Available online at www.sciencedirect.com

SCIENCE DIRECT®

Cement and Concrete Research 35 (2005) 637–640

CEMENT AND CONCRETE RESEARCH

Pore structure of cement/pozzolan composites by X-ray microtomography

U. Rattanasak*, K. Kendall

School of Chemical Engineering, University of Birmingham, Birmingham, B15 2TT, UK

Received 18 December 2003; accepted 26 April 2004

Abstract

Microtomography, a nondestructive X-ray analysis test, was used to study the pore structure of cement/pozzolan composites. The three-dimensional picture of the overall porosity was created by stacking together sequential slices of two-dimensional microtomographic images. This picture revealed all the pores above 5 μm in diameter, showing that the largest pore was 0.51 mm in diameter and that the volume of large pores was around 2% by volume.

Keywords: Microtomography; Pore structure; Cement/pozzolan composite; X-ray

1. Introduction

Microstructure has been widely studied to understand and improve cement/pozzolan properties [1]. Porosity is one major component of microstructure that governs several properties, such as permeability, shrinkage, elastic modulus, toughness, and strength. At present, the scanning electron microscope (SEM) can clearly show the large pores in two-dimensional images. Although, in principle, a three-dimensional picture could be built up by sequential polishing and SEM study, only two-dimensional micrographs are usually presented.

It is desirable to know the three-dimensional microstructure, especially of the pore distribution, because this would enable the properties and potential improvement of cement products to be quantified. In the last decade, significant studies [2–8] have been made in the development of X-ray microtomography to reveal cracks and damage in cement samples. X-ray microtomography has not been used to show pore sizes and distribution in cement products, although it has been used to reveal pores in cellulose granules [9]. This paper shows that X-ray microtomography is an excellent method for revealing the porous microstructure in cement products, allowing improved process to be developed.

2. Background

X-ray microtomography measures the three-dimensional internal structure of a material's X-ray absorption. Ordinary X-ray microtomography produces shadow graphs without depth information. To gather the depth information, a large number of X-ray shadow graphs must be produced at different angles. Attenuation images are collected from many angles by rotating the specimen through small, evenly spaced angular increments between 0° and 180° using a rotation stage. The number of images needed is about $N\pi/2$, where N is the width of the specimen image, in CCD pixels, as shown in Fig. 1. A computer is used to control both the beamline configuration and the microtomography apparatus. The control software collects the data and performs the reconstruction process.

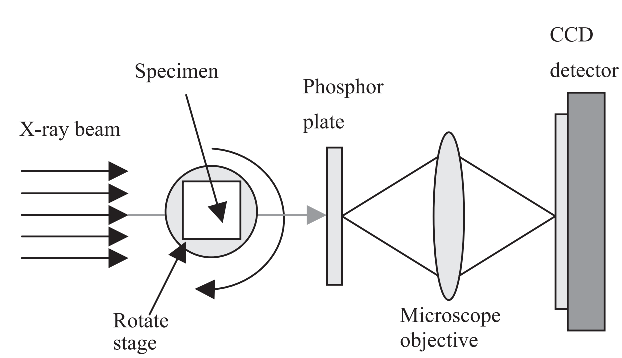

Fig. 1. A schematic diagram of microtomography.

* Corresponding author. Tel.: +44-121-4145082; fax: +44-121-4145324.
E-mail address: uxr105@bham.ac.uk (U. Rattanasak).

0008-8846/$ – see front matter © 2004 Elsevier Ltd. All rights reserved.
doi:10.1016/j.cemconres.2004.04.022

Table 1
Mix proportions and results of cement/pozzolan composites

Sample	%PC	%FA	%MK	%SP	W/C	S/V (mm^{-1})	Porosity (%)	Mean pore size (μm)	Largest pore size (μm)
PC	100	–	–	1	0.25	82.060	1.36	48.67	508
FA	75	25	–	1	0.25	49.655	2.35	33.51	232
MK	90	–	10	1	0.29	87.273	1.41	25.34	207

PC = Portland cement; FA = fly ash; MK = metakaolin; S/V = surface–volume ratio of pores.

Up to now, microtomography has been used to study several properties of cement-based composites, including the measurement of internal damage [2,5–7] and the progression of sulfate attack [4,8], but not to study porosity. This paper emphasizes the use of X-ray microtomography for identifying the largest pore size and the volume of large pores, parameters that determine strength, modulus, and permeability.

3. Materials and methods

The objective of this work was to show that improvements in cement composites obtained by pozzolan addition could be related to microstructure changes. A series of cylindrical cement-based composites was prepared and cast in plastic molds. Portland cement (PC) was individually replaced with 25% by weight of fly ash (FA) and 10% by weight metakaolin (MK). Additionally, melamine-formaldehyde superplasticizer was added at 1% by weight to maintain the same normal consistency as the control sample. The mix proportions are given in Table 1. After removal from the mold, the specimens were cured in a water bath for 28 days and then cut into small pieces with a diamond saw. All specimens were oven dried for 24 h prior to testing and then examined visually using the SkyScan-1072 (Skyscan, Belgium), whose X-ray microfocus tube operates at 20–150 kV/0–500 μA, with a special X-ray camera based on high-resolution (1024×1024 pixels) cooled CCD-sensor with fibre optic coupling (3:7:1 image reduction) to X-ray scintillator or 768×560 pixels CCD-sensor with lens coupling to X-ray scintillator. The reconstructed cross-sections have a 1024×1024 pixels format, and the voxel size of the sample is 5 μm. A typical cycle of data collection for reconstruction contains 200 to 400 views over 180° or 360° of object rotation.

4. Results and discussion

Fig. 2 shows the microtomographic scan produced on the specimens. The shadow X-ray image of the specimen is presented in Fig. 2a, while typical reconstructed cross-sectional images of different types of specimen are shown in Fig. 2b and c. Black areas represent dense material and white areas

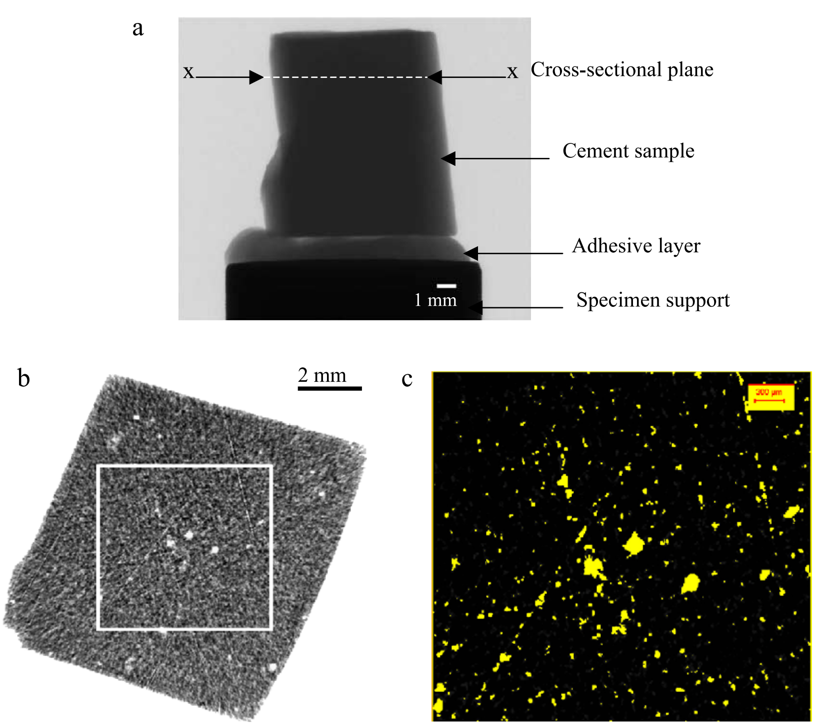

Fig. 2. Tomographic images: (a) X-ray shadow image of a typical cement sample; (b) typical image of X-ray microtomography data from cross-section x–x showing the white square selected area; and (c) processed data from selected plane of sample to show only the pore regions.

are pores. A three-dimensional volume of the boxed area was created by stacking together sequential slices of two-dimensional microtomographic images and is shown in Fig. 3. The pores determined from the cross-sectional images and reconstructed three-dimensional image were about 1.36% by volume for the control sample and 2.35% by volume for the FA–cement composite. In the case of the MK–cement composite, the porosity was substantially decreased to 1.41% by volume (Table 1). Large pores were defined by entrapped air during the mixing process for cement formation. The largest pore detected by the machine was 0.5mm in diameter for neat cement composite. However, the largest pores were smaller in the pozzolan–cement composites. The largest pore was 232 μm for FA–cement composite and 207 μm for MK–cement composite.

For the determination of the pore size distribution greater than 5 μm, the LEICA QWin software was used to mark each pore and to count the number of pores at a certain size. The pore size distributions of these samples are plotted in

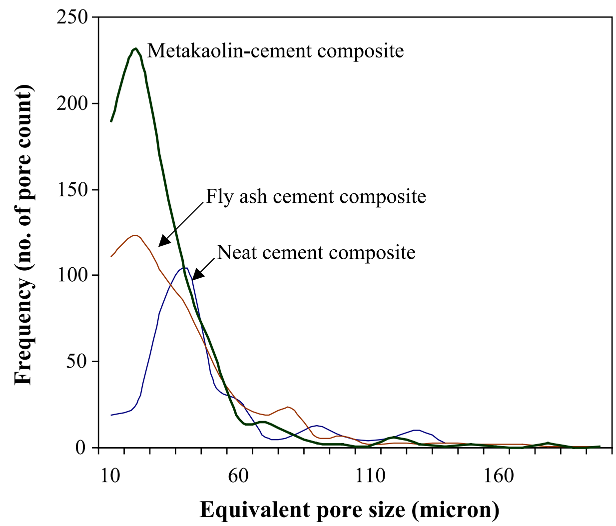

Fig. 4. Distribution of large pore sizes of cement/pozzolan composites measured by X-ray microtomography.

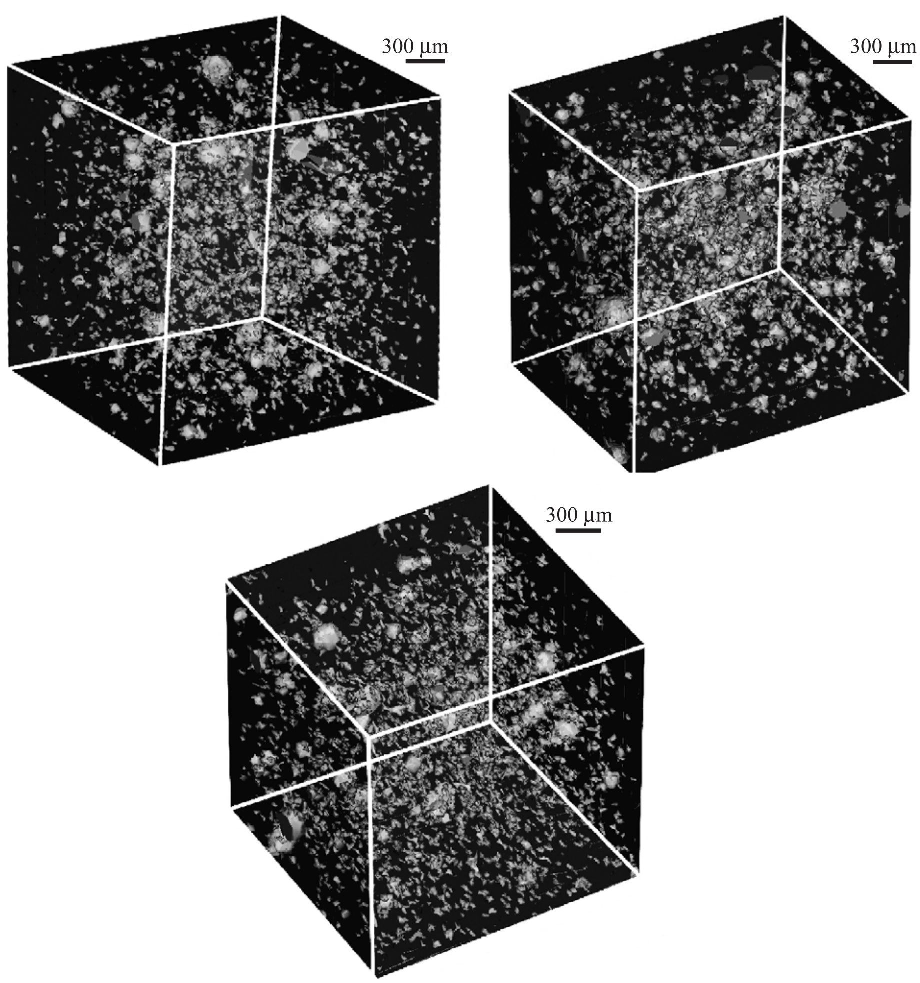

Fig. 3. Three-dimensional renderings of microtomography data: (a) neat cement paste; (b) FA–cement-based composite; (c) MK–cement-based composite.

Fig. 4. Equivalent pore size was defined as the diameter of the sphere of volume equal to the irregular shaped pore. The mean pore size of neat cement composite was 48.67 μm, while those of FA–cement and MK–cement composites were 33.51 and 25.34 μm, respectively. It can be seen that the pozzolan reduced the mean pore size of the products.

Microtomography cannot detect the finer pores, such as gel pores less than 10 nm in diameter [10], which are usually measured by mercury intrusion porosimetry (MIP) at high pressure. However, MIP cannot measure large pores accurately, and so X-ray microtomography is a useful new technique [11].

5. Conclusions

X-ray tomography is a promising tool for the nondestructive investigation of the porosity and morphology of cement-based composites. It can provide the three-dimensional volumetric images presenting the morphological information such as pore shape and connectivity. This new method shows that pozzolan additions give smaller sizes of the largest pores found in the samples, showing how such composites can display higher strengths.

Acknowledgements

The authors would like to thank the Royal Thai Government for financial support.

References

[1] K. Kendall, Molecular Adhesion and its Applications: the Sticky Universe, Kluwer Academic Publishing, New York, 2001.

[2] E.N. Landis, E.N. Nagy, D.T. Keane, Microstructure and fracture in three dimensions, Eng. Fract. Mech. 70 (2003) 911–925.

[3] D.P. Bentz, S. Mizell, S. Satterfield, J. Devaney, W. George, P. Ketcham, J. Graham, J. Porterfield, D. Quenard, F. Vallee, H. Sallee, E. Boller, J. Baruchel, The visible cement data set, J. Res. Natl. Inst. Technol. 107 (2002) 137–148.

[4] D.P. Bentz, N.S. Martys, P. Strutzman, M.S. Levenson, E.J. Garboczi, J. Dunsmuir, L.M. Schwartz, X-ray microtomography of an ASTM C109 mortar exposed to sulfate attack, Mater. Res. Soc. Symp. Proc. 307 (1995) 77–82.

[5] E.N. Landis, E.N. Nagy, D.T. Keane, Microtomographic measurements of internal damage in Portland-cement-based composites, J. Aerosp. Eng. 10 (1997) 2–6.

[6] J.S. Lawler, D.T. Keane, S.P. Shah, Measuring three-dimensional damage in concrete under compression, ACI Mater. J. 98 (2001) 465–475.

[7] S.P. Shah, S. Choi, Nondestructive techniques for studying fracture processes in concrete, Int. J. Fract. 98 (1999) 351–359.

[8] S.R. Stock, N.N. Naik, A.P. Wilkinson, K.E. Kurtis, X-ray microtomography (microCT) of the progression of sulfate attack of cement paste, Cem. Concr. Res. 32 (1992) 1673–1675.

[9] L. Farber, G. Tardos, J.N. Michaels, Use of X-ray tomography to study the porosity and morphology of granules, Powder Tech. 132 (2003) 57–63.

[10] S. Mindess, J.F. Young, Concrete, Prentice-Hall, New Jersey, 1981.

[11] S. Diamond, Mercury porosimetry: an inappropriate method for the measurement of pore size distributions in cement-based materials, Cem. Concr. Res. 30 (2000) 1517–1525.