Crystallization Kinetics of High-Pressure Phases from Amorphous Plagioclase

Time-resolved X-ray diffraction measurements have revealed that amorphization pressures in plagioclase decrease with increasing temperature, and that the crystallization kinetics in the amorphous plagioclase is different among the high-pressure phases. Jadeite first crystallizes, and nucleation of the other phases is significantly delayed. These findings have important implications for the plagioclase breakdown observed in shocked meteorites.

The evolution of asteroids by collisions was a main process of planetary formation in the early solar system. The physical conditions of the collisional processes are recorded in shocked meteorites, resulting in amorphization and high-pressure transformations of plagioclase, a major constituent mineral. The amorphous plagioclase and high-pressure phases of plagioclase commonly observed in meteorites must be important clues for understanding the collision conditions. However their study has been faced with problems. The experimental conditions for plagioclase amorphization in previous shock and diamond anvil cell (DAC) experiments have been limited to time scales much shorter and to lower temperatures compared to the conditions of natural shock events, resulting in unrealistically high experimental estimates of amorphization pressure [1]. It is well known that sodic plagioclase decomposes into jadeite and silica phases (coesite or stishovite) at high pressures. However in shocked meteorites, only iadeite has been found without the crystalline silica phase even in grains with the chemical composition of plagioclase [2]. Equilibrium phase relations of plagioclase can not explain the presence of unusual assemblages of high-pressure phases in meteorites. In order to solve these problems, we have conducted in-situ X-ray diffraction experiments on the plagioclase amorphization and crystallization kinetics of high-pressure phases from amorphous plagioclase using a Kawai-type high-pressure apparatus and synchrotron radiation.

Two kinds of plagioclase feldspars with different contents of NaAlSi₃O₈ albite (Ab), CaAl₂Si₂O₈ anorthite (An), and KAlSi₃O₈ Orthoclase (Or) were used as the starting materials in the present study. One of the samples was natural labradorite (Ab45.0An51.8Or3.2) powder, the other was natural albite (Ab98.0An0.4Or1.6) powder. Pressure was generated using a double stage system in the high-pressure apparatus MAX-80 and MAX-III installed at AR-NE5C and BL-14C2, respectively. The pressure was calculated from the unit cell volume of gold, and temperature was monitored by recording W3%Re-W25%Re thermocouple X-ray diffraction patterns every 10 sec at high pressures and high temperatures using the white X-ray energy dispersive method.

We carried out a total of 9 experiments for pressures between 8 and 20 GPa and temperatures between 25 and 1500°C. Figure 1 shows an example of the changes in the X-ray diffraction pattern of plagioclase during cold compression and heating. Cold compression of the samples caused broadening of the X-ray diffraction



Figure 1

Changes in the X-ray diffraction patterns of plagioclase (labradorite) during cold compression and heating. Cold compression caused broadening of the X-ray diffraction peaks in plagioclase. Full amorphization occurred by heating, and amorphous plagioclase survived without crystallization at relatively high temperatures. g = graphite capsule; au = gold pressure marker. All other peaks are from plaqioclase.



Figure 2

Changes in the X-ray diffraction patterns during crystallization of high-pressure phases from amorphous plagioclase (labradorite). g = graphite capsule; au = gold pressure marker; st = stishovite; jd = jadeite; gt = garnet.

peaks in plagioclase, however full amorphization was not observed at room temperature even at a pressure of 20 GPa. The X-ray peak broadening in plagioclase became more significant with increasing temperature after the cold compression. Finally plagioclase amorphization occurred by heating at high pressure. We found that the amorphization pressure decreases with increasing temperature. For example, we observed that labradorite becomes amorphous at 16.4 GPa and 470°C. This is a much lower pressure than was reported in previous shock experiments (30-32 GPa) [1]. Negative slopes of the amorphization boundaries are inferred from our experimental results. This may indicate the metastable extension of the melting curve of plagioclase, as suggested in other materials such as water ice and silica phase [3]. The amorphous plagioclase survives kinetically without crystallization even at relatively high temperatures of around 1273 K.

Figure 2 shows an example of the changes in the X-ray diffraction pattern during crystallization of highpressure phases. The crystallization kinetics from (partially) amorphous plagioclases is rather different for the different minerals. Jadeite first appears from amorphous plagioclases of both chemical compositions. Nucleation of other minerals such as garnet and stishovite is significantly delayed. Analysis of the kinetic data based on the Avrami rate equation indicates that the n-value for the formation of Jadeite is around 0.5, whereas the n-values for other phases are relatively large, at around 3. This may suggest fast nucleation and diffusion-controlled growth of jadeite, and slow nucleation rates for the other phases. Our results imply that jadeite can be metastably present without stishovite in certain temperature ranges on the timescale of the shock events. It may be possible to explain the missing silica mineral problem as being due to the differences in the crystallization kinetics of the high-pressure phases from amorphous plagioclase.

The plagioclase amorphization induced by heating and the crystallization kinetics of the high-pressure phases from the amorphous plagioclase must be considered when interpreting the shock conditions of meteorites. These findings, along with further quantitative studies on the P-T conditions of amorphization and the P-T dependencies of crystallization kinetics will make it possible to constrain the P-T-t conditions during shock events and asteroidal collision in the early solar system based on the unique nature of the plagioclase breakdown [4].

REFERENCES

- D. Stöffler, R. Ostertag, C. Jammes, G. Pfannschmidt, P.R.S. Gupta, S.B. Simon, J.J. Papike and R.H Beauchamp, *Geochim. Cosmochim. Acta*, 50 (1986) 889.
- [2] M. Miyahara, A.E. Goresy, E. Ohtani, T. Nagase, M. Nishijima, Z. Vashaei, T. Ferroir, P. Gillet, L. Dubrovinsky and A. Simionovici, *Proc. Natl. Acad. Sci.*, **105** (2008) 8542.
- [3] R.J. Hemley, A.P. Jephcoat, H.K. Mao, L.C. Ming and M.H. Manghnani, *Nature* 334 (1988) 52.
- [4] T. Kubo, M. Kimura, T. Kato, M. Nishi, A. Tominaga, T. Kikegawa and K. Funakoshi, submitted to *Nature Geoscience*.

BEAMLINES

AR-NE5C and 14C2

T. Kubo¹, M. Kimura², T. Kato¹, M. Nishi¹, A. Tominaga¹, T. Kikegawa³ (¹Kyushu Univ., ²Ibaraki Univ., ³KEK-PF)