In situ observation of crystallization of materials with high solubilities

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The crystallization from an aqueous solution is an important process in many systems, such as the creation of nanomaterials, the crystallization of protein crystals for molecular structure analysis, the production of minerals from living organisms, etc. To understand its physical picture, numerous studies has been performed by in situ observations using various microscopies. However, the understanding of early stages of crystallization, especially appearance of crystal nuclei (nucleation) and subsequent crystal growth under sub-micrometer scale, is still limited because of limitation of techniques to observe these rapid, unexpected and small scale phenomena in real time. Recently, several types of the liquid cells have been developed for putting liquids in the high vacuum environment of transmission electron microscopy (TEM). These techniques allow us to observe the crystallization processes inside the liquid and have potential for understanding the early stages of crystallization. In fact, several groups succeeded in observing the crystallization processes of some materials using the liquid cell TEM, and these observation suggests that there are multiple pathways in the nucleation of the stable crystalline phase (J. J. De Yoreo, *Prog. Cryst. Growth Charact. Mater.* **62**, 69, 2016). However, behind the mechanisms in the selectivity of the pathways are still incompletely understood. To investigate the early stages of crystallization in liquid, we employed the liquid cell TEM to observe the crystallization of the materials with relatively high solubilities, such as sodium chlorate and lysozyme protein (T. Yamazaki *et al.*, *Proc. Natl. Acad. Soc.* **114**, 2154, 2017.) which are relatively sensitive to electron irradiation, and, therefore, more difficult to observe by liquid cell TEM. Here we will introduce our recent results of in situ observation of the crystallization of these materials.

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