Identification of trigger sites in crack-formation during heterogeneous reduction of iron-ore sinters using persistent homology

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[Introduction]

Trigger sites are specific regions or features of heterogeneity in a material where key reactions initiate and take place in systems. 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. These 'empirical' approaches successfully identified trigger sites in a simple system such as metals. However this is not the case 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 such as investigated by XAS, are substantially different depending on their locations in a material. In this study, we introduce a new persistent homology approach for identifying trigger sites and apply it to the heterogeneous reduction of iron ore sinters.

[Experimental methods]

Sinter specimens were prepared by liquid sintering from iron ore and limestone. Then, the specimens were heated up to 1473 K in reductive gas for reduction, simulating the iron-making process. Chemical state mapping was carried out using XAFS at the synchrotron undulator beamline BL-15A1 of the Photon Factory, IMSS, KEK in Japan[1]. The crack formation and phase mapping of larger volumes were investigated using X-CT measurements performed.

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.

[Results & discussions]

The mapping of the valence states of iron oxidation reveals 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, resulting in an increase in the local stress and then to crack formation. The change in the microstructure (i.e. the heterogeneity of the phase mapping) is 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. Four 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.

[Conclusion]

We have proposed a new approach to identify trigger sites determine macroscopic properties in a case where heterogeneous reactions progress microscopically. The identification of these trigger sites can provide a design rule for reducing mechanical degradation during reduction. Furthermore, this approach is expected to apply to deal with multi-dimensional data obtained by spectroscopic imaging techniques such as TXM and STXM.

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