Advancing Concrete Durability Research through X-ray Computed Tomography

Takafumi Sugiyama¹ and Michael Angelo B. Promentilla²

Received 6 January 2021, accepted 13 June 2021 doi:10.3151/jact.19.730

Abstract

Radiation technology such as X-ray computed tomography (X-ray CT) is a powerful tool for materials research. Such non-destructive imaging technology allows us to visualize in three dimensions (3D) the internal structure of materials without damaging the specimen. This paper thus presents our studies on the application of microfocus X-ray CT, synchrotron radiation X-ray CT, and the integrated computed microtomography and X-ray diffraction (CT-XRD) method to advance cement and concrete research. CT images coupled with 3D image analyses allow us to identify and measure the air voids, pore scale microstructure and crack geometry. Image-based computational simulations provide us to estimate transport parameters such as diffusion tortuosity and water permeability in the digitized pore space. With the introduction of novel experimental techniques, deterioration or microstructure changes in hydrated cement systems, for example, attributed to calcium leaching, freeze-thaw cycles, elevated temperature, and steel reinforcement under tension are further elucidated. This review paper then ends with speculation of the future directions of concrete durability research via X-ray CT.

1. Introduction

Concrete is one of the most ubiquitous infrastructure materials in the built environment. Concrete made from Portland cement binder accounts for at least 30% of all material flow and is also considered second to freshwa- ter as the most widely used commodity on earth (Aitcin 2000; Bentur 2002). The global demand for concrete thus remains high and increases with urbanization and industrialization. For example, the global production of cement is expected to be around 5.5 billion metric tons in 2030 (Worrell et al. 2009). However, a significant carbon footprint is also associated with the production of these cement clinkers as these cement plants emit about two billion metric tons of CO₂ per year, which is about 5-7% of the global anthropogenic CO₂. Cement industries are also energy-intensive as they consume around 4 to 6 gigajoules (GJ) per metric tons of cement clinker produced (Taylor et al. 2006). Hence, there is a growing interest in future concrete which is envisioned as a sustainable and durable material to reduce its natu- ral resource and energy consumption, and consequently to reduce its life cycle’s environmental impact.

The durability of Portland cement-based concrete plays an important role in its sustainability, serviceability, and performance. A durable concrete structure which meets the requirement for functionality, strength, and stability through its design lifetime, rehabilitation and restoration cost will also become lower. This also lowers concrete consumption and accordingly, reduces its carbon footprint and environmental impact in the long term. Moreover, cement-based materials have varied applications, as they are not only used in the building and construction industry but also as an engineered barrier for radioactive waste disposal facilities of nuclear power plants. The durability of these barriers is very critical for considering a design lifetime of at least a thousand years or more. Thus, the future of concrete with improved and controlled durability will not only be advantageous from the cost perspective, but also for a sustainable carbon-constrained society.

Researchers worldwide have looked at the microstructure of concrete with great interest to understand and control its durability. The pore structure which ranges from few nanometers to micrometers including the cracks development in the cementitious matrix play a fundamental role in the physicochemical properties of concrete such as the compressive strength, permeability, and other transport properties that will affect its durability. The durability of the material is affected, for example, as the microstructure changes when in contact with water or exposed to an aggressive environment like elevated temperature and various climatic conditions such as freeze-thaw cycles.

The pore structure and cracks in concrete are typically characterized by a variety of experimental techniques such as liquid intrusion and gas adsorption porosimetry, and two-dimensional (2D) image microscopy analyses. Recently, X-ray computed tomography (X-ray CT) has been attracting more attention to many researchers to study the microstructure of cement-based materials (Monteiro et al. 2019; Brisard et al. 2020).
Why is X-ray computed tomography becoming then a mature technique to characterize and understand the microstructure of cementitious materials?

X-ray computed tomography is a non-destructive three-dimensional (3D) imaging technique that can provide morphological information of the microstructure of the cementitious matrix such as pore size distribution, pore connectivity, crack tortuosity, among others. In combination with the other experimental techniques, X-ray CT can provide additional data with valuable insights on the changes of microstructure before and after some tests.

Bossa et al. (2015) consider X-ray CT as a powerful tool to assess spatial heterogeneity of the internal microstructure because of the following features:

- No constraining specific specimen preparation requirement such as drying or surface polishing
- Non-destructive 3D imaging of microstructure with sufficient spatial resolution that is non-bulk dependent
- Direct measurement without requiring a hypothesis on pore geometry or interpolation

Figure 1 describes the result from Scopus-based bibliometric analysis, indicating an increasing trend of studies on X-ray CT applications to cement and concrete research in the last ten years (2009-2019). Scopus search of papers published from 2009 to 2019 with query (“X-ray” AND “computed” OR “tomography” OR “microtomography”) results to 227893 documents from its database. Filtering the search to Portland cement or concrete leads to 784 documents out of the initial 227893 documents (see Fig. 1). Furthermore, limiting the query (“X-ray” AND “computed” OR “tomography” OR “microtomography” AND (“Portland cement” or “concrete”) AND “durability” would result to 70 documents in the last ten years (2009-2019). This implies an opportunity to advance more research on concrete durability through X-ray computed tomography applications. Six of these 70 papers were co-authored by Prof. Sugiyama of Hokkaido University, i.e., the highest count in terms of number of documents by author.

1.1 Scope, purpose, and significance of the paper

The objective of this paper is to outline and review the previous and recent studies done by the authors on X-
ray computed tomography applications to cement and concrete research. The principle of the techniques based on synchrotron microtomography and microfocus X-ray computed tomography is presented with emphasis on both qualitative and quantitative illustrations to advance our understanding of the microstructure-related durability of concrete. This also includes the novel application of synchrotron-based integrated microtomography and X-ray diffraction analysis (CT-XRD) of cementitious materials.

2. Background of methodology

2.1 The foundation of X-ray computed tomography

The foundational concepts of X-ray computed tomography (CT) can be traced back to the mathematics established by the Austrian mathematician J. Radon in 1917, and the theoretical underpinning of computer-assisted tomography (CAT) scan developed by the South African physicist A. M. Cormack. Cormack showed the feasibility of using X-rays and a finite number of radiographic viewing directions to reconstruct the distribution of X-ray absorptivity within a cross-section of an object in the early 60s (Cormack 1963, 1964). And in 1971, the English electrical engineer G. N. Hounsfield built the first CT scanner for brain imaging at E. M. I. (Electric and Musical Industries) Laboratories in the United Kingdom, bringing Cormack’s theoretical calculation into a real application (Hounsfield 1973, 1980). Hounsfield also invented a quantitative scale of measuring radiodensity used by radiologists in interpreting CT images. Cormack and Hounsfield who both worked independently on the development of CAT were jointly awarded by Nobel prize for Medicine in 1979 (https://www.nobelprize.org/prizes/medicine/1979/press-release/).

The impact of X-ray computed tomography on medical diagnostics and health care has been growing since then. Advanced tomographic systems have also been developed for the medical practice, with a lower radiation dose, quite fast reconstruction times, and good image resolution (Stock 2020). More recently, X-ray CT at higher spatial resolutions (e.g., in nanometers and micrometers scale) has also been used to examine biological tissues, porous media, rocks, composite materials, and to many other industrial components. High resolution X-ray CT is also referred to microtomography or micro-CT. Though the division between the conventional CT and micro-CT is an artificial distinction, industrial CT scanners are considered micro-CT that include CT images obtained at least 10 to 100 micrometers spatial resolution.

How does X-ray computed tomography (CT) “see” the internal structure of the material like concrete without physically cutting the specimen as typically done in serial sectioning? It is a non-destructive imaging method where individual projections, also known as radiographs, recorded from different viewing directions are used to reconstruct the internal structure of the object of interest. This technique uses the fundamental principle of the interactions between X-rays and the materials when the specimen is bombarded by an X-ray beam and the transmitted beam is recorded on the detector. The X-ray CT scanning and imaging process is shown in Fig. 2.

The typical mode of scanning is attenuation-based where the X-ray intensity $I$ is attenuated following the Beer-Lambert’s law:

$$ I = I_0 e^{-\mu x} $$

where $I_0$ is the incident X-ray intensity and $x$ is the material thickness in the direction of X-ray transmission. The symbol $\mu$ is referred to as the linear attenuation coefficient (LAC), which is dependent on the X-ray energy and the atomic number of the material. Thus, CT allows the volumetric measurement of the X-ray attenuation or absorption within the object, creating images that map the variation of attenuation coefficient within the volume.

---

![Fig. 2 Schematics for X-ray CT scanning and imaging process.](image-url)
Charge-coupled device (CCD) or complementary metal-oxide semiconductor (CMOS) technologies are typically used as detectors to measure the transmitted X-ray intensities. The resulting image or radiographs is a superimposed information or projection of a volume in a two-dimensional (2D) plane. These projections are then taken at many angles or projection view to obtain the three-dimensional (3D) information. In medical CAT scan, this is commonly done by turning the X-ray source and the detector around the specimen (which in this case is a human patient). On the other hand, the specimen which is an inanimate object is rotated while the X-ray source and detector are fixed in positions in industrial CT scanners. Hence, the X-ray intensity projections are measured around the sample with incremental rotation.

These projections are then used to produce the contiguous 2D images using a reconstruction algorithm such as that of filter back-projection or Fourier transform-based methods. This two-dimensional image is typically referred to as “slice” because it is the virtual cross-section of what would be seen if the specimen were sliced along the scan plane. Each slice is a matrix of voxels (volume pixel) in which each voxel has grayscale value (GSV) related to the spatially-resolved linear attenuation coefficient (LAC). For visualization and storage purposes, these images are typically 8- or 16-bit grayscale values obtained through linear transformation of the LAC values that are mapped onto the reconstructed images (tomograms). Reconstructed cross-sectional images are then stacked in the perpendicular direction to obtain 3D microstructures.

In principle, we can identify the different features from these images since different material features and phases will have different X-ray absorption or attenuation properties. The volumetric image from CT scanning can thus provide valuable 3D structural information. The quality, resolution, and acquisition time of the CT scan would depend on the micro-CT setup, X-ray sources, detector system, and reconstruction algorithms including pre- and post-processing of the acquired data. However, the required spatial resolution needed for a particular application would depend on the microstructural features of interest. We can scan concrete specimen of size on the order of 10 cm, and the constituents of concrete such as binders, aggregates, and pores, air voids or cracks can be captured up to the resolution limit at the order of 10 micrometers to sub-millimeter scale. The sub-micrometer spatial resolution will be needed to “see” the capillary pore structures at the constituents of mortar and pore/solid-phase microstructures of the cement paste. Specimens or volume of interest (VOI) of the size on the order of 1 mm are used for scanning at higher spatial resolution.

There are two X-ray sources in CT systems that are typically used to achieve the high spatial resolution requirement. The first one uses the polychromatic divergent beam generated from a micro-focus X-ray tube. The second one uses the monochromatic parallel beam from synchrotron radiation (see Fig. 2). In the next section, description of the XRCT systems used in our studies including the basic image processing and analysis workflow are presented.

### 2.2 Micro-focus X-ray computed tomography

Industrial CT scanner is becoming popular for laboratory-based nondestructive testing of materials for various applications providing spatial resolutions higher than medical CT scanners. These micro-CT scanners uses microfocus X-ray tube as X-ray source. As the spatial resolution of the CT image is primarily decided by the effective focal spot size, the microfocus X-ray tube is one of the essential components of the XRCT system. Note that the spatial resolution of conventional CT systems is typically limited by the geometry of the X-ray beam along with the characteristics of the detector (Landis and Keane 2010). For a fan or cone beam, the spot size of the X-ray source is important since the smaller the spot size, the more accurate the projected image with less penumbral blurring. In other words, the larger spot could result to photons hitting a particular pixel which are traced back through different ray paths through the specimens. This leads to significant noise in the tomographic reconstruction.

At Hokkaido University, we use the microfocus CT system of TOSCANER-3000µhd (Toshiba IT & Control Systems Corporation, Japan). It consists of a micro-focus X-ray source, a specimen manipulator, an image intensifier (I) detector coupled to CCD camera, and an image processing unit (see Fig. 3). The X-ray source has a 4-micrometer focal point with an X-ray tube voltage range from 20-225 kV. The source-to-specimen distance and source-to-detector distance can be varied to obtain the desired geometric magnification. The micromanipulator precisely positions the specimen table in the X-ray beam, and rotates it under computer control through 360 degrees for projection data acquisition. The host computer receives the projection images and performs the data processing, and the image reconstruction using a built-in program for cone beam geometry.

For example, a power setting of 130 kV and 124 µA was used typically for our experiment to study the frost damage in mortar (Promentilla and Sugiyama 2010a). The specimen of 1 to 10 cm in size was set in a holder mounted on a precision rotation table, and then the table position was adjusted to fit the image within the field view. Scanning parameters and conditions are adjusted to obtain high resolution images, and to reduce the noise and artifacts in the images.

During scanning, a conical X-ray beam is emitted as the specimen rotates through 360°. The X-ray CT raw data were then collected every 0.24°, for a total of 1500 projection images that were recorded by a CCD camera with an array of 1024 x 1024 pixels. Using the in-house reconstruction algorithm of the system, a set of contiguous slices or cross-sectional images in x-y plane was obtained wherein each slice is an array of voxels (vol-
ume pixel). Stacked up in z-direction, these contiguous slices form the volumetric image.

The unit voxel size will depend on the scanning parameters set during image acquisition. For example, each slice of 40 micrometer thickness with a matrix size of 1024 x 1024 pixels and an in-plane resolution of 10 µm/pixel will have anisotropic voxel dimension of 10 µm x 10 µm x 40 µm. Each voxel is also associated with a raw CT number (CTN). CT number or Hounsfield unit measures X-ray linear attenuation of component inside the voxel.

2.3 Synchrotron microtomography
Using synchrotron radiation as an X-ray source has led to significant development in microtomographic imaging. There are several advantages of using synchrotron-based X-ray CT system to enhance the imaging process (Landis and Keane 2010).

First, the X-ray source provides the high flux that can resolve very subtle variations in absorptivity for better imaging of internal structure. Synchrotron radiation results from the bending of a high-energy electron beam due to a magnetic field. Thus, the emitted light is many orders of magnitude greater in brightness than that emitted by conventional X-ray sources. The second advantage pertains to its X-ray collimation beam in a parallel beam configuration. This simplifies the tomographic reconstruction algorithm, and the tunability of the X-ray energy to a narrow energy band. The third advantage pertains to its monochromatic X-ray beam. This improves the accuracy of the reconstructed tomographic images by eliminating the issue of energy dependence on X-ray absorption. Noted that to realize most of the advantages of synchrotron X-ray sources, imaging is typically limited to relatively small specimens of mm in size. With these high-quality imaging capabilities of synchrotron radiation, many researchers use this technique to better “see” the microstructure of cementitious materials in three dimensions at a sub-micron level of spatial resolution.

For our studies that require sub-micron spatial resolution, we use the X-ray CT system (BL20XU) in SPring-8 (Super Photon ring-8 GeV). SPring-8 is located at Hyogo, Japan and it is one of the world’s largest third generation synchrotron radiation facility. A schematic illustration of synchrotron microtomography is shown in Fig. 4. The X-ray CT system in SPring-8 consists of an X-ray light source from the beam line, double crystal monochromator, high precision rotation stage, and high resolution X-ray image detector (Promentilla et al. 2008b, 2009).

For example, the X-ray energy was tuned to X-ray beam energy of 15 keV to study the microstructure of deteriorated cement paste (Sugiyama et al. 2010). Using a high precision rotation stage, the image was taken at different views through 180 degrees rotation. The transmitted images are then detected by X-ray imaged detector which consists of thin scintillator, optic system and CCD camera. Tomographic reconstruction is done using a public domain computer program in use at SPring-8, which employs the convolution back projection algorithm to generate the slice images. The stack of these slices provides the reconstructed volumetric data of the scanned object. The reconstructed 3D-image data set was composed of 1300 contiguous grayscale images where each slice image contained 2000 x 2000 voxels (volume element or 3D pixel). The effective size of a cubic voxel in the CT image is 0.50 µm. Each voxel has grayscale value (GSV) that correspond to CT values, i.e., obtained through linear transformation of LACs.

2.4 Nondestructive integrated CT-XRD method
A non-destructive integrated computed microtomography and X-ray diffraction (CT-XRD) method was developed in 2013, and since then, the CT-XRD system has been improved while being applied to explore new aspects in the hydrated cement system of concrete (Sugiyama et al. 2014; Takahashi and Sugiyama 2016). This innovative measurement system is constructed in the beamline of 28B2 at SPring-8 in Japan. The schematic illustration of its setup is shown in Fig. 5. First, X-ray computed tomography (CT) is conducted to ob-
tain three-dimensional (3D) images followed by the determination of the regions of interest in the sample under measurement based on the reconstructed image. Then, localized X-ray diffraction analysis (XRD) is conducted on the assigned region. These operations are continuously implemented in-situ without the removal of the sample from the stage inside the beamline hutch.

Beamline in 28B2 permits to emit a white X-ray so that the diffraction spectrum with energy-dispersive X-ray is obtained. As compared with the angle dispersive

---

**Fig. 4** An example of synchrotron X-ray CT system.

**Fig. 5** Schematic illustration of the non-destructive integrated CT-XRD method.
X-ray diffraction, this can reduce the measurement time in the analysis. However, a monocrystal silicon is used to convert to monochromatic X-ray to enhance the image contrast for CT measurement. With an updated version of the overall arrangement of devices as shown in Fig. 5, the silicon is in the upstream position, and therefore the monochromatic X-ray is directly emitted on the sample. Typically, the X-ray energy is 25 keV to study the microstructure of cement-based materials. After the completion of the CT measurement, the arrangement is changed to direct white X-ray to the sample on the optical axis. Then the diffracted X-ray is collected at a specified angle by the solid-state detector. The angle is set at 10 degrees (two theta). These changes are all automated using a control program. In addition, the region (gauge volume) in the sample for emitting an X-ray signal is limited by using the slits on the incident side and the light receiving side.

2.5 Digital image processing and analysis

Image processing is usually the first step to prepare the 3D dataset obtained from stacking the contiguous slices. There are public domain programs such as ImageJ (Rasband 2007) and SLICE (Nakano et al. 2006), which were used in our studies for processing and analysis of CT images. For example, in the case of an 8-bit slice image, each voxel in the image has a grayscale value (GSV) between 0 and 255 (total of 2^8 values), whereas a 16-bit image is composed of grayscale values ranging from 0 to 65535 (total of 2^16 values); these voxel values can be presented by a histogram as shown in Fig. 6.

Note that the histogram is the frequency distribution of intensity values (GSV) in the image. In an 8-bit grayscale image, voxels with 0 GSV is imaged as black which corresponds to the material feature or phase with minimum LAC (e.g., air). Voxels with 255 GSV is imaged as white which corresponds to the material feature or phase with maximum LAC (e.g., unreacted cement clinker). Prior to extraction and quantification of microstructural feature of interest, a typical workflow in preparing the data sets include enhancing image through contrast stretching, filtering and thresholding.

(1) Normalization

Contrast stretching or normalization is a simple image enhancement technique that attempts to improve the contrast in an image by 'stretching' the range of intensity values. Such range contains to span a desired range of values, i.e., the full range of voxel values that the image type concerned allows. For example, normalization was applied to the whole stack of reconstructed slices from the synchrotron-based X-ray CT described in Promentilla et al. (2008b).

Figure 7 describes an example of a histogram of LAC in a reconstructed image. By setting the minimum (Min) and maximum (Max) cut-off for LAC values, these LAC values are transformed linearly to voxel’s GSV using the following equation:

\[ GSV = 0 \quad \text{if} \quad LAC \leq \text{Min} \]

\[ GSV = \frac{255}{\text{Max} - \text{Min}} (LAC - \text{Min}) \quad \text{if} \quad \text{Min} < LAC < \text{Max} \]

\[ GSV = 255 \quad \text{if} \quad LAC \geq \text{Max} \]

This process of normalization even out the brightness and contrast variation among the slices while enhancing the contrast in the said image. In addition, the set of images was also down-sampled from 16-bit to 8-bit grayscale images resulting in a smaller image file size.

(2) Filtering

Filters are image processing techniques applied to each voxel and its neighbors to remove noise or enhance further the image. These filters use mathematical algorithm to replace any voxel value in grayscale images that is inconsistent with its neighbors. One common approach is by convoluting a kernel (matrix) to the image. The kernel holds some values that would modify the image, and depending on the values, several tasks such as denoising, blurring, sharpening, and edge detection, can be performed (Burger and Burge 2010). For example, Gaussian and mean filters are used for denoising but drawback of these filters is that it blurs the image, including the phase boundaries which may be critical in the segmentation process. In our study, we typically use
edge-preserving filters such as median filter and anisotropic diffusion filter to avoid such drawback (Sheppard et al. 2004). Sharpening and edge detection filters such as Laplacian filters, Sobel, and Canny filter are another set of filters that can also be used prior to pore or crack detection and feature extraction.

(3) Segmentation

Segmentation is an image processing and analysis technique that partitions the image into multiple segments. There are many image segmentation methods reviewed in the literature for their performance and applications (e.g., see Abera et al. 2017). For concrete, we are typically interested to investigate the void spaces in the cementitious matrix. These void spaces include the capillary pores, air voids, and cracks that can be resolved at a given spatial resolution. Thus, segmentation becomes synonymous to binarization, i.e., segmenting the image into two to classify the void and solid phase. Typically, the segmentation process replaces voxel values of the phase of interest with a binary value of “1” (in physical terms these voxels are treated as void space) and the values of all other voxels with “0” (solid space).

Figure 8 illustrates how segmentation works using thresholding technique. Details of the specimen are described elsewhere (Promentilla and Sugiyama 2007a). Thresholding is a common term to describe the introduction of threshold value for binarization, which segments the features that are below the threshold and others that are above the threshold value. The most widely used approach is the Otsu method (Otsu 1979). This algorithm was originally invented to binarize images by using a weighted sum of variances of the two classes. However, Otsu thresholding may not work properly for heterogeneous samples and partial volume effects are significant.

Another useful approach is to assign the threshold value with the grayscale intensity of the voxel within the valley that separate the peaks based on histogram analysis. In the absence of a well-defined valley between histogram peaks, one common segmentation approach is assignment by inspection by selecting the threshold which best preserves the important fine-scale features of both the solid and the pore structure. Given
that this is a subjective process, we proposed in our previous studies a simple and yet robust method in assigning a threshold based on the porosity-threshold dependency curve (Promentilla et al. 2008b).

The image segmentation method using machine learning such as that of deep convolutional neural network have improved significantly in the recent few years that could address the noise and artifacts found in CT images. For example, recent techniques in image analysis of cementitious materials that employ machine learning can precisely segment microcracks (Dong et al. 2020) or the void, fibers and sand in a cement-based composite (Lorenzoni et al. 2020). Recently, novel techniques were also introduced that separate the cracks or pore from the solid by combining filtering and morphological operations in the 3D images of concrete (Mac et al. 2021).

3. Microstructure characterization of hydrated cement systems

3.1 3D pore microstructure and its quantification

Consider the pore microstructure of a deteriorated mortar as described by its CT image in Fig. 9a. The deterioration was accelerated with an electrical migration method to leach out calcium (Hitomi et al. 2007). The 3D image dataset consists of 1300 contiguous slices with dimensions of 2000 x 2000 voxels. One voxel has a physical size of 0.5 µm x 0.5 µm x 0.5 µm. The volume of interest (VOI) was selected as we were interested in the pore microstructure of cement pastes in the deteriorated mortar. The size of VOI is 300 x 300 x 300 voxels. In physical terms, the VOI is a cubic cement paste specimen of 150 µm size.

These contiguous slices containing selected region of interest produces 3D image such as that of the VOI (e.g., see Fig. 9b). Each slice (see Fig. 9c) is a matrix of voxels in which each voxel is associated with a grayscale value (GSV) that is related to the measured linear attenuation coefficient (LAC). In this figure, the pore or void space with lower attenuation coefficient is imaged as darker voxel compared with that of solid matrix. Segmentation of the image then resulted to binary image as shown in Fig. 9d to separate the pore voxels (black) from the solid voxels (white).

(1) Pore connectivity

To extract the largest percolating pore cluster, the pore connectivity in three dimensions was determined from the stack of images described in Fig. 9. Note that in voxel value-based thresholding, the voxels are selected primarily on the basis of their value and not on their location. We are more interested in the connected porosity volume as this property will play an important role in the transport process in the cement paste. Thus, we use the multiple cluster labeling technique from percolation theory (Ikeda et al. 2000; Stauffer and Aharony 1994) to identify individual connected pore voxels and provide each with a unique label. Labeling indicates that any pore cluster with a distinct label is disconnected from any other pore clusters in the binary image. The algorithm of Hoshen and Kopelman (1976) for multiple cluster labeling technique was used with the 6-point connectivity rule. Using this rule, when a pore voxel is sharing a common face with another voxel, the two voxels are connected; whereas those pore voxels that are in contact only at the vertex or edge are disconnected.

The labeled largest pore cluster in 2D slice image (imaged as white in Fig. 9e) seems to be disconnected, but these pore voxels are actually connected in three dimensions. Figure 10 illustrates the rendered volume of the pore space including the largest percolating pore cluster extracted from the VOI. For example, about 94% of the extracted pore space from the VOI of deteriorated mortar are connected to each other. The cluster labeling analysis thus allows us to separate the isolated and poorly connected pore clusters from the largest percolat-

---

Fig. 8 Application of thresholding technique in segmenting CT image of cement paste (left). The binary image (right) contains void space (black) and solid matrix (white) (Promentilla and Sugiyama 2007a).
ing pore cluster.

In physical terms, this well-connected pore space in the selected VOI would most likely contribute to the macroscopic transport property of the material. Note that the percolation defined here in three dimensions refers to this largest pore cluster that contains voxels that are connected to the six faces of the cubic VOI. In other words, a continuous path through the pore space exists when a fluid enters in one of the faces and exit to any of the other faces of the VOI.

Proper segmentation of the void space thus plays an important task in the workflow to characterize the pore microstructure and quantify the micro-geometry of the void space. In some hydrated cement pastes such as pervious concrete, the macropores or void spaces are relatively easier to segment wherein global thresholding will do to separate the pore from solid in the gray-scale image. Figure 11 describes the binarization of the CT image of pervious mortar.

For example, the histogram of the sample slice of pervious mortar (Fig. 11a) depicts a bimodal distribution (Fig. 11b) wherein two peaks suggest two major
phases, i.e., the void space and the solid matrix. Global thresholding was applied by setting a threshold value at the valley or the lowest point between the two peaks as shown in Fig. 11d. If the grayscale value of a voxel is less than or equal to the threshold value (e.g., 40), it will be set to zero (black voxel) to segment the void space. Otherwise, the grayscale value of the voxel is set to 255 (white) to segment the solid matrix. The resulting binary image in Fig. 11c can then be subjected multiple cluster labeling to identify the largest percolating pore space. Likewise, the rendered volume of the pore space including the largest percolating pore cluster from the CT images of pervious mortar is shown in Figs. 11e-11f.

(2) Pore volume, specific surface and mean pore size

Once the largest percolating pore cluster is identified from the VOI, further image analysis can be used to characterize this connected pore space. For example, it is easy to calculate the total volume of this cluster by summing up the number of voxels in that cluster, and multiplied by the physical dimension of a unit voxel size. On the other hand, interfacial area between the pore and solid matrix can be calculated using a simple face counting algorithm. However, the voxel staircase effect in the digitized 3D image (e.g., see Fig. 12) would result an overestimation of surface area using this algorithm (Nakashima and Kamiya, 2010).

A more accurate technique for obtaining surface area from a 3D image is by getting the isosurface based on marching cube algorithm (Lorensen and Cline 1987). This algorithm approximates the isosurface between the two phases from a generated triangular mesh. The said image analysis routines were checked by running them on a digitized sphere of known geometry and comparing the results to the expected results. Figure 12 summarizes the computed specific surface or surface-to-volume ($S/V$) ratio of a digitized sphere of 20-pixel radius. With the use of a simple face counting algorithm, the surface area and $S/V$ of the said sphere are overestimated by a factor of 1.5 while the values obtained from the marching cube algorithm provide a better estimate.

As for the mean pore size of the percolating pore cluster, the maximal sphere algorithm can be used as
this method does not make any assumptions of underlying geometry (Hildebrand and Ruegsegger 1997). The definition of pore size here is the thickness or the maximum diameter of a sphere that would fit in the pore volume. This model-independent algorithm estimates the “local thickness” of the pore structure by fitting maximal spheres to every point in the structure and from this local thickness, the volume-weighted mean pore size is then computed.

Table 1 summarizes the results obtained from the image analysis and quantification of pore structure micro-geometry. Despite the challenges in characterizing the pore microstructure in cement pastes, quantification of such pore micro-geometry from CT images can provide an insight into the transport properties of cementitious matrix through a connected porous network.

### (3) Diffusion tortuosity

Diffusion tortuosity is a parameter that can be used to characterize the complex pore micro-geometries that are relevant to many transport properties of fluids confined in a porous medium. It has been suggested that the time-dependent self-diffusion coefficient \(D(t)\) associated with the random Brownian motion of molecules can be used to probe the geometry of porous media (Latour et al. 1995; Sen 2004).

In bulk fluid, the mean square displacement of molecules due to Brownian motion is linearly proportional to the diffusion time and that proportionality constant is commonly referred to as bulk self-diffusion coefficient. On the other hand, the diffusion is restricted by the solid surface bounding the pore space when the fluid is confined in a porous medium. In the context of self-diffusion, tortuosity can be used to describe the longer connecting path imposed by obstacles (solid matrix) within the porous media relative to that motion in unconstrained free space or bulk fluid. As diffusion is described by the random thermal motion of molecules and atoms, the diffusion tortuosity is defined as follows (Nakashima and Kamiya 2007; Promentilla et al. 2009):

\[
\tau_{D} = \frac{D_0}{D_{\infty}}
\]

Here \(D_0\) refers to the time-independent bulk diffusion coefficient in free space (100% porosity) while \(D_{\infty}\) is the limiting value of the apparent diffusion coefficient in a well-connected pore space at long diffusion time. In this long-time limit, the walkers fully experience the connectivity and high tortuosity in the system and the self-diffusion coefficient reaches a constant value. For unrestricted diffusion, \(D_0\) is equal to \(D(t)\). Thus, the diffusion tortuosity in free space is expected to be one.

Diffusion tortuosity in a restricted space such as that pore structure in cement paste can be determined by employing random walk simulation in the digitized pore

| Table 1. Summary of pore structure parameters obtained from VOI of deteriorated mortar. |
|---------------------------------------------------------------|
| **Pore structure micro-geometry** | **Deteriorated mortar** |
| Total volume of pores (V) | 0.000862 mm³ |
| Porosity | 0.26 |
| Volume of the largest percolating pore cluster (V_c) | 0.000813 mm³ |
| Degree of connectivity (V_c/V) | 0.94 |
| Surface area (S) | 0.34 mm² |
| Specific surface (S/V) | 400 mm⁻¹ |
| Volume-weighted mean pore size (d_p) | 956 ± 327 µm |

Fig. 12 An illustration of staircase effect on measuring the surface to volume ratio of a digitized sphere.
space in three dimensions. The time-dependent self-diffusion coefficient $D(t)$ of a number non-sorbing walkers can be estimated from the time derivative of the mean square displacement $\langle r^2 \rangle$ such that:

$$D(t) = \frac{1}{6} \frac{d\langle (r^2(t)) \rangle}{dt}$$

where $t$ refers to the dimensionless lattice walk time. Note that the symbol $\langle \rangle$ denotes the average over all the initial and final positions of the walkers.

**Figure 13** describes how diffusion tortuosity can be computed from the mean square displacement vs time obtained from random walk simulation. At this long-time limit (e.g., one million of lattice walk time), the slope of MSD vs time approaches a value that is equivalent to the reciprocal of diffusion tortuosity. In a free space (100% porosity), the slope approaches 1.0, i.e., tortuosity is 1.0. In restricted space (open pore network), the slope is less than 1 but greater than zero, resulting to a diffusion tortuosity of greater than 1.0. On the other hand, in an isolated pore, the slope approaches zero resulting to an infinite diffusion tortuosity. This random walk simulation technique has been applied to digitized three-dimensional pore space in cement paste extracted from CT images. Details of implementation of this technique is described elsewhere (Promentilla et al. 2008b, 2009).

**Figure 14** illustrates a sample trajectory of a walker in a random walk simulation. **Table 2** summarized the result of random walk simulation in percolating pore space in three dimensions using 100,000 walkers at

| Pore Space                  | Free space in VOI (100% porosity) | Percolating pore space in VOI (24% porosity) | Isolated sphere in VOI |
|-----------------------------|----------------------------------|---------------------------------------------|------------------------|
| Slope of MSD vs time after $t = 1$ million | 1.0                              | 0.13                                        | 0.0                    |
| 3D Diffusion Tortuosity ($\tau_D$) | 1.0                              | 7.70                                        | $\infty$              |

**Table 2. Summary of diffusion tortuosity parameters in different pore space geometries.**

---

**Mean square displacement vs time**

Fig. 13 Quantification of tortuosity from random walk simulation.

**Inverse of Diffusion Tortuosity vs time**

Fig. 14 A sample trajectory of a walker in a 3D digitized pore space.
maximum lattice walk time of 2 million.

3.2 3D distribution of entrained air and its quantification

Entrained air voids are known to play a beneficial role in freeze-thaw durability of concrete. Air-void diameter and air-void system parameters such as the spacing factor and air content is generally computed from image obtained from an optimal microscope using the procedure described in ASTM C457. This study uses micro-focus X-ray CT coupled with 3D image analysis as an alternative approach to measure such parameters.

Figure 15 describes the region (ROI), and volume of interest (VOI) extracted from the CT images of an air-entrained fly ash mortar (Promentilla et al. 2008a). This mortar has a sand-to-cement ratio of 3.8, a water-to-binder ratio of 0.50, with fly ash replacement of 30%. Prior to the extraction and quantification of air voids, contrast enhancing, denoising, smoothing of the images in the VOI were done. A supervised thresholding for region-based segmentation was applied to obtain a set of binary images wherein white and black voxels represent the air void and solid matrix, respectively.

Watershed algorithm is then used to separate the overlapping air voids. The term “watershed” comes from the analogy wherein the grayscale image is treated as topographic surface, i.e., the pixel intensity corresponds to the height of the point in the map (Vincent and Soille 1991). Accordingly, the resulting watershed lines mark the boundaries of “catchment basins” in the image, Fig. 16 illustrates in two dimensions this watershed-based method of splitting the overlapping air voids.

Connectivity analysis through multiple cluster labeling technique was applied to identify and measure each individual air void, with the exclusion of those voids that are connected to the edges or face boundaries of the VOI. The local entrained air content and air voids number density can be computed from this analysis. This also includes the computation of the equivalent diameter, which is the diameter of the sphere having a volume equivalent to that of the said air void. Figure 17 shows the visualization and quantification of air voids system including its corresponding air void equivalent diameter distribution.

3.3 3D crack geometry

Characterizing the cracks in cement-based materials could play an important role to our understanding of the post-damage behavior of these materials. Such damage can be caused by various factors that result in different levels and configuration of cracks. This study illustrates how X-ray CT can be used to describe crack geometry in concrete.

Figure 18 illustrates the visualization of cracks from a frost-induced damaged mortar described in Promentilla and Sugiyama (2010a). The crack width distribution (see Fig. 19) was measured using the same algorithm to measure pore size, i.e., based on local thickness calculation from the direct distance transformation method developed by Hilderbrand and Ruegsegger (1997).
Crack tortuosity (see Fig. 20) can also be measured using the 3D medial axis method (3DMA) developed by Lindquist (1999). The 3D medial axis transform of the digitized crack, often referred to as a ‘skeleton’, refers to a simplified representation of the original 3D data in which important information about the topology and geometry of the original structure is maintained. The geometric path tortuosity ($\tau_g$) was calculated as the shortest paths along the void medial axis for x, y, and z-directions using $\tau_g = l/\Delta$ where $l$ is the actual path length and $\Delta$ is the linear separation of the two parallel planes of the VOI.

4. Alteration and damage evaluation of hydrated cement systems

4.1 Effect of calcium leaching on pore structure

Durability of concrete through time can be compromised through its exposure to water during its service life. When water percolates through an open crack or connected pore space and transports aggressive ions such as chlorides and sulfates, this causes deterioration. Progressive deterioration of concrete used in dams, underground storage, and radioactive waste repository also begins at the surface where abrasion occurs when in contact with water for a long time due to calcium leach-

![Fig. 17 Visualization and quantification of air void distribution from CT images of air-entrained fly ash mortar (Promentilla et al. 2008a).](image)

![Fig. 18 Visualization of crack geometry from CT images of a frost-induced damaged mortar (Promentilla and Sugiyama 2010a).](image)
ing out from the matrix. Investigation of the changes of
the microstructure of hydrated cement systems is thus
important to assess the long-term durability of the mate-
rial.

The material studied was an ordinary Portland cement
mortar with a water-to-cement ratio of 0.50, and a sand
(maximum particle size of 210 µm) to cement ratio of
2.0. The test specimen was cured further for about 20
weeks before it was subjected to an accelerated leaching
test for 13 weeks. This leaching test based on the princi-
ple of electrochemical migration is shown to simulate
the long-time degradation of cementitious material due
to leaching of calcium ions when in contact with water
(Sugiyama et al. 2010). The dissolution of hydrated
cement products such as portlandite is accelerated
through application of electrical field, as calcium ions
move rapidly to the cathode side.

Figure 21 describes the experimental set-up for ac-

Fig. 19 Measurement of crack width distribution from CT images of a frost-induced damaged mortar.

Fig. 20 Measurement of crack tortuosity from CT images of a frost-induced damaged mortar.

Fig. 21 Experimental set-up to study the deterioration of cement paste due to calcium leaching (Promentilla et al. 2016).
celerated leaching and the locations where the specimens were obtained for CT scanning. Synchrotron-based X-ray CT systems described in section 2.3 was used for image acquisitions. CT images were then processed and analyzed as described in Promentilla et al. (2016). Such analyses include not only the characterization of pore scale microgeometry but also the image-based transport modeling to compute diffusion tortuosity and water permeability in the digitized 3D pore structure.

Figure 22 summarizes the results of the analysis for the 5 samples representing the different regions of specimens exposed to accelerated leaching test. The specimen with the highest effective porosity and mean pore size was from OPC_de1. Moreover, the OPC_de2 was observed to less porous than OPC_de1, and the least porous was OPC_de3 which is located at the center of the specimen. Indications suggest that the rate of deterioration is significant to the region where the surface of the specimen was mostly exposed to water, and also nearest to the cathode side of the accelerated leaching test. Likewise, OPC_de5 was also found to be the second most porous microstructure which is the other end of the specimen wherein the surface was also exposed to water. Although OPC_de5 was also exposed to water, the OPC_de1 is more deteriorated since the movement of calcium ions toward the cathode side results in an increased dissolution rate of calcium hydroxide and other hydrated cement products in the cement matrix.

These changes of pore microstructure results to changes in transport properties such as tortuosity which is computed from square root of diffusion tortuosity. Intrinsic permeability was also computed through simulation in the digitized percolating pore space using a 3D linear Stokes solver (Promentilla et al. 2016). As expected, deterioration of the hydrated cement systems leads to a porous microstructure that is less tortuous and more permeable to mass and fluid transport.

4.2 Damage in microstructure due to freezing and thawing action

It is known that normal concrete is not resistant to cyclic freezing-thawing action which may result in surface scaling and internal cracking. Development of internal cracks can be visualized through the aid of X-ray CT. For example, microfocus X-ray CT described in section 2.2 was used to observe the formation of cracks at different freeze-thaw (FTC) cycles. Deterioration of the mortar through observation of the internal structure suggests how cracks developed inside the specimen as the number of FT cycle increases (see Fig. 23). Most of these cracks formed along the weaker interfacial transition zone (ITZ) of the sand and cement paste in the frost-induced mortar. This crack development results in an increase of void fraction as shown in Fig. 24. Details of the analysis are described in Promentilla and Sugiyama (2010a, 2010b).

4.3 Damage in microstructure due to elevated temperatures

Alteration of hydrated cement system due to elevated temperatures was investigated with the non-destructive integrated CT-XRD method (Takahashi and Sugiyama 2019) described in section 2.4. Cement paste with the Ordinary Portland cement (OPC) as the binder was used in this experiment. The water to cement ratio was 0.60. The cement paste was mixed and molded into a metal frame in the size of 40×40×160 mm. The sample was

Fig. 22 Microstructure-transport parameters obtained from CT images of deteriorated cement paste due to accelerated leaching (Promentilla et al. 2016).
de-molded after 24 hours of molding, and then cured under water until the cutting of specimen. After about 12 months of water-curing, the hardened cement paste was cut into the shape of a prism with the dimensions of 2.5×2.5×5.0 mm for the CT-XRD method. Inner part of the paste was selected for cutting so as not to disturb the quality of the prism during water curing.

Elevated temperature test was carried out using electric furnace with a temperature control program as shown in Fig. 25. The rate of temperature increase was set at 10°C per minute until the target maximum temperature of 200°C was reached. The maximum temperature was maintained for 2 hours and the rate of heat decrease was also set at 10°C per minute until room temperature. After heating the specimen, it was removed and set on the rig for the CT-XRD measurement.

Setup conditions of the CT-XRD method are as follows: the extracted energy of X-ray CT measurement was 25 keV. Angle steps during CT measurement were 0.12° with an exposure time of 0.4 s. Image resolution was 2.44 μm/voxel. The beam size was 0.05 mm in width and 0.3 mm in height. The angle of diffraction (θ) was fixed at 5° and the preset time was 300 s for XRD measurement to determine the crystalline phases of the heated cement paste.

Powder X-ray diffraction (P-XRD) with the monochromatic X-ray using a laboratory X-ray diffractometer apparatus (SmartLab, Rigaku) was also carried out. This P-XRD result was compared with that of the XRD measurement from the CT-XRD method. For P-XRD analysis, heated sample was grinded into powder in a diameter of less than 100 μm, and was tested immediately after grinding. XRD data were recorded in which the scanning range was 2θ = 10° to 60° degrees with a 0.01 step scan and a scanning speed of 5.5 degrees per
The diffraction profile was acquired and analyzed using powder diffraction analysis software “PDXL” from Rigaku.

Figure 26 shows microtomographic images of the heated sample by the CT measurement from the CT-XRD method. The cross section is located in the middle height of sample. Light gray indicates the cement paste and dark color represents the air and void space. The 3D dataset consists of contiguous 300 slices wherein each slice has a matrix size of 1280 × 1280 voxels. Examining the CT image, no crack is observed with the given temperature regime. Accordingly, little physical transformation in the heated cement paste occurs. Indication suggests that the strength reduction is minimal.

In the cross section obtained from the CT measurement, the ROIs (Region of Interests) of the XRD measurement in the CT-XRD were determined for the X-ray diffraction profiles, and labeled as Point 1 to Point 3 from the corner to the center (as shown as circles in Fig. 26). The results of XRD profiles are listed in the order of P-XRD, Point 1, Point 2, and Point 3 that are shown from top to bottom.

Examining the powder X-ray diffraction result (P-XRD), the peaks of CH (calcium hydroxide) and CaCO₃ were distinct. It is considered that the presence of CaCO₃ is the result of carbonation influenced by the atmosphere of the high temperature chamber. In addition, peaks of C₃S (belite) and CaO were observed. C₃S is derived from the presence of anhydrated cement grains. The powder X-ray diffraction is the result obtained from the heated hardened cement paste that were grinded to powder prior to XRD analysis. Therefore, it shows the average alteration within the hardened cement paste when subjected to elevated temperature for two hours.

On the other hand, the X-ray diffraction analysis from the CT-XRD method at three different positions of the hardened cement paste show the following results. First, there are many CaCO₃ peaks especially in the surface layer (Point 1). Second, CH is identified to be present from the surface to the inside (Points 2 and 3). Whether or not that the CH formation in the inner region is due to the rehydration with evaporating water remains to be seen. This phenomenon has been suggested initially to explain the crack growth observed from another X-ray CT study (Henry et al. 2016) of cement paste subjected to heating and water re-curing. On the other hand, CH can remain in the inner regions of the heated sample as hydrated cement product initially.

In the CT-XRD method, the lower X-ray energy would be absorbed within the sample during transmitting and diffracting while doing XRD measurement. Accordingly, the results associated with the lower angles were difficult to measure. Nonetheless, the CT-XRD method has verified that the chemical transformation of the hydrated cement pastes progresses inward.
according to the high temperature process. Therefore, it can be said that the CT-XRD method using a small sample can improve the spatiotemporal resolution of the altered state of the cement paste when subjected to elevated temperature.

4.4 Crack formation around steel reinforcement by tension

Bond between concrete and deformed steel bar is a key for assuring structural performance of reinforced concrete structures. The reduction of the bond strength can thus result in decreased performance and must be avoided. However, it has been reported that cracks develop around the bar in concrete. For example, Goto (1971) observed internally developed cracks in the vicinity of deformed tension rebar using a red ink method. In addition, X-ray radiography has been used to detect cracks inside reinforced concrete specimen when subjected to either pull out test or bending test. Although several attempts have been made, few studies have been conducted on the direct observation of the development of cracks around a reinforced steel bar embedded in concrete using X-ray CT technique. The following problems were also pointed out in using X-ray CT for such purpose:

- Closure of the (opening) internal cracks during CT measurement in unloaded specimen
- Necessity of a sophisticated loading system for in-situ loading test with X-ray CT machine.

In this study, these problems are solved by a simple and inexpensive system using a sleeve bar in concrete. Sleeve bar is designed to sustain tensile stress in concrete as shown in Fig. 27. The sleeve of a diameter of 13mm was processed using a regular deformed steel bar (D13) according to JIS G3112 (Takahashi et al. 2017).

The bar has a hole in the center with an inner diameter of 9mm while a hexagon nut with an inner diameter of 8 mm is welded on the upper end of the bar (hereinafter referred to as sleeve reinforcing bar). The hole does not penetrate through the full length of the sleeve but a 10 mm thick bottom on the rebar remains. In addition, a bolt that is 10 mm longer than the hole length is used and inserted into the sleeve rebar (see Fig. 27). A tensile force is applied by rotating the bolt. When the bolt is rotated further after it comes into contact with the bottom of the hole in the rebar, the nut at the top of the sleeve rebar and the bottom of the hole are pushed by the screw in the nut and the bottom tip of the bolt, respectively.

Concrete cylinder of a diameter of 55 mm and a length of 200 mm was tested. The water to cement ratio was 0.55. River sand and gravel with saturated surface dry density of 2.64 and 2.71 g/cm³, respectively were used in the concrete mix. Maximum size of the coarse aggregate was 15 mm. The compressive strength was 31.2 N/mm². The tensile force was applied to the sleeve rebar and increased in a step wise manner. At each load level, the X-ray CT measurement was carried out. It is noted that during the measurement, the tensile force was maintained due to the tension of the sleeve bar. Micromatrix X-ray CT was employed in this study using a voltage of 200 kV. The detector of X-ray was a flat panel with 8.0 x 8.0 inches in the view size. Resolution was 0.057 mm per pixel and the slice thickness was also 0.057 mm.

Figure 28 shows the cross-sectional images in the axial direction of the concrete specimen at increased widths of open crack that was measured on the surface by a crack scale. With the open crack width of 0.20 mm, internal crack begins to develop in the vicinity of the rib on the right side of the deformed bar. With further increase in the open crack width, the corresponding internal crack is greatly enlarged, and at the same time, the crack is connected from the next rib on the same side. The crack also develops around the coarse aggregate near the bar on the rib of the left side of the deformed bar. Moreover, the crack develops around the coarse aggregate near the bar.

It appears that the stress is concentrated on the adjacent ribs. Furthermore, the primary crack width has not
expanded so much, but the secondary crack width has expanded significantly. Indications suggest that the expansion of the secondary crack brought about the expansion of the surface crack width from 1.1 mm to 2.1 mm. The deformed bar near primary crack is separated from surrounding concrete. In this regard, the adhesion of the bar appears to be cut off at this stage.

4.5 Transport through crack in concrete

Through X-ray CT, it is possible to observe the real-time mass transfer inside concrete. Thus, we have developed the visualization technique of the mass transfer in concrete by using microfocus X-ray CT and an X-ray contrast medium such as the cesium carbonate aqueous solution (Darma et al. 2013).

Cylindrical mortar specimen having a diameter of 20 mm and a height of 40 mm was used in this study. Ordinary Portland cement was used with a water to cement ratio of 0.5 and sand to cement ratio of 2.5 by mass. In addition, the compressive strength was 57.2 N/mm². This specimen was repeatedly loaded in compression to generate cracks through 7 cycles between 15.6 to 40.6 N/mm². Then, to promote the permeation of the cesium carbonate aqueous solution into the specimen, it was dried in a drying oven at 105 degrees Celsius. At this time, the process was terminated when the mass at a certain point in time and the amount of mass loss after drying became less than 0.5% of the total mass.

The mass percent concentration of cesium carbonate used was 40%. Table 3 shows the immersion method and mass changes of specimens. Correspondingly, the specimen is immersed in an aqueous solution of cesium carbonate under atmospheric pressure, and then is immersed in the solution under a vacuum environment. The immersion time is shown cumulatively from the time when the immersion is started.

During the CT scanning of the specimen, tube voltage of X-ray is 130 kV and the current is 124 µA. The reconstructed CT image has a resolution is 23µm/pixel while each slice thickness is 40 µm. Figure 29 shows an example of a cross-sectional image of each immersion method.

It shows the cross section of three different layers along the vertical direction. In the cross-sectional image before immersion, cracks have occurred inside (the vertical direction in the figure). There are also entrapped air voids as shown in the CT images. Next, in the image of atmospheric pressure immersion (NP-3a2), it can be seen that cesium carbonate did not immediately penetrate to the inside part but remained at the outer edge. It is presumed that it takes a little more time to penetrate

| Specimen Code Name | Immersion methods | Mass g |
|--------------------|-------------------|--------|
| NP-3              | Before immersion (dry state) | 25.9   |
| NP-3a2            | Atmospheric pressure for 23 hours | 29.7   |
| NP-3a3            | Atmospheric pressure for 30 hours followed by vacuum for 2 hours | 30.0   |
into the cracks, and that the crack width will control the penetration rate. On the other hand, it can be seen from Fig. 29 that the cesium carbonate aqueous solution permeates deep into the cracks and to the air voids connected to the cracks after vacuum immersion.

Figure 30 shows a cross-sectional image of NP-3a3 seen from the longitudinal direction. This is the upper surface and the lower surface when the specimen is installed in the immersion container, coinciding with the top and bottom in the figure. Results show that an aqueous solution of cesium carbonate permeates into the air voids connected to the cracks. In addition, there are other air voids in which the cesium carbonate aqueous solution permeates and some air voids in which the cesium carbonate aqueous solution does not permeate. But in the former case, it is possible that they are connected to cracks that cannot be recognized in this cross section.

When it does not permeate the entire air void, it can be seen that the cesium carbonate aqueous solution is accumulated in the direction of gravity. Furthermore, it can be seen that the cesium carbonate aqueous solution may not immediately permeate the isolated air voids that are not in contact with the cracks even when their distance from the surface layer is near.

With the aid of X-ray CT, indication suggests mass transfer in concrete would not immediately occur into cracks under atmospheric pressure while vacuum condition can cause the penetration into deep internal cracks, and even into the air voids in contact with cracks.

4.6 Alteration of cracked cement paste by flowing water

Non-destructive integrated CT-XRD method was used to investigate the alteration of cracked cement paste due
to flowing water (Sugiyama et al. 2015). Ordinary Portland cement (OPC) paste was used in this study. Hardened OPC paste was made with a water to cement ratio of 0.30. After curing in water, a cylindrical specimen of 5 mm in the diameter and 5 mm in height was split into two parts to simulate crack. Aluminum tape was used to wrap the outer specimen so that each half could stay together having the artificial crack at the center. Then, a plastic tube of 2 mm inner diameter was connected at both ends. Next, leaching test was conducted with demineralized water as shown in Fig. 31.

The flow of demineralized water occurred through the crack space via a tube pump. The flow rate was 50 cc/h and continued for a period of nine months. An increase in the pH of running water and elution of calcium ions were confirmed for this study.

Figure 32 shows the results from the nondestructive integrated CT-XRD method. A total of 68 regions of interest including around the vicinity of the crack were specified on the cross-sectional image. Localized XRD profiles were then obtained. The observed diffraction peak energy was compared with the calculated diffraction peak energy on portlandite and calcite. The calculated diffraction peak energy was obtained using Bragg’s law, specific wavelength, different combination of possible miller indices (hkl) and lattice parameters of these crystals.

Figure 33 shows the regions of rich portlandite and calcite. Portlandite is identified at locations farther from the crack boundary while calcite is present near the crack area. However, less distinct peaks were obtained on some XRD profiles that resulted in the difficulty to identify crystals to describe the alteration of cracked cement paste due to flowing water. This warrants further investigation to improve the analysis with the use of this newly developed method.
5. Conclusion and prospects

The desire to see through what is invisible is the desire of many. Radiation technology is a non-destructive method that allows us to “see through” the internal structure of opaque objects. Advances in this area greatly contribute to the development of concrete engineering. Among the electromagnetic radiations, X-rays are familiar and common such as that of X-ray photography. This device is simple, consisting of a radiation source, a sample table, and a detector. Likewise, X-ray computed tomography (X-ray CT) is becoming more available for nondestructive 3D imaging of concrete and various materials.

The upgrade of X-ray CT equipment is remarkable, and such micro-CT facilities are available in many materials testing and development institutions, as well as in medical clinics in cities. New generation of X-ray CT systems is also recently being developed to provide nanometer-scale resolution (nano-CT). These advances are in parallel with the exponential development in the field of computing and information engineering. The development of image analysis methods necessary for data processing has led to superior tool for 3D imaging, identification and quantification of microstructure. Thus, it will be interesting to see more and more of cement and concrete research using this technology coupled with 3D image-based computational simulations.

We think that the challenges ahead lie on our perspective of actual structures in the built environment. Although we will be constrained by radiation regulations, we can expect that the progress will move toward a non-destructive diagnostic method that can visualize deterioration and damage inside concrete. For that purpose, it is important to understand the potentials and limitation of this radiation technology, and conduct more research to build knowledgebase on its application to concrete durability. Moreover, future studies will incorporate more elaborate testing apparatus and environmental chambers that can be coupled with X-ray CT systems. This will explore new applications of nano-CT and micro-CT as being integrated with other spectroscopic techniques such as X-ray microbeam diffraction map-
ping, among others. Hence, the number of scientific publications on concrete research that actively uses X-ray computed tomography will continue to increase. And it is more surprising to see how such research will expand the frontier of concrete engineering in the coming years.

Acknowledgement
The authors acknowledge SPring-8, a large synchrotron radiation facility, by allowing them to use the world leading apparatus, with the approval of the Japan Synchrotron Radiation Research Institute (JASRI). In addition, the authors express sincere gratitude to their former students and colleagues in the laboratory of Environmental Materials Engineering at Hokkaido University who were involved for their efforts in the related research works.

References
Abraha, K. A., Manahiloh, K. N., Nejad, M. M., (2017). “The effectiveness of global thresholding techniques in segmenting two-phase porous media.” Constr. Build. Mater. 142, 256-267.
Aitcin PC (2000). “Cements of yesterday and today: concrete of tomorrow.” Cement and Concrete Research, 30, 1349-1359.
Bentur, A., (2002). “Cementitious materials-Nine millennia and a new century: Past, present, and future.” Journal of Materials in Civil Engineering, 14, 2-22.
Bossa, N., Chaurand, P., Vicente, J., Borschneck, D., Levard, C., Aguerre-Chariol, O. and Rose, J., (2015). “Micro- and nano-X-ray computed-tomography: a step forward in the characterization of the pore network of a leached cement paste.” Cement and Concrete Research, 67, 138-47.
Brisard, S., Serdar, M. and Monteiro, J. M. P., “Multiscale X-ray tomography of cementitious materials: A review.” Cement and Concrete Research, 128, 2020.
Burger, W. and Burge, M. J., (2010). “Principles of digital image processing.” New York: Springer.
Cormack, A. M., (1963). “Representation of a function by its line integrals, with some radiological applications.” J. Appl Phys, 34, 2722.
Cormack, A. M., (1964). “Representation of a function by its line integrals, with some radiological applications II.” J. Appl Phys, 35, 2908.
Darma, I. S., Sugiyama, T. and Promentilla, M. A. B., (2013). “Application of X-ray CT to study diffusivity in cracked concrete through the observation of tracer transport.” Journal of Advanced Concrete Technology, 11(10), 266-281.
Dong, Y., Su, C., Qiao, P. and Sun L., (2020). “Microstructural crack segmentation of three-dimensional concrete images based on deep convolutional neural networks.” Constr. Build. Mater., 253, 119185.
Goto, Y., (1971). “Cracks formed in concrete around deformed tension bars.” ACI Journal, 68, 244-251.
Henry, M., Hashimoto, K., Darma, I. S. and Sugiyama, T., (2016). “Cracking and chemical composition of cement paste subjected to heating and water recuring.” Journal of Advanced Concrete Technology, 14(4), 134-143.
Hildebrand, T. and Rüegsegger, P., (1997). “A new method for the model-independent assessment of thickness in three-dimensional images.” J Microsc., 185, 67-75.
Hitomi, T., Takeda, N. and Iriya, K., (2007). “A study of CA leaching analysis with concrete pore structure.” Proceedings of the Japan Concrete Institutes, 29(1), 915-920, (in Japanese).
Hoshen, J. and Kopelman, R., (1976). “Percolation and cluster distribution. I. Cluster multiple labeling technique and critical concentration algorithm.” Phys. Rev. B, 14, 34-38.
Hounsfield, G. N., (1973). “Computerized transverse axial scanning (tomography): I. Description of system.” British Journal of Radiology, 46, 1016-1022.
Hounsfield, G. N., (1980). “Computed medical imaging.” Journal De Radiologie, 61, 459-468.
Ikeda, S., Nakano, N. and Nakashima, Y., (2000). “Three-dimensional study on the interconnection and shape of crystals in a graphic granite by X-ray CT and image analysis.” Mineralogical Magazine, 64(5), 945-959.
Landis, E. N. and Keane, D. T., (2010). “X-ray microtomography.” Materials Characterization, 61, 1305-1316.
Latour, L. L., Kleinberg, R. L., Mitra, P. P. and Sotak, C. H., (1995). “Pore-size distributions and tortuosity in heterogeneous porous media.” J. Magn. Res. A, 112, 83-91.
Lindquist, W. D., (1999). “3DMA general users manual.” New York: State of New York at Stony Brook.
Lorenzen, W. E. and Cline, H. E., (1987). “Marching cubes: A high resolution 3D surface construction algorithm.” Comput. Graph., 21(4), 163-169.
Lorenzoni, R., Curosu, I., Paciornik, S., Mechtcherine, V., Oppermann, M. and Silva, F., (2020). “Semantic segmentation of the micro-structure of strain-hardening cement-based composites (SHCC) by applying deep learning on micro-computed tomography scans.” Cem. Concr. Compos., J., 108, 103551.
Mac, M. J., Yio, M. H. N., Wong, H. S. and Buenfeld, N. R., (2021). “Analysis of autogenous shrinkage induced microcracks in concrete from 3D images.” Cement and Concrete Research, 144, 106416.
Monteiro, P. J. M., Geng, G., Marchon, D., Li, J., Alapati, P., Kurits, K. E. and Qomi, M. J. A., (2019). “Advances in characterizing and understanding the microstructure of cementitious materials.” Cement and Concrete Research, 124, 105806.
Nakano, T., Tsujiyama, A., Usugi, K., Usugi, M. and Shinohara, K. (2006). “SLICE—Software for basic 3-D image analysis [online].” Japan Synchrotron
Sen, P. (2004). “Time-dependent diffusion coefficient as a probe of geometry.” Concepts in Magnetic Resonance Part A, 23(1), 1-21.

Sheppard, A. P., Sok, R. M. and Averdunk, H., (2004). “Techniques for image enhancement and segmentation of tomographic images of porous materials.” Physica A, 339(1-2), 45-151.

Stauffer, D. and Aharony, A., (1994). “Introduction to percolation theory.” Revised 2nd ed., London: Taylor and Francis.

Stock, S. R., (2020). “Micro-computed tomography: Methodology and applications.” Boca Raton FL: CRC Press, Taylor & Francis Group.

Sugiyama, T., Promentilla, M. A. B., Hitomi, T. and Takeda, N., (2010). “Application of synchrotron microtomography for pore structure characterization of deteriorated cementitious materials due to leaching.” Cement and Concrete Research, 40(8),1265-1270.

Sugiyama, T., Hitomi, T. and Kajiwara, K., (2014). “Nondestructive integrated CT-XRD method for research on hydrated cement system.” In: J. Olek and J. Weiss Eds, Proceedings of 4th International Conference on the Durability of Concrete Structures, Purdue University West Lafayette Indiana USA, 24-26 July 2014. Purdue Scholarly Publishing Services, 298-303.

Sugiyama, T., Hitomi, T. and Kajiwara, K., (2015). “Non-destructive integrated CT-XRD method developed for hardened cementitious material.” In: Proceedings of 2nd International Conference on Tomography of Materials and Structures (ICTMS 2015), Quebec Canada 29 June-3 July 2015, 560-564.

Takahashi, H. and Sugiyama, T., (2016). “Investigation of alteration in deteriorated mortar due to water attack using non-destructive integrated CT-XRD method.” In: Proceedings of the 11th fib International Ph.D. Symposium in Civil Engineering, Tokyo Japan 29-31 August 2016. Lausanne: fib, 445-451.

Takahashi, H., Shimura, K., Sugiyama, T., and Tanaka, H., (2017). “Observation of cracks around deformed tensile reinforcement by X-ray CT.” In: Proceedings of the Japan Concrete Institute, 39(1), 307-312. (In Japanese).

Takahashi, H. and Sugiyama, T., (2019). “Application of non-destructive integrated CT-XRD method to investigate alteration of cementitious materials subjected to high temperature and pure water.” Construction and Building Materials, 203, 579-588.

Taylor, M., Tam, C. and Gielen, D., “Energy efficiency and CO₂ emissions from the global cement industry.” Energy Efficiency and CO₂ Emission Reduction Potentials and Policies in the Cement Industry, Paris 4-5 September 2006. IEA.

Vincent, L. and Soille, P., (1991). “Watersheds in digital spaces: An efficient algorithm based on immersion simulations.” IEEE Transactions on Pattern Analysis and Machine Intelligence. 13, 583-585.

Worrell, E., Bernstein, L., Roy, J., Price, L. and Harnisch, J., (2009). “Industrial energy efficiency and climate change mitigation.” Energy Efficiency, 2, 109-123.