

The equations of state of nine materials up to 0.43 TPa for extreme pressure science

Corresponding Author: Dr Takeshi Sakai

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

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Dear Dr Sakai,

Thank you for submitting your manuscript, "The equations of state for nine materials up to 0.5 TPa as internally consistent pressure standards for extreme pressure science", to Communications Materials. It has now been seen by 3 referees, whose comments are appended below. You will see that while they find your work of potential interest, some very important points are raised by all three reviewers which we require you to address in order to reach the publication standard of Communications Materials.

We are interested in the possibility of publishing your study in Communications Materials, but would like to consider your response to these concerns in the form of a revised manuscript before we make a decision on publication. We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

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

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type 'Revised Manuscript - Marked Up' when uploading the manuscript file to our online system.

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Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Rostislav Hrubíak, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-1489-2156

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

This paper presents material that is important from several points of view: (i) ultra-high pressure metrology; (ii) behavior of elements and compounds under compression and comparison with models; (iii) static high pressure technologies; (iv) equations of state formalism. Raw data shown in supplementary material is of high quality, and data discussion is sound. The geometry of the toroidal diamond anvils is precisely described. I thus support publication in a journal with a wide readership such as Communication Materials.

However, I believe that there is room for improvement in the form of the manuscript. Below is a list of missing or incorrect information (there is a lot of material presented in this manuscript, I hope I did not miss anything):

- There is missing information on the preparation of the toroidal anvils: geometry of the starting anvil; FIB characteristics (current etc.); method for the measurement of the profile provided in Fig. S1b.
- Lattice parameters are not provided for Fe and Re (hexagonal lattice) in tables S1, S2 and S6, and the volume alone does not allow to calculate diffraction angles as expected for a pressure calibrant
- I found missing references: Tange et al 2009, Zhang et al 2023, Rech+2019. There are mistakes in the references listed lines 120-121. The reference list is not provided in alphabetical order which makes it difficult to find them.
- Please provide the following information for runs 1b, 2b, 4b: loaded materials (sample and pressure calibrant)? Geometry?
- Table 1: missing reference for NaCl-B2 data; which Fe-hcp data have been included in the fit? The present body of data is not sufficient to constrain low P behavior. Same remark for molybdenum.
- Fig. 1: a part of the caption is missing

- Fig. 2: indicate, for each material, the calibration path used to establish the Volume-Volume relation (and subsequent compression curve). I found relevant information in Table S9 but this would deserve to be in the main text to avoid any confusion.
- Fig. 2a: plotting raw data points on the graph would be useful as it would indicate in which range the volume-volume relation has been actually measured, and where it is extrapolated.
- Fig. 4: conventional DAC data (Sakai et al. 2014) is plotted in this Figure. Is it also used to constraint the parameters listed in Table 2? Then why is Sakai et al. 2014 not mentioned in the caption of Table 2? It would be useful to plot other literature data such as Mao et al 90 (JGR 95 21737) and Dewaele et al 2006 (PRL 97 215504) – possibly others - so that the reader knows how the current compression curve differs from these classical measurements.
- Fig. 5 and corresponding text: please provide an analytical definition for Delta P to avoid any confusion (I guess it is $P_{\text{metal}} - P_{\text{Cu}}$ assuming volume_{metal}-volume_{Cu} relation in fig. 2a)
- Fig. S4, a, b and c exhibit differences between experimental runs. Can you comment? Please provide the forms for the Volume-volume relationships plotted as dashed lines. Could you plot the data from Dewaele et al. PRL 97 215504 2006 on the Fe-W graph?
- Fig. S7: I don't get this graph. How can the 2 MgO pressures taken from Tange et al 2009 differ that much?

I have further suggestions to improve the discussion part:

- The authors could consider re-fitting their NaCl-B2 EoS, the set of parameters they obtain (Table 2) differ significantly from what is found in the literature for other B2 alkali halides phases, where K'_0 is around 6.
- I found the first paragraph of the "Discussion" part rather confusing. It needs to be re-written and clear conclusions should be highlighted. The authors could provide a calibration of the diamond Raman scale based on the present work. Also, it seems to me that they show that platinum is not a reliable pressure calibrant, could they give an advice?
- The introduction of generalized Rydberg-Stacey Equation of state is a very interesting point of this work, as well as the suggestion to constraint its K'_{inf} parameter with ab initio calculations. The authors find that a compression to 700 GPa is necessary to obtain a reliable K'_{inf} for Re (l. 255-270). How would the K'_{inf} value converge for a more compressible material such as NaCl or KCl? A rare gas solid?

Reviewer #2 (Remarks to the Author):

This manuscript describes the experimental results of unit cell measurements for a set of materials at high pressures up to 430 GPa. These experiments are challenging, and the authors have successfully used very small culet anvils to reach pressures exceeding 300 GPa in 7 runs. Additionally, the use of FIB (focused ion beam) for sample loading is a commendable approach for arranging the involved samples as desired.

The measured volume data serve the purpose of testing the internal consistency of the equations of state (EOSs) across nine different materials. This consistency is crucial for understanding the various pressure scales commonly used in the high-pressure research community. Notably, this manuscript represents the first instance of an EOS internal consistency test above 300 GPa involving such a diverse range of materials.

While I support the publication of this manuscript in Communication Materials, I recommend considering the following improvements for its presentation:

1. In lines 121-122, it is mentioned that the V-V data are used to derive the internally consistent equations of state (EOSs), but without further details. Since this is the main focus of the paper, a more detailed description of the derivation process should be included.
2. When presenting the V-V relationships (lines 118-119), it is important to include a discussion on error analysis, including the overall uncertainty level in such relationships. Are there any outliers? How are errors determined and propagated from one material to the next when different pathways are used?
3. In lines 175-176, it is stated that there is a good agreement between the results from two different paths in Fig. 4. However, such information cannot be found in the figure or anywhere in the manuscript.
4. The sample loading process was only described for the t-DAC. What are the sample configurations for the low-pressure runs using the conventional DAC?
5. In Fig. S3, I suggest including XRD patterns at low pressures (at the starting points) for direct comparisons of stress conditions in the 300-400 GPa range.
6. Figure 1B in the main text is informative for illustrating the differences in the V-V relationship between the fitted curve and the measured volumes. I suggest plotting such differences in Fig. S4 for all other materials. For the V-V plots of W-Cu and W-Au, some measured volumes are far off the fitted curves, which requires an explanation.
7. Were the pressure gradients measured by 2D grids, or just line scanning across the cutlets? If the former, Fig. S6 may be replaced by those of 2D mapping.

Overall, this work represents a valuable contribution to the field, and addressing these points will enhance its impact in understanding pressure scales at extreme conditions.

Reviewer #3 (Remarks to the Author):

Please see the attached pdf file.

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Dear Dr Sakai,

Thank you for resubmitting your manuscript, "The equations of state for nine materials up to 0.43 TPa as internally consistent pressure standards for extreme pressure science", to Communications Materials. It has now been seen again by the 3 referees, whose comments are appended below. You will see that while they find your work much improved, some important points are still raised. We remain interested in the possibility of publishing your study in Communications Materials, but would like to consider your response to these concerns in the form of a revised manuscript before we make a decision on publication.

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In particular, the Data availability statement should include:

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- At a minimum, a statement confirming that all relevant data are available from the authors
- If applicable, a statement regarding data available with restrictions
- If a dataset has a Digital Object Identifier (DOI) as its unique identifier, we strongly encourage including this in the Reference list and citing the dataset in the Data Availability Statement.

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If a community resource is unavailable, data can be submitted to generalist repositories such as [figshare](https://figshare.com/) or [Dryad Digital Repository](http://datadryad.org/). Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

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Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Rostislav Hrubíak, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-1489-2156

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The manuscript has been significantly improved in the current version, in particular with the addition of Figure 1. It is now much easier to track which data and path have been used to calibrate each material discussed in the manuscript.

The errors I noticed in the form of the previous version have been corrected. Most of my remarks and questions have been addressed. My remaining comments (that need to be addressed and relevant corrections made in the manuscript) are the following:

1- One important point (point #11 in my first review), now discussed only in the caption of Fig. S6, deserves to be in the main text.

In their reply to referee, the authors write that: "The volume deviation from the V-V curve and/or each EoS is also found in t-DAC experiments at lower pressure conditions especially. This phenomenon was also discussed by Dewaele et al. (2018), and it is thought to be caused by the inhomogeneous stress distribution during the rapid pressure increase in "Stage II" of the t-DAC experiment. Although the data of Dewaele et al. (2018) was used for the EoS fitting of gold, the low-pressure data below 200 GPa was excluded as mentioned in the caption of supplementary Figure S6."

The authors have thus established a systematic way to select, gather and analyze various EoS data. Under moderate pressure (0<P<150 GPa), they use data collected under quasi-hydrostatic compression. Above 150-200 GPa, they use non-hydrostatic compression as the various datasets seem to converge (and above 250 GPa, only non-hydrostatic compression

data are available to date). This method has to be shared in the main text.

2-Similarly, raw data presented for epsilon-Fe in Fig. 3ab show that relative scatter of data collected in different studies under various pressurizing conditions decreases above ~150 GPa. This validates the strategy adopted by the authors but again this should be discussed.

3-It should be noted in the main text (for instance in the caption of Table 1) that no pressure medium is used in t-DAC experiments.

4-In Fig. S6, data collected at the end of Run #3 present a high scatter. Is it caused by the low Cu diffraction signal? Could the least constrained V-V points be removed from the dataset (or error bars indicated)?

5- I. 62 please remove "is a better technique". Ramp-compressed data also have issues such as non-hydrostatic compression bias.

6- Fig. 3 could you please comment in the manuscript on the deviation of Smith et al. data from static data below 150 GPa? Is it due to a lack of intermediate pressure data?

Remaining errors in the form:

5- IMPORTANT: Raw data are crucial for this publication. I have made a short survey of the material presented in supplementary figures: the data in Figures S5, S6, S7abc do not correspond to the pressure range in Table 1, please present relevant data.

6- please check the references in the legend of Fig. 3a, I am not sure that they are all correct?

7-In Fig. 5, please replace NaCl with NaCl-B2 in the legend and/or erase the pressure/volume region where NaCl-B2 phase is not observed. As such it is slightly misleading.

Reviewer #2 (Remarks to the Author):

The authors have reasonably addressed the reviewers' comments. The current revision is acceptable for publication. I have a few minor suggestions/comments related to clarification in presentation.

1. Lines 237-239: "Volume-Volume (V-V) relationship between copper and rhenium is shown in Figure 4. This V-V relationship is calculated using both Cu and Re EoS, as the volumes at the same pressure." I would suggest: "The experimental volume-volume (V-V) data of copper and rhenium are shown in Figure 4, together with a V-V relationship (solid line in Fig. 4) between copper and rhenium. This V-V relationship is obtained using EoS parameters of both Cu and Re in Table Sxx."

2. Line 271: Add units to lattice parameters

3. Line 297: Change "Compression curves of each material ..." to "Compression curves of 9 individual materials ..."

4. Line 344: Change "Pressure differences of ramp-compression scales compare to the copper scale" to "Pressure differences between the ramp-compression scale and the internally consistent EoS data based on the copper scale".

Reviewer #3 (Remarks to the Author):

The authors have answered all of my questions and addressed my concerns, I only have a few additional comments.

In supplementary figure S2 caption could the authors just clarify that these sample configurations are just for the toroidal anvil runs.

It would also help to reduce confusion if the authors state in the main text somewhere where supplementary figure S2 is introduced (line 118 of the revised manuscript) that the sample configurations shown in this figure are for the toroidal anvil runs similar to how it was described further down in the more detailed experimental method on line 488 of the revised manuscript.

My question about the tail of the X-ray beam was confusing. I was asking what the dimensions of the tail of the X-ray beam used in these runs was? I see that the FWHM are very small but what is the dimension of the tail of the beam?

There has been a recent paper on Mo that was published while you were revising this manuscript that would be worth looking at and likely referencing. They discuss why the uniaxial stress of Mo cannot be estimated and they also discuss why the 200 reflection is least influenced by uniaxial stress. So this new Mo EOS is probably worth comparing to in some way.

Zurkowski, C. C., Lim, R. E., Pardo, O. S., O'Bannon, E. F., Glazyrin, K., Söderlind, P., & Jenei, Z. (2024). Improving equations of state calibrations in the toroidal DAC—The case study of molybdenum. *Journal of Applied Physics*, 136(7).

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Dear Dr Sakai,

Your manuscript titled "The equations of state for nine materials up to 0.43 TPa as internally consistent pressure standards for extreme pressure science" has now been seen again by our referees, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials.

We therefore invite you to edit your manuscript to comply with our journal policies and formatting style in order to maximise the accessibility and therefore the impact of your work.

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Best regards,

Rostislav Hrubíak, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-1489-2156

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The paper is ready for publication under its current form.

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Reviewer #1 (Remarks to the Author):

This paper presents material that is important from several points of view: (i) ultra-high pressure metrology; (ii) behavior of elements and compounds under compression and comparison with models; (iii) static high pressure technologies; (iv) equations of state formalism. Raw data shown in supplementary material is of high quality, and data discussion is sound. The geometry of the toroidal diamond anvils is precisely described. I thus support publication in a journal with a wide readership such as Communication Materials.

However, I believe that there is room for improvement in the form of the manuscript. Below is a list of missing or incorrect information (there is a lot of material presented in this manuscript, I hope I did not miss anything):

- There is missing information on the preparation of the toroidal anvils: geometry of the starting anvil; FIB characteristics (current etc.); method for the measurement of the profile provided in Fig. S1b.

[Reply]

The information on the preparation of the t-DACs was added to the Supplementary Figure S1. The anvil profiles shown in Figure S1c are designed values, not measured ones. After FIB processing, we confirmed the height of the convex part by an interferometric measurement. The information of original anvil shape and FIB current was also added in the caption as follows.

[Fig. S1, caption]

These anvil profiles are designed values, not measured ones. The height of the convex part was measured by interferometric measurement after FIB processing. The measured height agrees with the design value within $\pm 0.1 \mu\text{m}$. Original anvil shapes were the culet-(bevel angle)-bevel size with 30-(8.5)-300 μm (Type Ia) for runs #1a,

#3, #4, 60-(10)-350 μm for run #2 (Type Ia), and 20-(9.5)-300 μm (Type IIas) for runs #5, #6. In run #5, the conical support type of anvil seat was used. Typical ion beam current during FIB processing was 7 nA.

- Lattice parameters are not provided for Fe and Re (hexagonal lattice) in tables S1, S2 and S6, and the volume alone does not allow to calculate diffraction angles as expected for a pressure calibrant

[Reply]

The values of a, c and c/a for Re and Fe were added to Tables S1, S2 and S6.

- I found missing references: Tange et al 2009, Zhang et al 2023, Rech+2019. There are mistakes in the references listed lines 120-121. The reference list is not provided in alphabetical order which makes it difficult to find them.

[Reply]

We apologize for the lack of information. All references were carefully checked and added in the revised version.

- Please provide the following information for runs 1b, 2b, 4b: loaded materials (sample and pressure calibrant)? Geometry?

[Reply]

The information about run #1b (and also #7abc) was added to the “Detailed experimental method” section as follows. And the results of previous compression studies with He pressure medium were used as

alternative data of run #2b and #4b. For details, please see the next section.

[Line 428- in the revised main text]

In run #1b, glycerin was used as a pressure medium. Cu and Re discs (thickness values were 12 μm for Cu and 6 μm for Re, diameter was 24 μm for both Cu and Re, respectively) were prepared and stacked by using FIB. Sample chamber diameter was 90 μm and thickness was ~ 50 μm . In run #7a, #7b and #7c, platinum foil (thickness ~ 5 μm) was embedded in magnesium oxide which act as a thermal insulator during the laser-annealing at about 1500~2000 K. Sample chamber diameter was 1/3 of culet, and thickness was 15~40 μm .

- Table 1: missing reference for NaCl-B2 data; which Fe-hcp data have been included in the fit? The present body of data is not sufficient to constrain low P behavior. Same remark for molybdenum.

[Reply]

In the revised manuscript, the calibration path and the experimental data used for each EoS fitting were summarized in Figure 1 and Table S8. To consider many previous studies, we supplemented the data under low pressure not only for NaCl but also for the other materials. For molybdenum, we additionally used the data from Dewaele et al. (2008) with using the IPPS-Ruby2020 scale, and from Dorfman et al. (2012) with MgO calibrant. For detail, please see Figure 1 and the related text in main body.

- Fig. 1: a part of the caption is missing

[Reply]

Fig. 1 of the original version moved to Fig. 4 in the revised version. The

symbols in (b) are the same as in (a).

- Fig. 2: indicate, for each material, the calibration path used to establish the Volume-Volume relation (and subsequent compression curve). I found relevant information in Table S9 but this would deserve to be in the main text to avoid any confusion.

[Reply]

Thank you for the comment. The calibration path was summarized in Figure 1 which was newly added in this revision, as an alternative to “Table S9” in the original version. We believe this figure will help readers to understand the calibration path. The experimental data used for each EoS fitting was also summarized in Table S8. We added the explanation in main text as follows.

[Line 116-127 in the revised main text]

Each EoS was calibrated according to the calibration path as shown in Figure 1. The copper scale²¹ was used as a primary scale. In addition to present results, the compression data of previous studies^{19,33,36,38,39-49} described in Figure 1 were also used in the EoS fitting (These are also summarized in supplementary Table S8). The results of experiments using the ruby fluorescence method^{33,34,41-44} were included as data under relatively low-pressure conditions. All pressures by Ruby-scale were recalculated based on IPPS-Ruby2020²⁴. Consistency between the copper scale and IPPS-Ruby2020 can be confirmed by the compression data of Dewaele et al.³³ as shown in supplementary Figure S5. As described above, the EoS of NaCl in B2 phase (NaCl-B2) was also calibrated using simultaneous compression data of Pt-NaCl^{36,39,38}, Au-NaCl³⁶ and MgO-NaCl³⁶. The compression data of the other combinations, W-Fe⁴⁵, Pt-MgO⁴⁶, NaCl-Fe^{47,48}, MgO-Au⁴⁹ by the conventional DAC, and Re-Au¹³ by t-DAC are also included.

- Fig. 2a: plotting raw data points on the graph would be useful as it would indicate in which range the volume-volume relation has been actually measured, and where it is extrapolated.

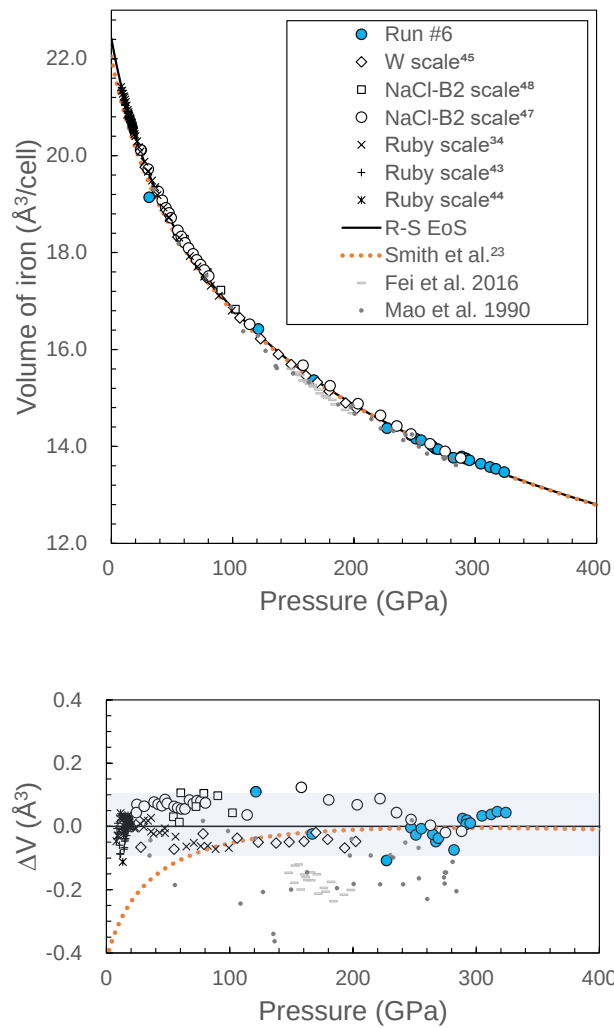
[Reply]

As it would be too complicated to include all the raw data, we added markers (open circles) indicating the minimum volume (maximum pressure) reached in the experiment (Figure 5a and 5b). The revised figure clarifies the pressure ranges of extrapolated EoS curves. Each EoS and V-V relationship with raw experimental data were shown in supplementary Figure S6 and S7.

- Fig. 4: conventional DAC data (Sakai et al. 2014) is plotted in this Figure. Is it also used to constraint the parameters listed in Table 2? Then why is Sakai et al. 2014 not mentioned in the caption of Table 2? It would be useful to plot other literature data such as Mao et al 90 (JGR 95 21737) and Dewaele et al 2006 (PRL 97 215504) – possibly others - so that the reader knows how the current compression curve differs from these classical measurements.

[Reply]

We added various previous experimental data as shown in Figure 1 and Table S8. Here the data of Dewaele et al. (2006) and Sakai et al. (2014) were included in the fitting of Fe EoS. In the case of Mao et al. (1990), the data (symbol “•”) deviates from the compression curves of this study and the reported experiments as shown in the following figure. The data of Fei et al. (2016) with MgO calibrant (symbol “-”) also showed systematically lower volume. Thus, these data were excluded in the EoS fitting.



- Fig. 5 and corresponding text: please provide an analytical definition for ΔP to avoid any confusion (I guess it is $P_{\text{metal}} - P_{\text{Cu}}$ assuming $\text{volume}_{\text{metal}} - \text{volume}_{\text{Cu}}$ relation in fig. 2a)

[Reply]

Thank you for the comments. Fig. 5 in the original version moved to Fig. 6 in the revised version. The definition of ΔP and the example of the calculation of ΔP were described as follows.

[Line 272- in the revised main text]

Here the pressure difference relative to the copper scale²¹ (ΔP) was calculated as follows. $\Delta P = P(V_{(P)}, Pt/Au/Fe \text{ EoS}) - P(V_{(P)}, Cu \text{ EoS})$, where $V_{(P)}$ is the volume at pressure of P which based on the copper scale, and $P(V_P)$ is the pressure at the volume of $V_{(P)}$ based on each EoS. For example, when the volume of Cu is $V/V_0=0.600$, $V/V_0=0.694$ is given for the relative volume of Pt according to the V-V relationship of present result (see Table S9). In this case, the pressure is 250 GPa by the Cu R-S EoS, whereas the Pt scale of ref.22 with $V/V_0=0.694$ yields 262 GPa, and ΔP becomes 12 GPa. Note that if you use the Pt R-S EoS, it gives the same pressure value as the copper scale because the V-V relationship is derived from the R-S EoS itself.

- Fig. S4, a, b and c exhibit differences between experimental runs. Can you comment? Please provide the forms for the Volume-volume relationships plotted as dashed lines. Could you plot the data from Dewaele et al. PRL 97 215504 2006 on the Fe-W graph?

[Reply]

Fig. S4 (V-V relationship of each run) moved to Fig. S7 in the revised version. As described above, we supplemented the data under low pressure by including the various published data. As you mentioned, the data of run #2b and #4b shows that the values systematically deviate from the data under good hydrostatic conditions realized by He pressure medium (Dewaele et al. 2004), so these values were excluded. Actually, the EoS fitting result was not affected so much by these data because V_0 and K_0 were fixed in most cases.

The data from Dewaele et al. (2006) were also plotted in the Fe-W V-V graph (see Figure S7i and S7j), and it is in good agreement with the present result.

The volume deviation from the V-V curve and/or each EoS is also found in t-DAC experiments at lower pressure conditions especially. This phenomenon was also discussed by Dewaele et al. (2018), and it is thought to be caused by the inhomogeneous stress distribution during

the rapid pressure increase in “Stage II” of the t-DAC experiment. Although the data of Dewaele et al. (2018) was used for the EoS fitting of gold, the low-pressure data below 200 GPa was excluded as mentioned in the caption of supplementary Figure S6.

[Caption of supplementary Figure S6]

In (e) and (f), because the low-pressure data below 200 GPa of Dewaele et al.¹³ shows large deviation from the EoS which is thought to be caused by the inhomogeneous stress distribution during the rapid pressure increase in “Stage II” of the t-DAC experiment, these data (crosses) was excluded for the Au R-S EoS fitting.

- Fig. S7: I don't get this graph. How can the 2 MgO pressures taken from Tange et al 2009 differ that much?

[Reply]

It was just the result of Tange et al. (2009), and it thought to be due to the lack of the experimental data at high pressure conditions. In the present result, the pressure difference between 3BM and Rydberg-Vinet (R-V) EoS decreased significantly as shown in the supplementary Figure S9.

I have further suggestions to improve the discussion part:

- The authors could consider re-fitting their NaCl-B2 EoS, the set of parameters they obtain (Table 2) differ significantly from what is found in the literature for other B2 alkali halides phases, where K'_0 is around 6.

[Reply]

As described above, the results of various previous studies were included in the revised version, and we obtained K'_0 of about 4.6. In the

previous study, for example Fei et al. (2007), they reported K'_0 value of 4.33 and 5.25 for 3BM and R-V EoS, respectively. As mentioned in main text, 3BM and R-V EoS correspond to the K'_∞ value of 3 and 2/3, respectively. In this study, we adopted the R-S EoS with the K'_∞ value of 5/3. Considering the tradeoff relationship between the EoS parameters, the present result seems to be reasonable and is not significantly different from previous works.

- I found the first paragraph of the “Discussion” part rather confusing. It needs to be re-written and clear conclusions should be highlighted. The authors could provide a calibration of the diamond Raman scale based on the present work. Also, it seems to me that they show that platinum is not a reliable pressure calibrant, could they give an advice?

[Reply]

The first paragraph of the “Discussion” in the original manuscript was about the diamond-Raman scale. As a result of including previous works in the EoS fitting, the pressure difference between the copper and gold scales significantly decreased as shown in Figure 6 of the revised version. We constructed the diamond-Raman scale calibrated with the Au R-S EoS, and the results were very consistent with those of Eremets et al. (2023). The conclusion of present result is that the diamond-Raman gauge proposed by Eremets et al. (2023) is supported by the copper scale. And we moved the argument of the phase diagram of hydrogen to discussion part from conclusion part as follows.

[Line 331- in the revised main text]

Thus, the phase diagram of hydrogen proposed by Eremets et al.²⁵ does not change so much even using the copper scale, because the R-S EoS for gold agree with the gold scale proposed by the ramp compression experiment²² within a few GPa as shown in Figure 6.

- The introduction of generalized Rydberg-Stacey Equation of state is a very interesting point of this work, as well as the suggestion to constraint its K'_{inf} Parameter with ab initio calculations. The authors find that a compression to 700 GPa is necessary to obtain a reliable K'_{inf} for Re (l. 255-270). How would the K'_{inf} Value converge for a more compressible material such as NaCl or KCl? A rare gas solid?

[Reply]

Thank you for your comment. We are also very interested in this point, and we hope to clarify it in future research. Another interesting point of view is that the relationship between K'_{∞} and γ_{∞} (the Grüneisen parameter at infinite pressure) proposed as Slater's equation, $\gamma=K'/2-1/6$. The relationship between thermal properties and compression behavior at extreme conditions should be investigated in the next step.

Reviewer #2 (Remarks to the Author):

This manuscript describes the experimental results of unit cell measurements for a set of materials at high pressures up to 430 GPa. These experiments are challenging, and the authors have successfully used very small culet anvils to reach pressures exceeding 300 GPa in 7 runs. Additionally, the use of FIB (focused ion beam) for sample loading is a commendable approach for arranging the involved samples as desired.

The measured volume data serve the purpose of testing the internal consistency of the equations of state (EOSs) across nine different materials. This consistency is crucial for understanding the various pressure scales commonly used in the high-pressure research community. Notably, this manuscript represents the first instance of an EOS internal consistency test above 300 GPa involving such a diverse range of materials.

While I support the publication of this manuscript in Communication Materials, I recommend considering the following improvements for its presentation:

- 1. In lines 121-122, it is mentioned that the V-V data are used to derive the internally consistent equations of state (EOSs), but without further details. Since this is the main focus of the paper, a more detailed description of the derivation process should be included.*

[Reply]

In the revised manuscript, the calibration path and the experimental data used for each EoS fitting were summarized in Figure 1 and Table S8. The derivation process is as follows. 1. Measure the volume-volume data by the static compression experiment. 2. Calibrate the equation of state using the primary scale. 3. The V-V relationship is calculated using both primary and secondary EoS as the volumes at the same pressure. 4. By the same procedure, the internally consistent EoSs provide us the V-V relationship between these materials. The derivation process of the V-V relationship was described in the main text as follows.

[Line 213- in the revised main text]

This V-V relationship is calculated using both Cu and Re EoS, as the volumes at the same pressure.

2. When presenting the V-V relationships (lines 118-119), it is important to include a discussion on error analysis, including the overall uncertainty level in such relationships. Are there any outliers? How are errors determined and propagated from one material to the next when different pathways are used?

[Reply]

Thank you for pointing that out. The pressure error from EoS parameters and the actual fluctuation of the experimental data were mentioned as follows:

[Line 284-, in the revised main text]

Although the error propagated from present EoS parameter's error (the error of K'_0) is a few GPa under extreme pressure condition, the fluctuation of the actual measured volume values with respect to the EoS curve corresponds to the pressure difference of 5~10 GPa (~2% in pressure) as shown in Figure 2b, 3b.

The problem is the evaluation of the error propagation through the 2nd, 3rd ... calibration shown in Figure 1. One method is to compare data from different calibration passes, which is discussed in the Fe EoS section. The other method is to compare the present result and the V-V data obtained directly by simultaneous compression. A good example of the simultaneous compression data of gold and platinum was recently reported by Zurkowski et al. (2024). The validity of our V-V relationship is supported by the consistency with their data within a few % in pressure at extreme conditions. Another example is the Mo-Pt reported by Akahama et al. (2014). Their result shows good agreement with present V-V relation of Mo-Pt. About these points, following sentences were added:

[Line 239-, in the revised main text]

It is important to evaluate the error propagation through the 2nd, 3rd ... calibration shown in Figure 1. One method is to compare data from different calibration passes, which is discussed in the Fe EoS section. The other method is to compare the present result and the V-V data obtained directly by simultaneous compression. One example is the simultaneous compression result of molybdenum and platinum⁶⁰. Akahama et al.⁶⁰ reported the unit cell parameters of $a_{\text{(Mo)}}=2.6530(12)$ and $a_{\text{(Pt)}}=3.3777(14)$ at highest pressure which originally reported as 410 GPa. According to present R-S EoSs, these values correspond to 375 ± 2 GPa and 377 ± 2 GPa, respectively, they are consistent within the error, although the pressure dropped in the present case of the copper scale calibration.

From the present V-V table, we can obtain the V-V relationship between any two materials. Especially, the combination of gold and platinum is important because these have been used as pressure reference materials in many studies to date. In this study, the EoS for gold and platinum were calibrated through independent calibration paths, but the V-V relationship between gold and platinum can also be drawn. The validity of this relationship is supported by recent direct volume simultaneous measurement results⁶¹ as shown in the supplementary Figure S8ab. The volume differences of gold from the V-V curve were about $-0.1 \text{ \AA}^3/\text{cell}$ at 200-300 GPa (Figure S8b). It corresponds to the pressure difference of about 4-5 GPa.

3. In lines 175-176, it is stated that there is a good agreement between the results from two different paths in Fig. 4. However, such information cannot be found in the figure or anywhere in the manuscript.

[Reply]

As mentioned above, the calibration path and the experimental data used for each EoS fitting were summarized in Figure 1 and Table S8. We believe this figure will help readers to understand the calibration path.

4. The sample loading process was only described for the t-DAC. What are the sample

configurations for the low-pressure runs using the conventional DAC?

[Reply]

Sample configurations of run #1b and #7abc were described in “Detailed experimental method” section as follows:

[Line 428-, in the revised main text]

In run #1b, glycerin was used as a pressure medium. Cu and Re discs (thickness values were 12 μm for Cu and 6 μm for Re, diameter was 24 μm for both Cu and Re, respectively) were prepared and stacked by using FIB. Sample chamber diameter was 90 μm and thickness was ~ 50 μm . In run #7a, #7b and #7c, platinum foil (thickness ~ 5 μm) was embedded in magnesium oxide which act as a thermal insulator during the laser-annealing at about 1500~2000 K. Sample chamber diameter was 1/3 of culet, and thickness was 15~40 μm .

5. In Fig. S3, I suggest including XRD patterns at low pressures (at the starting points) for direct comparisons of stress conditions in the 300-400 GPa range.

[Reply]

“Representative XRD profiles of each run at low pressure.” were shown in supplementary Figure S3. XRD profiles at high pressure were shown in supplementary Figure S4, and the parasitic scattering peak was removed in Figure S4d for run #4 at 368 GPa.

6. Figure 1B in the main text is informative for illustrating the differences in the V-V relationship between the fitted curve and the measured volumes. I suggest plotting such differences in Fig. S4 for all other materials. For the V-V plots of W-Cu and W-Au, some measured volumes are far off the fitted curves, which requires an explanation.

[Reply]

All EoSs and V-V relationships with the volume differences were shown in supplementary Figure S6 and S7.

7. Were the pressure gradients measured by 2D grids, or just line scanning across the cutlets? If the former, Fig. S6 may be replaced by those of 2D mapping.

[Reply]

“Pressure distribution in t-DAC experiments, run #1-#6” were shown in supplementary Figure S13. There is no 2D mapping data.

Overall, this work represents a valuable contribution to the field, and addressing these points will enhance its impact in understanding pressure scales at extreme conditions.

[Reply]

Thank you very much for your constructive feedback.

Reviewer #3 (Remarks to the Author):

This is a nice contribution to establishing internally consistent pressure scales for use in high pressure DAC work, specifically by calibrating EOS's over larger pressure range than has previously been done. In this study the authors have compressed nine materials between 310 and 440 GPa which is at least 50 GPa higher than previous studies that compared several materials and reported new EOS parameters. Below are some comments on the paper.

My main issues with the paper are (1) the title is misleading the authors did not report any experimental runs that reached 500 GPa this title needs to be changed, (2) the introduction should focus more on previous static compression studies where multiple materials were studied and EOS parameters are reported (3) was NaCl actually compressed in this study? Or were these data taken from Sakai et al. 2011 Equation of state of NaCl-B2 phase up to 304 GPa? If they were taken from another study perhaps this should be only eight materials and (4) equations of state that are derived from non-hydrostatic diamond anvil cell experiments are known to be unreliable, and the burden of proof of the reliability of these new reported EOS's lies with the authors.

Title

The title needs to be changed the authors state 0.5 TPa in the title and this is misleading since the maximum pressure reported is only 0.43 TPa (abstract) 0.44 TPa (table 1 run #1) really the only thing that can be truly said is that all of the runs exceeded 300 GPa (this is stated by the authors in the first sentence of the results section) which is not higher than the pressure at the center of the Earth.

[Reply]

The title was changed as follows.

“The equations of state for nine materials up to 0.43 TPa as internally consistent pressure standards for extreme pressure science”

Introduction

This paper focuses on establishing a new internally consistent pressure scales for 9 different materials, but the introduction focuses on a discussion about ultrahigh static pressure technique development and science cases for ultrahigh static pressure. I think that there should be a larger focus on previous pressure scale calibration a few studies come to mind Dorfman et al. (2000) Intercomparison of pressure standards (Au, Pt, Mo, MgO, NaCl and Ne) to 2.5 Mbar which is referenced by the authors, Dewaele et al. (2004) Equations of state of six metals above 94 GPa which is referenced in table 2 but does not appear in the reference list, and also Fei et al. (2007) Toward an internally consistent pressure scale (looking at Au, Pt, NaCl B-2, and Ne) this is not referenced by the authors.

Overall, I think the introduction needs to focus more on the previous published DAC work where multiple materials were investigated which is the same that was done in this work.

[Reply]

We added the following sentences in the introduction section.

[Line 67-, in the revised main text]

Many primary pressure scales for various materials have been proposed based on shock experiments^{e.g., 26-28}, ultrasonic and/or Brillouin scattering measurements²⁹⁻³¹, and *ab initio* calculations³². On the other hand, mutual consistency of these scales of different materials can be confirmed by the static compression experiment. Simultaneous static compression experiment of two or three materials combining with the X-ray diffraction (XRD) measurements give us an information on the volume-volume (V-V) (or density-density (ρ - ρ)) relationship of the sample. The V-V data enable us to check the consistency of each EoS, and to calibrate a new EoS for a given primary scale. At pressures up to 100~250 GPa, many efforts have been made to construct an internally consistent scale through simultaneous compression for metals, oxides, alkali-halides, and noble gas³³⁻³⁶. However, even these internally consistent equations of state become inconsistent when extrapolated beyond the pressure range of the experiment. Even for a single material, for example magnesium oxide (MgO), large discrepancies can arise in extrapolated pressure regions (>300 GPa) due to the lack of experimental data, depending on the type of equation of state

chosen³⁷.

Brief experimental method

The authors state “In run #7a-c, the platinum...” do the authors mean that there were actually three #7 runs? Meaning did they compress MgO and Pt in three separate runs and they are calling them runs 7a, 7b, and 7c? if this is the case table 1 needs to be changed to reflect this. The authors should also add which ones were toroidal and which ones were beveled and/or flat anvils. I also see in Figure1a that run #1a and run #1b are plotted and run #1b stops at ~0.75 V/V0. In the supplement it looks like run 2 and run 4 also had additional runs. Since these are used in the study for fitting EOS’s the specifics of these runs need to be listed in either table 1 or in a table in the supplement.

[Reply]

I apologize for the confusion caused by my inadequate description. Each run’s setup was summarized individually in the revised Table 1. And we rewrite “Brief experimental method” section as follows:

[Line 95-, in the revised main text]

Static extreme high-pressure experiments were conducted by using t-DAC for run #1a-#6, conventional DAC for run #1b, and the LHDAC for run #7a, #7b, and #7c. Culet size and sample combination of each run is summarized in Table 1.

Details of the sample loading for runs #4-6 should be clarified, what does similar to run #3 mean? Does it mean that W is always the thinner disk on the bottom and that Au, Mo, and Fe just replace the Cu? What was the sample configuration for run #7a-c? embedded in MgO so the Pt was surrounded by MgO so that it was not in direct contact with the diamonds? Anything else in the sample chamber for runs #7a-c?

The authors discuss laser annealing of the Pt sample, specifically run #7a-c, was laser annealing performed for any of the other samples? I see a nice uniaxial stress analysis for

run #2 and #7 but was this analysis performed for any of the other runs?

[Reply]

All of the sample configurations of t-DAC experiments were shown in supplementary Figure 2a-l with the SEM and the schematic images. About the conventional DAC experiments, the following sentences were added in “Detailed experimental method” section as follows:

[Line 428-, in the revised main text]

In run #1b, glycerin was used as a pressure medium. Cu and Re discs (thickness values were 12 μm for Cu and 6 μm for Re, diameter was 24 μm for both Cu and Re, respectively) were prepared and stacked by using FIB. Sample chamber diameter was 90 μm and thickness was ~ 50 μm . In run #7a, #7b and #7c, platinum foil (thickness ~ 5 μm) was embedded in magnesium oxide which act as a thermal insulator during the laser-annealing at about 1500~2000 K. Sample chamber diameter was 1/3 of culet, and thickness was 15~40 μm .

Table 1

More runs are discussed in the text than shown in the table, which runs were toroidal anvils and which were conventional beveled anvils? The details of the run on NaCl are not listed in this table?

[Reply]

Each run's setup was summarized individually in the revised Table 1. For the NaCl EoS, we used the previous compression result of our group and others. In the revised manuscript, the calibration path and the experimental data used for each EoS fitting were summarized in Figure 1 and Table S8.

Table 2

For the references to the values for K_0 the authors must cite the primary source for these values, for example for Pt they cite Dewaele et al. (2004) but that paper the value is from Ref 24: G. Simmons and H. Wang, *Single Crystal Elastic Constants and Calculated Aggregate Properties: A Handbook* (MIT Press, Cambridge, MA, 1971). For Cu, Fratanduono et al. (2020) fit the value of K_0 but there is a well-known and accepted acoustic value of 133 GPa for Cu, references are: W. Holzapfel, M. Hartwig, and W. Sievers, *J. Phys. Chem. Ref. Data* **30**, 515 (2001) & W. Daniels and C. Smith, *Phys. Rev.* **111**, 713 (1958).

My main point is that generally K_0 is fixed to a value that has been determined from acoustic measurements, this is what was done by Dewaele et al. (2004) and Dorfman et al. (2012). Additionally, please cite primary references for any value used in these fits.

[Reply]

Thank you for your accurate comment. The reference for each parameter was listed in Table 2. Following references were added.

- [53] Holzapfel, W., Hartwig, M. & Sievers, W. Equations of state for Cu, Ag, and Au for wide ranges in temperature and pressure up to 500 GPa and above. *J. Phys. Chem. Ref. Data* **30**, 515 (2001).
- [54] Daniels, W. & Smith, C. Pressure derivatives of the elastic constants of copper, silver, and gold to 10 000 bars. *Phys. Rev.* **111**, 713 (1958).
- [55] Kamada, S., et al. Elastic constants of single-crystal Pt measured up to 20 GPa based on inelastic X-ray scattering: Implication for the establishment of an equation of state. *C. R. Geoscience* **351**, 236–242 (2019).
- [56] Steinberg, D. Observations regarding the pressure dependence of the bulk modulus. *J. Phys. Chem. Solids* **43**, 1173 (1982).
- [57] Simmons, G. & Wang, H. *Single Crystal Elastic Constants and Calculated Aggregate Properties. A Handbook* MIT Press, Cambridge, MA (1971).
- [58] Katahara, K. W., Manghnani, M. H. & Fisher, E. S. Pressure derivatives of the elastic moduli of BCC Ti-V-Cr, Nb-Mo and Ta-W alloys. *J. Phys. F: Metal Phys.* **9**, 773-790 (1979).

Diamond anvil and gasket

More details of the toroidal anvils are needed, like milling time, current, what type diamonds were used, for the toroidal anvils did the authors start with beveled or flat anvils what were the dimensions of the starting anvils.

Were conventional 100-300 um anvils gas loaded? What was their sample configuration? A table with these details should be included in the supplement along with the maximum pressure achieved in these runs.

[Reply]

The information of t-DAC anvil was summarized in supplementary Figure S1. Here, the information of the original anvil shapes and FIB beam current etc. were added in caption.

About the conventional DAC experiments, the explanations were added in “Detailed experimental method” section as described above.

XRD and lattice volume measurements

What was the tail of the X-ray beam?

You mention using the 111 reflection for fcc metals which is reasonable (however a uniaxial stress analysis should still be presented). For the bcc metals W and Mo you used 3-5 peaks what is the spread in volume found from these different peaks? For Mo it has been reported that the 200 is least affected by non-hydrostatic compression (Akahama et al. 2014 Journal of Applied Physics, 116), I am not sure this has been found for W, but a uniaxial stress analysis could reveal if this is also the case for W.

[Reply]

Here “Tail” means the X-ray intensity outside of the FWHM in the intensity profile. In the case of t-DAC and ds-DAC experiments, the X-ray diffraction from the outside of culet was often measured

simultaneously even adjusting X-ray position to the center of culet, as shown in supplementary Figure S4a. The discussion about this point can be found in Yagi et al. (2020, HPR) “3.2. Anvil culet and X-ray beam size” section.

Thank you for the information about molybdenum. In the revised manuscript, we used 200 reflection of Mo for the determination of the unit cell volume. On the other hand, molybdenum and tungsten are not suitable for the estimation of uniaxial stress. Akahama et al. (2014) did not determine the uniaxial stress value also. We added the following sentences in “Detailed experimental method” section.

[Line 465-, in the revised main text]

The S values for molybdenum and tungsten can be estimated by using the elastic stiffness constant C_{ij} ⁵⁸ with using Birch extrapolation formulation³⁶, and these values are quite small above 100 GPa (10^{-4} ~ 10^{-5}). Thus, the volumes of molybdenum and tungsten are not sensitive to the non-hydrostatic effect, and in other words, not suitable for the estimation of the uniaxial stress t .

Conclusions

I think the Authors are on the right track for the last paragraph where they discuss an outlook for toroidal anvils (yes only one study Dewaele et al. 2018 was ~600 GPa achieved with toroidal anvils but all other runs in that study achieved much lower in pressure), I would like to emphasize two points (1) while the conventional beveled anvil can reach pressures of 400 GPa the review paper by O'Bannon et al. (2018) shows that this is not routine and in fact beveled anvils seem to more routinely reach pressures of ~250 GPa, and (2) I think the big advantage of the toroidal anvils is they can more routinely achieve pressures of ~400 GPa. Therefore, what toroidal anvils provide is not a significantly higher pressure range but the ability to achieve ~400 GPa routinely which is commonly quoted as the pressure limit for single crystal diamond anvils in 110 or 100 orientation.

[Reply]

Thank you for the comment. We added the second point in “conclusion” section as follows:

[Line 412-, in the revised main text]

The advantage of t-DAC is that it can routinely generate ~400 GPa exceeding the Earth's central pressure and the limit of the conventional DAC.

Stress analysis and pressure distribution

This is a very important part of this work as it is the proof that these non-hydrostatic data are reliable, was a uniaxial stress analysis performed for runs other than #2 and #7? If not I strongly suggest it is done and included in the supplement.

[Reply]

We added the analysis of the uniaxial stress of Pt and Au. In the case of Au in run #4, the elastic anisotropy parameter S was estimated from estimated from the elastic stiffness constant C_{ij} reported by Tsuchiya and Kawamura (2002) with using Birch extrapolation formulation (Dorfman et al. 2012). the uniaxial stress at highest pressure (368 GPa) was $t=4.5$ GPa. The results were summarized in supplementary Figure S12. And, as mentioned above, molybdenum and tungsten are not suitable for the estimation of uniaxial stress.

Reviewer #1 (Remarks to the Author):

The manuscript has been significantly improved in the current version, in particular with the addition of Figure 1. It is now much easier to track which data and path have been used to calibrate each material discussed in the manuscript.

The errors I noticed in the form of the previous version have been corrected. Most of my remarks and questions have been addressed. My remaining comments (that need to be addressed and relevant corrections made in the manuscript) are the following:

1- One important point (point #11 in my first review), now discussed only in the caption of Fig. S6, deserves to be in the main text.

In their reply to referee, the authors write that: “The volume deviation from the V-V curve and/or each EoS is also found in t-DAC experiments at lower pressure conditions especially. This phenomenon was also discussed by Dewaele et al. (2018), and it is thought to be caused by the inhomogeneous stress distribution during the rapid pressure increase in “Stage II” of the t-DAC experiment. Although the data of Dewaele et al. (2018) was used for the EoS fitting of gold, the low-pressure data below 200 GPa was excluded as mentioned in the caption of supplementary Figure S6.”

The authors have thus established a systematic way to select, gather and analyze various EoS data. Under moderate pressure ($0 < P < 150$ GPa), they use data collected under quasi-hydrostatic compression. Above 150-200 GPa, they use non-hydrostatic compression as the various datasets seem to converge (and above 250 GPa, only non-hydrostatic compression data are available to date). This method has to be shared in the main text.

[Reply]

Thank you for pointing that out. We added the following sentences in the main text. The t-DAC experiments' data points in present study (Run #1a, #2, #3, #4, #5, #6) at relatively lower pressure were included in EoS fittings but the EoS fitting result was not affected so much by these data because V_0 and K_0 were fixed in most cases, and due to the lower data density relative to the data set from previous studies. Thus, we have left these data as “fact-based results” of the t-DAC experiment.

[Line 240~]

The volume deviation from the V-V curve and/or each EoS is also found in t-DAC experiments at lower pressure conditions especially. This phenomenon was also discussed by Dewaele et al.¹³, and it is thought to be caused by the inhomogeneous stress distribution during the rapid pressure increase in “Stage II” of the t-DAC experiment. Although the data of Dewaele et al.¹³ was used for the EoS fitting of gold, the low-pressure data below 200 GPa was excluded as mentioned in the caption of supplementary Figure S6. The t-DAC experiments’ data points in present study (Run #1a, #2, #3, #4, #5, #6) at relatively lower pressure were included in EoS fittings but the EoS fitting result was not affected so much by these data because V_0 and K_0 were fixed in most cases, and due to the lower data density relative to the data set from previous studies.

2-Similarly, raw data presented for epsilon-Fe in Fig. 3ab show that relative scatter of data collected in different studies under various pressurizing conditions decreases above ~150 GPa. This validates the strategy adopted by the authors but again this should be discussed.

[Reply]

The following sentence was also added to the above.

[Line 249~]

On the other hand, at relatively higher pressure, the relative scatter of data collected in different studies under various pressurizing conditions decreases above 200 GPa in the case of Fe (Fig. 3a,b). It shows that the stress distribution in the t-DAC is getting relatively homogeneous in “Stage III”¹³ of the t-DAC experiment.

3-It should be noted in the main text (for instance in the caption of Table 1) that no pressure medium is used in t-DAC experiments.

[Reply]

The following sentence was added in the main text / caption of Table 1.

[Line 101~]

No pressure medium was used in t-DAC experiments.

4-In Fig. S6, data collected at the end of Run #3 present a high scatter. Is it caused by the low Cu diffraction signal? Could the least constrained V-V points be removed from the dataset (or error bars indicated)?

[Reply]

There is no significant difference between the data at the end of Run #3 (especially, 3 data of $\Delta V=0.29\sim0.37$ Å³/cell at 335~356 GPa) and the others in the point of view of the Cu-111 diffraction peak intensity. As you pointed out, the diffraction line intensity of Cu was very weak, and no peaks except the 111 diffraction could be observed. Therefore, it is not possible to evaluate the error in the Cu lattice constant for each data point. Thus, we cannot eliminate only this data. Securing a larger sample volume in t-DAC experiments is one of the issues we need to address in the future.

5- l. 62 please remove “is a better technique”. Ramp-compressed data also have issues such as non-hydrostatic compression bias.

[Reply]

We modified the sentence as follows.

[Line 62~]

... is one of the techniques ...

6- Fig. 3 could you please comment in the manuscript on the deviation of Smith et al. data from static data below 150 GPa? Is it due to a lack of intermediate pressure data?

[Reply]

The main reason for the difference at lower pressure is due to the difference in zero pressure volume. Smith et al. used the result of ref.21

in their paper. On the other hand, we determined the zero-pressure volume by EoS fitting. Our EoS fitting result reasonably reproduces the static compression data with He pressure medium and ruby scale.

Remaining errors in the form:

5- IMPORTANT: Raw data are crucial for this publication. I have made a short survey of the material presented in supplementary figures: the data in Figures S5, S6, S7abc do not correspond to the pressure range in Table 1, please present relevant data.

[Reply]

Source data for Figure S5 was provided in Table S14. “Pressure range” and the values in Table 1 were replaced by “Max P.”, as follows. The minimum measured volumes are also shown below. These volumes can be found in Table S1~S7.

Table 1 | Experimental setting.

Run	Sample	Type	Max. P (GPa)	culet (μm)
#1a	Cu + Re	t-DAC	431	20
#1b	Cu + Re	DAC	88	300
#2	Pt + Re	t-DAC	394	20
#3	Cu + W	t-DAC	356	16
#4	Au + W	t-DAC	368	16
#5	Mo + W	t-DAC	395	20
#6	Fe + W	t-DAC	324	20
#7a	MgO + Pt	LHDAC	314	40
#7b	MgO + Pt	LHDAC	144	100
#7c	MgO + Pt	LHDAC	55	300

[Scale and volumes]

Run	Table 1	Revised	Unit cell volume ($\text{\AA}^3/\text{cell}$)
Run #1a :	~430	431 (Cu)	V(Cu)=24.907
Run #1b :	~88	88 (Cu)	V(Cu)=34.87
Run #2 :	~390	394 (Re)	V(Re)=19.06
Run #3 :	~340	356 (Cu)	V(Cu)=26.09
Run #4 :	~370	368 (W)	V(W)=20.02
Run #5 :	~400	395 (W)	V(W)=19.69
Run #6 :	~320	324 (W)	V(W)=20.614
Run #7a :	~310	314 (Pt)	V(Pt)=40.05
Run #7b :	~144	144 (Pt)	V(Pt)=46.22
Run #7c :	~55	55 (Pt)	V(Pt)=52.513

6- please check the references in the legend of Fig. 3a, I am not sure that they are all correct?

[Reply]

Thank you for checking the details. Reference #43 was incorrect. It was corrected as follows.

[incorrect]

[43] Dewaele, A. et al. High pressure-temperature phase diagram and equation of state of titanium. *Phys. Rev. B* **91**, 134108 (2015).

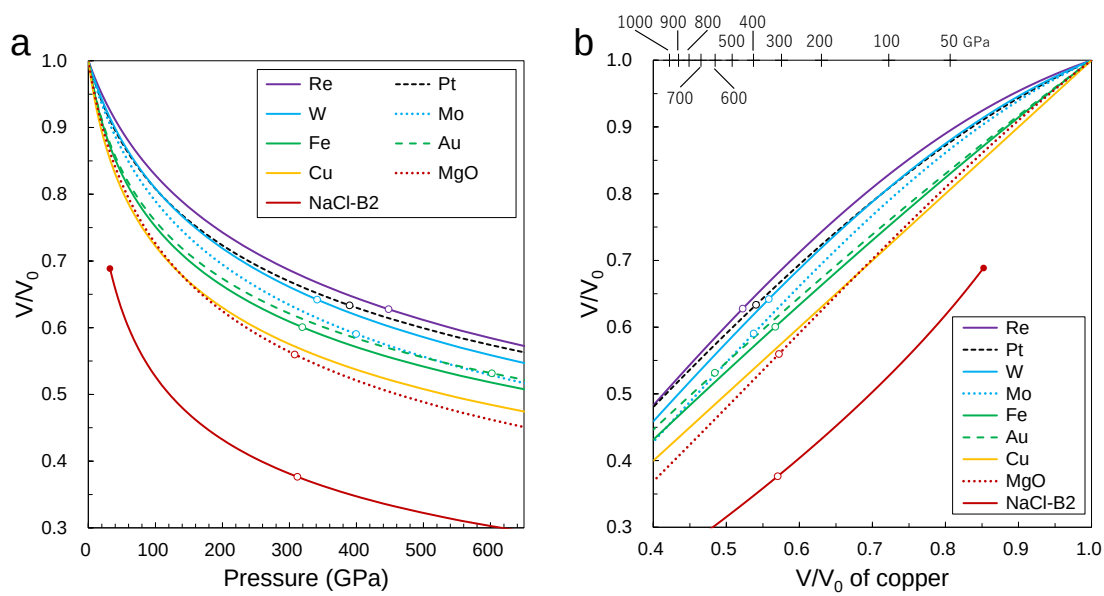
[revised]

[43] Dewaele, A. et al. Mechanism of the α - ϵ phase transformation in iron. *Phys. Rev. B* **91**, 174105 (2015).

7-In Fig. 5, please replace NaCl with NaCl-B2 in the legend and/or erase the pressure/volume region where NaCl-B2 phase is not observed. As such it is slightly misleading.

[Reply]

Figure 5a and 5b were replaced as follows.



Reviewer #2 (Remarks to the Author):

The authors have reasonably addressed the reviewers' comments. The current revision is acceptable for publication.

I have a few minor suggestions/comments related to clarification in presentation.

1. Lines 237-239: "Volume-Volume (V-V) relationship between copper and rhenium is shown in Figure 4. This V-V relationship is calculated using both Cu and Re EoS, as the volumes at the same pressure." I would suggest: "The experimental volume-volume (V-V) data of copper and rhenium are shown in Figure 4, together with a V-V relationship (solid line in Fig. 4) between copper and rhenium. This V-V relationship is obtained using EoS parameters of both Cu and Re in Table Sxx."

[Reply]

Thank you for your suggestion. The revised expression is more accurate. I have rewritten the main text as you suggested.

[Line 211~]

The experimental volume-volume (V-V) data of copper and rhenium are shown in Figure 4, together with a V-V relationship (solid line in Figure 4) between copper and rhenium. This V-V relationship is obtained using EoS parameters of both Cu and Re in Table 2.

2. Line 271: Add units to lattice parameters

[Reply]

Line 259, "Å" added.

3. Line 297: Change "Compression curves of each material ..." to "Compression curves of 9 individual materials ..."

[Reply]

Line 282~, Fig. 5 caption, changed

4. Line 344: Change *"Pressure differences of ramp-compression scales compare to the copper scale"* to *"Pressure differences between the ramp-compression scale and the internally consistent EoS data based on the copper scale"*.

[Reply]

Line 326~, Fig. 6, title was changed.

Reviewer #3 (Remarks to the Author):

The authors have answered all of my questions and addressed my concerns, I only have a few additional comments.

In supplementary figure S2 caption could the authors just clarify that these sample configurations are just for the toroidal anvil runs.

[Reply]

The following sentence was added to Figure S2 caption.

These sample configurations were just for the t-DAC runs.

It would also help to reduce confusion if the authors state in the main text somewhere where supplementary figure S2 is introduced (line 118 of the revised manuscript) that the sample configurations shown in this figure are for the toroidal anvil runs similar to how it was described further down in the more detailed experimental method on line 488 of the revised manuscript.

[Reply]

The sentences were modified as follows.

[Line 98~]

In all t-DAC experiments (run #1a-#6), metal samples (reagent grade, 99.9~99.99%) in disc shape were made by a focused ion beam (FIB) system and stacked each other, and then filled in the sample chamber made on gasket as shown in supplementary Figure S2a-l.

[Line 457~]

Sample configurations for all t-DAC experiments (run #1a-#6) are summarized in supplementary Figure S2a-l.

My question about the tail of the X-ray beam was confusing. I was asking what the dimensions of the tail of the X-ray beam used in these runs was? I see that the FWHM are

very small but what is the dimension of the tail of the beam?

[Reply]

Intensity profile of the X-ray beam at SPring-8 BL10XU can be found in Figure 4 in Hirao et al. (2020). It is difficult to determine the precise intensity profile corresponding to 1~2 % of the peak intensity, thus it is also difficult to say the tail size of the beam. The discussion about the tail has been also mentioned in Sakai et al. (2015, RSI) and Yagi et al. (2020, HPR). These relate to ds-DACs, but the same can probably be said for t-DACs.

**(Ref.) Hirao et al. (2020), Matter Radiat. Extremes 5, 018403
<https://doi.org/10.1063/1.5126038>**

There has been a recent paper on Mo that was published while you were revising this manuscript that would be worth looking at and likely referencing. They discuss why the uniaxial stress of Mo cannot be estimated and they also discuss why the 200 reflection is least influenced by uniaxial stress. So this new Mo EOS is probably worth comparing to in some way.

Zurkowski, C. C., Lim, R. E., Pardo, O. S., O'Bannon, E. F., Glazyrin, K., Söderlind, P., & Jenei, Z. (2024). Improving equations of state calibrations in the toroidal DAC—The case study of molybdenum. Journal of Applied Physics, 136(7).

[Reply]

Thank you for pointing out the new paper, Zurkowski et al. (2024). Their compression data (Supplementary Table S1~S6 in the paper) were plotted in newly added supplementary Figure S9 with our R-S EoS of molybdenum. Here pressure values are recalculated by using Cu-scale of Fratanduono et al.²¹ In their experiment, Cu and Mo were not arranged in a stacked configuration, but they showed good agreement with our EoS in the range of 150~340 GPa. Especially their run tDAC-Cu1 (orange circles in Figure S9) is a good support data of our Mo EoS. The following paragraph was inserted at line 272~. (The citation number has also been

corrected.)

[Line 272~]

Furthermore, simultaneous compression data for copper and molybdenum using t-DAC has been reported⁶². Figure S9 shows a comparison of the Mo R-S EoS in present study and the compression data from their six experiments. Here, pressure values have been recalculated by the copper scale²¹. There is some scatter in the data below 150 GPa, but above 150 GPa their data is in good agreement with our EoS. Note that for pressures below 150 GPa our Mo R-S EoS have adopted compression data using the ruby or MgO scale in a He pressure medium^{34,36}.

[Reference]

[62] Zurkowski, C. C. et al. Improving equations of state calibrations in the toroidal DAC—The case study of molybdenum. *J. Appl. Phys.* **136**, 075901 (2024).

[Figure S9]

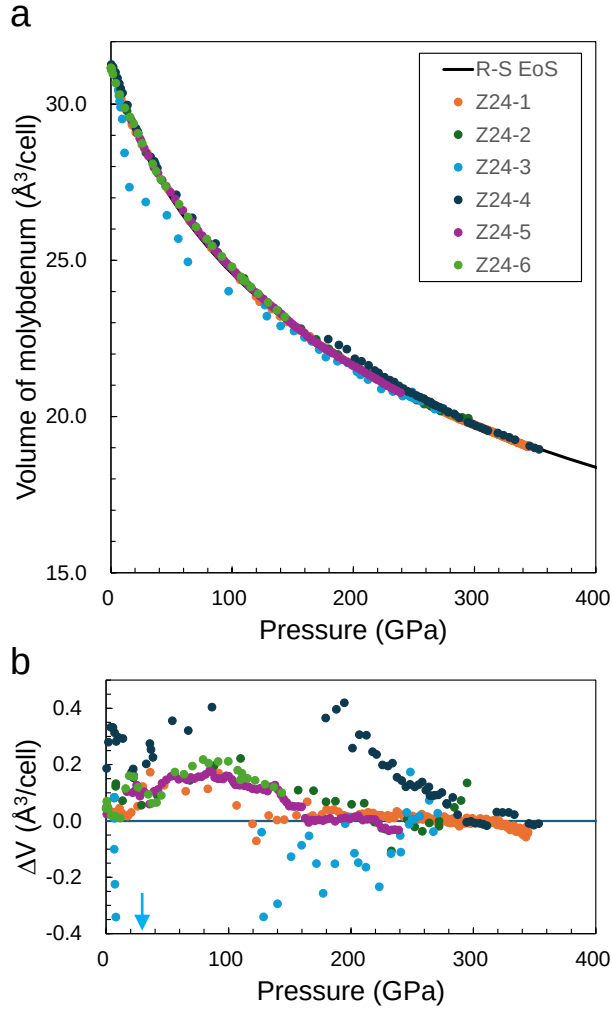


Fig. S9 | Compression curve of molybdenum comparing with the compression data of Zurkowski et al.⁶² **a** Solid curve shows the R-S EoS of molybdenum of this study. Z24-1~6 are the compression data of copper and molybdenum (run1-4, t-DAC; run5-6, Ne pressure medium) in Zurkowski et al.⁶² Here pressure values are recalculated by using the copper scale²¹. **b** the volume differences relative to the R-S EoS. The symbols are the same in Figure S9a. Arrow means that some data points exist at the out range of the figure in run Z24-3.

The other corrections

(Typo) Table 2, NaCl-B2, K_0 38.12 $\rightarrow$ 37.12