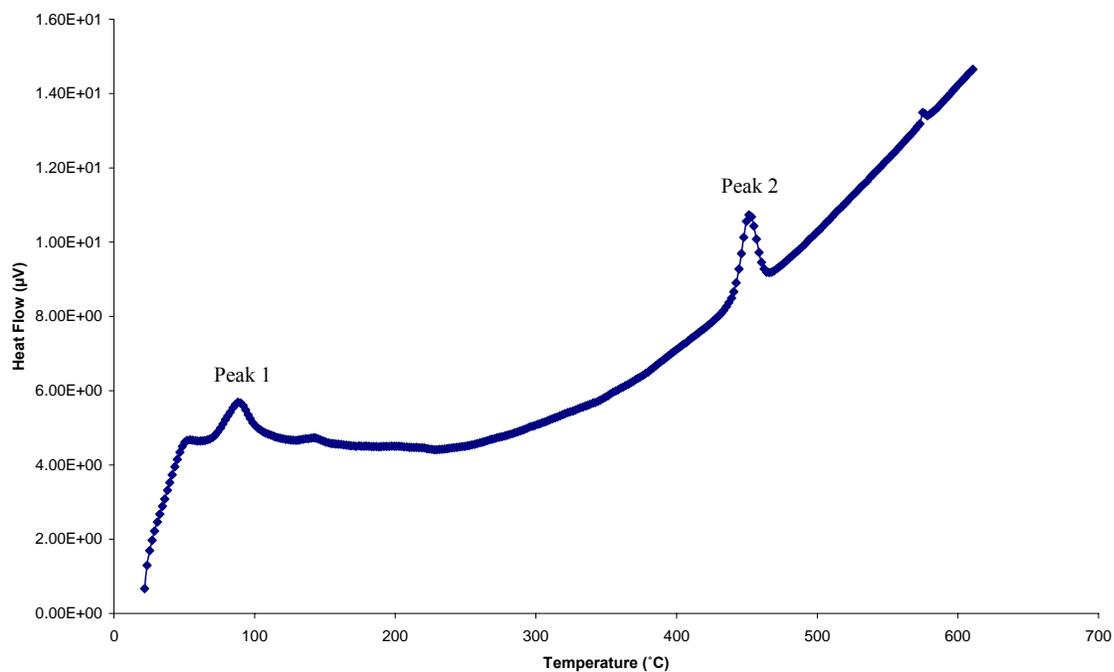


(i) NMA

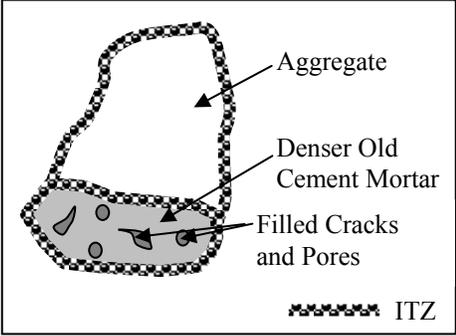


(ii) TSMA

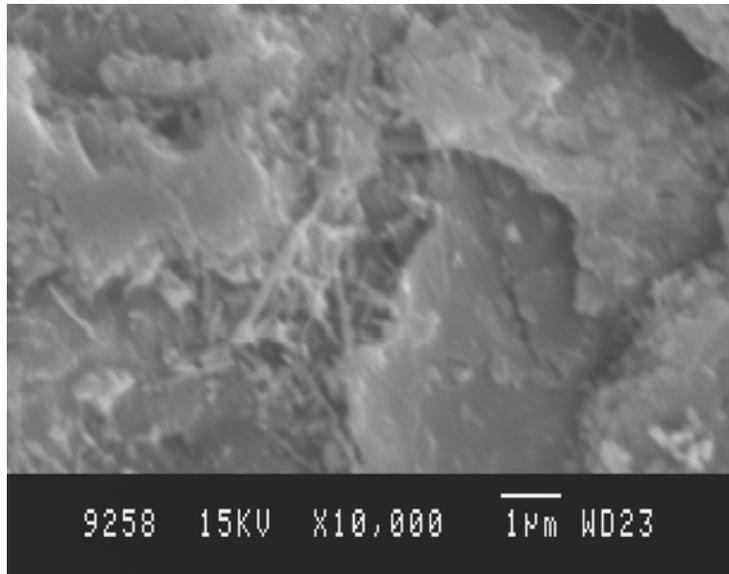
**Figure 4.** Mixing procedures of the (i) Normal mixing approach and  
(ii) Two-stage mixing approach



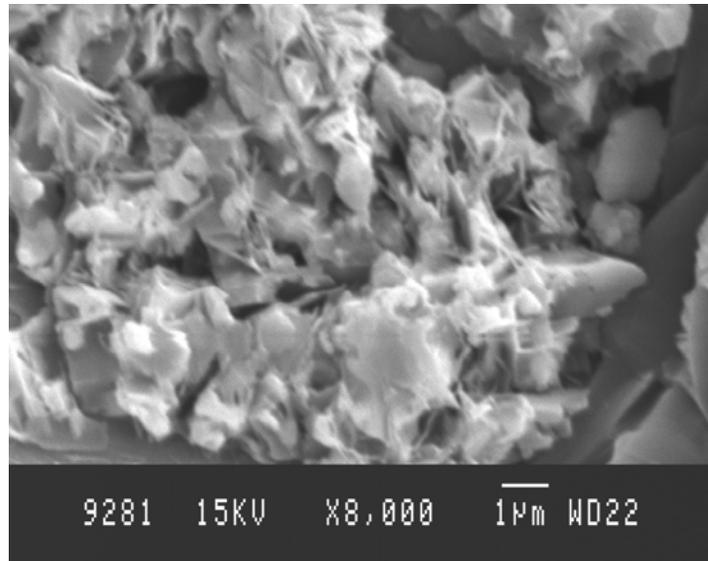
**Figure 5.** DSC curve for NMA with 0% RA substitution in 28-day curing.



**Figure 6.** RA's structure after adopting TSMA.

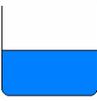


**Figure 7.** Crystallization of 20 percent RA substitution in TSMA.



**Figure 8.** Crystallization of 20% RA substitution in TSMA.

**Table 1.** Symbols used for representing various materials.

		
Fine Aggregate	Water	Recycled Aggregate + Natural Coarse aggregate
		
Half of the required Water	Cement	Concrete

**Table 2.** Summary of endothermic peak and peak area of DSCs results.

Curing Condition (days)	Sample		<i>CSH</i>		<i>CH</i>	
	Mixing Approach	RA Substitution (%)	Endothermic Peak (°C)	Peak Area ( $\mu\text{V}^\circ\text{C}$ )	Endothermic Peak (°C)	Peak Area ( $\mu\text{V}^\circ\text{C}$ )
28	NMA	0	88.19	21.85	453.15	25.35
		20	88.21	18.38	446.14	25.30
		100	88.60	12.97	453.18	21.23
	TSMA	0	86.46	23.07	442.39	27.35
		20	93.48	34.21	446.01	48.31
		100	87.95	20.45	444.18	28.61
56	NMA	0	86.46	24.24	442.39	26.45
		20	87.95	15.85	447.70	18.96
		100	84.87	7.98	443.23	11.35
	TSMA	0	89.80	25.92	446.07	29.33
		20	95.50	25.44	447.69	44.59
		100	88.04	9.19	449.55	20.16

**Table 3.** Summary of the results on compressive strength testing.

Mixing Methods		NMA (days)		TSMA (days)	
		28	56	28	56
Compressive Strength (MPa)	0%	55.7	67.6	56.0	68.0
	20%	53.7	68.8	65.1	72.0
	100%	48.3	52.7	55.6	69.6