Mechanistic Understanding of Ceramic Cold Sintering with In Situ Synchrotron-based High-Energy X-Ray Scattering and Diffraction

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Ceramic cold sintering processing represents a novel and environmentally friendly sintering technology capable of producing fully dense ceramic parts under a uniaxial compression typically lower than 500 MPa and at low temperatures less than 500 °C [1]. A mechanistic understanding of cold sintering processing is lacking, hindering its technology development and commercial maturation [2].

To meet this challenge, we developed an integrative in situ monitoring methodology consisting of simultaneous measurements of macroscopic temperature, pressure, volumetric shrinkage, microstructure, and atomic structure, utilizing synchrotron-based high-energy small-angle X-ray scattering and X-ray diffraction techniques. Using ZnO, the most widely studied materials system for ceramic cold sintering [3,4], as a model system, we investigated the phase transition and grain growth mechanism of cold sintering. We demonstrated that cold sintering could occur at unusually low pressure (50 MPa), circumventing the hydrothermal condition previously considered necessary for cold sintering.

We comprehensively characterized the morphological, structural, and interfacial evolution under an isothermal and isobaric condition. Our results revealed three distinct stages. The first stage, associated with heating ZnO to a target temperature, sees a significant volumetric shrinkage and creates the surface contact necessary for sintering. The intermediate stage features the most structural and morphological changes. We observed a kinetic evolution of two nanocrystalline phases and the ZnO, accompanied by systematic changes in the surface area, pore volume, and interfacial roughness. The final stage features a continued evolution of nanocrystalline phases and a reduction of the surface roughness, with a clear presence of residual nanocrystalline phases in the final cold sintered ZnO.

Our results unveil the structural transformation kinetics of cold sintering of ZnO at an unprecedented level and provide previously unattainable data for model validation. Our methodology, extended to other cold-sintering systems, will enable a comprehensive understanding of the transient structural transformations necessary for producing cold-sintered materials with desired mechanical and functional properties.

![Figure 1: (a) in situ SAXS and (b) XRD data revealing the phase and microstructural transformation during ceramics cold sintering.](image)

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