

1 Investigation of self-sealing in high-strength and ultra-low-permeability concrete
2 in water using micro-focus X-ray CT

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28 strength and ultra-low-permeability concrete

29 **Abstract**

30

31 High-strength and ultra-low-permeability concrete (HSULPC) is thought to be
32 useful as a radioactive waste package. Thus, a high confining ability is desirable.
33 For cementitious materials, sealing of cracks may occur in water due to the
34 precipitation of calcium compounds. This can affect the confining ability. In this
35 study, the sealing of a crack in HSULPC in water was investigated using micro-
36 focus X-ray computed tomography (CT). The sealing by precipitation occurred
37 only around the end of the specimen. Sealed regions of the crack were identified
38 using three-dimensional image registration and CT image subtraction of images
39 obtained for the specimen before and after it was immersed in water to evaluate
40 temporal changes of the sealing deposits in the crack. The sealing deposits
41 increased as the HSULPC specimen was kept in water longer. It was concluded
42 that cracks in HSULPC in water are sealed by precipitation.

43 **1. Introduction**

44

45 For the geological disposal of radioactive wastes, the radioactivity intensity of
46 the radionuclides can be reduced by engineered barriers, such as bentonite buffers,
47 and natural barriers, such as rock mass. If a repository of radioactive waste is
48 located in an area where the hydraulic gradient and the permeability are high, the
49 retardation of radionuclide migration by these barriers may not be sufficient. To
50 retard the migration, several alternative concepts of radioactive waste packages
51 are being developed. High-strength and ultra-low-permeability concrete
52 (HSULPC) is planned for radioactive waste packages for the geological disposal
53 of transuranic (TRU) waste [1–3] to confine the radionuclides with low adsorption
54 by the engineered barriers, such as C-14 included in TRU waste in Japan.

55 Generally, water migrates through networks of cracks and pores in a solid. In
56 cementitious materials, precipitation, mainly of calcium compounds, occurs in
57 water. Thus, the sealing of cracks and pores by precipitation can occur, and this
58 may affect permeability. This phenomenon has been investigated by various
59 researchers [4–12]. Edvardsen [7] showed that sealing of a crack occurs by
60 precipitation of calcium carbonate, generated from CO_3^{2-} in water and Ca^{2+} in
61 cement paste. The crack width and applied water pressure affected the sealing
62 significantly, but the composition of the surrounding water had little effect [7].
63 Yang et al. [11] showed that sealing occurs if the crack width is less than 0.15
64 mm. Reinhardt and Jooss [8] investigated the temperature dependence of the
65 sealing and showed that sealing is enhanced with increasing temperature, up to
66 80°C.

67 Because a high confining ability is required for HSULPC, detailed investigation
68 of the sealing of cracks and pores in water is of considerable importance. It is thus
69 important to observe the sealing of cracks and pores directly. X-ray computed
70 tomography (CT) scanners are an effective tool for observing the sealing of cracks
71 and pores, because we can observe not only the surface but also the interior of the
72 material with this non-destructive technique. Specifically, we can observe the
73 details of the sealing of cracks and pores in HSULPC using micro-focus X-ray
74 CT, because images with high resolution can be obtained. However, to date,
75 observations of this sort have not been reported. Additionally, the temporal
76 behavior of sealing has not yet been clarified.

77 In this study, we investigated the sealing of cracks in HSULPC in water using
78 micro-focus X-ray CT.

79 **2. Sample**

80

81 HSULPC made by Taiheiyo Co., Ltd., was used. Because detailed information
82 about the material properties of HSULPC has been reported previously by Nara et
83 al. [13], we provide only the composition of the HSULPC in Table 1.

84 Fig. 1 shows a photograph of the cracked HSULPC specimen used in this
85 study. An initially intact cylindrical specimen of HSULPC was split in half axially
86 using the Brazil test technique, and was set in an acrylic cylinder tube. The initial
87 crack width was approximately 0.1 mm throughout the specimen. The height of
88 the acrylic cylinder tube was 35 mm. The diameter and height of the HSULPC
89 specimen were approximately 13 mm and 15 mm, respectively.

90 3. Observation by X-ray CT

91

92 3.1. Observation method

93

94 In Japan, salt water is often found underground [14–16]. Thus, considering the
95 condition of groundwater in Japan, the HSULPC specimen was kept in simulated
96 seawater inside a plastic bottle. Table 2 shows the chemical composition of the
97 simulated seawater used. When the specimen was kept in water, a vacuum
98 desiccator was used to ensure that water would fill the entire crack. The amount
99 (weight) of water used was ten times larger than the weight of the HSULPC
100 specimen. To avoid the undesirable dissolution of CO₂ in the water, the air in the
101 plastic bottles was replaced with nitrogen gas. The specimen was kept in a
102 thermostatic chamber at a temperature of 293 K. Because X-rays are more or less
103 attenuated by the presence of water and this causes undesirable error in the image
104 analysis, as discussed below, the HSULPC specimen was dried for a day at room
105 temperature before each X-ray CT observation so as to remove the water from the
106 specimen. The observations were conducted at the start and after keeping the
107 specimen in water for 1, 3, and 7 weeks.

108 For the observation by X-ray CT, a micro-focus X-ray CT scanner,
109 TOSCANER 31300μhd (Toshiba IT & Control Systems Co., Ltd.), installed at
110 Hokkaido University, Japan, was used. The applied tube voltage and maximum
111 tube current were 130 kV and 62 μA, respectively, which were used in each scan
112 in this study. For X-ray CT scanning, cone-beam scanning mode [17] was used in
113 which multiple CT images, up to several hundred cross sections, were provided
114 through one scan, and quick three-dimensional (3D) reconstruction of specimen
115 was possible. The number of pixels in each cross section was 1024 × 1024
116 corresponding to 16 μm × 16 μm for each pixel. The notion of a “voxel” is used in
117 X-ray CT, defined as a pixel having some thickness, named the slice thickness.
118 Here, the slice thickness was 24 μm.

119 During scanning, the observed specimen is placed on a table and the table
120 rotates continuously with the relative positions between the X-ray tube and
121 detector, named an X-ray image intensifier, being fixed. The number of projection
122 directions used in all scans was 1500 (i.e., the interval between each projection
123 angle was $2\pi/1500$ radians). In each projection direction, 20 consecutive scans

were conducted and averaged projection data were used for image reconstruction to reduce statistical noise caused in the X-ray image intensifier. The linear attenuation coefficient [18], μ , for each voxel was computed from the projection data with an image reconstruction algorithm, based on a filtered back-projection method [17, 19] in which a Ramachandran-Lakshminarayanan filter function [20] was used. Then, CT images were reconstructed through the CT values, calculated as follows:

$$V_{ct} = S\mu + B \quad (1)$$

where V_{ct} is the CT value, which is a signed integer taking a value between -8192 and 8191 , and S and B are both constants. In this study, S and B were 200 and 0 , respectively. The CT values in Eq. (1) are not defined in Hounsfield units, as found in many industrial X-ray CT scanners. For noise reduction measures, which are quite important in X-ray CT, a reduction in undesirable effects of scattered X-rays on the X-ray image intensifier was achieved using an X-ray collimator set in front of X-ray tube. Additionally, two notorious artifacts, the cupping effect, due to beam hardening, and the ring artifact [17, 18], were minimized using a copper filter with a thickness of 0.1 mm in a so-called “gain calibration” [18].

Based on the considerations above, 680 CT slice images in total were obtained to reconstruct the entire crack in the HSULPC specimen by X-ray CT scanning and to identify the precipitated regions.

3.2. Results

A typical CT slice image at one cross section of the specimen before it was kept in seawater is shown in Fig. 2. The cross section also corresponds to the top in the image analysis described in Section 4. Based on a window level of 400 and a window width of 1200 , this image is presented in gray-scale with a range of 256 shades from black to white. High CT values, shown in white or brighter colors, correspond to regions with higher density. Regions of low CT values, shown in black or darker colors, correspond to areas of lower density. The black zigzag line found in the central part of the image is the crack, and the dark circular shapes of various sizes are pores. Small white granular regions having higher density are also found over the whole slice and these correspond to metal included in the

158 silica fume as a byproduct of metal refining. The other regions, shown in gray
159 colors, are the matrix of HSULPC, consisting of cement and aggregates.

160 It was observed from the CT images that most of the pores with their diameters
161 approximately greater than ten micrometers were isolated from each other,
162 although some of the pores were on the surface of the specimen. Additionally,
163 some pores were connected to the crack. It may be possible that pores of much
164 smaller sizes ($< 1 \mu\text{m}$) are connected, which are not observable at the resolution of
165 the used CT scanner. Fig. 3 shows CT images of the crack, for approximately the
166 same cross section, extracted from seven CT images of a specimen before it was
167 immersed in seawater and after it was kept in seawater for 7 weeks. These images
168 are presented at intervals of $48 \mu\text{m}$ from one end to the interior of the specimen. In
169 the region close to the end, most parts of the crack were sealed in the HSULPC
170 specimen kept in seawater for 7 weeks. In contrast, less sealing was observed
171 towards the interior of the specimen. Although most parts were sealed in the crack
172 closer to the end, the CT image of the corresponding part showed blurring due to
173 the so-called “partial volume effect” [18], where voxels are affected by including
174 both precipitates and air within them. Blurring is also caused by the tilted surface
175 of the specimen at the ends. From these results, sealing of the crack by
176 precipitation generally occurred only near the end of the specimen.

177 Fig. 4 shows the temporal change in sealing of the crack for one cross section.
178 These cross sections are the same sections as Fig. 2 and those indicated by dotted
179 lines in Fig. 3(a). The occurrence of precipitation was observed after the specimen
180 was kept in water for 1 week. Additionally, the sealed area increased with elapsed
181 time. Because no re-opening of the sealed crack was found, dissolution of the
182 precipitates did not occur over the period of observation. No swelling of the
183 specimen was found during the period of observation.

184 The following observations were clarified by the X-ray CT. The occurrence of
185 significant precipitation was found from the end to the interior of the specimen
186 within a range of approximately 0.2 mm, and the total amount of precipitate
187 increased with elapsed time in the region closer to the end. Most parts of the crack
188 were sealed after 7 weeks due to precipitation near the end. Once such sealing was
189 achieved, no further precipitation was observed in the crack below the sealed part.

190 A photograph of the end of the specimen kept in seawater is shown in Fig. 5.
191 Precipitation occurred in such a way that the precipitates covered over the end.

192 Based on these results, image analyses were conducted to evaluate the sealing
193 process only near the end of the specimen, where significant precipitation was
194 observed in the crack.

195 4. Image analysis

197 4.1 Image processing method

198
199 To investigate temporal changes in the sealed regions of the crack, it was
200 necessary to extract the sealed regions by image processing. For the cross sections
201 where sealing was observed, we sought to extract precipitates by applying an
202 image subtraction technique between the CT images before and after the specimen
203 was kept in water. However, due to the need to set the specimen on the table in
204 the CT scanner manually before each scan, an alignment gap between the
205 comparison images occurred. Thus, before conducting the image subtraction,
206 image registration was used to minimize the alignment gap. For this purpose, an
207 affine linear transformation, expressed by the following equation, was used:

$$208 \quad u_i = \alpha_{i1} + \alpha_{i2} \cdot x + \alpha_{i3} \cdot y + \alpha_{i4} \cdot z \quad (i = 1, 2, 3) \quad (2)$$

209 where u_i indicates the alignment gap of corresponding voxels between comparison
210 images in three directions, defined in Cartesian coordinates, and (x, y, z) indicate
211 the integer coordinates of each voxel in the reference CT images. CT images
212 before the specimen was kept in water were used as the reference images, and CT
213 images after the specimen was kept in water were mapped onto the reference
214 integer coordinates. In each direction, four unknown coefficients, α_{ij} , need to be
215 determined.

216 Here, we explain how to determine α_{ij} . In Fig. 6, a 3D image of the upper half
217 of a HSULPC specimen kept in seawater is shown. Distinctly white granular
218 regions are included in the image. We used these granular regions to determine α_{ij} .
219 The region of interest (ROI) was set as a rectangular parallelepiped. The thickness
220 corresponding to 30 slices from the end to the interior of the specimen was
221 included. The matrix size of the ROI was 342×342 in the center of each slice. In
222 the ROI of reference images, 11 sampling points were selected where the white
223 granular regions took their local maximum values. Then, approximate values of u_i
224 in Eq. (2) for the 11 points were obtained by a least squares method. Then, to
225 obtain more precise values of α_{ij} , the volumetric cross-correlation, C , defined in
226 the following equation was introduced:

$$227 \quad C = \iiint_{\text{ROI}} f_0(x, y, z) \cdot f_R(x + u_1, y + u_2, z + u_3) dx dy dz \quad (3)$$

228 where f_0 is the spatial distribution of CT values in the reference images and f_R is
229 that of comparison images, transformed by u_i of the given α_{ij} . We considered that
230 α_{ij} became optimal when C took the maximum value in the ROI. Because the
231 nearest cross section to the end resulted in blurred CT images (see Fig. 3) and they
232 might cause significant error in the image analysis, blurred CT images were
233 excluded from the top of the ROI. Consequently, the top cross sections of the ROI
234 were those surrounded by dashed lines in Fig. 3(a).

235

236

237 4.2 Results

238 The results of the image subtraction (i.e., subtraction images) are shown in Fig.
239 7. The reference images correspond to those obtained for the initial specimen
240 before immersion in seawater. Comparison images were obtained after 1, 3, or 7
241 weeks in seawater. In the figure, only the cracks in the ROI are presented at an
242 interval of 48 μm from the top to the interior of the ROI. These images are
243 displayed in gray-scale, with a range of 256 shades from black to white, for a
244 window level and width of 0 and 1000, respectively. In regions where no change
245 occurred, the difference between the CT values was approximately zero, displayed
246 as a gray color in the subtraction images. In regions where precipitation occurred,
247 the difference between the CT values was positive, displayed as a white color.
248 Fig. 7 shows that the region of precipitation observed in Fig. 3 was successfully
249 extracted from the crack.

250 In the subtraction images, reasonable segmentation between precipitated and
251 non-precipitated regions was required. If a voxel includes both phases (i.e.,
252 precipitated and non-precipitated regions), the CT value, and, accordingly,
253 differences in CT values, of this voxel takes an intermediate CT value between
254 these two phases. Such a voxel is called a “mixel” [21-24]. Based on this, a
255 maximum likelihood thresholding method considering the effect of mixels [21–
256 23] was used to set an appropriate threshold, t . The threshold was determined
257 from the histogram obtained in the boxed area A_t in Fig. 8(a). Fig. 8(a) shows the
258 same cross section surrounded by dashed lines in Fig. 7(c). The histogram of the
259 probability density function is shown in Fig. 8(b). The threshold determined from
260 the maximum likelihood thresholding method was $t = 182$. Binarized images
261 obtained from the subtraction images in Fig. 7 using this threshold are shown in

262 Fig. 9. The images show the cross sections from the top to the interior of the ROI
263 at an interval of 48 μm , and demonstrate that the geometry of the precipitate was
264 successfully segmented using the threshold.

5. Discussion

The X-ray CT observation results in Section 3 showed that sealing of the crack by precipitation occurred only near the end of the crack in the specimen. In this region, most parts were sealed. This suggests that the crack near the ends was in an environment where precipitation of calcium compounds, such as calcium carbonate, was enhanced by the calcium ions dissolved from the HSULPC and both the carbonate and bicarbonate ions found in the seawater.

If the crack is completely sealed near the end, the network of cracks inside the HSULPC can be isolated from the surrounding environment, which can cause a decrease in permeability [25] and retard degradation in terms of the confining ability of the HSULPC. Because the CT observations suggest that the sealed regions spread with elapsed time, an investigation of the temporal change of the sealing deposits could provide important information to evaluate the effect of the precipitates on limiting water flow into HSULPC. Thus, we investigated the temporal change of the sealing deposits in the crack.

The 30 binarized images obtained in the previous section were used to evaluate the sealing. Fig. 10 shows a schematic illustration of precipitation in the crack. The percentage of sealing deposits in the crack, P_{seal} , in each slice, having slice thickness T_{slice} , was calculated. P_{seal} was calculated as follows:

$$P_{\text{seal}} = \frac{T_{\text{slice}} S_{\text{pre}}}{T_{\text{slice}} S_{\text{crack}}} \times 100 = \frac{l^2 N_{\text{pre}}}{l^2 N_{\text{crack}}} \times 100 = \frac{N_{\text{pre}}}{N_{\text{crack}}} \times 100(\%) \quad (4)$$

where T_{slice} is the slice thickness, S_{pre} is the total area of precipitates, S_{crack} is the total area of the crack, l is the pixel size (16 μm), and N_{pre} and N_{crack} are the numbers of pixels for precipitates and the crack, respectively, in ROI₁ in Fig. 9. N_{pre} in ROI₁ was obtained from the extracted precipitates in the binarized images. N_{crack} was obtained by counting the number of pixels corresponding to the crack in ROI₁ by applying the maximum likelihood thresholding method to the reference CT images before the specimen was kept in water, where the regions of the crack and matrix of the HSULPC were segmented. Because the mean value of the crack width in ROI₁ was approximately constant, the temporal change in P_{seal} , denoted as V_{seal} , was computed using the following equation:

$$V_{\text{seal}} = \frac{\Delta P_{\text{seal}}}{\Delta t} (\% \cdot \text{day}^{-1}) \quad (5)$$

297 where ΔP_{seal} is the change in P_{seal} over the given immersion period, Δt .

298 P_{seal} in ROI₁ is shown in Fig. 11 with respect to the depth from the top to the
299 interior of ROI₁. From this figure, P_{seal} became larger towards the end in each
300 period, and P_{seal} within a depth of 0.05 mm also increased with elapsed time.

301 Similarly, V_{seal} in ROI₁ is shown in Fig. 12 with respect to the depth from the
302 top to the interior of ROI₁. From this figure, V_{seal} towards the end became larger
303 than in the inner region. The maximum V_{seal} was found at 3 weeks, after which
304 V_{seal} started to decrease. This suggests that little sealing could be expected to
305 occur after 7 weeks in ROI₁.

306 Based on the investigations described above, when HSULPC is kept in water,
307 precipitation, such as of calcium compounds, occurs in the crack near the end, and
308 the sealed regions or sealing deposits increase with elapsed time. Considering the
309 application of HSULPC, it is desirable that such sealing should occur over a short
310 period time. Thus, identifying optimum water conditions and crack widths is of
311 considerable importance. To achieve this, more detailed investigation regarding
312 the influence of water conditions on sealing and the change in water content due
313 to precipitation could also be important. Additionally, considering that the
314 temperature in the surrounding environment changes and that heat release occurs
315 due to the exothermic reactions of radioactive waste, the temperature-dependency
316 of the sealing behavior also needs to be further investigated. However, these
317 investigations are beyond the scope of the present study, and we regard them as
318 future work. The results reported in this paper could be important for engineering
319 projects such as the geological disposal of radioactive wastes, and further
320 accumulation of relevant information is important.

321 **5. Conclusions**

322

323 Sealing of crack in HSULPC was investigated in this study. A cracked
324 HSULPC specimen was prepared and kept in simulated seawater for up to
325 7 weeks. The surface and interior of the cracked HSULPC specimen were then
326 observed using micro-focus X-ray CT.

327 The results revealed that sealing of the crack occurred only near the end of the
328 specimen. The occurrence of significant precipitation was found within
329 approximately 0.05 mm from the end.

330 Temporal changes in sealing in the crack showed that the sealing deposits
331 increased with the time the specimen was kept in water. Additionally, the sealing
332 deposits increased towards the end of the HSULPC specimen.

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338 **References**

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417 **Figure legends**

418

419 Fig. 1. Photograph of a cracked HSULPC specimen. The height and diameter of
420 the HSULPC were 15 mm and 13 mm, respectively. The crack width was
421 0.1 mm. (a): General view, (b): top view.

422 Fig. 2. X-ray CT sliced image of a cracked HSULPC specimen before it was kept
423 in seawater.

424 Fig. 3. Comparison of corresponding CT images of the crack: (a): before
425 immersion in seawater, (b): after it was kept in seawater for 7 weeks. The
426 height and width of each image are 11 mm and 1 mm, respectively.

427 Fig. 4. Comparison of CT images for a particular section. The height and width of
428 each image are 11 mm and 1 mm, respectively.

429 Fig. 5. Photograph of precipitation that occurred over the end of the HSULPC
430 specimen when it was kept in seawater. The regions with the white
431 coloration correspond to precipitates.

432 Fig. 6. 3D CT image of the upper half of the HSULPC specimen used in the
433 image analysis, where the ROI used in the image registration given by
434 Eqs. (2) and (3) is indicated by the rectangular parallelepiped.

435 Fig. 7. Subtraction between the initial reference image and ones obtained after the
436 specimen was kept in seawater for (a) 1, (b) 3, or (c) 7 weeks. The height
437 and width of each subtraction image are 3.04 mm and 0.64 mm,
438 respectively.

439 Fig. 8. Subtraction image and histogram for probability density of CT values used
440 to determine a threshold. (a) Subtraction image. Region A_t was used to
441 determine the threshold segmenting the precipitated and non-precipitated
442 parts. (b) Histogram obtained from A_t , where the threshold was
443 determined from the maximum likelihood thresholding method
444 considering the effect of mixels.

445 Fig. 9. Binarized images obtained for the differences between reference images
446 and images after the specimen was kept in seawater for (a) 1, (b) 3, and
447 (c) 7 weeks. The height and width of each subtraction image are 3.04 mm
448 and 0.64 mm, respectively. The crack width in ROI_1 was 0.06 mm.

449 Fig. 10. Schematic diagram of precipitation on the surface of the crack in each
450 slice.

451 Fig. 11. Relationship between the percentage and position of the sealing deposits
452 in specimen kept in seawater for 1, 3, and 7 weeks.
453 Fig. 12. Temporal change of the percentage of the sealing deposits for specimen
454 kept in seawater for 1, 3, and 7 weeks.

455 **Table legends**

456

457 Table 1. Composition of HSULPC (after Nara et al. [13]).

458 Table 2. Chemical composition of simulated seawater (mol/L).

459 **Tables**

460

461 Table 1. Composition of HSULPC (after Nara et al. [13]).

462

Amount [kg/m ³]	
Low-heat Portland cement	744 – 1014
Silica fume	158 – 496
Fillers (fly ash, blast furnace slag, etc.)	225 – 541
Aggregates	631 – 947
Water-reducing admixture	24
Water	180

463

464

465

466 Table 2. Chemical composition of simulated seawater (mol/L).

467

Ca ²⁺	10×10 ⁻³
SO ₃ ²⁻	29×10 ⁻³
Na ⁺	45×10 ⁻²
K ⁺	19×10 ⁻³
Cl ⁻	56×10 ⁻²
Mg ²⁺	55×10 ⁻³
HCO ₃ ⁻	24×10 ⁻⁴

468

469

470

471

472

473

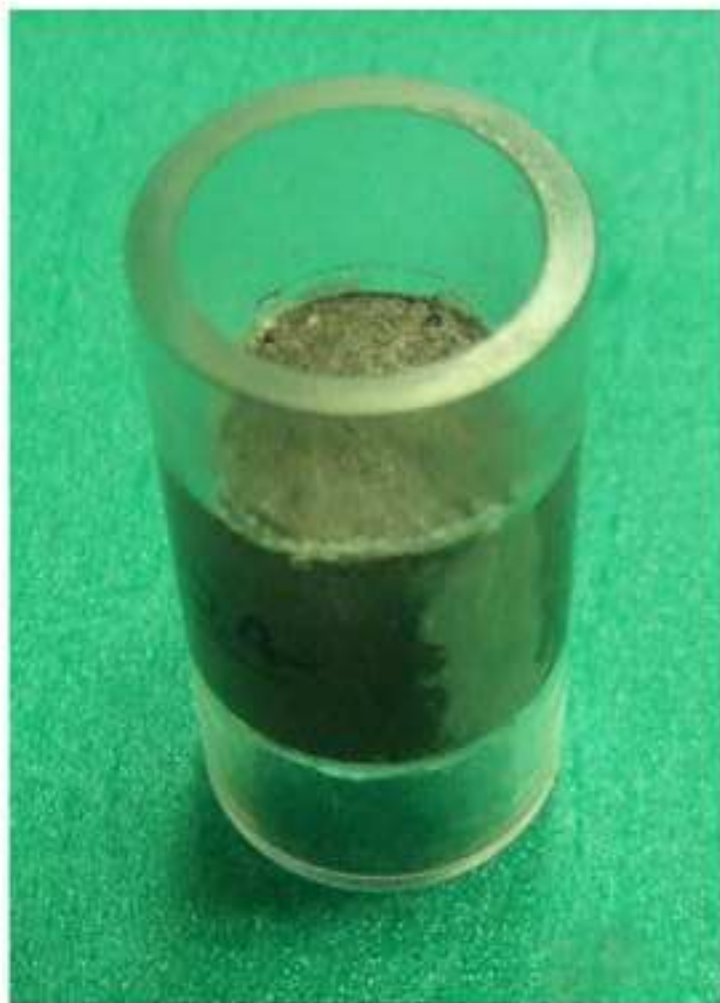
474

475

476

Figure1

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(a)



(b)

Figure2
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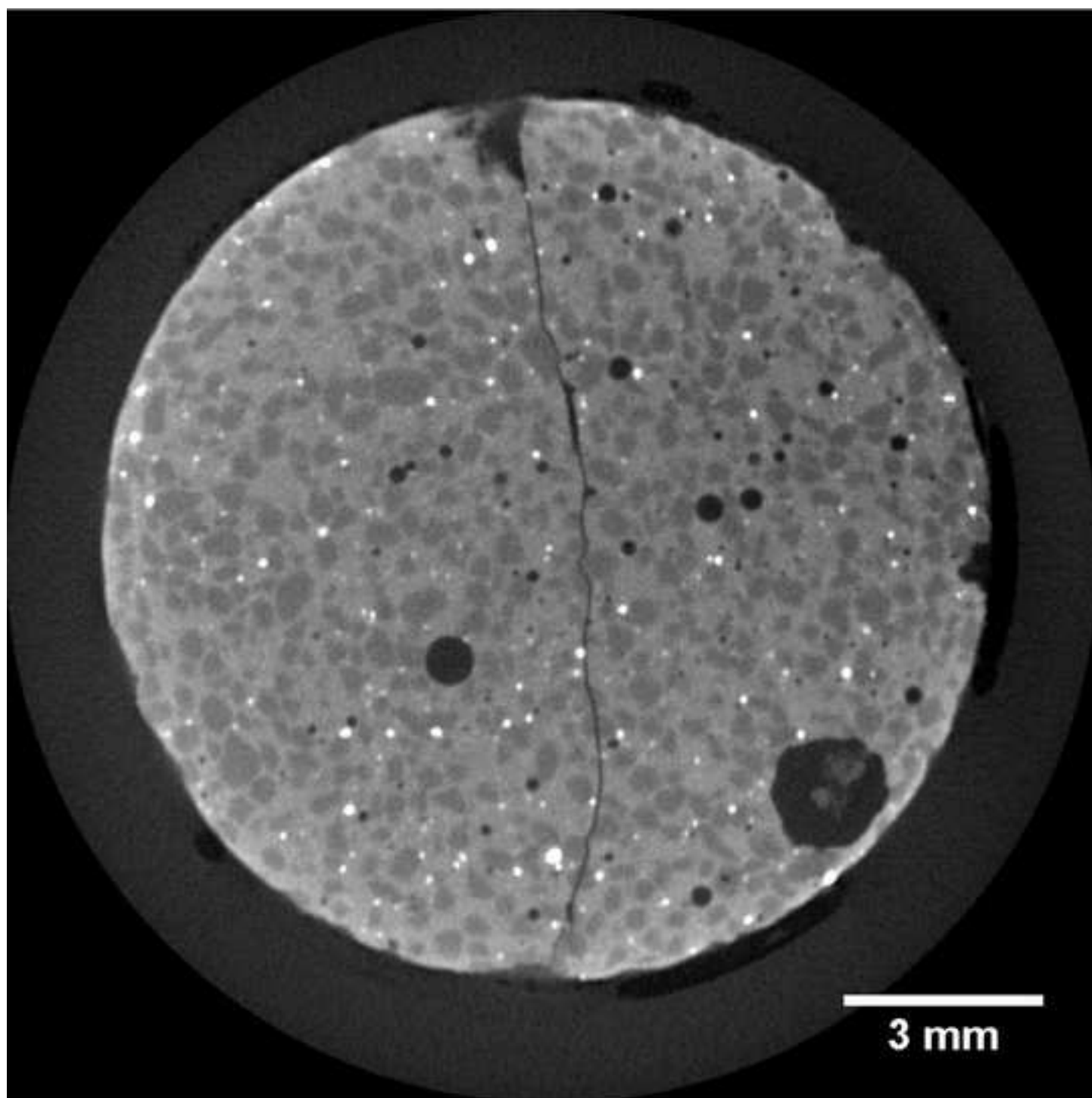


Figure3a
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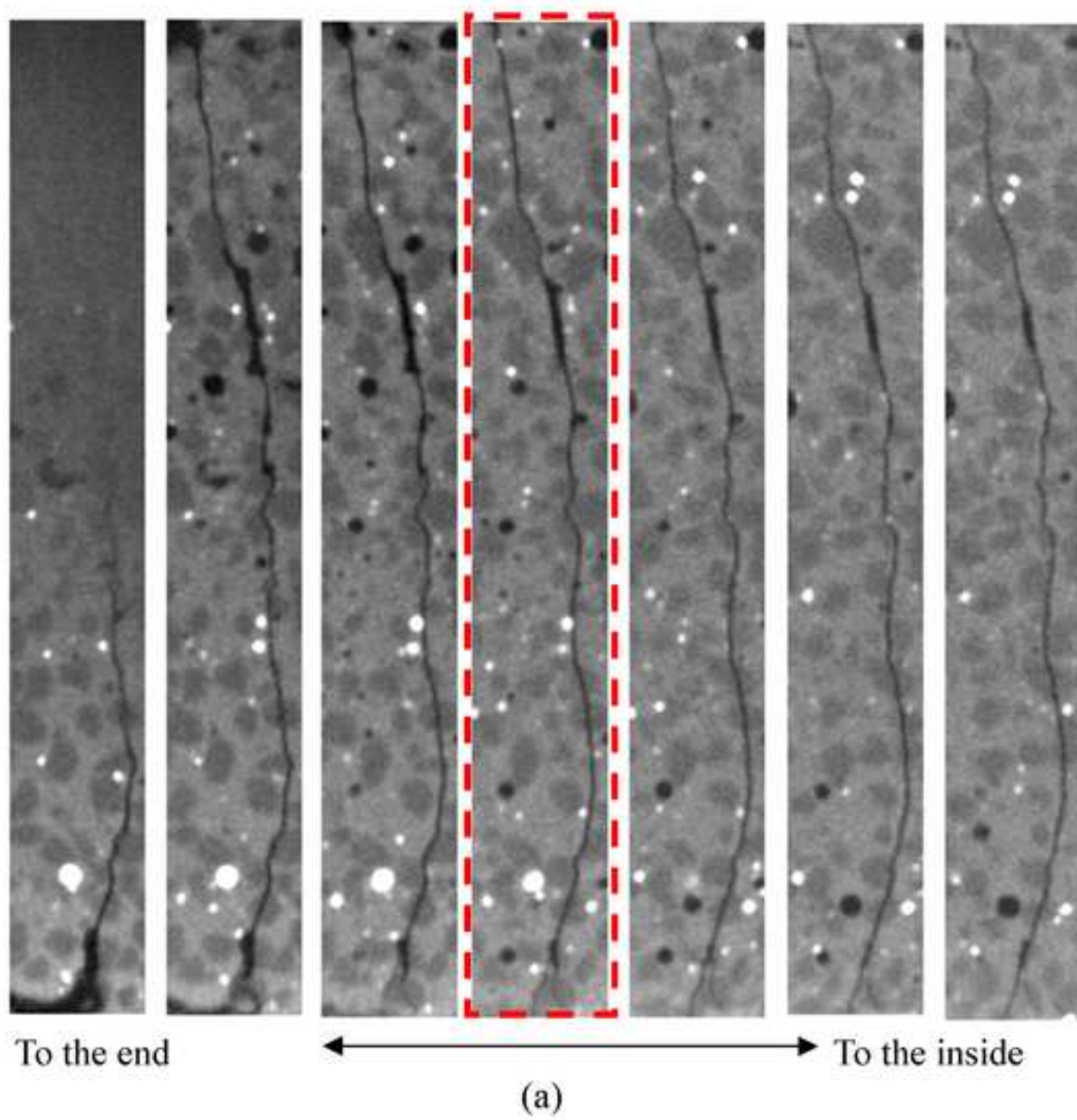


Figure3b
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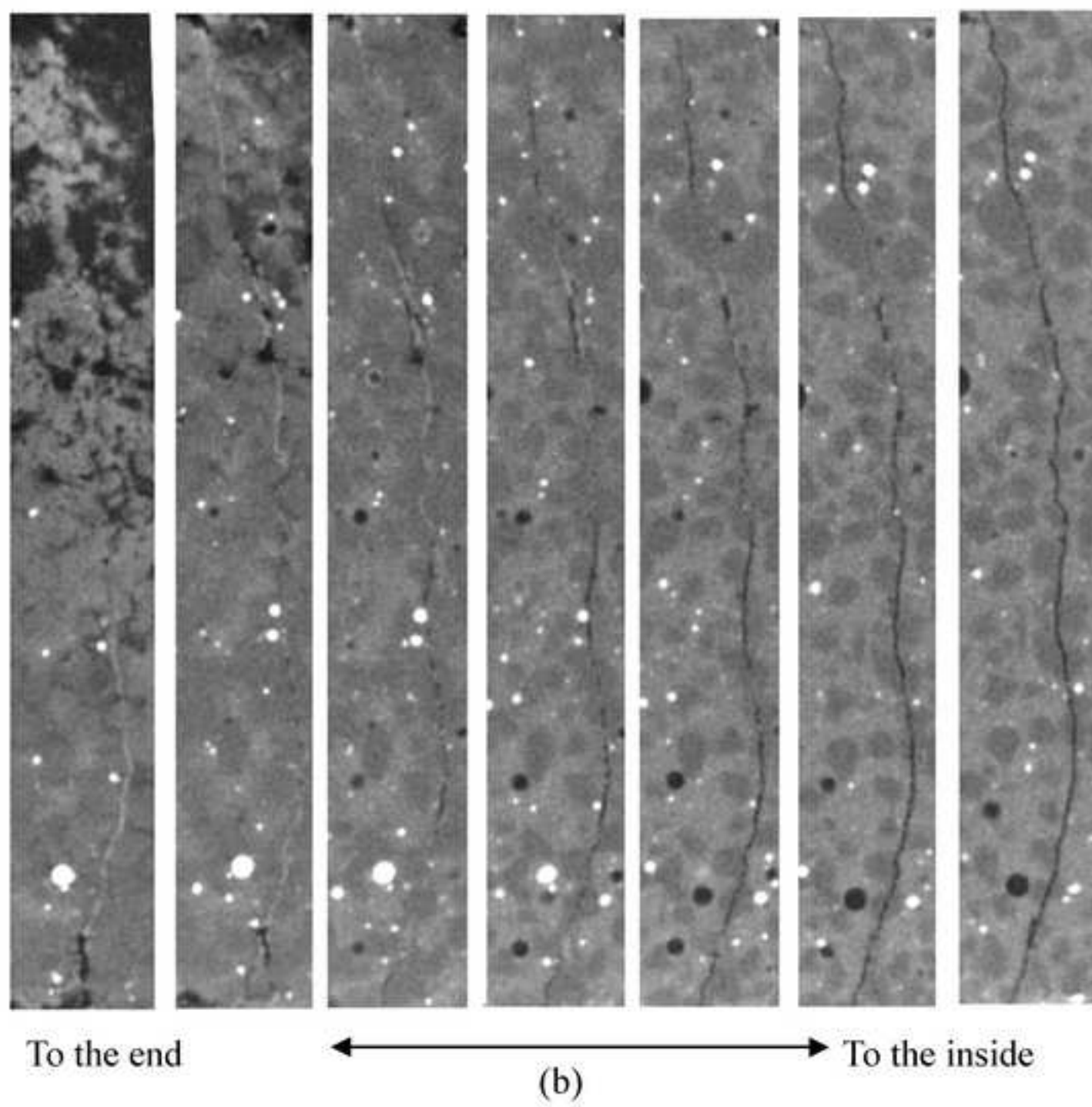
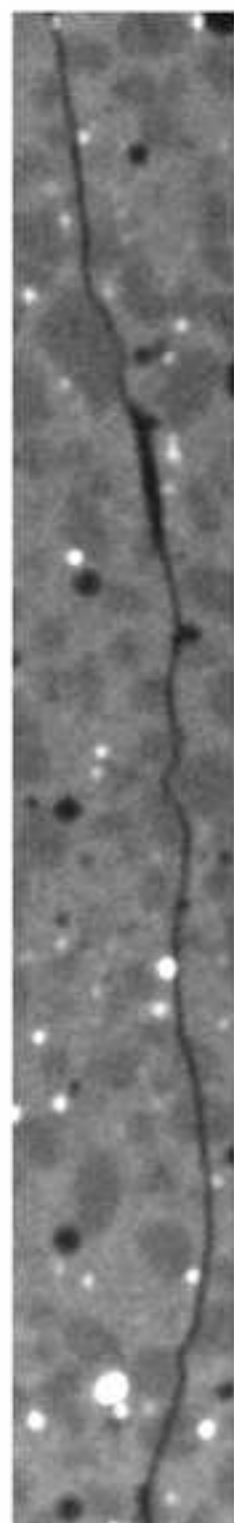
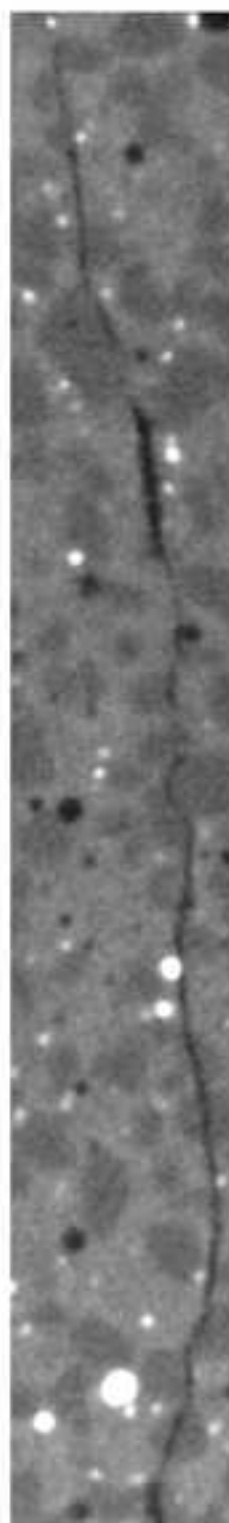


Figure4
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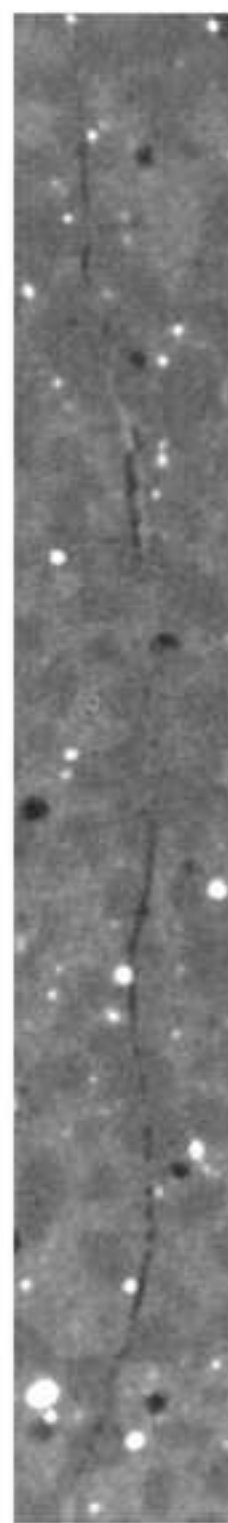
Before
immersion



1 week



3 weeks



7 weeks

Figure5
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Figure6
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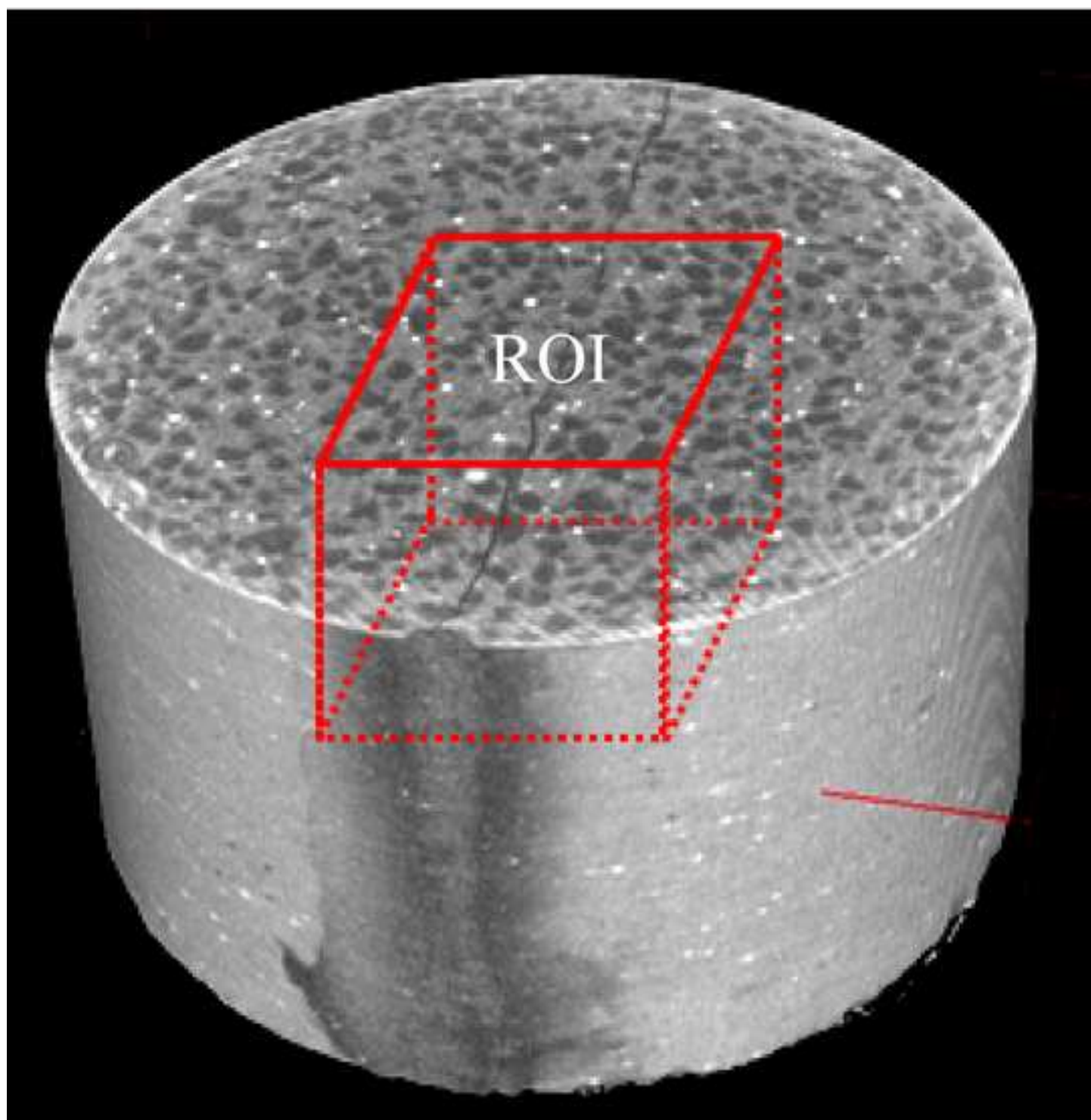


Figure7
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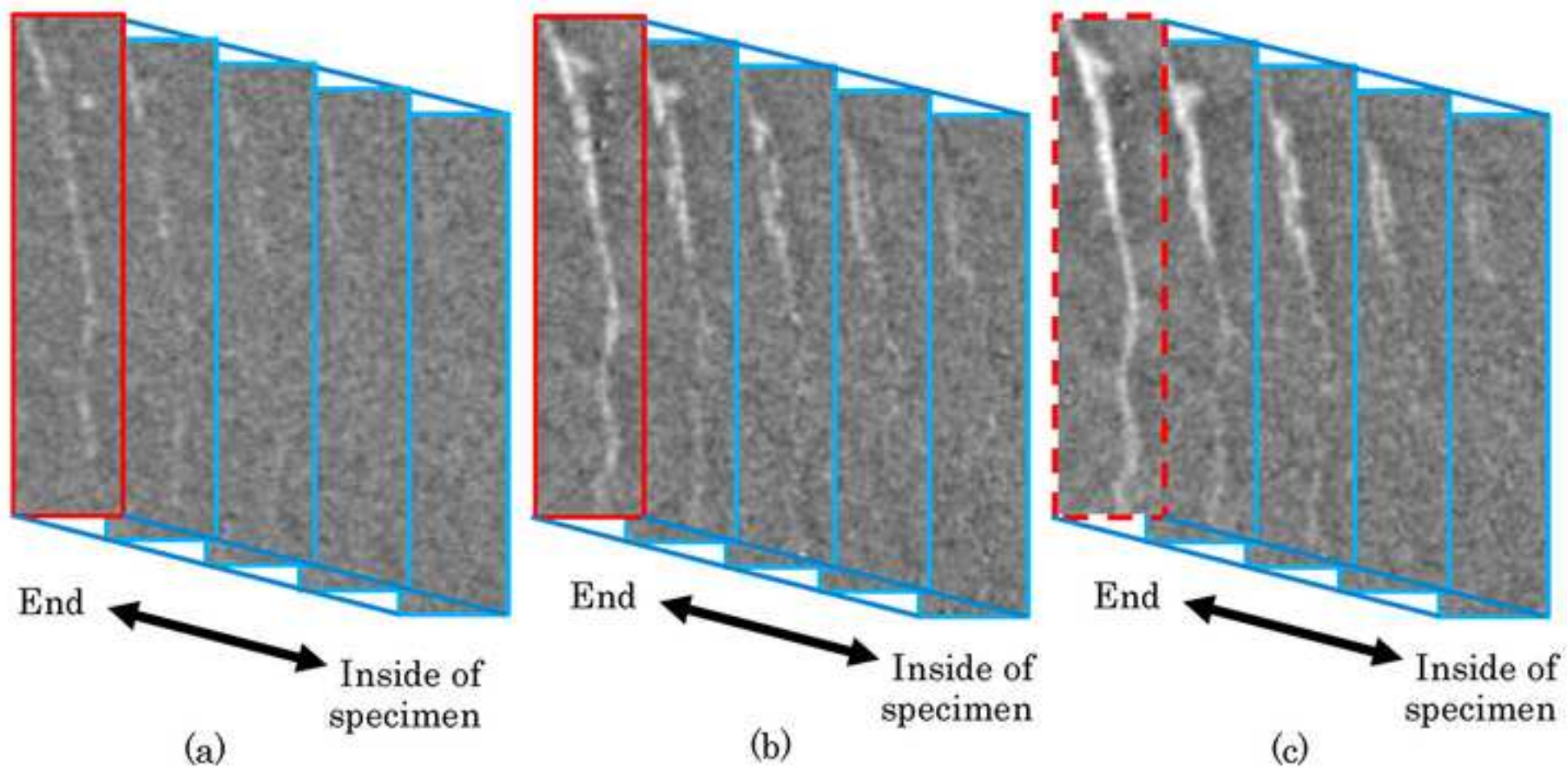
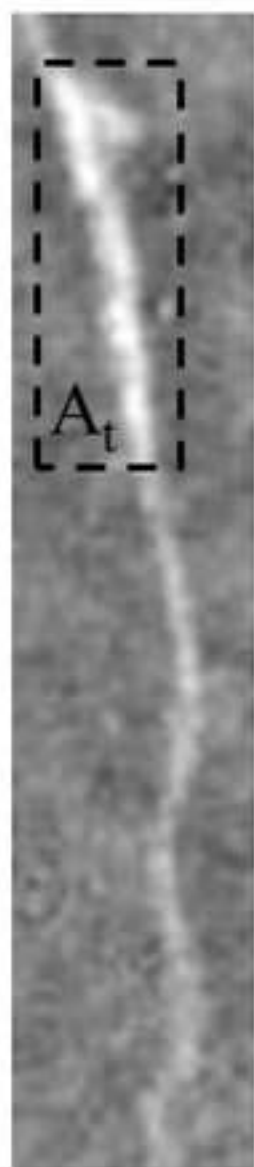
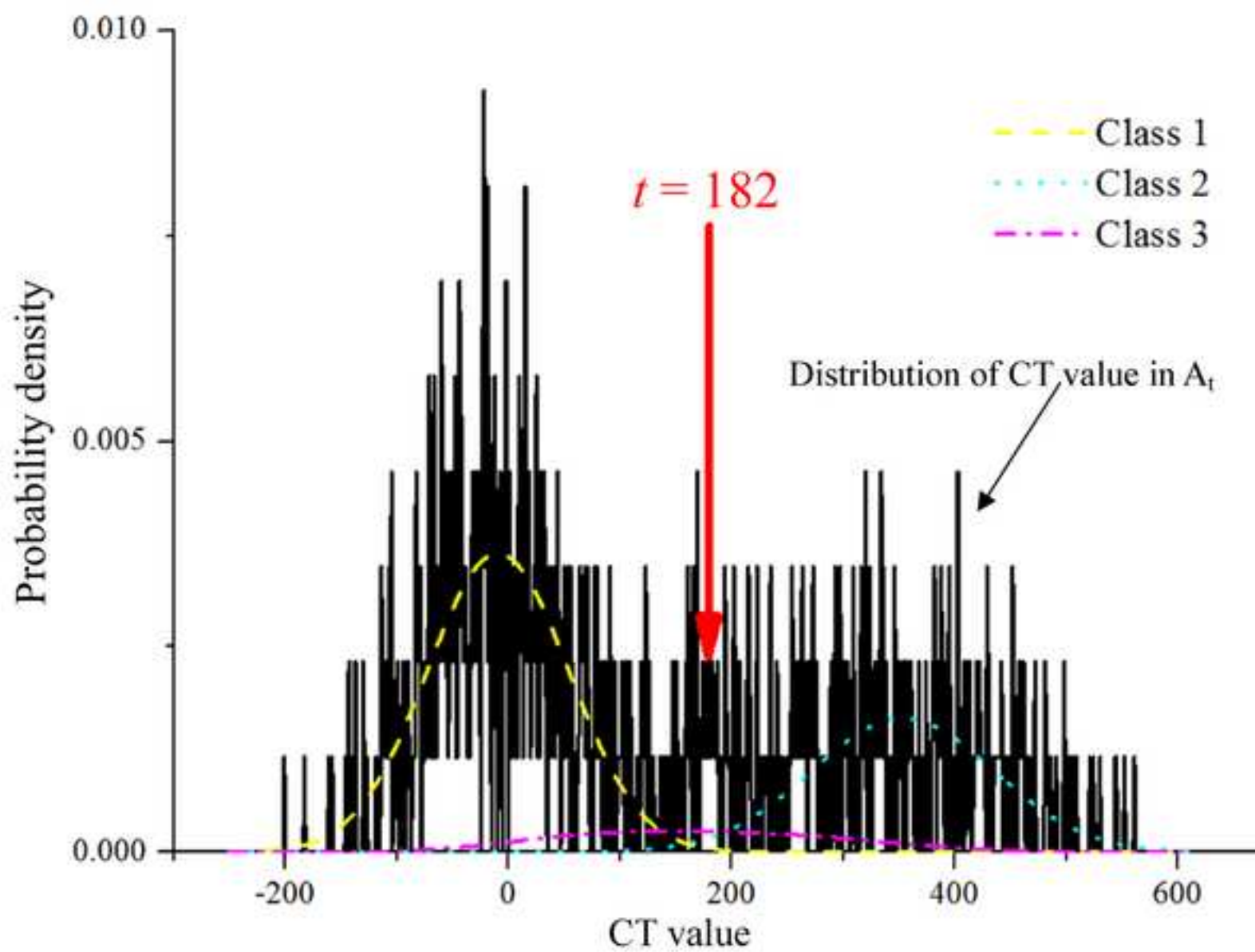


Figure8
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(a)



(b)

Figure9
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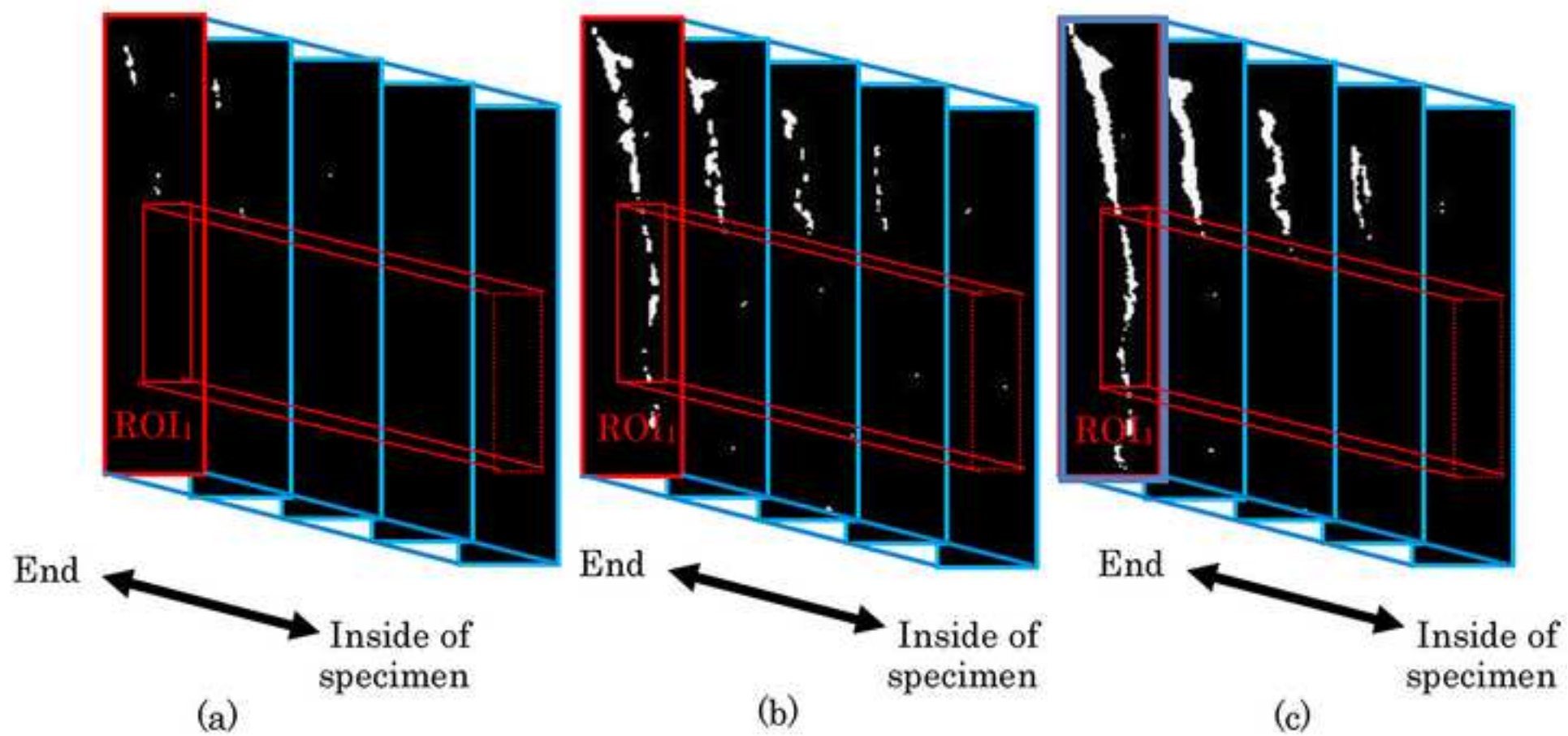


Figure10
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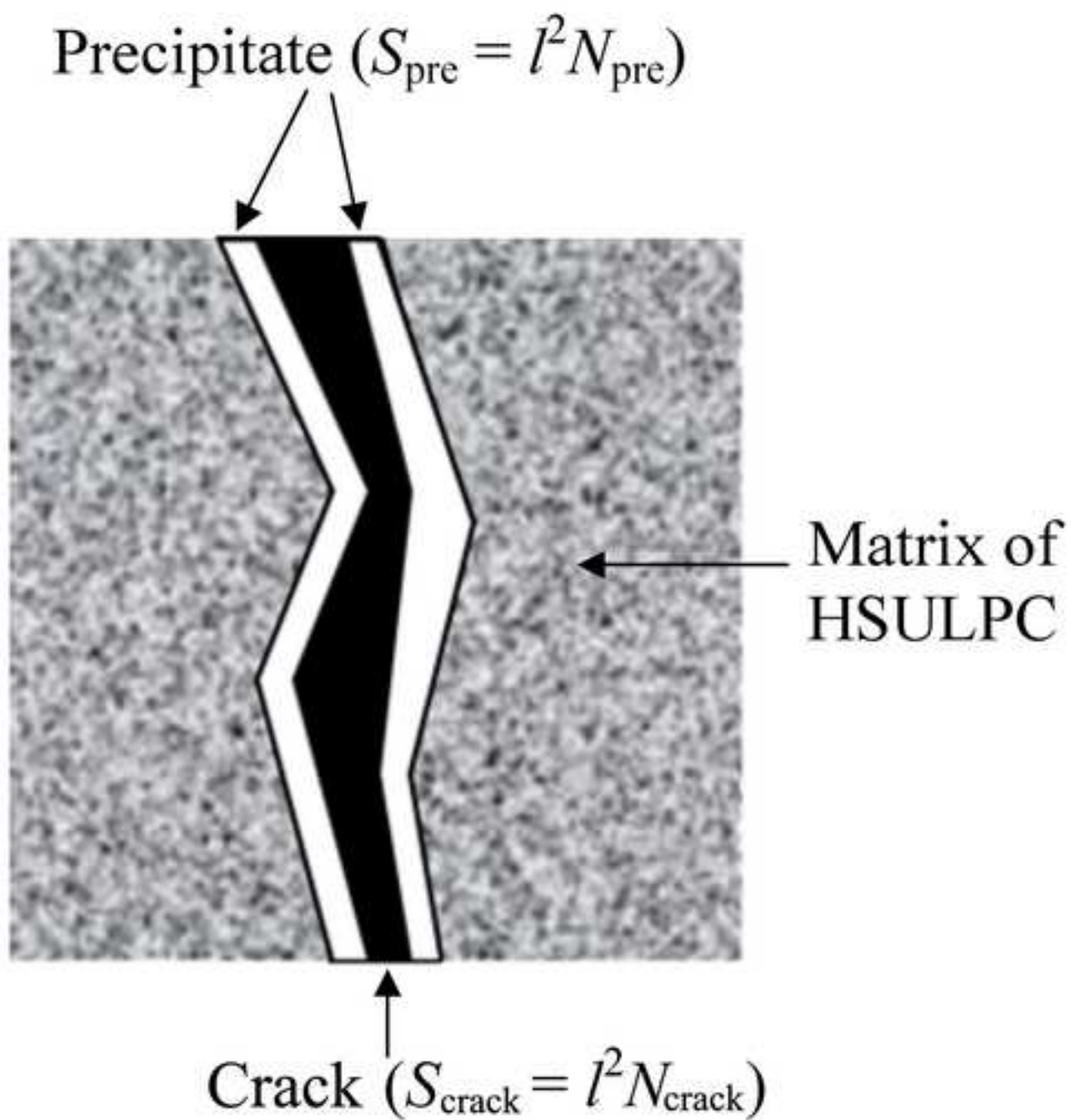


Figure11

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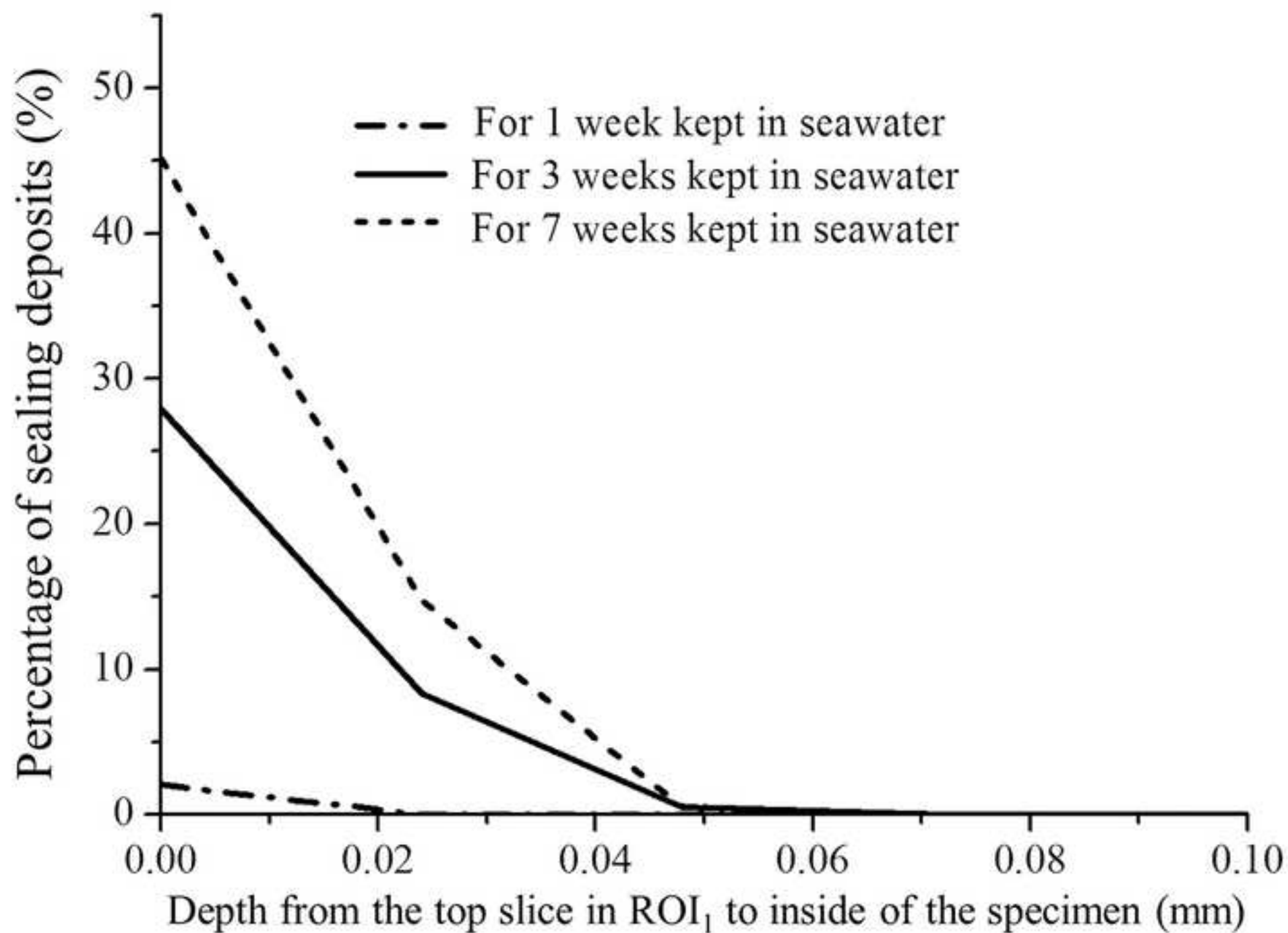


Figure12

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