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The effect of waste oil-cracking catalyst on the compressive strength of cement pastes and mortars

Jung-Hsiu Wu, Wan-Lung Wu, Kung-Chung Hsu*

Department of Chemistry, National Taiwan Normal University, Taipei 116, Taiwan

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Abstract

Epcat, one of the spent fluid catalytic cracking (FCC) catalysts from oil-cracking refineries, shows pozzolanic activity. In this study, pastes and mortars with Epcat were prepared and cured, and their compressive strengths after 3, 7 and 28 curing days were measured. The water/binder (W/B) ratios were 0.2, 0.25 and 0.3, and the replacement levels of cement by Epcat were 0, 5, 10 and 15 wt.%. Proper amount of superplasticizer was added into each mix to ensure similar workability. The results indicate that the presence of Epcat would increase the compressive strength of mortars substantially, but increase the compressive strength of the related pastes only slightly. Epcat mortars with W/B = 0.25 show more strength-enhancing effect than those with W/B = 0.3, and this effect increases with the catalyst content. Therefore, the mix (W/B = 0.25) incorporated 15% Epcat exhibits the greatest compressive strength (92.3 MPa). For mortars with W/B = 0.2, the strength-enhancing effect occurs only for those containing 5% catalyst; this effect becomes unclear when mixes containing 10% Epcat or more because high dosage of superplasticizer was added in obtaining proper workability and that affects the strength development. The improvement in the mechanical properties of mortars is attributed to the increase in the hydrated cement paste itself and, more importantly, improved bonds between the cement paste and aggregate.

Keywords: Waste catalyst; Pozzolan; Cement paste; Mortar; Compressive strength

1. Introduction

As the world economy continues to grow and technology continues to advance, more and more industrial wastes would be produced and the disposal or treatment of these wastes would become a severe challenge. One possible way to resolve this situation is to use wastes as mineral admixtures in concrete. It would not only reduce the consumption of energy in the production of structural materials by the partial substitution of cement, but would also contribute to the environmental protection. Among the industrial wastes, silica fume, fly ash and blast-furnace slag are some that can be used for such a purpose. These materials have cementitious or pozzolanic properties. Incorporation of these in concrete would not only solve the disposal problem, but also improve some properties of concrete. Physically, these materials act as microfillers in enhancing the packing in cement paste itself and at the cement paste-aggregate particle interface, and forming a denser and more homogeneous microstructure in the transition zone. Chemically, they undergo the pozzolanic reaction by reacting with calcium hydroxide (CH) in the cementitious system and accelerate the hydration of cement. The finer and the more vitreous the pozzolans, the faster is their reaction with CH. Therefore, concretes and mortars that incorporate these materials show improved compressive strength and durability [1–4].

Spent fluid catalytic cracking (FCC) catalyst, the waste material from the FCC unit of oil refineries, is composed mainly of silica and alumina. This catalyst is initially used in the FCC unit to cause hydrocarbon molecules to break into two or more smaller molecules. The FCC unit

* Corresponding author. Tel.: +886-2-2930-9088; fax: +886-2-2932-4249.

E-mail address: kchsu@cc.ntnu.edu.tw (K.-C. Hsu).

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Table 1
Basic properties of cement and Epcat

	Cement	Epcat
Composition (%)		
SiO_2	20.0	47.0
Al_2O_3	5.4	38.0
Fe_2O_3	3.4	< 1.0
CaO	63.2	–[a]
MgO	2.3	< 1.0
K_2O	< 1.0	< 1.0
TiO_2	< 1.0	< 1.0
V_2O_3	–	< 1.0
SO_3	2.0	5.6
Ignition loss (%)	0.9	8.9
Mean particle size (μm)	5.2	1.7
BET specific surface area (m^2/g)	0.951	47.3
Specific gravity	3.11	2.38

[a] Not measurable.

usually includes three vessels, i.e., a converter, a stripper and a regenerator. During the catalytic operation in the converter, some cracked products or coke will deposit on the catalyst particles. However, they are removed or burned off either in the stripper or in the regenerator. Therefore, spent catalyst from the FCC unit contains minute inorganic oxides and almost no organic compounds. Currently, they are classified as nonhazardous materials, and most of them are solidified and disposed off as landfills [5]. However, interest in the utilization of spent FCC catalyst as construction material has been growing in recent years and so researches are in this area. For examples, Pacewska et al. [6] have examined the pozzolanic nature of this catalyst using thermal and spectroscopic techniques and have disclosed that its ability to react with CH is similar to microsilica. Paya et al. [7] have demonstrated the high activity of the previously ground catalyst. Su et al. [8,9] and Paya et al. [10] carried out a feasibility study of reusing this waste catalyst and their results indicate that the additive can replace up to 15–20% of cement or 10% of fine aggregate without sacrificing the quality of mortars.

It is clear that spent FCC catalyst, like silica fume, can increase the compressive strength of the resulting cementitious materials [6–10]. Regarding the mechanism for the improvement of strength by using silica fume in mortars or concretes, some results indicate that it is due to the improved bond strength between the cement paste and the aggregate, while others reveal that an increase in the strength of the cement paste itself is the main cause [11–18]. It would be of interest to understand the role of this waste catalyst in cementitious composites. This study aims at examining and comparing the compressive strengths of both pastes and mortars with or without the presence of the spent catalyst, so that the strength-enhancing mechanism by this industrial waste could be further understood.

2. Experimental

2.1. Materials

The materials used include Type I Portland cement, quartz sand, spent FCC catalyst (Epcat) as a mineral admixture and a superplasticizer. Cement was from the Taiwan Cement and complies with ASTM C150. The quartz sand used had a specific gravity of 2.6 and a fineness modulus of 2.16. Epcat, coming from the China Petroleum, is a waste catalyst in the form of catalyst fines, which had been generated in FCC units and removed from the regenerator off-gas, and collected by an electrostatic precipitator. Table 1 lists the basic properties of cement and the spent catalyst. Epcat consists mainly of SiO_2 and Al_2O_3, and other minute metal oxide impurities. However, this spent catalyst contains some SO_3 (5.6%) and has high ignition loss (8.9%). As will be shown later, the presence of SO_3 facilitates the formation of Aft in hydrated pastes. Fig. 1 shows the particle size of Epcat particles ranging from 0.1 to 10 μm, with a mean particle size of 1.7 μm. The BET specific surface area is huge (47.3 m^2/g). Fig. 2 shows the scanning electron micrograph (SEM) of Epcat, indicating that this catalyst has irregular shape. Fig. 3 shows the XRD pattern of an Epcat. This catalyst appears to be a crystalline material with some amorphous phase in the structure. The crystallized phases been identified include faujasite, quartz, kaolinite and mullite. In addition, the gypsum peak was also found, indicating that Epcat does contain some SO_3. A commercial superplasticizer (MTP), manufactured by the HI CON Chemical Admixture Taiwan, was used. MTP is a copolymer of multicarboxylic acid/salts with 58% in solid content and relative density of 1.04. The superplasticizer was used to adjust the workability of tested samples.

2.2. Preparations of cementitious samples

Both pastes and mortars with or without Epcat were mixed in a Hobart Mixer. Cement and one-half of tap water were

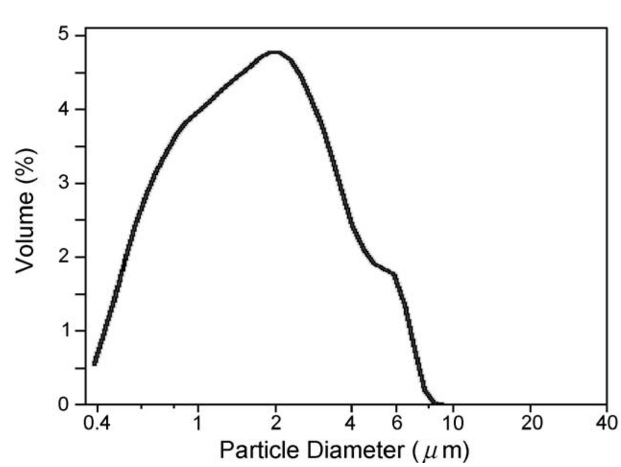

Fig. 1. Particle size distribution of Epcat particles.

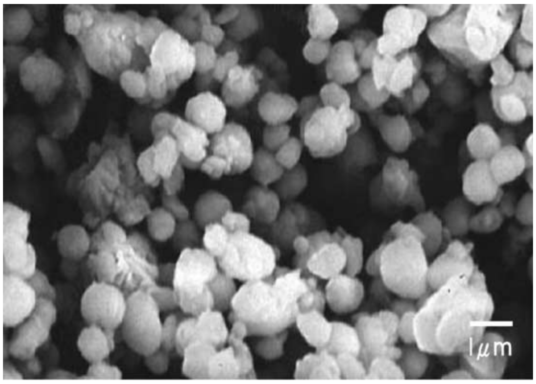

Fig. 2. SEM micrograph of Epcat particles.

placed in the mixer, mixed at a slow speed for 3 min. Epcat or sand was then placed and mixed at a slow speed for 1 min. Finally, the remaining water with superplasticizer were added and mixing was continued for 3 min at a medium speed.

The cementitious samples were prepared with three water/binder (W/B) ratios, i.e., 0.2, 0.25 and 0.3; replacement levels of cement by Epcat were 0, 5, 10 and 15 wt.%. The binder/sand ratio for mixed mortars was fixed at 1/1.25. Proper amount of MTP was added into mortars to obtain similar workability.

2.3. Particle size and surface area measurements

The particle size of Epcat particles was measured by a particle size analyzer (Coulter LS 230), and the BET specific surface area was measured by a surface area analyzer (Micromeritics ASAP 2010).

2.4. Setting time measurements

The setting time of cement pastes was measured by an automatic tester with the Vicat needle according to ASTM C191.

2.5. Scanning electron microscope (SEM) observation

The microstructure of Epcat particles and those of mortars were observed by an SEM (JEOL JSM-6300).

2.6. X-ray diffraction (XRD) analysis

The mineralogy of either an Epcat or cement pastes was analyzed by a powder X-ray diffractometer (JEOL JDX-8030).

2.7. Differential scanning calorimetry (DSC) measurements

Cement pastes were prepared and cured under saturated water vapor pressure at the ages of 3, 7 and 28 days. About 25 mg of cured samples were taken and ground thoroughly. They were then put in a sample holder and tested immediately in a differential scanning calorimeter (SHIMADZU TA-50). All measurements were conducted at a heating rate of 20 °C/min up to 550 °C.

2.8. Workability test

The workability of mortars was determined and indicated by the spread diameter of tested samples on the flow table according to ASTM C230.

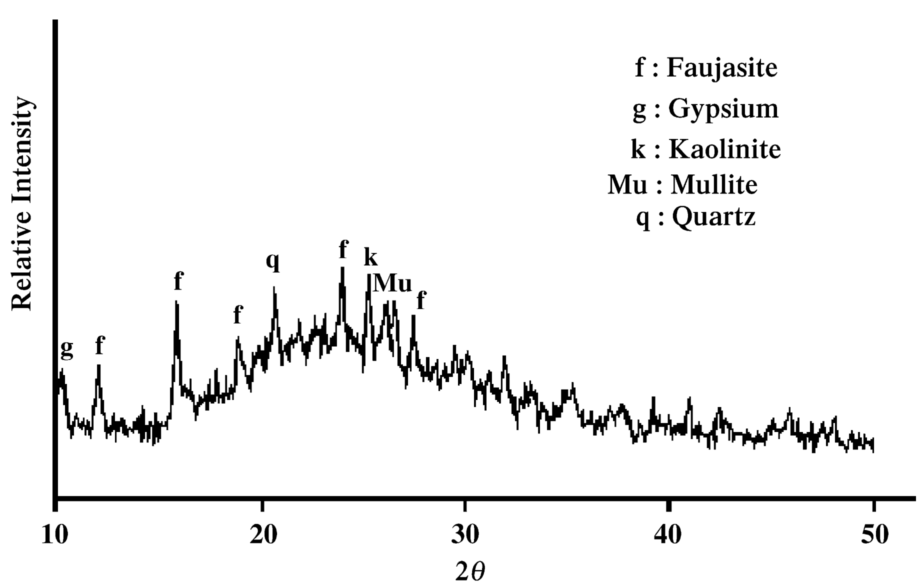

Fig. 3. XRD pattern of an Epcat.

2.9. Compressive strength test

Either paste or mortar samples of 5 × 5 × 5 cm were prepared, cured and their compressive strengths measured at the curing ages of 3, 7 and 28 days, according to ASTM C109. Each strength value is an average of three measured data.

3. Results and discussion

The workability and compressive strengths of both the cement pastes and mortars, with or without the presence of Epcat, are listed in Table 2. In order to determine the true effect of Epcat on the compressive strength of the cementitious mixes, all tested samples with or without catalyst incorporated were controlled to have similar workability or close spread diameter on the flow table so that their structures were initially homogeneous with similar packing. The spread diameter of either pastes or mortars was about 13–15 cm for samples with 0.2 or 0.25 W/B ratio and about 16–18 cm for those with W/B = 0.3. When W/B is decreased or Epcat content is increased, the required amount of superplasticizer increases. As mentioned earlier, the waste catalyst used is small in particle size and huge in surface area, and it would have high affinity to water. Samples containing it would require more water, or more chemical admixture with same W/B ratio, to achieve similar workability. As shown in Table 2, the required dosages of superplasticizer (MTP) are 0.05–0.20%, 0.15–0.40% and 0.20–2.5% for samples with W/B equal to 0.3, 0.25 and 0.2, respectively. It is noted that MTP would extend the setting time of cement pastes, as shown in Fig. 4. Furthermore, the setting time of cement pastes increases with the MTP content. According to ASTM C150, the initial setting time (T_i) for Type I Portland cement pastes should be no less than 45 min and the final setting time (T_f) no more than 375 min. As shown in Fig. 4, only the paste sample without superplasticizer has setting time meeting the requirement just mentioned. For samples containing 0.05% MTP or more, all their T_f values are more than 375 min, confirming that the added chemical admixture would delay the cement hydration. Fig. 5 shows the effect of Epcat content on the final setting time of cement pastes. The mix proportions of paste samples are listed in Table 2. Namely, all these samples contain both Epcat and MTP; pastes incorporated higher catalyst content or with lower W/B value clearly have more superplasticizer included. Compared the results from Fig. 5 to those from Fig. 4, it reveals that the presence of Epcat would accelerate cement hydration and shorten the setting time. For examples, the T_f value of pastes (W/B = 0.3) with Epcat = 5% and MTP = 0.10% is 4.25 h (Fig. 5), which is lesser than the value of those with MTP = 0.10% only (Fig. 4). For paste samples with W/B value equal to 0.25 or more, their T_f values (shown in Fig. 5) generally decrease with Epcat content, indicating that the accelerating effect by the catalyst predominates over the delaying effect by the chemical admixture. However, cement pastes with W/B = 0.2 show the opposite trend. Besides, incorporated 10% or 15% Epcat in the mixes require 10.25 h or more to reach the final setting

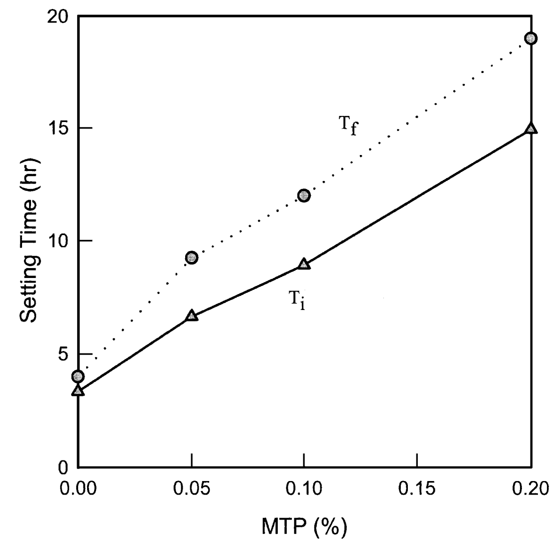

Fig. 4. The effect of MTP content on the setting time of cement pastes (W/B = 0.3).

Table 2
Workability and compressive strength of cementitious samples

Material	W/B	Epcat/B (%)	SP/B (%)	Workability (cm)	Compressive strength (MPa)		
					3 days	7 days	28 days
Cement paste	0.20	0	0.20	13–15	74.0	74.9	93.3
		5	0.60	13–15	74.7	83.9	89.8
		10	1.00	13–15	68.2	87.7	88.9
		15	2.50	13–15	5.5	69.8	72.1
	0.25	0	0.15	13–15	61.0	63.6	69.8
		5	0.20	13–15	62.2	66.4	73.3
		10	0.30	13–15	70.4	75.4	77.8
		15	0.40	13–15	72.3	77.6	78.0
	0.30	0	0.05	16–18	55.2	64.2	82.3
		5	0.10	16–18	59.6	65.1	87.2
		10	0.15	16–18	62.3	66.6	87.7
		15	0.20	16–18	63.1	70.6	83.5
Mortar	0.20	0	0.20	13–15	71.0	74.5	78.8
		5	0.60	13–15	75.9	87.9	91.9
		10	1.00	13–15	73.0	83.4	84.2
		15	2.50	13–15	15.9	49.9	55.4
	0.25	0	0.15	13–15	58.2	60.5	69.1
		5	0.20	13–15	65.9	67.3	78.9
		10	0.30	13–15	71.6	79.0	83.4
		15	0.40	13–15	73.0	79.8	92.3
	0.30	0	0.05	16–18	46.8	57.6	68.7
		5	0.10	16–18	61.9	69.8	77.9
		10	0.15	16–18	62.7	72.4	84.4
		15	0.20	16–18	65.9	75.9	90.4

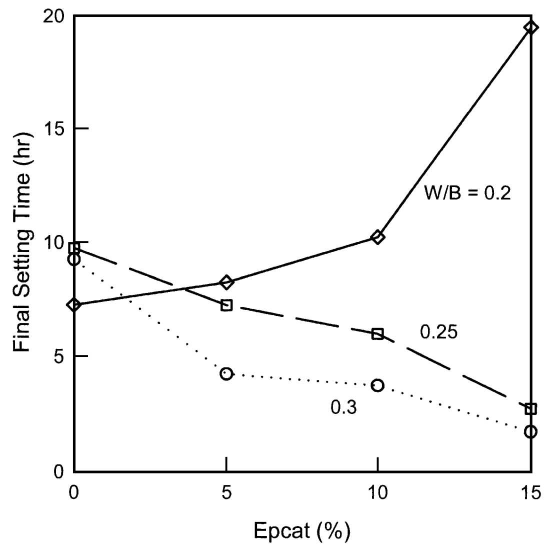

Fig. 5. The effect of Epcat content on the final setting time of cement pastes.

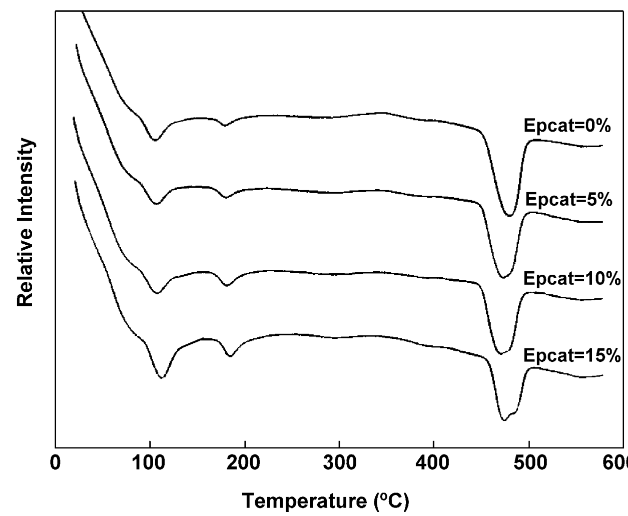

Fig. 7. DSC curves of hydrated pastes cured at 3 days (W/B = 0.3).

state. This discloses that strong set retardation would take place if too much superplasticizer is added to mixes, and this affects the strength development of cementitious composites seriously. As listed in Table 2, the compressive strength of mortars is generally increased with increasing catalyst content. However, mortars (W/B = 0.2) with 15% Epcat shows much less strength values than those with 0–10% Epcat cured at 3–28 days, for the former containing much more superplasticizer (MTP = 2.5%) than the latter.

Fig. 6 shows the effect of Epcat content on the compressive strength of cementitious samples with W/B = 0.3, cured at different ages. Apparently, pastes containing the spent catalyst exhibit slightly higher values in compressive strength than those without. As the replacement level of Epcat is increased, the strength value of the resulting pastes also becomes higher. Separately, the result from Pacewska et al. [6] also indicates that pastes with 25% spent catalyst have higher compressive strengths than those without. Two reasons are responsible for the higher strength of the Epcat paste. First, Epcat particles are very small and act as fillers for the spaces between cement grains. This results in a reduction in the pore sizes or voids in the paste. Second, the waste catalyst shows the pozzolanic activity [17]. Fig. 7 shows the DSC curves of hydrated cement pastes with different Epcat contents at the age of 3 days. Addition of Epcat would decrease the heat energy or intensity of CH peak (450–480 °C), but increase that of C–S–H peak (80–100 °C) and that of Aft peak (180–190 °C). As the replacement level of the catalyst increases, the corresponding CH peak becomes smaller but the C–S–H peak and Aft

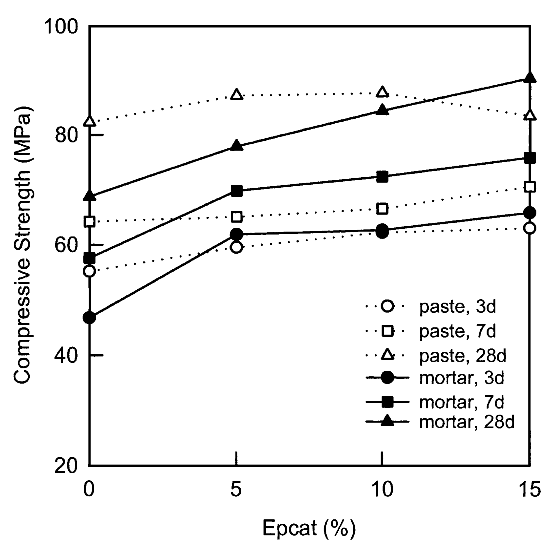

Fig. 6. The effect of Epcat content on the compressive strength of samples cured at different ages (W/B = 0.3).

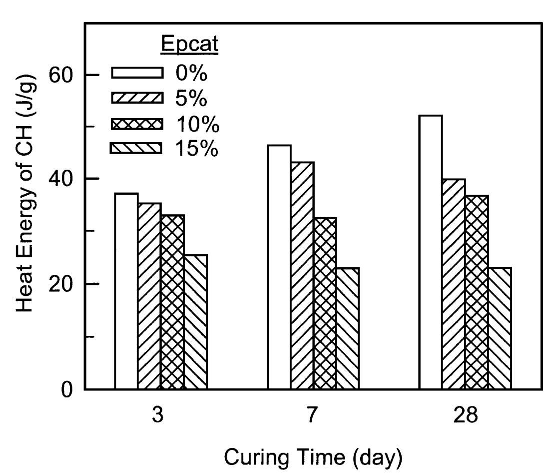

Fig. 8. The heat energy of CH of hydrated pastes cured at different ages (W/B = 0.3).

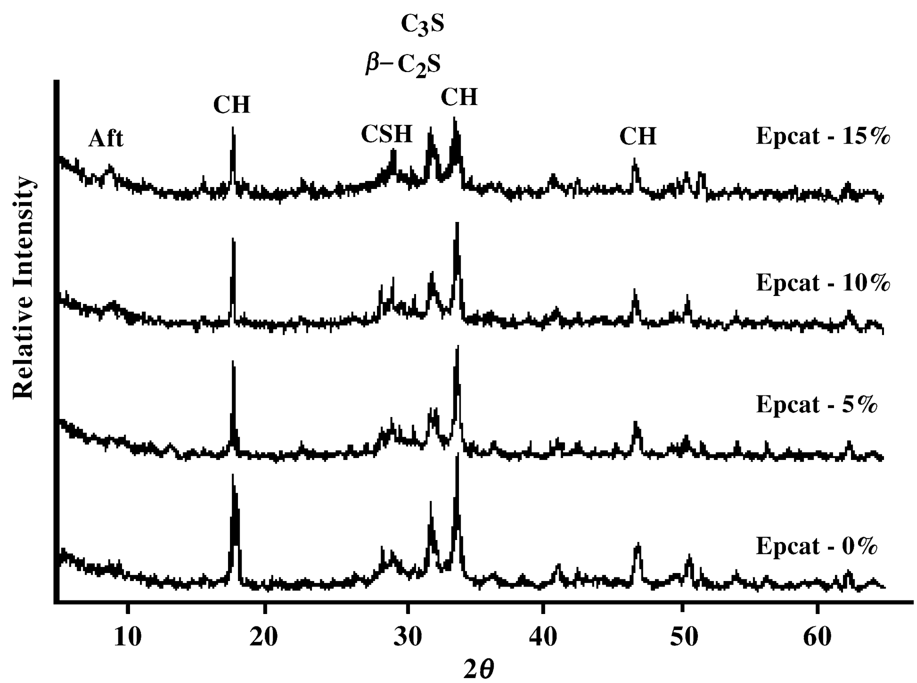

Fig. 9. XRD patterns of hydrated cement pastes containing 0–15% Epcat cured at 28 days (W/B = 0.3).

peak become larger, suggesting that more C–S–H gel and Aft but less CH produced for pastes with greater amount of Epcat present. Fig. 8 shows the heat energy of CH of hydrated pastes cured at different ages. The heat energy of CH clearly decreases with the catalyst content at same curing age. Fig. 9 shows XRD patterns of hydrated cement pastes containing 0–15% Epcat (W/B = 0.3) cured at 28 days. All patterns appear to be similar, but the intensities of cement components (C_3S, β-C_2S) and hydration products (CH and C–S–H) are different. It is clear that the intensity of C_3S + β-C_2S (2θ = 32.1°, 32.6°) and that of CH (2θ = 18°) decreases, but that of C–S–H (2θ = 29°) increases slightly with the Epcat content [19,20]. Both DSC measurements or XRD results indicate that the studied catalyst indeed shows good pozzolanic activity.

As shown in Fig. 6, the effect of Epcat content on the compressive strength of mortars is similar to that of pastes. However, the strength enhancement in mortars appears to be more significant than that in pastes, as the former was observed to have higher rate of strength gain than the latter. It is obvious that the pozzolanic materials act as microfillers in improving the interface and as a result of this, the strength increases. Fig. 10 shows SEM micrographs of mortars (W/B = 0.3) cured at 7 days and containing 0% and 15% Epcat. The mortar with no waste catalyst appears to be more porous and there is a separation between cement paste and aggregate. In contrast, the Epcat mortar is denser and more consolidated, and the separation in the interfacial region reduces drastically. This proves that the presence of Epcat results in an improved bond between cement paste and sand. As mentioned before, this waste catalyst is an active pozzolan, which would produce new C–S–H gel from the reaction between Epcat and CH in the interfacial area, generating the improved bond.

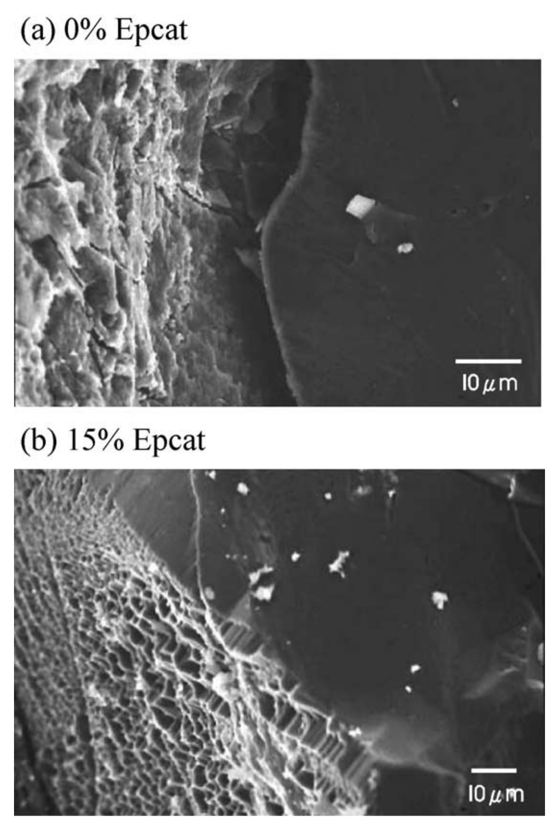

Fig. 10. SEM micrographs of mortars cured at 7 days with (a) 0 and (b) 15% Epcat (W/B = 0.3).

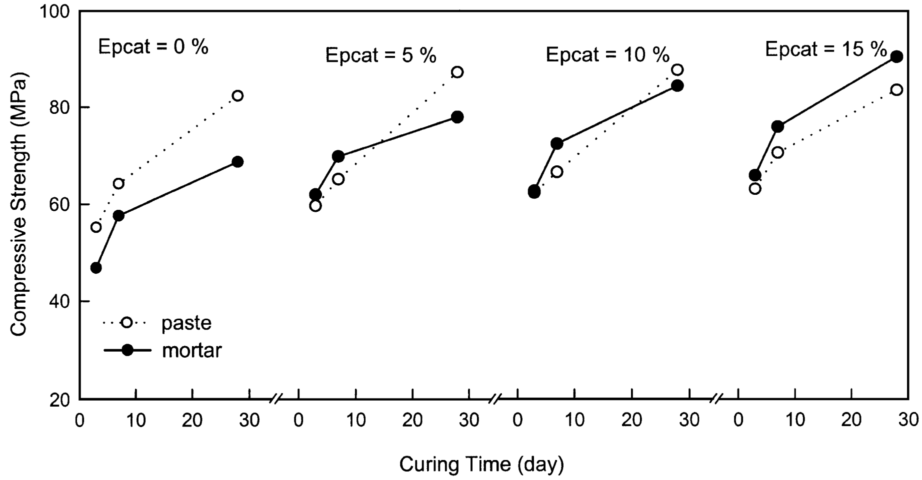

Fig. 11. The effect of curing time on the compressive strength of samples with different Epcat contents (W/B = 0.3).

On the other hand, Fig. 11 shows the effect of curing time on the compressive strength of cementitious samples (W/B = 0.3) with different Epcat contents. For samples with no waste catalyst, the compressive strength of mortars is expectedly less than those of pastes, as aggregate acts as an inert filler to induce a dilution effect and a weaker paste/aggregate interfacial zone. When the spent catalyst is present, the compressive strength of the resulting mortars is improved and is higher than that of pastes, especially at earlier curing period, i.e., from 3 to 7 days. In other words, a significant strength-increasing rate along with curing time was observed in mortar samples up to 7 days, followed by a period of low increment rate up to 28 days. This indicates that the contribution of Epcat by the pozzolanic reaction is more significant at earlier period. Furthermore, more catalyst in mortars would cause stronger pozzolanic reaction, result in more strength enhancement and last longer. Accordingly, mortars with catalyst content of 15% have better mechanical properties than pastes at whole tested curing period.

Fig. 12 shows the effect of Epcat content on the compressive strength of cementitious samples with W/B = 0.25, cured at different ages. The results are similar to those with W/B ratio of 0.3. Namely, the plain paste has higher strength than the plain mortar. Epcat generates more strength-improving effect in mortars than in pastes. Nevertheless, Epcat mortars always show greater strength values than the corresponding pastes, suggesting that the strength enhancement is more at lower W/B value. In addition, this strength-enhancing effect increases with the catalyst content. Therefore, the mix incorporated 15% Epcat exhibits the greatest compressive strength (92.3 MPa). Fig. 13 shows the effect of Epcat content on the compressive strength of cementi-

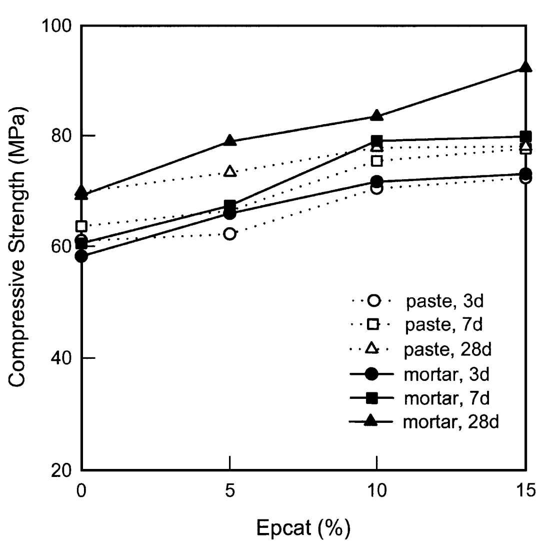

Fig. 12. The effect of Epcat content on the compressive strength of samples cured at different ages (W/B = 0.25).

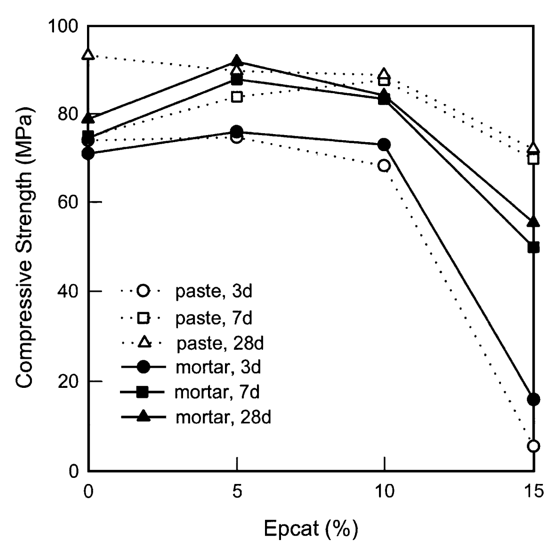

Fig. 13. The effect of Epcat content on the compressive strength of samples cured at different ages (W/B = 0.2).

tious samples with W/B = 0.2, cured at different ages. The results are unlike the previous. For either pastes or mortars, the compressive strength increases with Epcat content up to 5%, and then decreases subsequently. This is due to the occurrence of an extensive set retardation induced by the superplasticizer used. As shown in Table 2, superplasticizer of 1.0% or more was required in samples with Epcat >10% to achieve workability having spread diameter of 13–15 cm. When samples contain such high dosage of superplasticizer, strong retarding effect would take place. Consequently, the strength values for both pastes and mortars with 10% or 15% Epcat replacement levels become less than those with lower catalyst content.

Finally, Figs. 14 and 15 show the aggregate effect and interface response of samples at different curing ages and two W/B ratios. The relative strength is defined as the ratio of the compressive strength of Epcat mortars to that of Epcat cement pastes. It was seen from Fig. 14 that the relative strength (W/B = 0.3) is increased with curing time, reaches maximum value at 7 days and then decreases thereafter. The relative strength of samples with the catalyst is greater than one at either 3 or 7 curing ages, and less than one at 28 days except for 15% Epcat replacement level. As mentioned earlier, the addition of active pozzolans would improve the strength of cementitious materials by (1) making stronger cement pastes and (2) enhancing the paste/aggregate interfacial bond [1,16,17]. Apparently, the latter contributes more than the former for samples cured at earlier period. In contrast, the relative strength of mortars with reduced W/B ratio is always greater than one, indicating that the contribution by an improved interfacial bond is more significant in the whole tested curing period (Fig. 15). In the case of silica fume, it is still under debates as to which contribution is more prominent in improving the strength of mortars or concrete [11–18]. Regarding the case of spent catalyst studied, either one could contribute more than another, depending on mix proportions and curing ages.

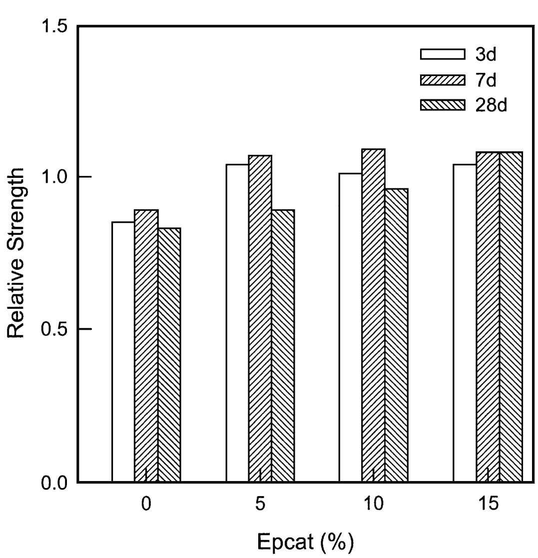

Fig. 14. The relative strength of samples at different curing ages (W/B = 0.3).

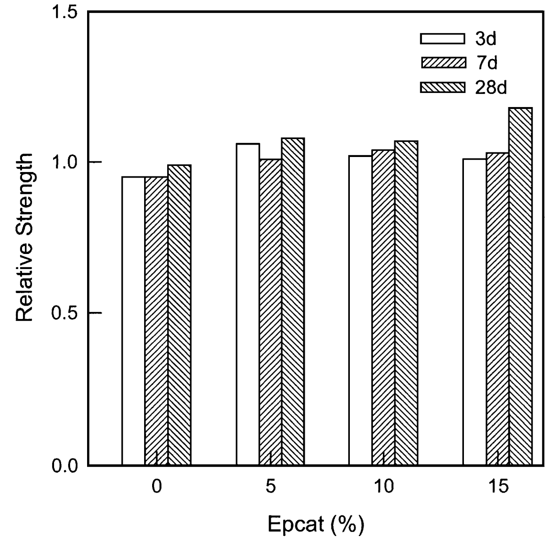

Fig. 15. The relative strength of samples at different curing ages (W/B = 0.25).

4. Conclusions

Epcat, one of the spent FCC catalysts from oil-cracking refineries, shows pozzolanic activity from both DSC and XRD analyses of hydrated cement pastes with this additive present. Addition of Epcat would increase the compressive strengths of mortars substantially, but increase those of the related pastes only slightly. Epcat mortars with W/B = 0.25 shows more strength-enhancing effect than those with W/B = 0.3, and this effect increases with the catalyst content. Consequently, the mix (W/B = 0.25) incorporated 15% Epcat exhibits the greatest compressive strength (92.3 MPa). For mortars with lower W/B ratio (W/B = 0.2), the strength-enhancing effect occurs only for those containing 5% catalyst. The aforementioned effect becomes unclear when mixes containing 10% Epcat or more, because high dosage of superplasticizer (MTP $\geq$ 1%) was added to achieve proper workability and that affects the strength development. The improvement in the mechanical properties of mortars could be attributed to the increase in the hydrated cement paste itself and, more importantly, improved bonds between the cement paste and aggregate. However, the former could contribute more than the latter, depending on mix proportions and curing ages.

Acknowledgements

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