DETECTING MICROSTRUCTURAL CHANGES IN CONCRETE CONDITIONED UNDER THERMO-MECHANICAL LOADING USING X-RAY COMPUTED TOMOGRAPHY

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Abstract

The combined action of thermal and mechanical loading in nuclear applications can affect the pore structure of concrete. It is important to characterise these changes in order to understand the effects they may have on material strength and durability. Common methods of microstructural analysis such as electron microscopy can fail to capture representative data. To overcome this, 3D X-ray Computed Tomography (XCT) has been used to investigate microstructural changes in concrete specimens from a research program on Advanced Gas-cooled Reactor (AGR) pressure vessels which were subjected to hydrostatic mechanical loading and thermal treatment. Cylindrical cores extracted from the specimens were characterised non-destructively using XCT at the Henry Moseley X-ray Imaging Facility (HMXIF), University of Manchester. Analysis of the reconstructed XCT data for 14.6 µm resolution scans has revealed significant changes in the pore size distribution of heated specimens with respect to that of unconditioned control specimens. Whilst changes in pore size distribution due to hydrostatic mechanical load cannot be clearly identified, those specimens conditioned under combined thermo-mechanical loading experienced a greater change than those conditioned using thermal treatment only.

Key words: thermo-mechanical loading, cementitious materials, porosity, X-ray computed tomography
1. INTRODUCTION

Concrete experiences a combination of thermal and mechanical loading in certain safety critical nuclear applications such as in the structure of AGR pressure vessels\(^1\) and interim/long term High Level radioactive Waste (HLW) storage facilities\(^2\). The effects of these loads on the pore structure of concrete are of particular importance as a result of their potential influence on material strength and durability. However, measurement of pore characteristics is complicated due to the range of length scales over which they are observed. In this paper, pore size distribution is described using concrete science terminology where porous structures between 1 nm and 10 µm in diameter are referred to as capillary pores and those larger than 10 µm as air voids\(^3\).

Mercury Intrusion Porosimetry (MIP) is often used to characterise the pore structure of cement based materials down to pores of the order of a few nanometres. MIP can provide a reliable estimation of the intrudable porosity of a specimen\(^4\), i.e. the total volume of pores which can be intruded within the pressure range of the MIP equipment and those pores which are not isolated from the pore network. With respect to individual pore quantification though, this technique systematically misallocates pore sizes\(^4,5\) as a result of the “accessibility effect”\(^4\) where the intrusion of smaller pores which lie before larger pores on the intrusion path of the mercury masks the presence of the larger pores because the intrusion pressure for these pores has already been overcome. This effect is particularly noticeable with air voids above 10 µm in diameter which are often unreported in MIP studies despite constituting a significant proportion of the total porosity of cementitious materials\(^4\).

Other methods of pore structure quantification such as the analysis of images from Optical and Scanning Electron Microscopy (SEM) provide valuable quantitative data on total porosity and specific surface area within the limit of their image resolution\(^6\). However, data on 3D properties such as pore size distribution are limited. A further drawback to MIP and SEM analysis is the need for specimen preparation. Damage induced during drying and polishing, in the case of microscopy, can significantly affect pore characteristics and properties\(^7\).

In this paper, non-destructive 3D X-ray Computed Tomography (XCT) imaging was carried out to characterise the air void structure of concrete specimens conditioned under differing regimes of loading and heating. The air void data obtained in our work was limited by image resolution, which is a function of (i) specimen size, (ii) distances between specimen, X-ray source and detector and (iii) detector attributes (number of pixels).

2. MATERIALS AND EXPERIMENTAL METHOD

2.1 Specimen preparation and conditioning

One concrete type with a mix typical of that used to construct AGR pressure vessels was considered with all constituents and mass proportions shown in Table 1. Specimens were prepared in 2003 as part of an experimental program on the strength of AGR pressure vessels\(^1\). The concrete was cast in slabs with dimensions of 740 x 620 x 150 mm., followed by immersion in water with a 10 day temperature matched curing cycle in which the temperature reached a maximum of 65°C. Following the 10 day cycle, the slabs were de-moulded and returned to the water tank for a further 60 days at 20°C. Each concrete slab was then cut and ground to form the 100 mm cubes used here. The machined cubes were stored at room temperature and humidity until conditioning with the unique mac\(^2\) apparatus for multiaxial compression of concrete at elevated temperature (Figure 1A).
The experimental program consisted of the eight tests, with individual conditioning regimes and two repeats each (Table 2). The conditioning regimes included (i) mechanical loading only, (ii) thermal treatment only, (iii) mechanical loading with thermal treatments and (iv) untreated control specimens. All thermally treated specimens experienced a heating/cooling rate of 0.2 °C/min. Specimens conditioned under mechanical load were subjected to a hydrostatic compressive stress of 0.6 \( f_{cu} \) in line with previous testing\(^{[1]} \) and in order not to induce damage associated with deviatoric loading. After completing the appropriate mechanical loading and/or thermal treatment, a steady state period of 24 hours was allowed before unloading/cooling. Following the conditioning treatments, a 28 mm diameter core was extracted from the centre of each cube for analysis using XCT (Figure 1B).

Table 2: Conditioning regimes of the concrete cubes for XCT analysis (\( f_{cu} \) = ultimate compressive strength of concrete)

<table>
<thead>
<tr>
<th>Conditioning Name</th>
<th>(i) L06</th>
<th>(ii) H250</th>
<th>(iii) L06H250</th>
<th>(iv) Control</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mechanical Loading</td>
<td>0.6 ( f_{cu} )</td>
<td>-</td>
<td>0.6 ( f_{cu} )</td>
<td>-</td>
</tr>
<tr>
<td>Thermal Treatment</td>
<td>-</td>
<td>250 °C</td>
<td>250 °C</td>
<td>-</td>
</tr>
</tbody>
</table>

Figure 1A: mac\(^2\)T apparatus at the University of Sheffield

Figure 1B: Concrete cube and extracted core with the two XCT scan regions. XCT data shows one scan (29 x 29 x 29 mm) with three orthogonal slices (orthoslices) of 3D pixels (voxels)
2.2 X-ray CT imaging

XCT allows the internal structure of an object to be imaged in three-dimensions through the computerised reconstruction of a set of 2D radiographs taken at different projection angles on a single axis of rotation. The radiograph is detected on a Charge Coupled Device (CCD) positioned in a plane normal to the x-ray beam (Figure 2A). Each pixel of the radiograph shows the total attenuation of x-rays by features within the corresponding area of the specimen.

Figure 2A: Nikon Custom Bay at the HMXIF with a schematic plan of the XCT equipment

Figure 2B: Grey-scale value histogram showing tri-modal distribution of 16-bit XCT data and approximate phase distribution for a control specimen

Like other image analysis techniques, pore characterisation using XCT is limited by image resolution. Image resolution in turn is governed by specimen thickness since it is necessary for the specimen to remain within the CCD field of view throughout the 360 degree rotation for image reconstruction. With the large aggregate size of the concrete considered here, a specimen diameter of 28 millimetres was chosen, which corresponds to a resolution of 14.6 μm on a 2000 x 2000 pixel CCD.

Specimens were analysed using the Nikon 225/320 kV Custom Bay at the HMXIF, University of Manchester (Figure 2A). The 225 kV static source with a tungsten target (3 μm spot size) was used in conjunction with a 1.5 mm thick copper filter in order to optimise the emitted x-ray beam energy spectrum. A power level of 160 kV and 200μA was selected to achieve good radiograph contrast. The cylindrical specimens were mounted on the precision 5-axis manipulator arm (Figure 2A) and positioned within the CCD detector field of view. Two scans were then taken, one above and one below the specimen centre line (Figure 1B). Once a scan had been commenced, the manipulator arm allowed radiographs to be taken at accurate angular steps of 0.115 degrees which equated to 3142 radiographs over a 360° rotation. The source to specimen distance was 73.5 mm, the source to detector distance was 1007 mm and the acquisition time per radiograph was 1000 ms.

After data acquisition, all 3142 projections were reconstructed using Metris CT pro software with corrections for the centre of rotation, beam hardening and noise reduction. The
reconstructed data in the form of 3D pixels or “voxels” was then down sampled from 32 bit to 16 bit grey-scale in order to reduce file sizes for image analysis.

2.3 Data processing

Image analysis was performed using the Avizo Fire software package. A Region Of Interest (ROI) was selected from each scan with dimensions 1700 x 1200 x 1200 voxels. This corresponds to a volume of 25.2 mm x 18 mm x 18 mm. The ROI was chosen from the centre of the cylindrical specimen in order to exclude voxels in the background regions and minimise effects from the cone shaped x-ray beam.

Various methods are proposed in the literature to facilitate the segmentation of data into distinct phases\cite{8,9}. Here, as a result of the distinct tri-modal grey-scale distribution shown in figure 2B, it was possible to use classic minima thresholding to differentiate porosity from cementitious and aggregate phases within the concrete. A sensitivity analysis of the grey-scale value (GSV) distribution was carried out at three increments of 10, 25 and 100 both above and below the GSV corresponding with the minima of one ROI. An error of less than two percent could be achieved by selecting a GSV within ±25 points of the actual minima. This accuracy was easily obtainable through sorting of the GSV data.

Segmented voxels were then analysed using the Avizo XQuant Quantification features in Avizo Fire to determine their connectivity. In this procedure, the six faces of each voxel in the image are evaluated to ascertain whether a voxel of the same segmented phase is connected\cite{10}. Voxels of the same phase which are connected using this criterion are then grouped into clusters or pores in our study. Data is presented with respect to the equivalent pore diameter which is calculated by assuming that all pores are spherical in shape.

Finally, the Nyquist-Shannon sampling theorem\cite{11} was used to determine the minimum size of features that could be resolved based on the image resolution of 14.6 µm. This theorem proves that a digital image must have more than two pixels per resolvable element in order to ensure that all features are sampled sufficiently to detect their presence. In this case, three voxels was chosen as the feature resolution limit meaning the smallest detectable porosity in this study has a diameter of 31.5 µm.

3. RESULTS AND ANALYSIS

A typical reconstructed data set from the ROI of a control specimen is visualised using 3 orthoslices in Figure 3A along with the segmented pore structure in Figure 3B. The orthoslices show the clear presence of large aggregate particles, cementitious matrix and porosity. Both igneous and sedimentary rock forms are present which is indicative of the Whinstone aggregate source used for material in this experiment.

The observed pores do not form a long range network and can be classified as air voids. No interaction of the porous structure with aggregate phases was observed. Again, this was also expected as any increase in porosity within the Interfacial Transition Zone (ITZ) between the cement paste and aggregate particles is known to occur within 20 µm of the aggregate surface\cite{12} which is slightly below the feature resolution of data presented here.

It is also clear from Figure 3B that a small number of very large pores exist in the control specimen considered, with typical diameters in excess of 2000 µm. Pores of this magnitude are observed to a varying degree in all specimens and their presence is thought to be a result of inherent variability in the material rather than a result of the conditioning regimes. Their size with respect to the ROI is sufficient so as to raise doubts over whether they may be
considered quantitatively and therefore equivalent pore diameter data is only presented for pores below 1000 µm.

Figure 3A: ROI showing orthoslices of the reconstructed data from a control specimen
Figure 3B: Segmented porosity within the same volume

Figure 4: Equivalent pore diameter distribution results for three conditioning regimes L06, H250, L06H250 and the unconditioned control. Error bars show standard error calculated for 4 XCT scans from 2 cubes repeated using the same conditions.

Figure 4 shows the equivalent diameter distribution of pores for each of the conditioning regimes. It can clearly be identified from Figure 4 that thermal treatment (sample H250 and L06H250) generally results in a decrease in the measured frequency of pores below the 100 µm diameter threshold and in parallel an increase in pores with a diameter greater than 200 µm compared to the control specimens. The presence of mechanical load for specimens which
also experience thermal treatment produces a further decrease in pores below 50 µm over those which have experienced thermal treatment only. Data for mechanical load only (L06) specimens indicate that, in this instance, pore size changes do not occur.

4. DISCUSSION

Results from the XCT analysis of thermo-mechanically loaded concrete indicate that thermal treatment causes permanent changes in the air void pore structure of concrete. However, pore size changes as a result of hydrostatic mechanical loading only cannot be identified. Whilst previous researchers have detected changes in the pore structure of cementitious materials due to mechanical load at the length scales considered here\cite{13}, loading was applied in the uniaxial orientation and the changes observed in these studies can possibly be attributed to “fracture of walls” between large pores. Such changes are unlikely under the relatively low levels of hydrostatic mechanical loading applied here.

Pore size distribution changes due to thermal treatment may be a consequence of the internal deposition of calcium hydroxide and/or ettringite\cite{14}. Whilst the results for pores below 100 µm in diameter support this, there is an increase in the frequency of thermally loaded pores above 200 µm. However, these results present normalised data using relative frequency percentage and thus a decrease in pores in the measured region below 100 µm would be compensated by an increase in the frequency of pores with larger diameters. This is because pores below 100 µm may have been reduced sufficiently in diameter to drop below the feature resolution of these XCT images. Also, for any uniform increase in cement hydration and therefore uniform deposition of hydration products, smaller pores would experience a greater change in diameter with respect to larger pores.

It also appears that the presence of load during heating increases the change in pore size distribution at diameters below 100 µm. It has been observed that the presence of load below a uniaxial stress of 0.6 $f_{cu}$ results in a decrease in the gas permeability of concrete\cite{15}. This would mean that the escape of water present in capillary pores as a result of heating above 100 °C was possibly restricted. One potential effect of this is that further hydration of the cement conditioned with load and heat over that which experiences thermal treatment only may be enhanced. However, further chemical testing would be required in order to ascertain with a greater certainty the likelihood of this hypothesis.

5. CONCLUSIONS

The XCT analysis of concrete specimens from a research program into the behaviour of AGR pressure vessel concrete conditioned under mechanical load and heat treatment has revealed the following:

- Laboratory based XCT is a useful technique for determining quantitative changes in the characteristics of the 3D air void structure in concrete. Experimental testing shows a good degree of repeatability between similarly conditioned specimens of batch cast concrete.
- A reduction in pores with an equivalent diameter of between 30 and 100 µm and an increase in pore diameters above 200 µm occurs as a result of thermal treatment at 250 °C with respect to an unconditioned control specimen.
- The presence of hydrostatic mechanical load at 0.6 $f_{cu}$ during thermal treatment appears to increase the change in equivalent pore diameter frequency below 100 µm.
There are no identifiable changes in equivalent pore diameter for specimens conditioned under hydrostatic mechanical load at 0.6 $f_{cu}$ only.

To provide further insight into the mechanisms involved, future sub-micron resolution XCT analysis of selected regions within these specimens will be carried out at the HMXIF. This work will allow the visualisation of changes in the capillary pore structure and any phase alterations in the cement itself thereby shedding new light on the microstructural mechanisms which act on the concrete under thermo-mechanical loading.

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