Meteoritics & Planetary Science 58, Nr 6, 897–900 (2023)
doi: 10.1111/maps.13996

Report

Is µCT irradiation nondestructive? A noble gas study on matrix samples from the CV3 chondrite Allende

Parastoo GHAZNAVI[1], Yogita KADLAG[1], David HABERTHÜR[2], Ruslan HLUSHCHUK[2], and Ingo LEYA[1]*

[1]Space Sciences and Planetology, University of Bern, Bern, Switzerland
[2]Institute of Anatomy, University of Bern, Bern, Switzerland
*Corresponding author.
Ingo Leya, Space Research and Planetology, University of Bern, Sidlerstrasse 5, Bern 3012, Switzerland.
Email: ingo.leya@unibe.ch

(Received 24 October 2022; revision accepted 05 May 2023)

Abstract–Micro-computed tomography (µCT) is a fast and powerful technology for studying textural, physical, and chemical properties of solid objects in three dimensions. While regularly used for sample documentation and curation, it is often assumed that µCT techniques are essentially nondestructive or at least very little destructive. However, there are very few studies proving or rejecting the assumption of nondestructiveness. Here we study whether X-ray tomographic imaging affects the noble gas budget of matrix samples from the CV3 carbonaceous chondrite Allende. We irradiated powdered and homogenized matrix samples in the Bruker SkyScan 1272 µCT instrument at three different X-ray tube acceleration voltages of 30, 70, and 100 keV. By comparing the noble gas concentrations and especially the elemental and isotopic ratios of the irradiated samples with data for two non-irradiated aliquots, we found no significant differences. Our study therefore demonstrates that X-ray tomographic imaging has no measurable effect on the noble gas budget and can therefore safely be used for sample characterization prior to noble gas studies.

INTRODUCTION

Most studies of early solar system processes, planet formation, and the dynamics of small bodies in the solar system are based on meteorites and other rare materials from sample-return missions. In recent years, the amount of sample material available for scientific studies is getting smaller, either due to more focused studies, for example, the investigation of certain minerals, or due to obvious limitations for materials from sample-return missions. At the same time, however, there is a requirement for performing as many studies as possible on the same sample. In this context, obtaining detailed physical and chemical information of the studied object, that is, chondrules, calcium–aluminum-rich refractory inclusions (CAIs), or other inclusions, before sample preparation and analysis provides valuable information not only on their origin and evolution, but it also helps optimizing sample preparation procedures.

Earlier studies established micro-computed tomography (µCT) techniques to investigate the overall petrography of the two CV3 chondrites Allende and Mokoja with the goal to better constrain their formation histories (Griffin et al., 2012; Hezel et al., 2013). In another study, Beitz et al. (2013) used µCT techniques to experimentally study the formation of chondrule rims and very recently, Barosch et al. (2020) discussed that chondrule classification based on two-dimensional cuts is not reliable and can result in a misclassification depending on where exactly the chondrule has been cut. They propose that 3D images, like those obtained by X-ray tomography, are better qualified for chondrule classification. In addition to the petrographic and mineralogical studies, there are also studies in which µCT imaging is used to provide a quality assessment of the sample preparation procedure. For example, we recently demonstrated that µCT studies are essential to demonstrate that separated chondrules are free of rim contamination. Even chondrules that have been treated in an abrasion cell and that appear round are not necessarily free of contamination and such contamination must be removed or at least corrected for before doing precise isotope studies. For example, Roth et al. (2016) and Roth and Leya (2018) demonstrated that even small amounts of remaining rim material or matrix contamination can seriously affect the measured noble gas concentrations. The same is true for stable isotope studies, such as for Cr, S, and Fe.

Whereas for petrographic and mineralogical studies, X-ray tomographic imaging can be considered nondestructive, the same is not yet proven for noble gas and/or stable isotope studies. Using X-ray energies of 90 and 190 keV, Hanna and Ketcham (2017) studied whether X-ray scanning can change the remnant magnetization of the sample. They found no resolvable change in the magnetic moment, indicating that there is no significant magnetic contamination. Ebel et al. (2009) also investigated the effects caused by X-ray scanning. In this study, by using synchrotron tomographic imaging, the authors found changes in the composition of polycyclic aromatic hydrocarbon. By using higher X-ray energies and longer irradiation times, Friedrich et al. (2016) tested for changes in the amino acids from the CM2 chondrite Murchison and found no changes. There are two studies demonstrating that X-ray scanning significantly affects the thermoluminescence characteristics of the studied sample (Sears et al., 2016, 2018). The authors stated that X-ray tomographic imaging should not be referred to as nondestructive and that it should not be performed without prior knowledge of any detrimental effects. Actually, detrimental effects of X-ray scanning are expected, because of the ionizing radiation used, more so for organic than for non-organic material. Here we study whether or not X-ray scanning can affect the noble gas budget of the studied samples.

EXPERIMENTAL

Sample Preparation

Chondrules and refractory inclusions were separated from a ~2 g fragment of the CV3.6 carbonaceous chondrite Allende. Remaining matrix material was inspected carefully for chondrule fragments and coarser objects and was purified using a binocular microscope and dental tools. The extracted matrix material was further powdered using an agate mortar, sieved using a nylon mesh, and the grain size fraction smaller than 33 µm was selected. We decided to use a very fine-grained fraction for this study because we expect that any losses of noble gases are more pronounced for smaller grain sizes than for bigger ones. In other words, if there are no measurable effects observed for the small grain sizes, there are no effects expected for bigger objects like chondrules, CAIs, and/or other inclusions that are usually studied using µCT. Thus, the prepared powdered matrix sample was separated into five aliquots, each with a weight of ~70 mg.

X-Ray Scanning

Three of the five prepared aliquots were selected for irradiation treatment using a SkyScan 1272 µCT system (Bruker microCT, Kontich, Belgium) located at the Institute of Anatomy, University of Bern. The X-ray source is a (non-serviceable and non-openable) microfocus source from Hamamatsu (Type: L11871_20) with tungsten as the target material. One sample was irradiated for 13 h with a X-ray acceleration voltage of 100 kV, one was irradiated for 14 h with an acceleration voltage of 70 kV, and one was irradiated for 14 h with an acceleration voltage of 30 kV (see Table 1). Note that the given energy is the acceleration voltage of the X-ray source, which is not equivalent to the peak energy in the X-ray spectrum. For example, with an acceleration voltage in the range of 10 kV, the Bremsstrahlung spectrum has an intensity maximum at ~6 keV. In this respect, classical X-ray sources are different to synchrotron sources, for which a given energy of, for example, 30 keV corresponds to the peak energy in the spectrum.

The exposure times are in line with exposure durations typically used for other µCT scans, especially if one aims for low voxel sizes or high-resolution scans of dense materials. However, the exposure durations are slightly longer than typical irradiation durations for, for example, chondrules, which are in the range of 10 h.

For calculating the irradiation dose, we followed the procedure described by Friedrich et al. (2016). Briefly, we calculated the photon spectrum using the program SpekCalc (Polundniowski et al., 2009; Polundniowski & Evans, 2007a, 2007b) by giving the peak energy, a thickness of 0.5 mm for the used Al filter, and assuming a distance of 2 cm between the X-ray tube and our samples. The program then gives the number of photons per keV, per cm^2, and per mAs as a function of photon energy. Multiplying the number in each energy bin with the beam current and the irradiation duration gives the number of photons per cm^2. Assuming a sample cross-section of ~1 cm^2 and ~70% absorption gives the energy delivered in each energy bin. Integrating now over all energies gives the total energy (in keV) absorbed by the sample. By changing the energy unit from keV to Joule and considering the sample mass (~70 mg for each aliquot), we can calculate the deposited energy in J kg^{-1}, which corresponds to the unit Gray (Gy). Thus, the calculated irradiation doses range between ~1.3 and ~3.1 Gy (Table 1).

TABLE 1. Sample parameters, irradiation conditions, and radiation doses.

Sample	Weight (mg)	Irradiation energy (kV)	Source current (μA)	Irradiation time (h)	Radiation dose (Gy)
1	2.66	No irradiation			
2	2.82	No irradiation			
3	2.57	100	100	13	3.1
4	2.57	70	142	14	1.8
5	2.49	30	210	14	0.6

Can we bring these values into context? With an ionization energy in the range of ~1000 kJ mol^{-1} for silicon, we calculate that the 100 kV irradiation (D ~ 61 J kg^{-1}) can fully ionize ~87 μg of silicon. In addition to ionization effects, there might also be effects due to sample heating. Assuming a specific heat capacity in the range of 1000 J kg^{-1} K^{-1}, we estimate a temperature increase of the order 0.003 K, which would be negligible for noble gas release.

Noble Gas Measurements

From the irradiated and non-irradiated samples, aliquots with masses in the range of 2–3 mg were taken for noble gas studies (weights are given in Table 1). The fine-grained powders were gently pressed into the sample holder to avoid material loss during ablation. Each sample holder can contain nine samples. The sample holder is made of aluminum, its high thermal conductivity helps to avoid heating neighbored samples while one sample is laser irradiated. Samples were degassed at a temperature of ~2000°C using an infrared diode laser with a continuous wavelength of 808 nm and a maximum power of 75 W (Type LM808, Dr. Mergenthaler, Germany). The laser is connected to a two-color pyrometer. The laser power is slowly increased until the sample is completely molten.

After extraction, gases were cleaned using various getters and cold traps in a dedicated low-volume low-blank extraction line connected to a MAP 215-50 magnetic sector field mass spectrometer with an improved detecting system. Helium and Ne isotope concentrations were measured in all five matrix aliquots together with signals from possible interferences, that is, $^{20}H_2O^+$ and $^{40}Ar^{++}$ on ^{20}Ne and $^{44}CO_2^{++}$ on ^{22}Ne. After each sample, re-extractions at slightly higher laser powers were performed. Corrections from re-extractions, blanks, and interferences are <2% for ^{3}He and ^{4}He and <1% for ^{21}Ne.

RESULTS: STATISTICAL INTERPRETATION OF THE DATA

We focus the following discussion on the (raw) $^{3}He/^{21}Ne$ and $^{3}He/^{4}He$ ratios because they are the most sensitive data for studying noble gas losses. The ratios given in Table 2 are not corrected for mass fractionation and counting efficiency, that is, the $^{3}He/^{21}Ne$ ratios are simply the ion counts for ^{3}He divided by the ion counts for ^{21}Ne (unit Hz/Hz). The $^{3}He/^{4}He$ ratios are given in the unit (Hz/V), they are simply the readings for the ion counter divided by the readings on the DVM for the Faraday detector. Doing the data interpretation this way avoids being limited by uncertainties due to counting efficiencies and fractionation, which affect all data the same way. The given uncertainties comprise the uncertainties from the counting statistics and from the extrapolation of the time-dependent noble gas readings to the time of gas inlet. The statistical interpretation of the data has been done using the statistical software package R.

TABLE 2. Noble gas isotope ratios for irradiated and non-irradiated samples.

Sample	Weight (mg)	Dose (Gy)	$^{3}He/^{21}Ne$	$^{3}He/^{4}He$
1	2.66	0	0.964 ± 0.005	117,006 ± 727
2	2.82	0	0.925 ± 0.010	113,494 ± 1471
	Average		0.945 ± 0.020	115,250 ± 2030
3	2.57	3.1	0.953 ± 0.004	114,923 ± 778
4	2.57	1.8	0.974 ± 0.006	115,773 ± 888
5	2.49	0.6	0.922 ± 0.007	111,104 ± 894
	Average		0.950 ± 0.021	113,933 ± 2030

Note: The noble gas data are not corrected for spectrometer sensitivities. For $^{3}He/^{21}Ne$, the data give the ratios of the counter readings in (Hz/Hz) and for $^{3}He/^{4}He$ the data give the counter readings divided by the Faraday readings (Hz/V).

The average $^{3}He/^{21}Ne$ ratio (raw) for the two non-irradiated samples is 0.945 ± 0.020. The uncertainty is the 1σ-standard deviation of the mean, which is larger than the internal uncertainty. For the irradiated samples, the average is 0.950 ± 0.021 (1σ-standard deviation). An independent t-test gives a p-value of 0.8082 (t = 0.265, df = 3), indicating that the two mean values are not significantly different.

The (raw) $^{3}He/^{4}He$ ratios for the irradiated and the non-irradiated samples are $1.153 \times 10^5 \pm 1.756 \times 10^3$ and $1.139 \times 10^5 \pm 2.030 \times 10^3$, respectively. The given uncertainties are the standard deviations. Performing the same t-test for the $^{3}He/^{4}He$ ratio gives p = 0.488,

$t = 0.789$, $df = 3$. The p-value indicates that the two mean values are not significantly different.

The statistical interpretation of the noble gas data indicates that the $^{3}He/^{4}He$ and $^{3}He/^{21}Ne$ ratios for the irradiated and the non-irradiated samples are indistinguishable. If any differences, the $^{3}He/^{4}He$ and $^{3}He/^{21}Ne$ ratios in the irradiated samples are even slightly (but not significantly) higher than the same ratios in the non-irradiated samples, the opposite to what is expected if there would be noble gas losses. From this finding, it is safe to conclude that 3D X-ray imaging, even when using irradiation doses, that is, energy, current, and time, higher than in typical micro-CT studies and for samples prone to noble gas loses due to their small grain size, can be considered "nondestructive" in terms of the noble gas budget.

CONCLUSIONS AND REMARKS

We demonstrated in a systematic study by using irradiation doses higher than usually applied in μCT studies and with samples of extremely small grain sizes (<33 μm) that there are no noble gases losses; the $^{3}He/^{4}He$ and $^{3}He/^{21}Ne$ ratios of irradiated and non-irradiated samples are indistinguishable. Therefore, 3D X-ray tomographic imaging can safely be considered nondestructive for noble gas studies.

Acknowledgments—This work was supported by the Swiss National Science Foundation (200020_196955) and the NCCR PlanetS (Swiss National Science Foundation Grant Number 51NF40-141881). We are grateful to Jan Wenger and Urs Geissbühler for assistance. We thank J.M. Friedrich for a very helpful review. Open access funding provided by Universitat Bern.

Conflict of Interest Statement—We declare no conflict of interest.

Data Availability Statement—The data that support the findings of this study are openly available in the Harvad Dataverse.

Editorial Handling—Dr. Marc W. Caffee

REFERENCES

Barosch, J., Hezel, D. C., Sawatzki, L., Halbauer, L., and Marrocchi, Y. 2020. Sectioning Effects of Porphyritic Chondrules: Implication for the PP/POP/PO Classification and Correcting Modal Abundances of Mineralogically Zoned Chondrules. *Meteoritics & Planetary Science* 55: 993–9.

Beitz, E., Blum, J., Mathieu, R., Pack, A., and Hetzel, D. C. 2013. Experimental Investigation of the Nebular Formation of Chondrule Rims and the Formation of Chondrite Parent Bodies. *Geochimica et Cosmochimica Acta* 116: 41–51.

Ebel, D. S., Greenberg, M., Rivers, M. L., and Newville, M. 2009. Three-Dimensional Textural and Compositional Analysis of Particle Tracks and Fragmentation History in Aerogel. *Meteoritics & Planetary Science* 43: 1445–63.

Friedrich, J. M., Glavin, D. P., Rivers, M. L., and Dworkin, J. P. 2016. Effect of a Synchrotron X-Ray Microtomography Imaging Experiment on the Amino Acid Content of a CM Chondrite. *Meteorite & Planetary Science* 51: 429–37.

Griffin, L. D., Elangovan, P., Mundell, A., and Hezel, D. C. 2012. Improved Segmentation of Meteorite Micro-CT Images Using Local Histograms. *Computers & Geosciences* 39: 129–34.

Hanna, R. D., and Ketcham, R. A. 2017. X-Ray Computed Tomography of Planetary Materials: A Primer and Review of Recent Studies. *Chemie der Erde* 77: 547–72.

Hezel, D. C., Elangovan, P., Viehmann, S., Howard, L., Abel, R. L., and Armstrong, R. 2013. Visualisation and Quantification of CV Chondrite Petrography Using Micro-Tomography. *Geochimica et Cosmochimica Acta* 116: 33–40.

Polundniowski, G. G., and Evans, P. M. 2007a. Calculation of X-Ray Spectra Emerging from an X-Ray Tube. Part 1. Electron Penetration Characteristics in X-Ray Targets. *Medical Physics* 34: 2164–74.

Polundniowski, G. G., and Evans, P. M. 2007b. Calculation of X-Ray Spectra Emerging from an X-Ray Tube. Part 2. X-Ray Production and Filtration in X-Ray Targets. *Medical Physics* 34: 2175–86.

Polundniowski, G. G., Landry, G., DeBois, F., Evans, P. M., and Verhaegen, F. 2009. A Program to Calculate Photon Spectra from Tungsten Anode X-Ray Tubes. *Physics in Medicine & Biology* 54: 433–8.

Roth, A., and Leya, I. 2018. No Cosmic-Ray Precompaction Exposure of Chondrules in CR2.7 MIL 090657. *Meteoritics & Planetary Science* 53: 2644–51.

Roth, A. S. G., Metzler, K., Baumgartner, L. P., and Leya, I. 2016. Cosmic-Ray Exposure Ages of Chondrules. *Meteoritics & Planetary Science* 51: 1256–67.

Sears, D. W., Sears, H., Ebel, D. S., Wallace, S., and Friedrich, J. M. 2016. X-Ray Computed Tomography Imaging: A Not-So-Nondestructive Technique. *Meteoritics & Planetary Science* 51: 833–8.

Sears, D. W., Sehlke, A., Friedrich, J. M., Rivers, M. L., and Ebel, D. S. 2018. X-Ray Computed Tomography of Extraterrestrial Rocks Eradicates their Natural Radiation Record and the Information it Contains. *Meteoritics & Planetary Science* 53: 2624–31.