Polished sample preparing and backscattered electron imaging and of fly ash-cement paste

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Abstract. In recent decades, the technology of backscattered electron imaging and image analysis was applied in more and more study of mixed cement paste because of its special advantages. Test accuracy of this technology is affected by polished sample preparation and image acquisition. In our work, effects of two factors in polished sample preparing and backscattered electron imaging were investigated. The results showed that increasing smoothing pressure could improve the flatness of polished surface and then help to eliminate interference of morphology on grey level distribution of backscattered electron images; increasing accelerating voltage was beneficial to increase gray difference among different phases in backscattered electron images.

1. Introduction
Fly ash is now applied widely in cementitious materials preparation as a kind of admixture and the hydration property of fly ash-cement pastes is always research hotpot[1,2].

Researchers have paid a lot of attention on investigation of micro-performance of fly ash-cement paste. However, the works were mostly limited to qualitative analysis other than quantitative analysis. It was due to the coupling effect between fly ash and cement increasing the difficulty of the quantitative test. From 1980s, the technology of backscattered electron imaging and image analysis(BSE-IA) has been concerned about by more and more researchers. Meanwhile, its superiority in research of blended cement is becoming more and more recognized. This technology was applied in cement paste study firstly by Scrivener in 1980s[3]. Then, it is increasingly applied in quantitative research of compound cement paste[4,5].

The technology of BSE-IA is composed of 3 parts-polished specimen preparing, backscattered electron imaging and image processing. Each part has great influence on accuracy of research results. Peiming wang et al. from Tongji university summarized several quality problems effecting on quantitative analysis including the flatness, scratches, holes, etc[6]. Polished specimen preparation has a complex working procedure, but there is few report about it could be referenced except for an article by K.O.Kjellsen which presented the rough procedure of polished sample preparation including resin inlays, grinding and polishing [7]. However, the effects of preparation procedure on specimen quality were not mentioned. The few works above are aimed at pure cement paste and the research work of BSE-IA aimed at fly ash-cement paste has yet to see. That limits the application of this technology seriously.

Above all, to provide certain guidance for application of BSE-IA technology, this work investigated effects of two factors including smoothing pressure and accelerating voltage on grey level distribution of backscattered electron image(BSE).

2. Materials and methods
2.1 Preparation of fly ash-cement paste
Hydration paste was prepared with 50% (by mass) Type I Portland cement replaced by fly ash. The chemical compositions of cement and fly ash were shown in table 1. Water-binder ratio of the paste was 0.5.
The pastes were cured at 20±1 °C water after 24 hours. When the pastes hydrated for 7 days, the hardened pastes were soaked in ethanol to terminate hydration. Then, they were dried in vacuum drying chamber at 40 °C.

|          | CaO  | SiO$_2$ | Al$_2$O$_3$ | Fe$_2$O$_3$ | MgO   | SO$_3$ | K$_2$O | TiO$_2$ | MnO   | SrO   |
|----------|------|---------|-------------|-------------|-------|--------|--------|---------|-------|-------|
| cement   | 64.50| 21.00   | 4.94        | 3.27        | 1.11  | 2.70   | 0.76   | 0.19    | 0.05  | 0.11  |
| fly ash  | 6.49 | 48.9    | 33.8        | 5.08        | 0.67  | 0.51   | 0.88   | 1.24    | 0.06  | 0.19  |

2.2 Preparation of Flat-polished sample
Firstly, samples were pre-smoothed by 240 grade sandpaper. Secondly, they were impregnated with epoxy under vacuum condition. Thirdly, the impregnated samples were smoothed using sandpapers from coarse to fine. Thirdly, the smoothed samples were polished on polishing cloth. For fear of excessive abrasion, the time of each smoothing and polishing step should be controlled in 2 min.

2.3 Acquisition of BSE image
BSE images of polished sample were acquired by a scanning electron microscope on which backscatter electron detector was installed. Before BSE imaging, the quality of polished sample was checked by secondary electron imaging.

3. Results and discussions
3.1. Effect of smoothing pressure on polished sample
Smoothing is the base of polished sample preparation. There are two main purposes of smooth process: firstly, remove coating resin layer on test surface to show the harden paste with resin inlays; secondly, smoothing test surface for the subsequent polishing operation. Grey level distribution of BSE figure is depended on both morphology and chemical composition of the test surface. Elimination of height difference among different phases can help to distinguish phases according to their chemical compositions, which has great influence on quantitative analysis by BSE-IA. There were few paper about preparation technology of polished harden cement paste sample at home and abroad. In our experiments about preparation of polished sample, it was found that pressure setting during smoothing process had influence on polished surface.

In this paper, experiment was designed to study on the effects of smoothing pressure on polished sample of fly ash-cement harden paste and the results were shown in fig.1.

The pressure of 5 N, 10 N, 15 N and 20 N were chosen in our work. Then, the flatness of polished surface was estimated through morphology observation by secondary electron(SE) images. It could be seen that the polished surfaces of the paste were uneven apparently when pressure of 5 N and 10 N were applied, unhydrated clinker and fly ash particles were raised obviously as shown in fig.1(a),1(b). With smoothing pressure increasing, the flatness of polished surface was improved. Unhydrated particles were almost even with hydrates at pressure of 15 N as shown in fig.1(c). However, continue increasing of pressure showed no obvious benefit on flatness improving (fig.1(d)).

From the above, it could be concluded that the sample showed uneven surface under secondary electron imaging when it was prepared with lower smoothing pressure, and the flatness of surface could be improved by increasing smoothing pressure properly. This is because the harden fly ash-cement paste is a kind of unisotropic body with unhydrated particles much harder than hydrates. Hydrates could be ground while harder unhydrates couldn’t be at lower pressure. Suitable smoothing pressure for fly ash-cement paste is 15 N according to our work. Need to point out that overlarge pressure increases the risk of excessive grinding and also should not be applied in sample preparation like lower pressure.
Figure 1. SE images showing polished surfaces of samples prepared with different smoothing pressure

Effects of flatness on grey level distribution of BSE images could be concluded by comparing the BSE images of two polished surfaces with low and high flatness respectively as shown in fig.2. Compared to BSE image of low flatness surface, the grey level of unhydrate clinker particles was higher in BSE image of high flatness surface. It was due to that raised morphology of unhydrate particles increased their grey level in BSE image. BSE figure of contrast was affected by both morphology and average atomic number. The average atomic number of cement clinker were much higher than hydrates. On polished surface with low flatness(fig.2(a)), unhydrate clinker particles was raised from hydrates and the raised morphology further increased intensity of backscattered electron bounced from unhydrate clinker particles. Then, difference of grey level between hydration particles and hydrates increased (fig.2(b)). While on polished surface with high flatness(fig.2(c)), there is no obvious difference in height between hard phases and soft phases (fig.2(c)) and the grey level distribution of phases in BSE image was decided by their average atomic number only. With no enhancement of their raised morphology, unhydrate clinker particles showed smaller grey level difference with hydrates(fig.2(d)).
3.2. Accelerating voltage

According to the principle of scanning electron imaging, accelerating voltage has an effect of both image resolution and image contrast. As shown in fig.3, different values of accelerating voltage were set during backscattered electron imaging to investigate the effects of that on grey level distribution of BSE image.

There is no significant difference of grey level distribution between BSE image taking with 15 kV (fig. 3(a)) and 20 kV (fig. 3(b)). In both BSE images, several different phases could be identified by their grey feature such as ferrite particles from anhydrate fly ash, anhydrate cement clinker and porosity. There are other phases that can’t be identified by grey feature since their grey level overlap such as silica-acuminate spherical particles, CH and other hydrates. Some fly ash particles could be distinguished by their morphology features, which is helpful to qualitative analysis while helpless to quantitative analysis. As shown in fig.3(c), there is an obtuse angle peak of grey level ranging from 50 to 150 that consisted of different phases such as fly ash, CH and other hydrates. The grey level peak located in range between 150 to 200 refers to anhydrate clinker. It could be seen that the grey level peak of anhydrate clinker moves to the right and grey difference with other phase increases as the accelerating voltage increases. The results proved that increasing accelerating voltage could increase grey difference among different phases and width of grey distribution. These phenomenon could be explained by following formulas.

\[
i_b = k_1 \cdot \frac{1}{E_a^z}
\]

(1)

\[
i_b = k_2 \cdot E_a
\]

(2)

\[
E_a \geq k_3 \cdot \frac{1}{C^z}
\]

(3)

\(i_b, E_a, C\) mean intensity of backscattered electron, accelerating voltage, backscatter coefficient respectively. \(k_1, k_2, k_3\) are physical constant.
4. Conclusions

BSE-IA is a useful technology for research of fly ash blended cement paste. The operation of polished specimen preparing and backscattered electron imaging have great influence on grey level distribution of BSE images, which further affect accuracy of analysis result.

It was shown that that increasing smoothing pressure could help improve the flatness of polished surface and help to eliminate the effect of morphology on grey level distribution. For sample of fly ash-cement paste, polished surface with high flatness couldn’t be acquired with smoothing pressure lower than 15 N. Grey difference among different phases and width of grey distribution increased with increasing of accelerating voltage. For sample of fly ash-cement paste, accelerating voltage of 15 kV was enough for backscattered electron imaging.

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