

## Twinning in synthetic anorthite: A transmission electron microscopy investigation

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### ABSTRACT

Transmission electron microscopy (TEM) has revealed dense, complex twins in synthetic anorthite crystals formed by annealing  $\text{CaAl}_2\text{Si}_2\text{O}_8$  glass. The crystals are dominated by Carlsbad and Carlsbad-albite twins, with small amounts of albite twins. All the composition planes are parallel to (010). High-resolution TEM images show the twin boundaries to be coherent. Selected-area electron diffraction (SAED) patterns containing the  $b^*-c^*$  reciprocal plane are useful for distinguishing these twins. Extremely dense Carlsbad twins at the unit-cell scale indicate that additional energy caused by the Carlsbad twin boundaries in highly disordered anorthite is lower than that caused by albite twin boundaries. We propose that for anorthite with an initially disordered structure the total energy induced by Carlsbad twinning is lower than that caused by albite twinning and higher than that caused by albite twinning in anorthite with an initially ordered structure.

### INTRODUCTION

Twinning is common in feldspars. Twins can form during crystallization, cooling, deformation, or combinations thereof (Buerger 1945). There are many types of twins in feldspars and their details vary. Here, we consider Carlsbad, Carlsbad-albite, and albite twins that formed during crystallization.

Carlsbad and Carlsbad-albite twins are abundant in plagioclase from volcanic and high-temperature plutonic rocks and rare in metamorphic rocks, whereas albite twins commonly occur in triclinic feldspar (Gorai 1951; Wenk 1973, 1977, 1983). Carlsbad and Carlsbad-albite twins have been called complex twins or C twins by Gorai (1951), who suggested that the frequency of the C twins in calcic plagioclase may indicate the crystallization temperature of the feldspar crystals and their initial structural states (Al-Si order). Theoretical studies have shown that (010) is a reasonable composition plane for both Carlsbad and Carlsbad-albite twins, and that the composition plane does not produce serious lattice misfit between neighboring twin lamellae (Dowty 1980; Wooster 1981).

Figure 1 schematically illustrates these twins in anorthite. Twin lamellae 1 and 1' in Figure 1A are related by an albite twin law, that is, a (010) mirror operation. Twin lamellae 1 and 2 (or 1' and 2') are related by a Carlsbad twin law, that is, a [001] twin axis. Lamellae 1' and 2 (or 1 and 2') are related by a Carlsbad-albite twin law, even though they are not in contact with each other (Fig. 1A); they are related by both a (010) twin plane and a [001] twin axis (or, a twin axis normal to [001] and in the (010) plane). The Carlsbad-albite twin law results from a combination of Carlsbad and albite twin laws and is therefore called a compound twin (Luo 1985; Smith and Brown 1988).

Although there are numerous transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM) results for albite and pericline twins (Brown and Macaudière 1986; Brown and Parson

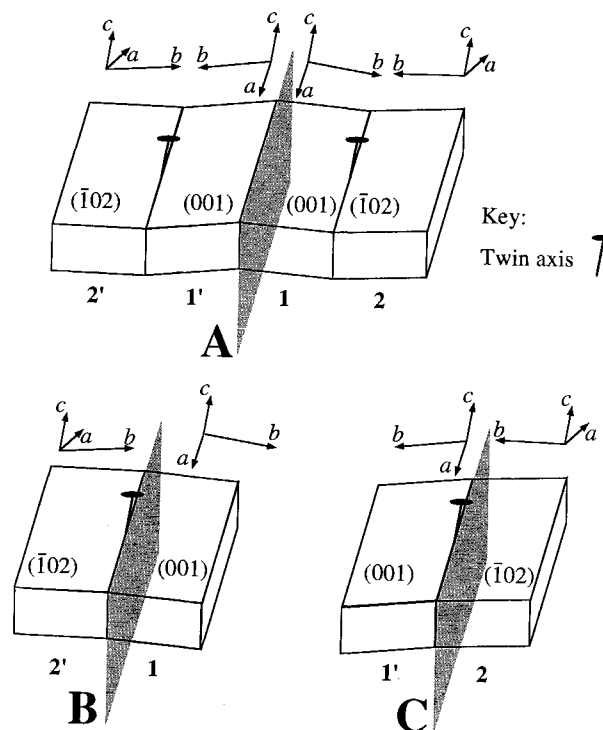
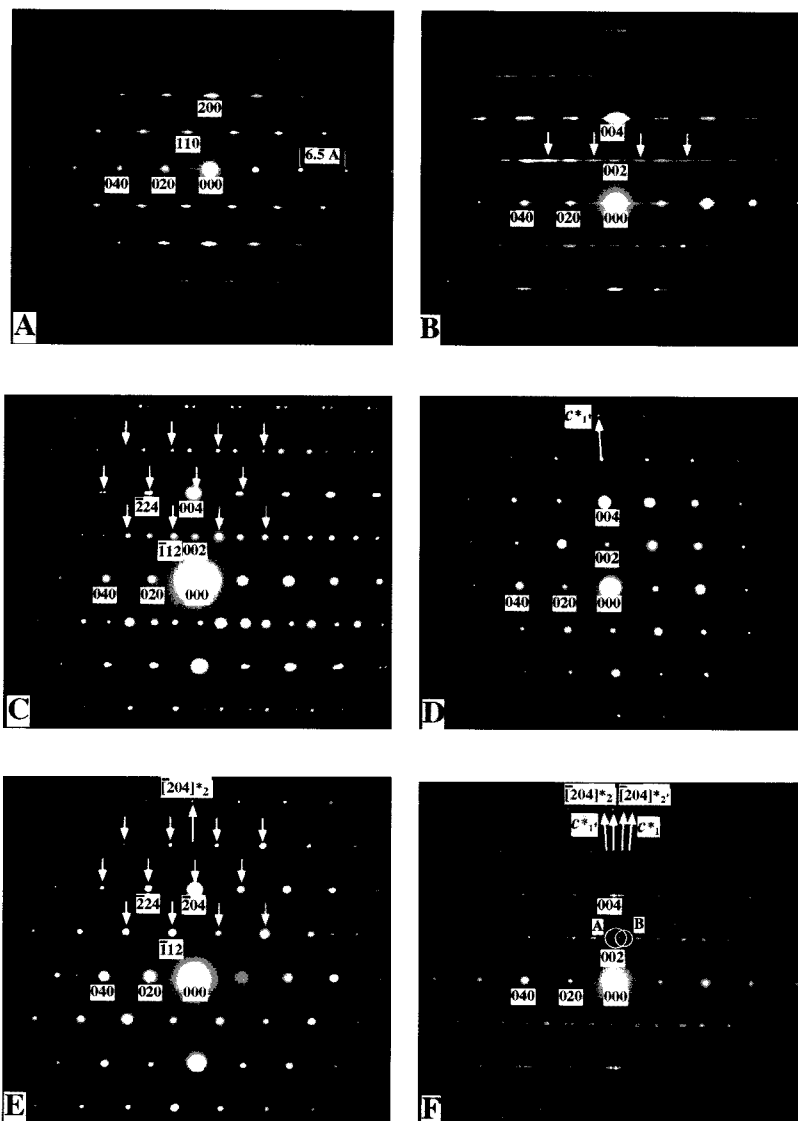


FIGURE 1. Diagram schematically illustrating albite (A), Carlsbad (A), and Carlsbad-albite (A–C) twin laws in anorthite. The gray (010) twin plane shows the albite twin mirror operation. See text for details.



**FIGURE 2.** SAED patterns of synthetic anorthite annealed at 1400 °C for 1 min. (A) A [001] orientation showing strong streaking along the  $b^*$  direction. (B) Overlapping [100] and [201] orientations showing Carlsbad and Carlsbad-albite twinning, with strong streaking along the  $b^*$  direction. White arrows indicate reflection maxima from the [201] orientation. (C) Overlapping [100] and [201] orientations from coarse anorthite lamellae in a Carlsbad-albite twinning relationship. SAED patterns of

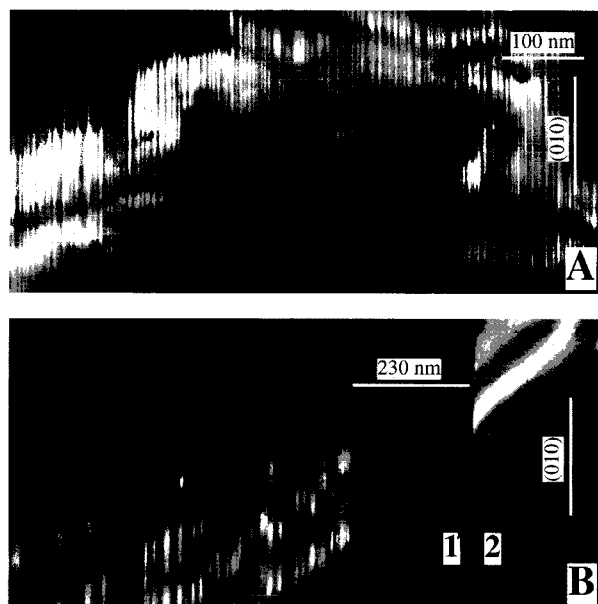
lamellae in [100] and [201] orientations are shown in D and E, respectively. (D) Diffuse “ $b$ ” and “ $c$ ” reflections. (E) Sharp “ $a$ ” reflections. Arrows in C and E indicate the same reflection spots for the [201] orientation. (F) SAED pattern of [100] and [201] orientations from the anorthite lamellae in albite, Carlsbad, and Carlsbad-albite twins. Two white circles (A and B) indicate objective apertures used for producing the DF images in Figures 4A and 3B, respectively.

1984; Brown et al. 1982; Eggleton and Buseck 1980; Fitz Gerald and McLaren 1982; McLaren 1984; Ried and Korekawa 1978; Xu et al. 1995a, 1995b), there are no reported HRTEM studies of Carlsbad and Carlsbad-albite twin boundaries in plagioclase and alkali feldspar. Twins in synthetic anorthite crystals were considered to be solely albite twins in previous TEM studies (Robie et al. 1978; Kroll and Müller 1980; John and Müller 1988; Carpenter 1991). In this paper, we show the Carlsbad and

Carlsbad-albite twin boundaries as revealed by HRTEM and explain possible structures at the twin boundaries.

#### SAMPLES AND EXPERIMENT

The synthetic anorthite crystals were prepared by annealing  $\text{CaAl}_2\text{Si}_2\text{O}_8$  glass at 1400 °C for 1, 4, and 15 min. Detailed procedures were described by Carpenter (1991). Specimens for TEM experiments were prepared by crushing the anorthite in an agate mortar and placing a drop



**FIGURE 3.** (A) A 200 DF image of the anorthite corresponding to the orientation of the SAED pattern in Figure 2A, showing dense (010) twin lamellae. (B) A 002 DF image of the anorthite corresponding to the orientation of the SAED pattern in Figure 2B, showing dense (010) twin lamellae and two coarse twin lamellae (1' and 2) in a Carlsbad twinning relationship.

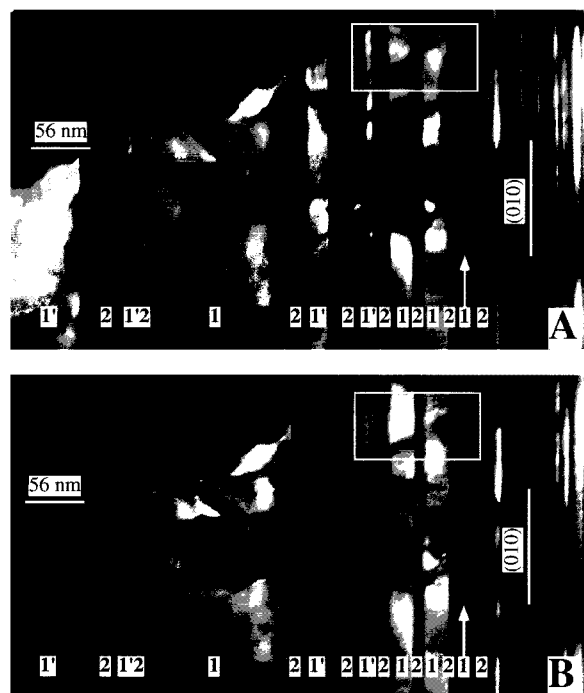
of crystal-alcohol suspension on holey-carbon-coated copper grids. Primary electron diffraction of the anorthite annealed at 1400 °C for 15 min was obtained at Johns Hopkins University with a Phillips 420ST transmission electron microscope equipped with an EDAX energy-dispersive X-ray detector and a Princeton Gamma Tech analyzer. HRTEM images and electron diffraction of other samples were obtained with a JEOL 2000FX transmission electron microscope operated at 200 keV at Arizona State University.

### RESULTS AND DISCUSSION

A selected-area electron diffraction (SAED) pattern with [001] orientation of a synthetic anorthite annealed at 1400 °C for 1 min shows strong streaking along the  $b^*$  direction (Fig. 2A). This pattern is similar to that from feldspar with dense albite twin boundaries. A dark-field (DF) image produced by choosing a 200 reflection and its nearby diffuse intensity shows dense twin lamellae (Fig. 3A). However, the image does not show whether they are albite or Carlsbad and Carlsbad-albite twins.

A [100] SAED pattern from the same anorthite also shows strong streaking along the  $b^*$  direction (Fig. 2B). The diffraction maxima (indicated by arrows) in the streaks could not be due to reflections resulting from albite twinning, but they could be due to Carlsbad twinning. A 002 DF image corresponding to this orientation shows dense twin lamellae with relatively coarse twin lamellae in local areas (Fig. 3B).

SAED patterns from regions with relatively coarse twin



**FIGURE 4.** (A) A DF image of the anorthite produced by two 002 reflections (i.e., aperture A in Fig. 2F), showing bright (010) lamellae related by albite twins. Dark lamellae are related by Carlsbad or Carlsbad-albite twins with respect to the bright lamellae. (B) A DF image of the anorthite produced by one 002 reflection (i.e., aperture B in Fig. 2F), showing only one set of bright twin lamellae. Lamellae of types 1 and 2, 1 and 1', and 1' and 2 are in Carlsbad, albite, and Carlsbad-albite twinning relationships, respectively. The boxed area corresponds to the area of the HRTEM image in Figure 5.

lamellae show overlapping sharp reflections. Figure 2C shows that there are two orientations of lamellae in a Carlsbad-albite twinning relationship. SAED patterns from each lamella show that the pattern in Figure 2C resulted from an overlapping of [100] (Fig. 2D) and  $[20\bar{1}]$  (Fig. 2E) SAED patterns. The neighboring lamellae with the orientations shown in Figures 2D and 2E are in a Carlsbad-albite twinning relationship. There are diffuse “ $b$ ” reflections ( $h + k$  odd, and  $l$  odd) and “ $c$ ” reflections ( $h + k$  even,  $l$  odd) in Figures 2C and 2D. More complex twinning occurs locally (Fig. 2F), where Carlsbad, albite, and Carlsbad-albite twins coexist.

Lamellae with orientations 1 and 1', and 2 and 2' in Figure 2F are in an albite twin relationship. Those with orientations 1 and 2, and 1' and 2' are in a Carlsbad twin relationship, and those with orientations 1 and 2', and 1' and 2 are in a Carlsbad-albite twin relationship (Figs. 1 and 2). On the basis of our investigation of SAED and DF images of the anorthite, crystals annealed at 1400 °C for 1, 4, and 15 min show similar concentrations of twin boundaries. Most are Carlsbad and Carlsbad-albite twins.

The crystal annealed for 1 min locally contains coarse twin lamellae. By placing an objective aperture around

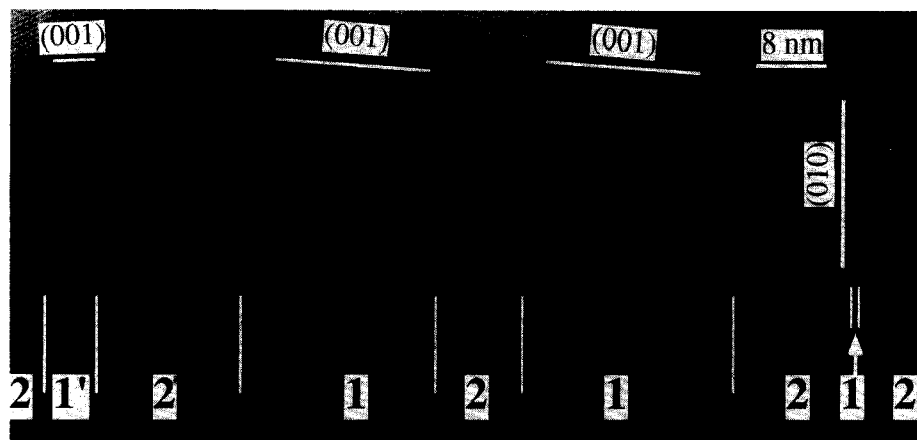


FIGURE 5. HRTEM image of the anorthite shown in the boxed area of Figure 4, showing albite, Carlsbad, and Carlsbad-albite twin lamellae with coherent twin boundaries. The lamellae labeled 2 are dark in both DF images, and those labeled 1 are bright in both DF images (Fig. 4). However, the lamella labeled 1' is bright in the DF image in Figure 4A and dark in the DF image in Figure 4B. All twin boundaries are marked by vertical bars. See text for details.

the two 002 reflections (aperture A in Fig. 2F), bright and dark lamellae are evident (Fig. 4A). These lamellae are in either Carlsbad or Carlsbad-albite twinning relationships. If we choose only one 002 reflection (aperture B in Fig. 2F), only some of the bright lamellae in Figure 4A remain bright (Fig. 4B). An HRTEM image of the boxed area in Figure 4 shows twin lamellae in Carlsbad (lamellae 1 and 2) and albite (lamellae 1 and 1') twinning relationships (Fig. 5). An HRTEM image of an area containing dense lamellae shows twins with thicknesses on the unit-cell scale in Carlsbad, albite, and Carlsbad-albite twinning relationships (Fig. 6). All the observed twin boundaries are coherent.

HRTEM images of anorthite crystals annealed at 1400 °C for 15 min also show the lamellae in Carlsbad, Carlsbad-albite, and albite twinning relationships (Figs. 7 and 8). The twin boundaries are coherent. In the HRTEM images, the (102) plane of anorthite is the same as the (101) plane of alkali feldspar because the dimension of anorthite along the *c* axis is doubled with respect to alkali feldspar (Ribbe 1983). Figure 8 shows the relationship among the albite, Carlsbad, and Carlsbad-albite twin laws illustrated in Figure 1A. Twin lamellae 1 and 1' are in an albite twinning relationship; that is, they are related by a (010) twin mirror or a twin axis normal to (010). Similarly, lamellae 2 and 2' are also in an albite twinning relationship, even though they do not contact each other. Lamellae 1 and 2 (or 1' and 2') are related by a twin axis parallel to the *c* axis and could be considered Carlsbad twins. Lamellae 1' and 2 (or 1 and 2') are in a Carlsbad-albite twinning relationship; that is, they are related by a twin axis normal to the *c* axis and in the (010) plane.

Previous studies of Carlsbad twinning in alkali feldspars have shown only possible atomic arrangements on (001) planes of one twin lamella (Dowty 1980; Wooster 1981). There is only a slight difference across the Carlsbad twin boundary in this projection, and therefore it is

difficult to identify Carlsbad twins in HRTEM images for this orientation. However, the difference between neighboring Carlsbad twin lamellae is obvious in views down the *a* axis of one of the twin lamellae. HRTEM images corresponding to this orientation clearly show lamellae in Carlsbad and Carlsbad-albite twinning relationships (Figs. 5–8). It is possible that some Carlsbad twins may be misidentified as albite twins because of feldspar orientation.

It can be inferred that relatively disordered calcic plagioclase crystallizing at high temperature favors the formation of Carlsbad and Carlsbad-albite twins, on the basis of the frequencies of the twins in both natural and synthetic anorthite crystals. The anorthite annealed at 1400 °C for 1 min shows a disordered structural state (i.e., Al-Si distribution) (Carpenter 1991). We think that the initial structural state in the anorthite annealed at 1400 °C



FIGURE 6. HRTEM image of the anorthite corresponding to the orientation of the SAED pattern in Figure 2B, showing twin lamellae at the unit-cell scale. Lamellae 1 and 1', 1 and 2, 1' and 2 are, respectively, in albite, Carlsbad, and Carlsbad-albite twinning relationships. Twin boundaries are marked by vertical bars.

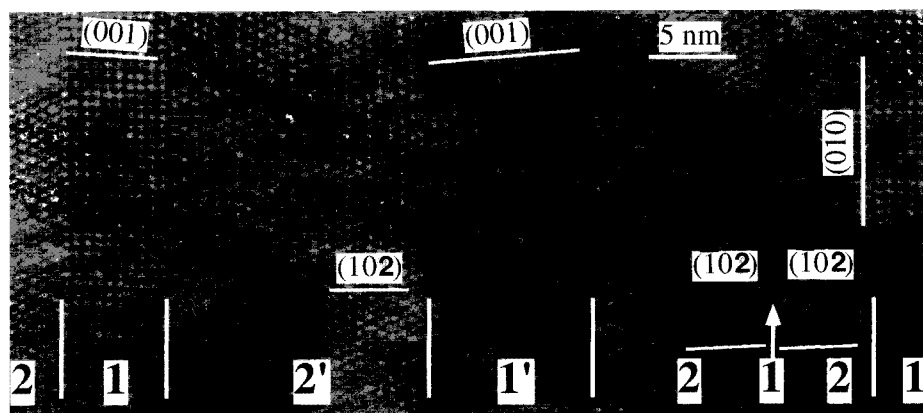


FIGURE 7. HRTEM image of the anorthite annealed at 1400 °C for 15 min, showing complex twin lamellae with coherent twin boundaries. All twin boundaries are marked by vertical bars. See text for details.

during the transformation from glass into anorthite is similar.

We propose that the strain energy that results from Carlsbad and Carlsbad-albite twin boundaries is lower than that resulting from albite twin boundaries in calcic plagioclase with disordered Al-Si, and that additional energy arising from Carlsbad and Carlsbad-albite twin boundaries increases as the Al-Si ordering increases in calcic plagioclase. Therefore, the concentration of Carlsbad and Carlsbad-albite twin boundaries, and the ratio of the concentration of such twin boundaries to albite and pericline twin boundaries, may be related to the initial structural state of the calcic plagioclase. Local Al-Al, Al-Si, and Si-Si interactions at twin boundaries affect the formation of the twins and the stability of the twin boundaries. Our TEM results confirm that high concentrations

of Carlsbad and Carlsbad-albite twin boundaries in calcic plagioclase indicate their relatively high crystallization temperature, on the basis of the dense Carlsbad and Carlsbad-albite twin boundaries in the anorthite. In conclusion, we presume there was extensive Si-Al disorder in the anorthite glass and melt, and rapid crystallization prevented Al-Si ordering and produced the dense Carlsbad and Carlsbad-albite twins we observed using HRTEM.

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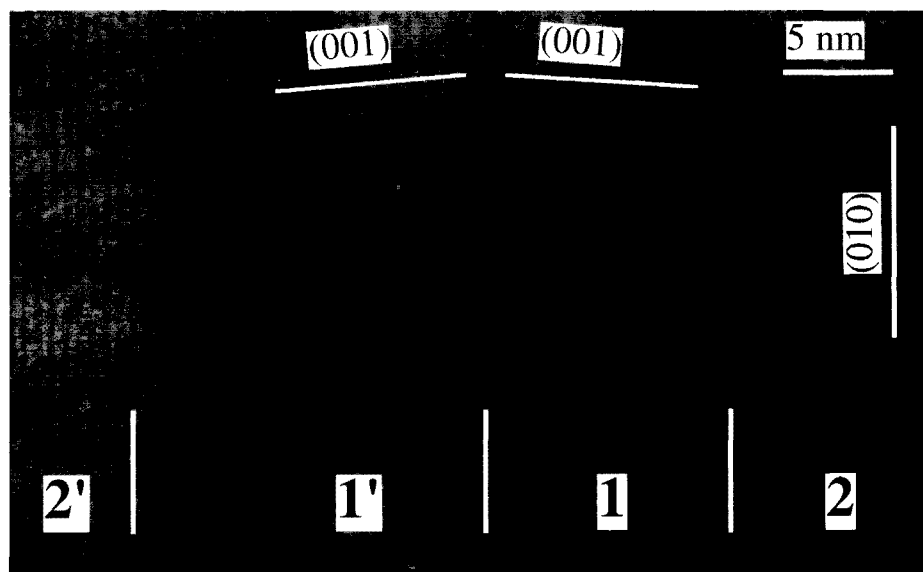


FIGURE 8. HRTEM image of the anorthite annealed at 1400 °C for 15 min, showing complex twin lamellae with coherent twin boundaries (see text for detail). The SAED pattern from this area is similar to the pattern in Figure 2F.

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