Non-Empirical Identification of Trigger Sites in Crack-Formation during Heterogeneous Reduction of Iron-Ore Sinters Using Persistent Homology

Macroscopic phenomena such as fracture, corrosion, and degradation in materials are often associated with chemical reactions and/or atomic diffusion, which progress heterogeneously among materials. Thus, material properties are generally determined not by their averaged characteristics but by specific features of heterogeneity (or 'trigger sites') of phases, chemical states, etc., where key reactions determining the macroscopic properties initiate and propagate. Therefore, in order to control macroscopic properties it is crucial to identify trigger sites, but this is a challenging task, especially when using 'big data'. Here, we introduce a new persistent homology approach for identifying trigger sites and apply it to the heterogeneous reduction of iron ore sinters [1, 2].

Previous studies have attempted to determine the locations of the trigger sites of heterogeneous processes on the basis of materials-science knowledge derived from experimental data. However, this approach is not valid for more complicated systems such as composite materials (e.g. iron ore sinters and carbon fiber reinforced plastics (CFRP)), batteries, and catalysts, where the heterogeneity of the microstructure and/or chemical states as investigated by X-ray absorption spectroscopy (XAS) are substantially different depending on their locations in a material. In this report, we introduce a new persistent homology approach for identifying trigger sites and apply it to the heterogeneous reduction of iron ore sinters.

Sinter specimens were prepared by liquid sintering from iron ore and limestone. Then, the specimens were heated up to 1473 K in a reducing gas, simulating the iron-making process. Chemical state mapping was carried out using X-ray absorption fine structure spectroscopy (XAFS) at the synchrotron undulator beamline BL-15A1 [3]. The crack formation and phase mapping of larger volumes were investigated by taking measurements using X-ray computed tomography (X-CT).

The X-CT datasets of the reduced sinter were deconvoluted into (a) the initial pores, (b) the microcracks formed during reduction, (c) calcium ferrite phases, and (d) iron oxide phases. The analysis using persistent homology involves (1) transforming each image into a persistence diagram (PD) and then (2) into a vector, (3) feeding the vectors together with the measured crack areas into the absolute shrinkage and selection operator (LASSO), (4) identifying the dominant birth–death pairs, and finally (5) mapping them back into the original image.

The mapping of the valence states of iron oxidation revealed the heterogeneous dynamic evolution of the chemical states from Fe(III) to Fe(II) during the reduction process. At an intermediate stage of the reduction process, the spatial distribution of the changes in the reduced areas was heterogeneous rather than homogeneous, leading to an increase in the local stress and then to crack formation. The changes in the microstructure (i.e. the heterogeneity of the phase mapping) are very complicated, and we cannot determine how the progress of heterogeneous reduction causes crack formation nor empirically identify trigger sites.

However, we have successfully determined the most representative topological features characterizing the reduction process by PD, and the trigger sites for crack formation using LASSO regression techniques. Different types of trigger sites, 'hourglass'-shaped calcium ferrites and 'island'-shaped iron oxides, were determined to initiate crack formation using only mapping data depicting the heterogeneities of phases and cracks without prior mechanistic information.

We have proposed a new approach to identify trigger sites determining macroscopic properties in a case where heterogeneous reactions progress microscopically. Identification of these trigger sites can provide a design rule for reducing mechanical degradation during reduction. Furthermore, this approach is expected to be used to deal with multi-dimensional data obtained by spectroscopic imaging techniques such as transmission X-ray microscopy (TXM) and scanning transmission X-ray microscopy (STXM).

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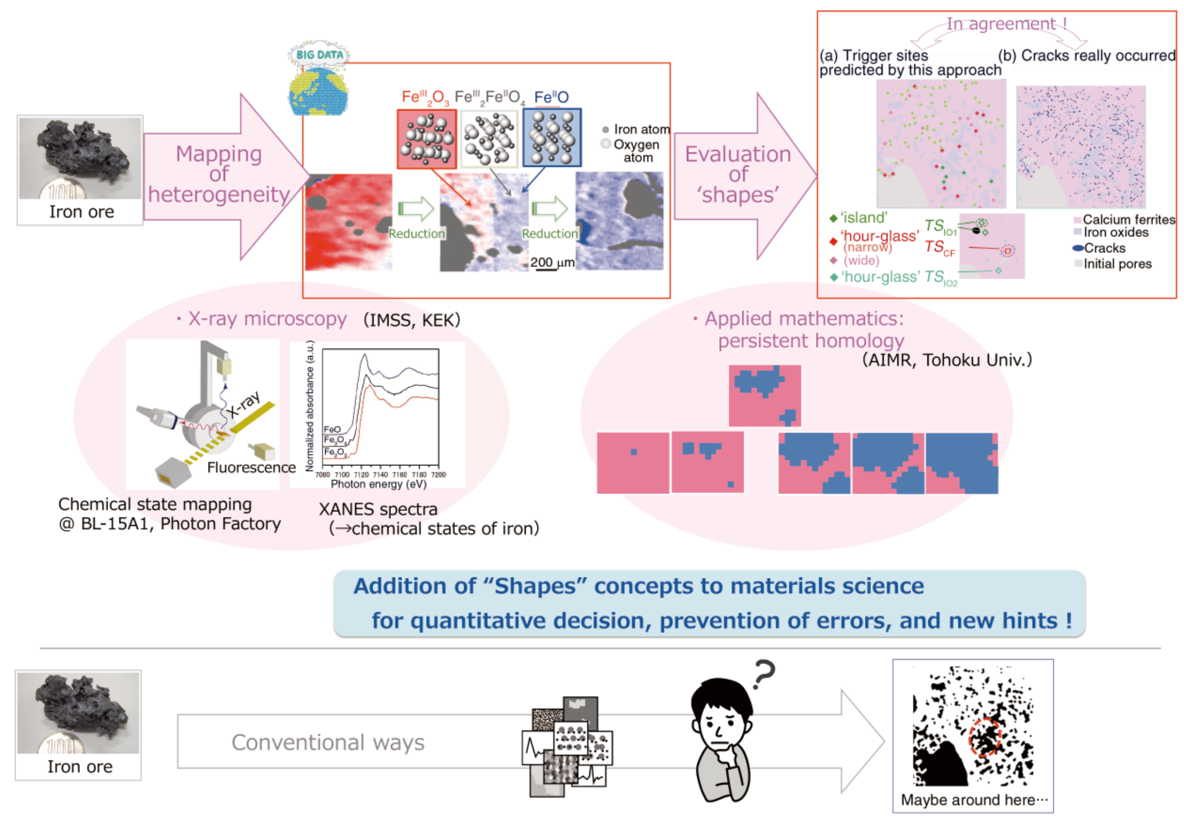

Figure 1: Outline of the new approach to non-empirical identification of trigger sites by combination of X-ray microscopy and persistent homology.

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