

Supernormal enrichment of cadmium in sphalerite via coupled dissolution-reprecipitation process



Open Access This file is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. In the cases where the authors are anonymous, such as is the case for the reports of anonymous peer reviewers, author attribution should be to 'Anonymous Referee' followed by a clear attribution to the source work. The images or other third party material in this file are included in the article's Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit <http://creativecommons.org/licenses/by/4.0/>.

Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality

18th Apr 23

Dear Professor Wang,

Please allow me to apologise for the delay in sending a decision on your manuscript titled "Supernormal enrichment of cadmium in sphalerite via coupled hydrothermal dissolution-precipitation process". It has now been seen by 2 reviewers, whose comments are appended below. You will see that they find your work of some potential interest. However, they have raised quite substantial concerns that must be addressed. In light of these comments, we cannot accept the manuscript for publication, but would be interested in considering a revised version that fully addresses these serious concerns.

In particular, we would need to see that any revised manuscript meets the following editorial thresholds:

- * Present a compelling conceptual model for the enrichment of cadmium in sphalerite which accounts for the limitations and uncertainties associated with your small sample size.
- * Provide compelling evidence for the presence of Greenockite as well as further evidence that your interpretations are robust and based on sound electron microscopy techniques.
- * Justify and explain your interpretation of episodic fluid pulses in the context of the Jinding and other deposits.

We hope you will find the reviewers' comments useful as you decide how to proceed. Should additional work allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. If you choose to take up this option, please either highlight all changes in the manuscript text file, or provide a list of the changes to the manuscript with your responses to the reviewers.

Please bear in mind that we will be reluctant to approach the reviewers again in the absence of substantial revisions.

If the revision process takes significantly longer than three months, we will be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

We are committed to providing a fair and constructive peer-review process. Please do not hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the reviewers' comments with a list of your changes to the manuscript text (which should be in a separate document to any cover letter) and any completed checklist:

[Link redacted]

** This url links to your confidential home page and associated information about manuscripts you

may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

Please do not hesitate to contact me if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Joe Aslin

Senior Editor,
Communications Earth & Environment
<https://www.nature.com/commsenv/>
Twitter: @CommsEarth

EDITORIAL POLICIES AND FORMAT

If you decide to resubmit your paper, please ensure that your manuscript complies with our editorial policies and complete and upload the checklist below as a Related Manuscript file type with the revised article:

Editorial Policy Policy requirements (Download the link to your computer as a PDF.)

For your information, you can find some guidance regarding format requirements summarized on the following checklist:(<https://www.nature.com/documents/commsj-phys-style-formatting-checklist-article.pdf>) and formatting guide (<https://www.nature.com/documents/commsj-phys-style-formatting-guide-accept.pdf>).

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

This paper presents a detailed study of hydrothermal-based sulfide mineral reprecipitation processes leading to metal redistribution and Cd enrichment. Electron-beam techniques were used to characterize material collected from the Jinding deposit, Yunnan Province, Southwestern China. The work shows the usefulness of nanoscale imaging in the study of ore genesis.

The manuscript is logically organized and generally well-written. I feel with some alterations, this will make a nice submission.

Major comments

While reading the paper, I wondered about the representativeness of results relying on a limited number of samples to explain a very complex and spatially heterogeneous natural process. I am aware that it is a general weakness of advanced, time-consuming studies. Still, the authors should indicate how they overcome the problem in their study. Why do they assume that using one hand specimen and one FIB specimen is enough to propose a model for a large-scale hydrothermal process? Showing three times the same part of one polished section (Figs 1a, 1b, and 1d) does not help. How do the authors know they have greenockite in the sample and not the cubic variety of CdS? Presumably, not based on their own results as the SAED pattern (Fig. 2a) is rather inconclusive (see

the comments below). Please provide clear evidence for the greenockite occurrence in the sample.

Minor comments

Abstract: Consider mentioning Ag in addition to Cd in the abstract.

Lines 58-95: what technique/instrument was used to obtain the results? BSE imaging within the EMPA system? SEM-FIB? Unclear; please explain.

Line 71: remove "(CMR)"; used only ones in the text.

Line 78-80. Oversimplification. I do not think that Fig. 1a proves increased crystallinity from the core to the rim. SEM imaging is not a proper method; the image reveals rather a multi-stage crystallization/reprecipitation process. Please rephrase.

Line 106: remove "Pioneer". Starting with "Discovery" is attractive enough.

Line 109: how do the authors know the aggregates are non-crystalline?

Line 112: the same aggregates are now polycrystalline? Please correct/explain adequately.

Line 113: the amorphous matrix is better seen in the SAED pattern (Fig. 2a)

Line 118: why do the authors think Ag is in the amorphous phase? Why not in the CdS NPs?

Line 119: probably better "in the middle"

Fig. 1. Please indicate the technique used to obtain the images.

Fig. 2. SAED patterns of CdS and PbS NPs and ZnS: please correct/supplement by providing measurements of the d-spacing of the hkl rings. There is probably more than one phase in the SAED patterns presented in Fig. 2a, 2c, and 2e. The hkl indices are incorrectly provided. For sphalerite should be: 111, 220, 311 and not: 111, 022, 113. The sequence for galena is: 111, 200, 220, 222. Please see: DOI: 10.1021/acs.inorgchem.8b02880 Inorg. Chem. 2019, 58, 672–678 for greenockite. Supplementary Table 1. There are two Tables in the supplement. Please provide descriptions for both tables. Add units in the first Table! The data provided in Table 1 are crucial for the ms, but difficult to understand as presented. JD-2, JD-4, ... are analyzed points and not samples. Please correct. Correlate the mineral description in the Table with results in the ms (sphalerite core, sphalerite mantle, ... ?). BSE image of the location of the point analyses is highly desirable.

Reviewer #2 (Remarks to the Author):

The manuscript entitled "Supernormal enrichment of cadmium in sphalerite via coupled hydrothermal dissolution-reprecipitation process" by Duan et al documented nano-scale textural and compositional characteristics of Cd-rich sphalerite from the Jinding Zn-Pb deposit and proposed a dissolution-reprecipitation model to explain the Cd-enrichment mechanism. The interpretations and conclusions are supported by the observations, and both of them are novel and have significance for understanding Cd enrichment in hydrothermal systems in general and for MVT or MVT-like Zn-Pb deposits in particular. However, there are a few problems that need to be addressed before it can be considered for publication. The main problems are listed below, and minor comments are annotated in the PDF. Both need to be addressed in the revision of the ms.

1) "Critical elements" or "critical metals" is a political term which means different elements for different countries. Thus, elements that have completely different geochemical behaviors can be included in the so-called "critical elements". It is appropriate to call Cd a critical element, but it is totally inappropriate to discuss how "critical elements" behave geochemically, which does not make any scientific sense. Therefore, delete such general uses or discussions about "critical elements" throughout the paper.

2) Although dissolution-reprecipitation process has been well explained as shown in Fig. 4, it is unclear how this process was repeated episodically as is required for the Cd enrichment in the core of the sphalerite. It is mentioned that this may be related to multi-pulsed fluid process, but it is unclear how many pulses might be involved, whether this is particular to Jinding or common to MVTs. If it is

particular to Jinding, could this be related to the episodic faulting related to the Pijiang fault? If this is common in MVT deposits, could this be related to the fluctuation of basinal-scale hydrodynamic regime? These are questions that need to be discussed in the paper.

3) Fig. 3 displays observations and should be part of the section of results, even though these observations are used to discuss about the process, just like Fig. 1 and Fig. 2. Therefore, part of the contents of discussion (those related to observations in Fig. 3) should be moved to the section of results.

Response (in blue) to Reviewer Comments

Manuscript Number: COMMSENV-23-0205
Manuscript Title: Supernormal enrichment of cadmium in sphalerite via coupled hydrothermal dissolution-reprecipitation process

Reviewer #1:

Major concerns:

(1) While reading the paper, I wondered about the representativeness of results relying on a limited number of samples to explain a very complex and spatially heterogeneous natural process. I am aware that it is a general weakness of advanced, time-consuming studies. Still, the authors should indicate how they overcome the problem in their study. Why do they assume that using one hand specimen and one FIB specimen is enough to propose a model for a large-scale hydrothermal process? Showing three times the same part of one polished section (Figs 1a, 1b, and 1d) does not help.

[Response 1] We totally agree with your concern about the representativeness of samples. Actually, this work is based on SEM-EDS studies on zinc concentrations derived from nearby concentrator plant, where CdS inclusions distributed within cadmiferous sphalerite are easily observed. CdS phases, majorly as mineral blebs and floccules can be recognized in both in-situ mineralization assemblage and zinc concentrates. Besides, the mineralization features in our studied polished section have already been reported in former researches, although the supernormal enrichment of Cd in these assemblages has not been recognized until this work/ We have provided a detailed explanation of this situation in the text line 58-64. Except from FIB cut #1, we add new FIB cuts #2 and #3, which crosscut all types of Cd-anomaly phases mentioned above, to minimize the uncertainty of results as much as possible. The illustration is added, and can be seen in Line 82-92.

(2) How do the authors know they have greenockite in the sample and not the cubic variety of CdS? Presumably, not based on their own results as the SAED pattern (Fig. 2a) is rather inconclusive (see the comments below). Please provide clear evidence for the greenockite occurrence in the sample.

[Response 2] Yes, we recognize that the evidence we provided is not sound. As a supplement for diffraction patterns, we performed direct measurements of d-spacing of both CdS NPs and discrete CdS minerals from high-resolution TEM images, and confirmed that they are hexagonal CdS, namely greenockite, rather than cubic forms. For cross validation purpose, we also assessed Raman spectrum on these CdS phases. However, the results are unsatisfactory for negligible difference in Raman spectra between CdS polymorphism and nanoparticles (Fig. 1), as well as intense fluorescence effect caused by sphalerite host (Fig. 2). Thus, we didn't add these studies in this work.

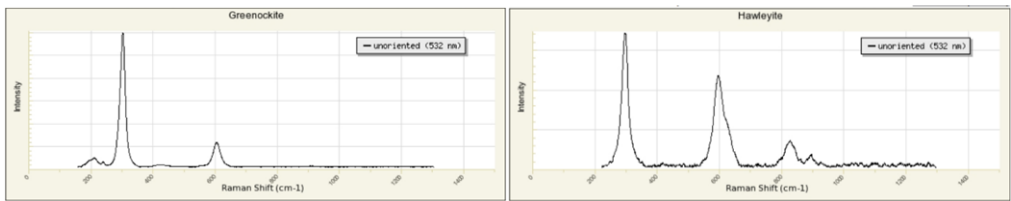


Fig. 1 Comparison between Raman spectra of greenockite and hawleyite form database RRUFF™ Project

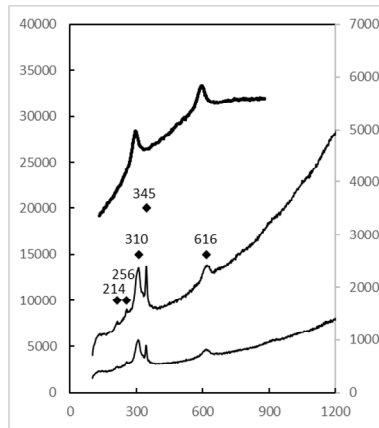


Fig. 2 Result of Raman spectra of CdS blebs in the cadmiferous sphalerite, compared with a typical greenockite Raman spectrum.

Minor concerns:

(3) Abstract: Consider mentioning Ag in addition to Cd in the abstract.

[Response 3] Thanks for your advice. Unfortunately, extreme Ag concentration detected by STEM-EDS are turn out to be artificial, because a major Cd peak at ~ 3 Kev in EDS spectrum are mistakenly classified as Ag signal due to their similarity. This is also confirmed by EPMA analyses, for less than 0.2 wt.% Ag is detected in CdS inclusions. We have revised the whole text in manuscript, especially point analyses and mappings by EDS, major revisions can be seen in revised Fig. 3 and Supplementary Fig. 1.

(4) Lines 58-95: what technique/instrument was used to obtain the results? BSE imaging within the EMPA system? SEM-FIB? Unclear; please explain.

[Response 4] We accept this criticism and have added descriptions of technique/instrument in Line 58, and 65.

(5) Line 71: remove “(CMR)” ; used only ones in the text.

[Response 5] Good suggestion. We have removed “(CMR)” as seen in Line 65-66.

(6) Line 78-80. Oversimplification. I do not think that Fig. 1a proves increased crystallinity from the core to the rim. SEM imaging is not a proper method; the image reveals rather a multi-stage crystallization/precipitation process. Please rephrase.

[Response 6] We accept this criticism and have accordingly removed over-interpretation on increased crystallinity, as seen in Line 72.

(7) Line 106: remove “Pioneer”. Starting with “Discovery” is attractive enough.

[Response 7] Yes, we have removed “Pioneer” in Line 82.

(8) Line 109: how do the authors know the aggregates are non-crystalline?

[Response 8] We have rephrased the aggregates as “poly-crystalline” sulfide nanoparticles based on high-resolution TEM image and SAED pattern, as seen in Line 97.

(9) Line 112: the same aggregates are now polycrystalline? Please correct/explain adequately.

[Response 9] We have confirmed the aggregates as “poly-crystalline” sulfide nanoparticles based on high-resolution TEM image and SAED pattern.

(10) Line 113: the amorphous matrix is better seen in the SAED pattern (Fig. 2a).

[Response 10] Thanks for your suggestion. The amorphous matrix is generally presented as clouds

rather than spotted in SAED patterns. Thus, we have rephrased CdS phases as polycrystalline hexagonal-CdS nanoparticle aggregate in Line 95-98.

(11) Line 118: why do the authors think Ag is in the amorphous phase? Why not in the CdS NPs?

[Response 11] As mentioned in [Response 3], we reconsider Ag signal as mis-classified Cd signal.

(12) Line 119: probably better “in the middle”.

[Response 12] Good advice. However, because of structural adjustment, we have removed such illustration in the manuscript, as seen in Line 93-118.

(13) Fig. 1. Please indicate the technique used to obtain the images.

[Response 13] Thanks for your suggestion. Correction can be seen in revised Fig. 1.

(14) Fig. 2. SAED patterns of CdS and PbS NPs and ZnS: please correct/supplement by providing measurements of the d-spacing of the hkl rings. There is probably more than one phase in the SAED patterns presented in Fig. 2a, 2c, and 2e. The hkl indices are incorrectly provided. For sphalerite should be: 111, 220, 311 and not: 111, 022, 113. The sequence for galena is: 111, 200, 220, 222. Please see: DOI: 10.1021/acs.inorgchem.8b02880 Inorg. Chem. 2019, 58, 672–678 for greenockite.

[Response 14] We accept this criticism and have provided standard hkl indices, as seen in Fig. 2.

(15) Supplementary Table 1. There are two Tables in the supplement. Please provide descriptions for both tables. Add units in the first Table! The data provided in Table 1 are crucial for the ms, but difficult to understand as presented. JD-2, JD-4, ... are analyzed points and not samples. Please correct. Correlate the mineral description in the Table with results in the ms (sphalerite core, sphalerite mantle, ... ?). BSE image of the location of the point analyses is highly desirable.

[Response 15] Yes, we have merged two tables as Supplementary Fig. 1, added units in the first row of the chart, corrected numbering method as Spot-1, Spot-2, ..., and provided analyzed positions of EPMA in BSE image in Excel files of supplementary Table. 1.

Reviewer #2

(16) “Critical elements” or “critical metals” is a political term which means different elements for different countries. Thus, elements that have completely different geochemical behaviors can be included in the so-called “critical elements”. It is appropriate to call Cd a critical element, but it is totally inappropriate to discuss how “critical elements” behave geochemically, which does not make any scientific sense. Therefore, delete such general uses or discussions about “critical elements” throughout the paper.

[Response 16] We agree with your concern on terminology usage of “critical elements” and we replaced it by “dispersed metals” that used to refer Cd, Ga, Ge, In, Tl throughout the paper.

(17) Although dissolution-reprecipitation process has been well explained as shown in Fig. 4, it is unclear how this process was repeated episodically as is required for the Cd enrichment in the core of the sphalerite. It is mentioned that this may be related to multi-pulsed fluid process, but it is unclear how many pulses might be involved, whether this is particular to Jinding or common to MVTs. If it is particular to Jinding, could this be related to the episodic faulting related to the Pijiang fault? If this is common in MVT deposits, could this be related to the fluctuation of basinal-scale hydrodynamic regime? These are questions that need to be discussed in the paper.

[Response 17] We appreciate your concern in these issues. Generally, Zn-Pb mineralization in MVT deposit involve mixing between deeply-derived, oxidative (SO_4^{2-} -dominant) and metal-bearing brine with shallow, H_2S -rich and reducible sedimentary formation waters. During brine ascending and cooling, the pH of the brine shifted towards more acidic and the oxidation increased to

conditions above the hematite-magnetite buffer (Pfaff et al., 2010). Thus, we speculate metalliferous brines are fully competent for serving as oxidative, acidic fluids for triggering coupled dissolution-reprecipitation reaction. When metalliferous brines meet reduced sulfur and are buffered by wall rocks, sulfide precipitation dominates, but how about brine injection into early-stage formed sulfide assemblages without mixing with sulfur or neutralized by carbonates? At such conditions, sulfide dissolution, rather than precipitation dominant. In this case, we propose hydrothermal overprinting by late-stage oxidative acidic brines on early-formed Cd-rich sphalerite during insufficient S supply as a main trigger for supernormal enrichment of Cd via coupled dissolution-reprecipitation reaction (CDR) at the core of sphalerite (most likely early-stage sphalerite relics). As you can imagine, deep brine injection by episodic faulting related to the Pijiang fault in the Jinding deposits, or fluctuation of basinal-scale hydrodynamic regime can indeed explain Cd variation in hydrothermal fluids as well as in compositionally zoned sphalerite, in this case, late-formed sphalerite mantles and rims. However, such mechanisms may not ensure CDR take place. Thus, we have emphasized intense hydrothermal overprinting at deposition site of sphalerite, rather than fluctuation in basinal-scale hydrothermal regime as a major mechanism for rare supernormal enrichment of Cd in rare paragenetic assemblages, veins and Zn-Pb deposits. These issues are also briefly discussed in the revised manuscript, as seen in Line 219-249.

Reference:

Pfaff, K. et al. Formation of the Wiesloch Mississippi Valley-type Zn-Pb-Ag deposit in the extensional setting of the Upper Rhinegraben, SW Germany. *Miner. Depos.* 45, 647–666 (2010).

(18) Fig. 3 displays observations and should be part of the section of results, even though these observations are used to discuss about the process, just like Fig. 1 and Fig. 2. Therefore, part of the contents of discussion (those related to observations in Fig. 3) should be moved to the section of results.

[Response 18] Good advice. We have moved this part into results and emphasized it in discussions, as seen in Line 119-125.

22nd Aug 23

Dear Professor Wang,

Your manuscript titled "Supernormal enrichment of cadmium in sphalerite via coupled hydrothermal dissolution-precipitation process" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

EDITORIAL REQUESTS:

Please review our specific editorial comments and requests regarding your manuscript in the attached "Editorial Requests Table".

*****Please take care to match our formatting and policy requirements. We will check revised manuscript and return manuscripts that do not comply. Such requests will lead to delays. *****

Please outline your response to each request in the right hand column. Please upload the completed table with your manuscript files as a Related Manuscript file.

If you have any questions or concerns about any of our requests, please do not hesitate to contact me.

SUBMISSION INFORMATION:

In order to accept your paper, we require the files listed at the end of the Editorial Requests Table; the list of required files is also available at <https://www.nature.com/documents/commsj-file-checklist.pdf> .

OPEN ACCESS:

Communications Earth & Environment is a fully open access journal. Articles are made freely accessible on publication under a [CC BY license](http://creativecommons.org/licenses/by/4.0) (Creative Commons Attribution 4.0 International License). This license allows maximum dissemination and re-use of open access materials and is preferred by many research funding bodies.

For further information about article processing charges, open access funding, and advice and support from Nature Research, please visit <https://www.nature.com/commsenv/article-processing-charges>

At acceptance, you will be provided with instructions for completing this CC BY license on behalf of all authors. This grants us the necessary permissions to publish your paper. Additionally, you will be asked to declare that all required third party permissions have been obtained, and to provide billing information in order to pay the article-processing charge (APC).

Please use the following link to submit the above items:

[Link redacted]

** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Joe Aslin

Senior Editor,
Communications Earth & Environment
<https://www.nature.com/commsenv/>
Twitter: @CommsEarth

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The manuscript has been greatly improved, all the comments adequately addressed, and is now a pleasure to read. I just have a few minor comments for the authors to consider. I recommend a minor revision.

I suggest carefully checking the punctuation (e.g., line 127) and spelling (e.g., line 227) in the text. I also have the impression that the authors sometimes confuse dissolution of metal sulfides (sulfides are highly insoluble) with oxidative dissolution (congruent decomposition/dissolution of the mineral by oxidation). Please check carefully.

Line 43: maybe better: "Mississippi-valley type (MVT) Zn-Pb deposits" and after that (e.g., lines 53), "MVT Zn-Pb deposits".

Line 61: the word "bonanza" is all right in the abstract but appears rather strange in the result section. Maybe microsites?

Line 62: replace "are firstly reported in this work" by "were found"

Lines 67 and 72: correct to "BSE imaging" or "in BSE image".

Line 137: There is no Fig. 2m in the ms. Please correct.

Line 216: fractionation?

Line 289: correct to "The thin sections for TEM study were prepared"

Line 291: what with Figs. 1 g-i?

Reviewer #2 (Remarks to the Author):

The authors have addressed my comments. The main remaining problem is English writing: there are too many grammatical problems and careless typos. I have marked a few in the first page, and will leave the rest for the authors or professional editing.

Response (in blue) to Reviewer Comments

Manuscript Number: COMMSENV-23-0205B

Manuscript Title: Supernormal enrichment of cadmium in sphalerite via coupled hydrothermal dissolution-reprecipitation process

Reviewer #1:

(1) The manuscript has been greatly improved, all the comments adequately addressed, and is now a pleasure to read. I just have a few minor comments for the authors to consider. I recommend a minor revision. I suggest carefully checking the punctuation (e.g., line 127) and spelling (e.g., line 227) in the text. I also have the impression that the authors sometimes confuse dissolution of metal sulfides (sulfides are highly insoluble) with oxidative dissolution (congruent decomposition/dissolution of the mineral by oxidation). Please check carefully.

[Response 1] Thanks for your advice. We rechecked the punctuation and spelling of the whole manuscript to make sure a better presentation of our manuscript. Your concern about terminology confusion between “dissolution” vs. “oxidative dissolution” is appropriate. We accepted this criticism and replaced controversial terms by terms that emphasize the role of decomposition/dissolution of the sphalerite by oxidation during CDR process (e.g., Line 159, 182, 189 in yellow).

(2) Line 43: maybe better: “Mississippi-valley type (MVT) Zn-Pb deposits” and after that (e.g., lines 53), “MVT Zn-Pb deposits”.

[Response 2] Yes, we have correct related description in Line 36, 46.

(3) Line 61: the word “bonanza” is all right in the abstract but appears rather strange in the result section. Maybe microsities?

[Response 3] Good advice. We replaced the word “bonanza” with “microsites” in Line 55, 259.

(4) Line 62: replace "are firstly reported in this work" by “were found”

[Response 4] Thanks for your suggestion. We understand that terms like “first” can lead to unproductive controversy and is unnecessary. Thus, we have checked the whole manuscript to avoid these wordings.

(5) Lines 67 and 72: correct to “BSE imaging” or ”in BSE image”.

[Response 5] Yes, we have corrected, as seen in Line 59, 61, 66. labelled using lower case letters

(6) Line 137: There is no Fig. 2m in the ms. Please correct.

[Response 6] I apologize for such a negligence. Correction can be seen in revised Fig. 2l.

(7) Line 216: fractionation?

[Response 7] Thanks, we corrected the spelling error, which can be seen in Line 216.

(8) Line 289: correct to “The thin sections for TEM study were prepared”

[Response 8] Yes, we have corrected.

(9) Line 291: what with Figs. 1 g-i?

[Response 9] Thanks for your correction. We have corrected it to “The sampling locations are shown in Fig. 1g-I”.

Reviewer #2

(10) The authors have addressed my comments. The main remaining problem is English writing: there are too many grammatical problems and careless typos. I have marked a few in the first page, and will leave the rest for the authors or professional editing.

[Response 10] Yes, we appreciated your careful review and sincerely accept your criticism. We have rechecked grammatical and spelling errors throughout the whole manuscript to make sure a better presentation of our works. We marked the correction in the manuscript in red.