

JoVE62015R2

Quantitative atomic-site-analysis of functional dopants/point defects in crystalline materials by electron-channeling-enhanced microanalysis
by Masahiro Ohtsuka and Shunsuke Muto

Reply to editorial and reviewers' comments

Thank you so much for useful comments and we would like to reply to each comment in the following, described in red underneath each corresponding comment. We hope the present manuscript is now appropriate for publication.

To Editorial comments:

1. Please take this opportunity to thoroughly proofread the manuscript to ensure that there are no spelling or grammar issues.

We thank Editage (www.editage.com) for English language editing.

2. Please revise the lines 21-23, 47-50, 79-88, 242-261 to avoid overlap from previously published work "High-precision quantitative atomic-site-analysis of functional dopants in crystalline materials by electron-channeling-enhanced microanalysis"

We rephrased the specified texts to avoid overlaps. Please confirm if they are appropriate.

3. Please ensure that all text in the protocol section is written in the imperative tense as if telling someone how to do the technique (e.g., "Do this," "Ensure that," etc.). The actions should be described in the imperative tense in complete sentences wherever possible. Avoid usage of phrases such as "could be," "should be," and "would be" throughout the Protocol. Any text that cannot be written in the imperative tense may be added as a "Note." However, notes should be concise and used sparingly.

4. Line 111: Please define the abbreviations before use (ASID)

5. Line 111: Please ensure that the figure number mentioned is correct (TCM is loaded as Figure 3)

6. Line 119: Please elaborate on the fluorescent viewing screen.

7. Line 118: Please ensure that the figure number mentioned is correct (Uploaded Figure 2 has LOP and ROP)

8. Line 131: Please clarify the step mentioned to understand how the condenser is introduced.
9. Line 141: Please mention how the sample and the pivot point height are matched using the Z control keys.
10. Line 146: Please mention how is the annular dark-field detector placed at the beam position.
11. Line 228: Please use the following format for the figure and table legends. Identify the panel label before the panel description. Figure 1: Figure heading. (A) Panel description. (B) Panel description.

We revised the text according to the comments #3-11.

12. Please obtain explicit copyright permission to reuse any figures from a previous publication. Explicit permission can be expressed in the form of a letter from the editor or a link to the editorial policy that allows re-prints. Please upload this information as a .doc or .docx file to your Editorial Manager account. The Figure must be cited appropriately in the Figure Legend, i.e. "This figure has been modified from [citation]."

We obtained copyright permission and uploaded the file to our account. The figures are cited in the corresponding legends, as suggested.

13. Please ensure that the figure numbers mentioned in the text and figure legends match with the uploaded figures.

We confirmed them.

To Reviewers' comments:

Reviewer #1:

I have no major concerns with this manuscript. It gives a comprehensive background for the methodology and describes the motivation and potential interest to other scientists well. The protocol is described in detail and demonstrated in a sufficient manner. The description on the "protocol" section is very detailed, technical, and specific to the JEOL 2100 instrument. This is appropriate, but to make it more relevant to users of other microscopes,

I would consider prefacing this with a brief but more general discussion of the experimental considerations needed for a successful experiment.

Thank you very much for the positive comments to our manuscript. We prefaced about the present protocol specific to a JEM2100 STEM and provide some information in the final paragraph of Discussion section how users of other microscopes should realize the present protocol.

Reviewer #2:

Manuscript Summary:

This manuscript uses a rocking-beam transmission electron microscopy technique to perform quantitative, site-specific microanalysis. The chemical composition and occupancy of various lattice sites is investigated via energy-dispersive X-ray spectroscopy. The technique is applied to investigate preferential site occupation of dopant atoms in co-doped Ca_2SnO_4 . This report appears useful, and I recommend publication. If the suggestions below can be incorporated, this work will be even more widely useful to the research community.

Major Concerns:

This manuscript could be improved by adding data that is mentioned but not shown. Specifically, I recommend to include a figure with the single-doped $\text{Eu}:\text{Ca}_2\text{SnO}_4$ sample in order for the reader to see the direct comparison with the data for the co-doped sample, which would more effectively illustrate the utility of the technique. Second, it is also mentioned that "The line scan function in the EDX controlling software enables 1D tilting measurements of a systematic row of reflections." It would be useful to show this in a figure so that the reader can see this directly. Finally, although not necessary for publication, adding an example using EELS (HARECES) would make this report even more useful to readers.

We thank so much reviewer#2 for the positive and useful comments. We can provide the ICPs of single-doped sample, though they were recorded around the different zone axis and the sample thickness was not the same, which we are afraid would not so effective nor obvious to show the differences between the two samples. This is another advantage of the present method because the analysis fully takes the dynamical effects into account, which ensures quantitative trustability of the results.

Secondly, we revised the protocol and associate figure to include description how 1D tilting experiment is achieved.

Finally readers can easily access several useful examples of HARECES by referring to our published papers cited in the text.

Minor Concerns:

The figure captions are mislabeled for Figs. 5 & 6.

Thank you so much for pointing out the mislabels. We revised the captions.