ASR-SENSITIVE SLOW-LATE-AGGREGATES; MACRODAMAGE AND MICROSTRUCTURE

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ABSTRACT

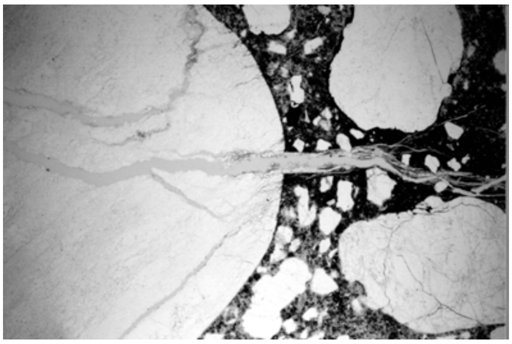
The reason for ASR-sensitivity of aggregates is the presence of amorphous or bad crystalline quartz. While the behaviour of opaline sandstone and chert is well known and the current test methods [1, 2] are sufficient to prevent ASR, some other aggregates make problems because they react slow and late [3]. Some deterioration cases by ASR with slow-late aggregates occurred in Germany during the last few years. 80 samples of materials, which lead to these ASR-problems, were investigated. The aim of these investigations were to detect the properties of these aggregates and to estimate the context between microstructure and ASR-sensitivity. Thin section microscopy, solubility tests, X-ray diffraction analysis of mineral content and microstructure and testing of mortar bars in a fog-chamber were done. In most cases there is a good interrelation between the results of the test methods. The Comparison of thin section microscopy and X-ray diffraction analysis of microstructure shows an advantage of the X-ray diffraction method. While thin section analysis gives only qualitative information about "strain" by undulatory extinction, via X-ray diffraction it is possible to detect "strain" and crystallite size quantitatively. Comparison with solubility tests shows a quantitative correlation between SiO_2-solubility, crystallite size and strain in most cases. Testing the mortar bars in a fog chamber leads to a higher length change of bars made with opaline sandstone and ground quartz compared to than the length change of bars only made with ground quartz. But using only ground quartz leads to length change too. It was shown, that all investigated materials are potential ASR-sensitive. The most sensitive material was siliceous limestone with small crystallite size of silica and calcite. Stressed quartz is least sensitive of all investigated aggregates, but there is a correlation between microstructure and ASR-sensitivity too.

1 INTRODUCTION

ASR is a complex sequence of chemical and physical processes. Its appearance depends on the kind of materials used for concrete production. ASR sensitive quartz variations as components of aggregates react with alkali from the pore solution of concrete (Dent Glasser [4]). The reaction products may damage the concrete by swelling, infiltration into the cement stone and reacting with the cement matrix. For avoiding ASR problems a lot of standard test methods were used [5]. Using this methods for estimating the sensitivity of aggregates containing chert and opal leads to good protection against ASR damage. The estimation of the behaviour of stressed quartz, quartzite and other quartz containing aggregates like siliceous limestone and greywacke is more difficult. This uncertainty leads to unexpected cases of concrete damage with aggregates, which are assumed to be innocuous by standard test methods. Knowledge of the microstructure and its influence to ASR sensitivity will help to understand the ASR mechanism and to estimate the behaviour of quartz with special (detectable) micro structural characteristic.

2 DAMAGE OF CONCRETE STRUCTURES

During the last years a few of concrete deterioration cases by ASR occurred in Germany. The damaged concrete were made with quartz containing aggregate (granite, greywacke, rhyolite, siliceous limestone, chert and stressed quartz). For the investigation of these deterioration cases, about 300 thin sections of the damaged concrete were made and gave the possibility to detect, the special characteristics of aggregate grains, which are deteriorated by cracking or solution processes. It is necessary to distinguish between stressed quartz (quartz content 80-100 %) and the other aggregates, because there are some other influence factors beside the microstructure of quartz if there are some other minerals present. When using solely stressed quartz, the deteriorated aggregates show undulatory extinction, veins, sometimes texture and areas with very fine quartz crystals (Figure 1).



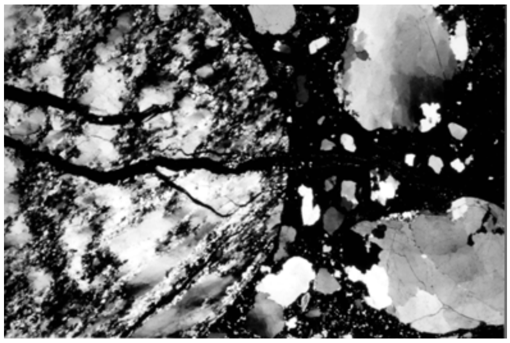


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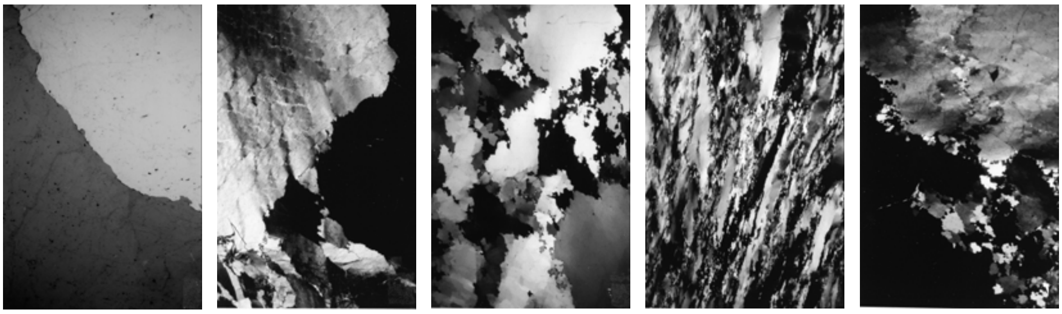
Figure 1: Concrete-Structure – Aggregate of stressed quartz with cracks and gel formation; Thin-sections, 30x, Pol || (a), + (b)

Estimating the context between microstructure of quartz and ASR sensitivity by aggregates containing some other minerals is more difficult. In all investigated cases, the quartz grains are smaller than in the stressed quartz aggregates. Often it is not possible to distinguish the micro-structural characteristic of these very fine quartz crystals. Cracks often occurred along veins with feldspar or clay minerals. Investigating the microstructure of quartz by thin section microscopy leads not to a sufficient result because there are a lot of other influences to ASR-sensitivity like the kind and quantity of minerals present.

3 MICROSTRUCTURE OF QUARTZ

There are a lot of investigations regarding the problem of estimation ASR sensitivity of quartz by examination undulatory extinction [6, 7, 8]. The measured parameter in this case is the undulatory extinction angle. But the opinion about the effectiveness and reproducibility of this method differs between different authors. While Gogte [6] saw a good correlation between the grade of undulatory extinction and the length change of mortar bars, stored for 9 month in a fog chamber, Mather [9] found no correlation between ASR sensibility – measured as SiO_2-solubility- and the undulatory extinction angle. One problem may be that the undulatory extinction is mainly influenced by strain. But crystallite size, which is not detectable by thin section microscopy, influences the ASR sensibility too. The second problem is, that it is very difficult to quantify undulatory extinction angle because it depends on the grain size of the investigated area and on the orientation of the investigated grain. Therefore a test of the reproducibility of these investigations was done by Andersen & Thaulow [10]. Six specialists for petrography determined 900 undulatory extinction angles. The result of this test was, no correlation between the results and none of the results was in good correlation to ASR tests made with the same kinds of stones. Quantification of undulatory extinction by measuring the extinction angle is difficult [10] but a qualitative estimation of the behaviour of quartz is possible according García del Amo [11]. Thereby 60 samples of stressed quartz and 25 samples of different other quartz containing aggregates (greywacke, quartzite, andesite, granite, siliceous limestone) were investigated. The materials were the same as in the deteriorated concrete structures explained before. Comparison of thin sections from the deterioration cases, thin section of the pure quartzes and results of solubility and mortar bar tests gave the possibility to grade the different stressed quartz aggregates into 5 classes [12]. The ASR sensitivity growth up with higher number (Figure 2).



a b c d e

Figure 2: Types of stressed quartz: thin sections, 30x, Pol +

a - quartz without strain. Homogeneous interference colour
b - quartz with a big area with undulatory extinction
c - quartz with sub grain formation, grains are indented
d - quartz with sub grain formation and texture
e - quartz with chaotic undulatory extinction and re-crystallisation respectively new formation of crypto-crystalline quartz

For quantitative estimation of ASR sensitivity it is necessary to find another investigation method to detect and quantify micro structural properties and its influence on ASR. The method used in this work -beside thin section microscopy- was microstructure analysis by X-ray diffraction. The detectable properties are “strain” and “crystallite size”, where “strain” means the deviation of the space between two layers in a crystal lattice (d) and the crystallite size (measured by X-ray diffraction) means a coherent diffracting domain size (defect free distance) Snyder [13]. The mineral content, crystallite size and strain were detected by X-ray diffraction analyses (Rietveld method) for all samples. In about 80% of all investigated stones, there is a good correlation between ASR sensitivity (mass loss in alkaline solution) - Berninger [14] - and microstructure. The smaller the crystallite size and the higher the strain the sensible is the stone (Figures 3 and 4).

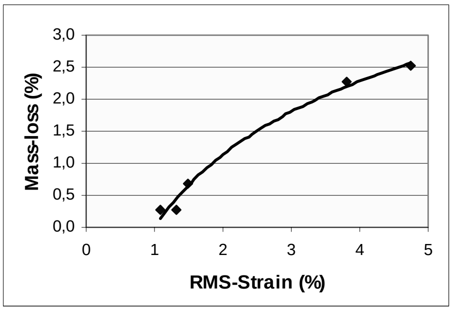


Figure 3: Correlation between mass loss and RMS Strain

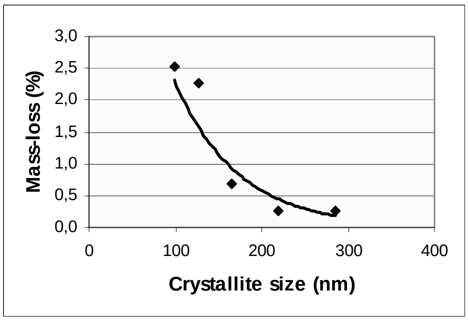


Figure 4: Correlation between mass loss and crystallite size

Also there are good correlations between the thin section analyses and microstructure analyses by X-ray diffraction. Comparison between crystallite size values and results from thin section microscopy seems to be impossible, because crystallite size is not detectable by light microscopy. Of course some indication for small crystallite sizes will be seen by the means of thin section. If there are re-crystallisations or new formations of cryptocrystalline quartz it can be assumed, that small crystallite sizes for quartz will be detected by X-ray diffraction. On the other side, if there are big areas without or with less undulatory extinction, bigger crystallite sizes and small values for strain will be expected. Regarding the stress values a good correlations could be seen. Estimation of crystallite size using thin section microscopy is more difficult; there are only indications. Figures 5 and 6 are two examples for the correlation between strain-values detected by diffraction and thin section microscopy.

Figure 5: Crystallite size: 226 mm
RMS-strain: 2,04 %

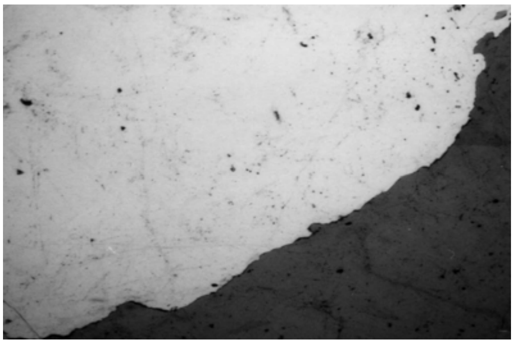


Figure 6: Crystallite size: 261 mm
RMS-strain: 0,12 %

The investigated siliceous limestones show big differences in the solubility and in crystallite size and strain. It was possible to calculate a theoretical solubility (as mmol/l SiO_2) using both micro structural factors (Figure 7).

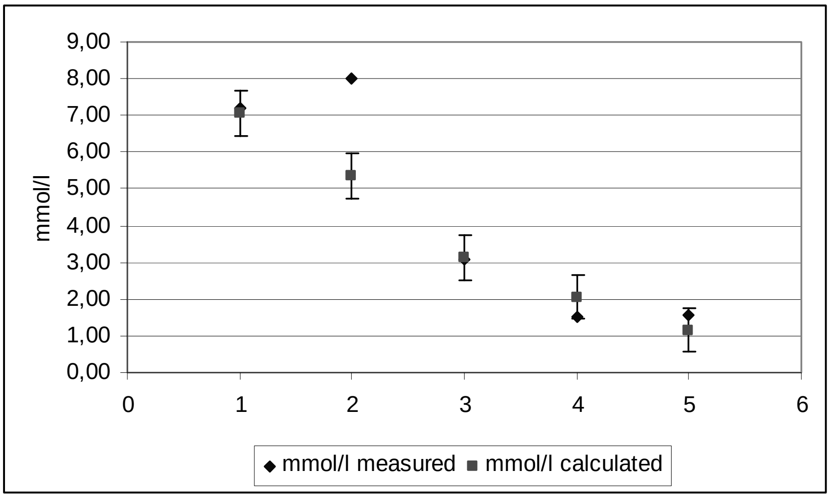


Figure 7: Comparison of measured and calculated values for dissolved SiO_2

The reason for different ASR-sensitivity of quartz variations is the great variety in microstructure. While thin section microscopy is a good method for the qualitatively estimation of the behaviour of quartz, a quantitatively estimation is possible using X-ray diffraction. Crystallite-sizes smaller than 100 nm lead to extremely increasing solubility. The differences in the behaviour of SiO_2 with crystallite sizes bigger than 200 nm are very small. In the area between 200 nm and 400 nm (it's a typical value for powder samples of rock crystal) the influence of strain predominates the influence of crystallite size. Using the RMS-strain values, it is possible to calculate a theoretical solubility of quartz with similar crystallite size [14]. The agreement with solubility experiments is more than 70 %. The behaviour of siliceous limestone can be calculated by knowing crystallite size and strain. In this case, the agreement with the solubility experiments is more than 80 %. All investigated stones are potential ASR reactive. Stressed quartz is finally less reactive than all other stones.

An x-ray diffraction analysis is a good method for quantitative estimation of ASR reactivity of stressed quartz and siliceous limestone. In all other cases some other factors like kind of minerals presenting a quantitative mineral content influence the ASR sensitivity too.

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