Influence of Recrystallized Silica Aggregates on Alkali-Silica Reactivity

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\textbf{ABSTRACT}

The potential reactivity of aggregates and the influence of recrystallized silica was studied using several test methods—namely, oxide composition analysis, accelerated mortar-bar testing (AMBT), accelerated concrete microbar testing (ACMT) and analysis of microscopic characteristics using a digital stereo microscope and a scanning electron microscope (SEM). Three different types of greywacke aggregate from the same source were investigated; greywacke aggregate (GRA), moderate quartz greywacke aggregate (MGA) and recrystallized aggregate (REA). The results indicated that all studied aggregates were quite similar in oxide composition. Thin-section analysis indicated that GRA and MGA were coarse-grained greywacke composed of quartz feldspar rock fragments. However, REA was high weathering chlorite with a matrix of re-crystallized quartz and rock fragments altered by tectonic forces. The findings indicated that the recrystallized aggregates increased the alkali silica reaction risk and may be considered as having a more deleterious effect than the other aggregates. The largest expansion was observed from both the AMBT and AMCT methods. The results from stereomicroscopy of concrete thin-section analysis and the SEM study confirmed this finding.

\textbf{Keywords:} alkali silica reaction (ASR), recrystallized silica, petrographic, ASR gel, accelerated test

\textbf{INTRODUCTION}

The first evidence of alkali silica reaction (ASR) (Sujjavanich \textit{et al.}, 2010; Sujjavanich \textit{et al.}, 2012a) and continued research works on local aggregates (Sujjavanich \textit{et al.}, 2012b) suggest a high risk of the occurrence of ASR problems in Thailand in the near future as its importance regarding concrete deterioration may be higher than was thought. Many studies have indicated a reaction due to the destructive effect of hydroxyl ions at the aggregate surface in the disintegration of the silicate network (Figg, 1983; Helmuth \textit{et al.}, 1993). This attack depended on the variability of the crystal structure of silica (Ferraris, 1995). However, there is still an unclear understanding of the effects of transformed silica, due to the recrystallization phenomena in aggregates causing changes in mineral properties and their influence on ASR performance.

This study investigated the influence of recrystallized silica on alkali silica reactivity, especially their presence in coarse aggregates as a deleterious component. The expansion behavior of samples using a similar aggregate type from the same area, with and without recrystallized particles, were also compared using various methods.
MATERIALS AND METHODS

Materials

Three different types of greywacke aggregates from the same source were studied: greywacke aggregate (GRA), moderate quartz greywacke aggregate (MGA) and recrystallized aggregate (REA). Normal Type I Portland cement was used.

Methods

Several techniques were used in this study. Aggregates were studied using the X-ray fluorescence technique (XRF) for oxide composition analysis. Thin sections were studied with an Olympus imaging petrographic microscope (BX-41; Olympus (GuangZhou) Industrial Co., Ltd; Guangzhou, China) equipped with plane and cross polarization light modes, with a wide magnification range from 1.25 to 100x for mineralogy and detailed investigation. The microscopic characteristics were also analyzed using a digital stereo microscope (Micropot-FXA; Nikon Corporation; Tokyo, Japan). Expansion tests of samples under different conditions were investigated using the accelerated mortar-bar test (AMBT) ASTM C 1260 (ASTM International, 2004), RILEM AAR-2 (Nixon and Sims, 2000) and the accelerated concrete microbar test (ACMT) RILEM AAR-5 (Grattan-Bellew et al., 2003; Sommer et al., 2005). Samples were cast using suggested graded aggregates for each type. Scanning electron microscopy (SEM) was used to image concrete samples after the AMBT and ACMT testing, using conventional SEM equipment (Philips XL 30; FEI Asia Pacific; Shanghai, China) with mouse-driven operation in a Windows computer operating system environment which provided both secondary electron and backscattered electron imaging.

RESULTS AND DISCUSSION

Oxide components analysis

The XRF results of the aggregates investigation are shown in Table 1. The major oxide compositions of GRA, MGA and REA are quite similar (SiO₂ 56–60%, Al₂O₃ 13–18% and Fe₂O₃ 5–6%).

Thin section analysis of aggregates

The results of the petrographical microscopy used to investigate the physical characteristics of the rocks are shown in Figure 1. Aggregate samples were collected from the same source but from three different zones in the pit. The GRA sample was composed of quartz crystalline/microcrystalline, feldspar, clay/graphite, rock fragments with less chlorite and was classified as Class II “alkali-reactivity uncertain” (Sims and Nixon, 2003). The high weathering stage was observed in the rock fragments of greywacke samples in both GRA and MGA. The MGA was coarse-grained greywacke, comprising quartz, feldspar, clay/graphite and rock fragments. The mineral sizes were larger than those of the GRA and the quartz crystalline particles observed in the matrix. This MGA was classified as Class II “alkali-reactivity uncertain”. REA was identified as quartz, clay/graphite and rock fragments,

Table 1 Oxide components of studied aggregates.

<table>
<thead>
<tr>
<th></th>
<th>SiO₂</th>
<th>Al₂O₃</th>
<th>Fe₂O₃</th>
<th>CaO</th>
<th>MgO</th>
<th>SO₃</th>
<th>Na₂O</th>
<th>K₂O</th>
<th>LOI</th>
</tr>
</thead>
<tbody>
<tr>
<td>GRA</td>
<td>58.22</td>
<td>13.93</td>
<td>6.11</td>
<td>6.96</td>
<td>2.53</td>
<td>0.24</td>
<td>3.08</td>
<td>1.32</td>
<td>6.77</td>
</tr>
<tr>
<td>MGA</td>
<td>60.21</td>
<td>13.41</td>
<td>5.99</td>
<td>7.46</td>
<td>2.45</td>
<td>0.18</td>
<td>3.46</td>
<td>0.83</td>
<td>5.17</td>
</tr>
<tr>
<td>REA</td>
<td>56.48</td>
<td>18.64</td>
<td>5.29</td>
<td>3.21</td>
<td>1.90</td>
<td>0.38</td>
<td>1.15</td>
<td>3.39</td>
<td>8.36</td>
</tr>
<tr>
<td>Cement</td>
<td>18.74</td>
<td>5.22</td>
<td>3.20</td>
<td>65.30</td>
<td>0.82</td>
<td>2.80</td>
<td>0.08</td>
<td>0.50</td>
<td>2.75</td>
</tr>
</tbody>
</table>

GRA = Greywacke aggregate; MGA = Moderate quartz greywacke aggregate; REA = Recrystallized aggregate; LOI = Loss on ignition.
Figure 1 Different characteristics from thin section analyses of aggregates of: (a) Greywacke aggregate; (b) Moderate quartz greywacke aggregate; (c) Recrystallized aggregate. (Qtz = Quartz; F = Feldspar; Ch = Chlorite; Rf = Rock fragment and C = Clay).
and the dominance of tectonic forces caused “recrystallization quartz grains”. The brittle deformation transformed the very fine quartz to a microcrystalline/cryptocrystalline matrix with 30% of clay/graphite matrix. This may be the reason for the highest ASR reactivity in this aggregate group. REA was classified as Class III “very likely to be alkali-reactive”.

**Accelerated expansion test**

The results of the accelerated expansion test in Figure 2 clearly reveal differences in the reactivity and expansion behavior of these aggregates. The relationship between expansion and age after casting of the mortar bars, according to AMBT, are shown in Figure 2a. Samples using REA aggregates showed the highest expansion rate during the early stage of the test. The average expansion of samples of REA MGA and GRA was 0.333, 0.238 and 0.279%, respectively, at 16 d after casting, and higher than the suggested limit of 0.20% (ASTM International, 2004) which indicated potentially deleterious ASR expansion. The ACMT results in Figure 2b show expansion at 28 d in all the accelerated concrete microbars greatly exceeded 0.1%, the suggested limit from previous studies (Nixon and Sim, 2000; Sommer et al., 2005). The largest expansion (0.387%) was observed with REA, compared to GRA and MGA (0.278 and 0.251%, respectively). The expansion results supported the findings from the petrographic analysis.

**Stereo microscope**

The stereo microscope investigation of the slab samples, cut from concrete microbars after ACMT testing (Figure 3), showed that the alkali–silica gel REA samples (which yielded the largest expansion) exhibited cracks inside some aggregates and some crust pitting on the surface of the aggregate.
Figure 3 Images from stereo microscopy of accelerated concrete microbar testing polished section of: (a) Greywacke aggregate; (b) Moderate quartz greywacke aggregate; (c) Recrystallized aggregate. (ASR = Alkali silica reaction)
**Thin-section analysis**

Photomicrography using a digital microscope with plane polarized mode provided the characteristics observation of AMBT samples as shown in Figure 4. The ASR gel (in cracks) was observed in all accelerated samples. These observations confirmed the ASR-affected concrete samples using these aggregates and the evidence of alkali–silica gel. The observed crack widths of REA samples were greatest and some cracks were

![Figure 4](image_url)

**Figure 4** Thin sections of accelerated mortar-bar testing samples: (a) and (b) Greywacke aggregate (c) and (d) Moderate quartz greywacke aggregate; (e) and (f) Recrystallized aggregate. (Figures 4a, c and e are at 40x magnification and Figures 4b, d and f are at 100x magnification; ASR = Alkali silica reaction).
also detected inside ASR gel and aggregates. Surface cracks on the ACMT concrete specimen were further investigated in thin-section analysis. ASR-induced cracks inside the aggregate extended into the surrounding cement-paste aggregate surface as well as the aggregate–paste interface, as shown in Figure 5. The large amount of ASR gel observed on the surface of all accelerated concrete samples with expansion greatly exceeded 0.1% at 28 d and expansive

**Figure 5** Thin sections of accelerated concrete microbar testing samples and analysis images, respectively of: (a) and (b) Greywacke aggregate (c) and (d) Mmoderate quartz greywacke aggregate; (e) and (f) Recrystallized aggregate. (gray color = Aggregate; dark color = Paste and white color = Cracking in analysis images; ASR = Alkali silica reaction).
micro cracking was seen in all aggregates. These confirmed that concrete using the investigated aggregate was generally affected by ASR and the occurrence of alkali–silica gel was detected. The largest area of cracking was attributed to REA with particles containing significant amounts of “shear re-crystalline strained grain”. These particles seemed to be recrystallized, leading to the formation of ASR gel inside and at aggregate-paste boundaries, as well as in the matrix. These may explain the significant observed expansion and deformation compared with that observed in the GRA and MGA samples. The largest expansion of REA samples (more than 0.3%) indicated the highest degree of deterioration. In Table 2, the largest values of three quantified amounts of length, area and width of cracks of REA, compared to those of MGA and GRA confirmed the most severe level of reactivity. The crack width of REA was about 3 and 5 times larger than that of GRA and MGA, respectively.

**Scanning electron microscope analysis**

All AMBT and ACMT samples were mounted in a plastic mold using an epoxy-based resin and cured at relatively low temperatures (Moreland, 1968) before conducting SEM with the back-scattered electrons image mode of SEM. The images of these concrete samples in Figure 6 and Figure 7 show details of aggregates, the matrix and cracks. It was also observed that all samples were cracked, which had initiated from gel-reactive products after the accelerated test and subsequently, cracks propagated into the matrices. Similarly, cracks in all samples occurred along the boundary of the aggregate and some were widespread into the cement matrix. Moreover, it was found from both accelerated methods that only REA samples showed numerous cracks surrounding the aggregate surface, aggregate–paste interface and inside aggregates. This crack pattern was different from the others. These cracks were expected as the result of the severe alkali-silica reactivity level of REA.

**CONCLUSION**

From this study, various conclusions were drawn.

1. All studied aggregates were similar in terms of oxide composition, but thin-section analysis indicated the difference most clearly. GRA and MGA were coarse-grained greywacke composed of quartz feldspar rock fragment. However, REA was high-weathering chlorite with a matrix of re-crystallized quartz and rock fragments of clay/graphite, altered by tectonic forces.

2. The recrystallized aggregates yielded the largest expansion from both the AMBT and ACMT methods. The results confirmed the findings from other investigations using stereo microscopy, thin section analysis and SEM.

3. Recrystallized aggregates appeared to increase the ASR risk and may be considered as more deleterious materials than those of normal aggregates of the same type, in terms of alkali–silica reactivity.

**ACKNOWLEDGEMENTS**

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Figure 6  Images from scanning electron microscopy of accelerated mortar-bar testing samples of (a) and (b) Greywacke aggregate; (c) and (d) Moderate quartz greywacke aggregate; (e) and (f) Recrystallized aggregate. (Figures 6a, c and e are at 200x magnification and Figures 6b, d and f are at 2,000x magnification ASR = Alkali silica reaction.)
Images from scanning electron microscopy of accelerated concrete microbar testing samples of (a) and (b) Greywacke aggregate; (c) and (d) Moderate quartz greywacke aggregate; (e) and (f) Recrystallized aggregate. (Figures 7a, c and e are at 200x magnification and Figures 7b, d and f are at 2,000x magnification; ASR = Alkali silica reaction).

**LITERATURE CITED**


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