FERROELECTRIC PHASE TRANSFORMATION ACCELERATED IN NANOLAMINATE HfO_2-ZrO_2 THIN FILMS

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Ferroelectric HfO_2 thin films have a potential to realize high-density nonvolatile memories and neuromorphic circuits for next generation computing. Thus, developments of ferroelectricity-based devices such as FeRAM, FeFET, and FTJ are in progress [1]. Among the dopant engineering studies in HfO_2 crystalline films to attain orthorhombic phase that shows ferroelectricity [2], HfO_2-ZrO_2 solid solution system is recognized as a convenient choice because good ferroelectric properties are achieved at the metallic element ratio of Hf:Zr = 50:50. This character has also promoted the investigations of nanolaminate films and superlattice films that consist of alternately deposited HfO_2 and ZrO_2 ultrathin films [3-6]. In this work, we present the advantage of nanolaminate HfO_2-ZrO_2 thin films from the viewpoint of phase transformation kinetics.
12-nm-thick $Hf_{0.5}Zr_{0.5}O_2$ solid solution and HfO_2-ZrO_2 nanolaminate films were deposited by a sputtering system at room temperature. The $Hf_{0.5}Zr_{0.5}O_2$ film was prepared by a $Hf_{0.5}Zr_{0.5}O_2$ solid solution target. The HfO_2-ZrO_2 nanolaminate film was prepared by HfO_2 and ZrO_2 targets, and the deposition thickness of each layer was controlled to 0.25 nm by mechanical shutters. TaN films were used as bottom and top electrodes, and the post metallization anneal (PMA) was processed at 700 ^oC in 1 atm N_2. Details of experimental conditions are described in Ref. [4].

P-V characteristics of capacitors are shown in Fig. 1. The PMA times were set to 1, 10, and 100 min. In case of the solid solution films, the P-V behaviors gradually changes form antiferroelectric to ferroelectric with long annealing time. It suggests that the crystalline structures in the films are transforming from tetragonal to orthorhombic slowly. In contrast, the nanolaminate films show defined ferroelectricity since the 1-min-anneal, and it is stable. It indicates that the ferroelectric phase formation in nanolaminate film is completed fast with a small thermal budget. In this way, phase transformation is drastically accelerated in nanolaminate films. The fast phase transformation is considered to come from the artificial interfaces introduced in the HfO_2-ZrO_2 nanolaminates. It is assumed that a small difference in the chemical bonding natures between Hf-O and Zr-O has worked as driving force to form non-centrosymmetric phase. Microstructural design at the deposition stage is thus effective to control the crystalline phases.

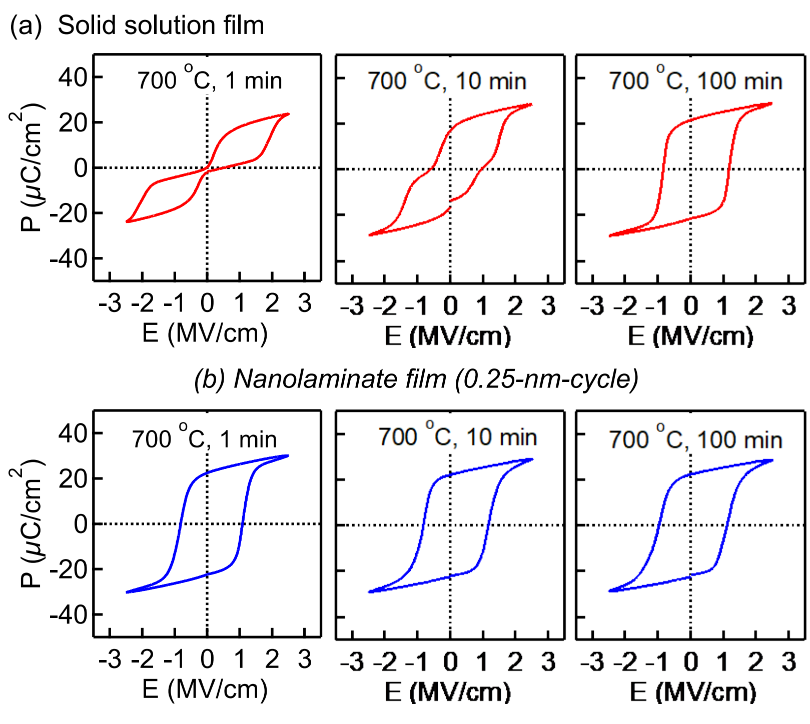

Fig. 1. Impact of annealing times on P-V characteristics of 12-nm-thick Hf-Zr-O capacitors, (a) solid solution and (b) nanolaminate films.

References
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