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A Peltier-based freeze-thaw device for meteorite disaggregation

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A Peltier-based freeze-thaw device for the disaggregation of meteorite or other rock samples is described. Meteorite samples are kept in six water-filled cavities inside a thin-walled Al block. This block is held between two Peltier coolers that are automatically cycled between cooling and warming. One cycle takes approximately 20 min. The device can run unattended for months, allowing for ~10 000 freeze-thaw cycles that will disaggregate meteorites even with relatively low porosity. This device was used to disaggregate ordinary and carbonaceous chondrite regoltih breccia meteorites to search for micrometeoroid impact craters. *Published by AIP Publishing.* https://doi.org/10.1063/1.5006261

I. INTRODUCTION

Freeze-thaw disaggregation of meteorites has been used for decades to separate constituents of the samples for further analyses.[1–3] Disaggregating rocks by freeze-thaw in the laboratory mimics the natural terrestrial "frost-wedging" processes that helped form, for example, the famous hoodoos in Bryce Canyon, Utah.[4] In laboratory freeze-thaw, a chip of a meteorite is placed into water, cooled until the water is frozen, and then warmed again until the water becomes liquid. Meteorites have a wide range of porosities[5,6]—stony meteorites are porous enough to allow liquid water to penetrate the rock through cracks. When the water freezes, its density decreases by 8% (from 1.00 to 0.92 g/cm^3, Fig. 1) and the expanding ice further enlarges the crack. Repeated freeze-thaw cycles can disaggregate the rock into its constituent grains as water penetrates along grain boundaries. Single crystals and other nonporous objects in the meteorite, like individual chondrules, do not get broken down by the freeze-thaw process (unless the object is structurally compromised, like a cracked chondrule). Stony meteorites have large variations in their crushing strengths (though only ~20 have been measured): 5–500 MPa for ordinary chondrites[7] and 0.3–30 MPa for carbonaceous chondrites.[8] Presumably, meteorites with lower material strengths and/or high porosity, like the Bells ungrouped carbonaceous chondrite,[9] will disaggregate faster than meteorites of higher petrographic grade, like type 4–6 ordinary chondrites, which are more coherent due to metamorphic processing.

Previously, freeze-thaw disaggregation has been done manually, e.g., by moving samples in a water-filled beaker into and out of a freezer. Only ~10 s of freeze-thaw cycles are required to sufficiently disaggregate some carbonaceous chondrites, such as Murchison for separation of calcium-aluminum inclusions.[10] This type of manual freeze-thaw disaggregation is tedious but sufficient to disaggregate the most friable meteorites. An automatic freeze-thaw device has been described[11] that uses a robotically controlled arm to move samples, sealed in a container with water, from liquid nitrogen to hot water. This device makes it feasible to disaggregate CR2 chondrites over ~250 cycles of freeze-thaw (100 h total duration) to separate millimeter-sized chondrules from the surrounding matrix. The porosity of the Y-793495 CR chondrite is ~11%,[5] which is a factor of two higher than porosities measured in most ordinary chondrites using similar techniques. Gentle crushing[12] and electric-pulse disaggregation[13] have also been used to disaggregate meteorites. Crushing can fracture grains, and electric-pulse disaggregation requires highly specialized and complicated equipment.

In this paper, I describe an automatic freeze-thaw device based on Peltier thermoelectric coolers that is inexpensive to build and run, can operate for weeks to months with almost no maintenance, and can subject a meteorite (or terrestrial rock) to ~10 000 freeze-thaw cycles. Using this device, it is possible to disaggregate even the most lithified stony meteorites. I will describe the disaggregation of some regolith breccia stony meteorites (type 4–6 ordinary chondrites) using this device, and the subsequent search of the samples for evidence of micrometeoroid impacts on space-exposed grains.

II. DESIGN AND PERFORMANCE

Water ice, at atmospheric pressure, has its minimum density at 0 °C (Fig. 1). Therefore, during freeze-thaw disaggregation, it is most efficient to cool the water just to freezing before warming it back up to liquid. Cooling the ice below 0 °C does not help with freeze-thaw disaggregation because the ice decreases in volume; no further pressure is applied on the rock from the ice, and no further disaggregation occurs. A Peltier thermoelectric device[14] is capable of achieving these temperatures without the need to continually refill a liquid nitrogen bath.

I use a Peltier "sandwich" (a sample block held between two Peltier cold plates) to cool and heat a 316 stainless steel block of six vessels (the "six-pack") that hold the meteorite samples and purified 18 MΩ water (Fig. 2). The top of each vessel is tapped (5/16 in.–24) for a 316 stainless steel bolt that serves as a lid. The bolt compresses a plastic O-ring (1/4 in. outer diameter) that makes the seal water-tight. The block that is cooled by the Peltier coolers has six vessels, but only five of these are used to hold samples. The sixth vessel is left open to monitor the temperature of the water/ice (Fig. 3).

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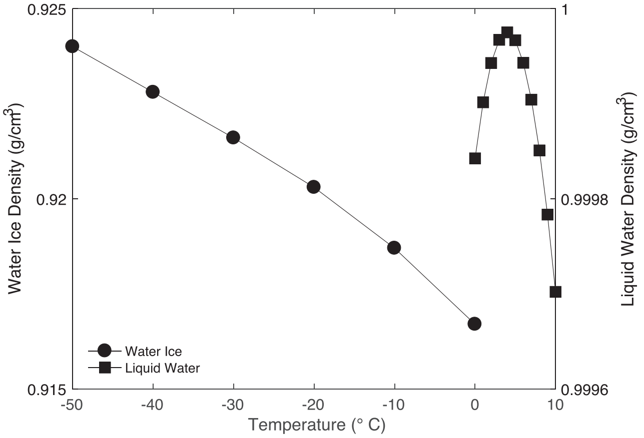

FIG. 1. Density of water ice (left axis) and liquid water (right axis) at atmospheric pressure; data from CRC Handbook of Physics and Chemistry.[15]

The cold plates and six-pack are held together by six bolts that can be tightened by hand (turquoise-handled bolts in the top-right of Fig. 6). This geometry is most efficient at cooling and heating the samples. The Peltier devices are Teca thermoelectric air-cooled aluminum cold plates (purchased from eBay for \$100–\$200 each). By switching polarity, the Peltier devices can be used for heating. A small bread pan sits underneath the sample block to collect water that condenses and eventually drips off the device.

The Peltier devices are controlled with a timer that allows for a user-determined duration of cooling and a separate user-determined duration of heating. The polarity supplied to the Peltier device is reversed whenever the timer reaches zero. During the heating cycle, a fan is turned on that removes water and prevents ice building up on the lids of the vessels. A green light-emitting diode (LED) is lit when the system is cooling, and a red LED is lit when it is heating. An electromechanical counter tracks the total number of freeze-thaw cycles. A circuit block diagram is shown in Fig. 4. All electronics are contained in a sealed metal box (Figs. 5 and 6).

I experimented with different cooling and heating times to arrive at an optimal temperature profile. About 18 min of cooling time followed by ~18 s of heating time was enough to freeze the water in the vessel completely, and keep it frozen for several minutes. The temperature inside each vessel varies from −4 °C to 7 °C; the temperature profile is shown in Fig. 7. A shorter (~15 min) cooling time could be used to speed up the freeze-thaw process, but I erred on the side of caution: cooling for an extra 3 min would increase the duration of the freeze-thaw cycle by only 3 min, but not cooling enough could result in no sample disaggregation at all if the vessel does not freeze all the way through.

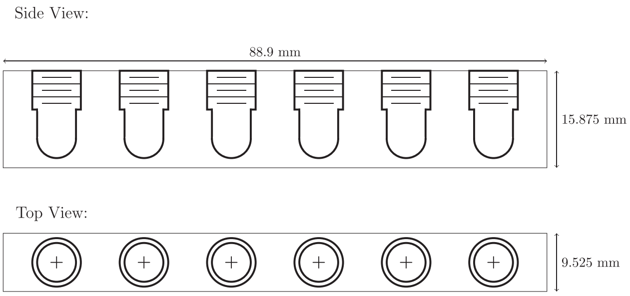

FIG. 2. Schematics of the "six-pack" sample holder. The top cylinder of each vessel is 7.9375 mm in diameter and 6.35 mm tall; the lower cylinder is 6.35 mm in diameter and 4.7625 mm tall. The total height of each vessel is 14.2875 mm.

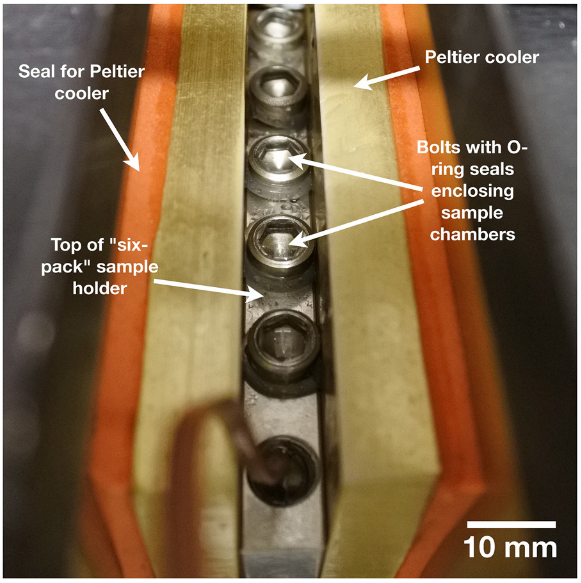

FIG. 3. Top-view of the "six-pack" sample holder (silver color) held between two Peltier cold plates (gold color). Five of the sample holders are in use (with bolts screwed into the top holes); one is used to monitor the freeze-thaw process either visually or with a temperature probe.

After ~8000 freeze-thaw cycles, the original Peltier devices (which were used when I bought them) required 25% more time to freeze the same volume of water. By leaving one of the vessels filled with water but no sample, and uncapped, I could monitor the temperature of the system and see when the Peltier devices began failing. I initially compensated for this loss in efficiency by increasing the cooling time. Eventually, when the device needed 100% more time to freeze water in the empty vessel, the Teca cold plate was dismantled and the individual Peltier junctions were replaced with new devices (Table I). After this replacement, the device performed at its original efficiency. The new Peltier junctions should last much more than 10 000 cycles because they are designed to be cycled.

III. APPLICATION

I used the described automatic freeze-thaw device to disaggregate regolith breccia meteorites.[16,17] I will discuss results from the Adzhi-Bogdo (stone) meteorite.[18]

Adzhi-Bogdo (stone) is an LL chondrite regolith breccia of petrologic type 3–6. Well-defined dark and light lithologies are visible in hand specimen (Fig. 8). This meteorite has affiliations with L chondrites: concentrations of Fe and Ni are intermediate between L and LL, and fayalite contents of olivines in some components are more consistent with L chondrites. Adzhi-Bogdo is a polymict breccia with sub-mm to cm-sized fragments embedded in a clastic matrix of

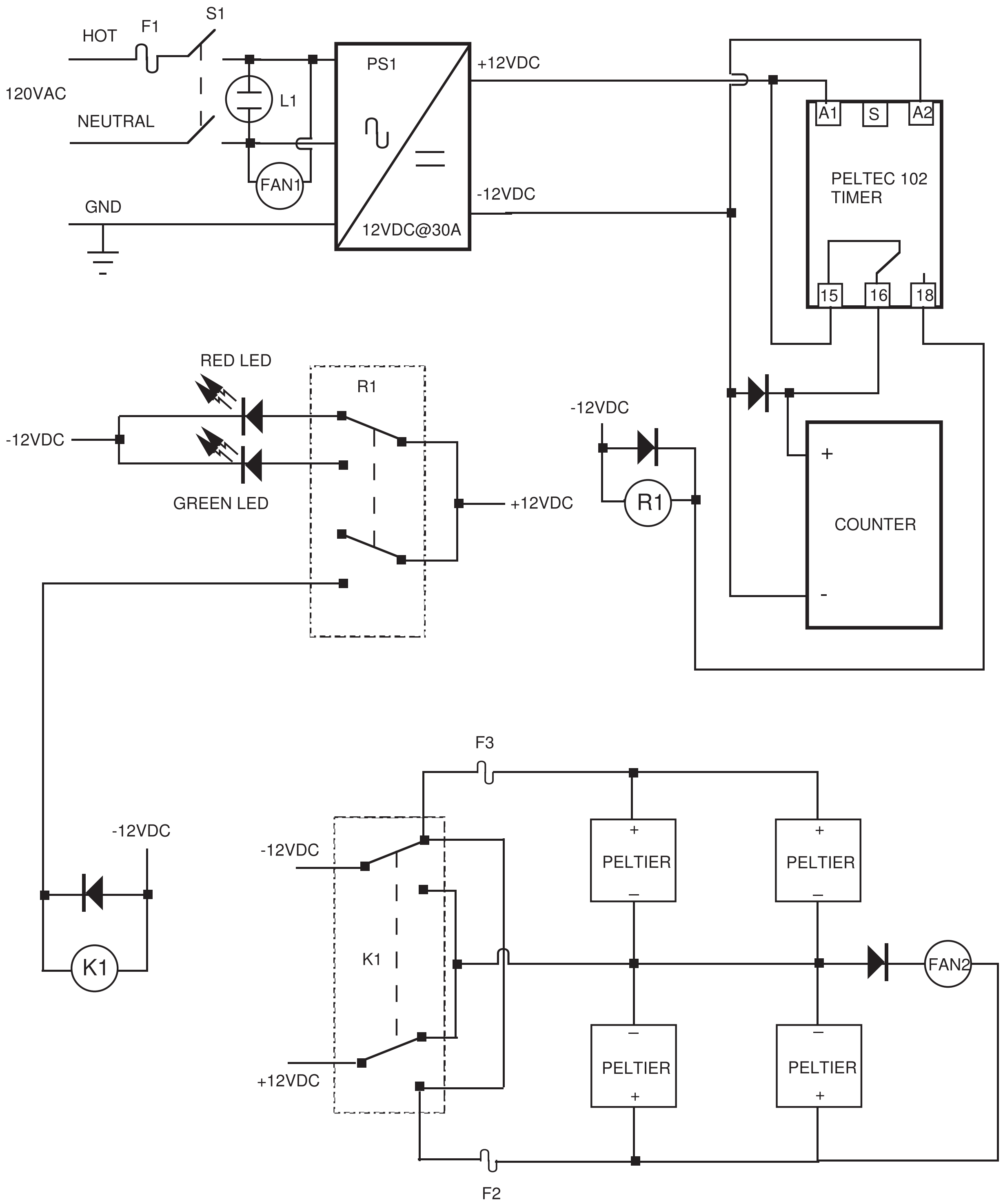

FIG. 4. Circuit block diagram for the freeze-thaw device.

<50 μm fragments. Chondrules, melt rock clasts, achondritic clasts, metamorphosed LL clasts, and fine-grained fragmental breccias are the majority of the components in the rock. Adzhi-Bogdo contains solar wind noble gases (solar ^{20}Ne $\approx 6 \times 10^{-8}$ ccSTP/g[18]), meaning that it is a regolith breccia meteorite that was exposed to the solar wind on the surface of its parent asteroid. Most regolith breccia chondrites are H chondrites—there are only five known LL-chondrites that contain solar gases. JAXA's Hayabusa mission to asteroid Itokawa returned regolith grains that were analyzed to be similar to LL4–6 chondrites.[19] Adzhi-Bogdo, a regolith breccia LL3–6 chondrite, is one of the closest meteorite analogs to the samples returned from asteroid Itokawa.

I acquired 1 g of Adzhi-Bogdo for freeze-thaw disaggregation. This sample clearly contained both dark and light lithologies (Fig. 8). The meteorite samples were subjected to ~3000 freeze-thaw cycles, which took 2 months to complete. It is expected that Fe-Ni metal and other phases would

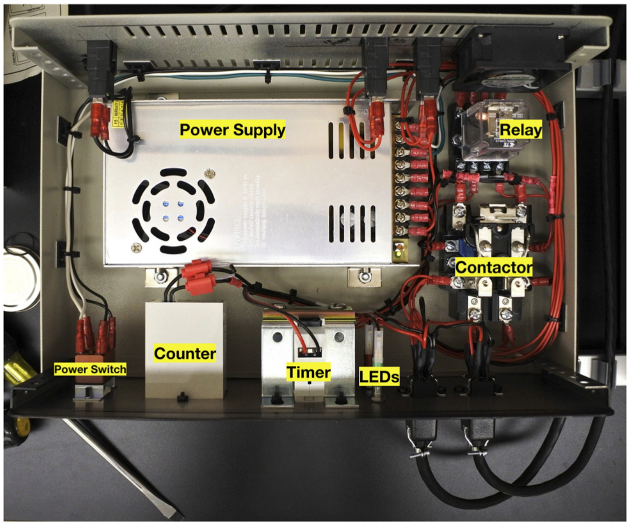

FIG. 5. Electronics and power supply of the freeze-thaw device.

oxidize during the disaggregation process, as the meteorite is submerged in water for weeks to months. A careful assessment of sample alteration during freeze-thaw disaggregation has not been performed. The study described here is concerned with physical features on mineral grains that are not likely to be affected by aqueous alteration during the disaggregation.

Surface features that mimic mechanical space-weathering processes (e.g., impact craters) may be created by the freeze-thaw disaggregation process. To understand this effect, I simultaneously disaggregated three ordinary chondrites that were not regolith breccias: Bath (H4), Tuxtuac (LL5), and Ankober (H4). Bath has a measured porosity of 6.1%.[5] Chips of these meteorites, 0.3–0.5 g in mass, were only partially disaggregated after ~3000 freeze-thaw cycles: a fraction of the original chip remained, although most of it had been disaggregated to sub-mm grains. In contrast, the entire Adzhi-Bogdo sample was reduced to powder (grain size: 10 μm–1 mm, Fig. 9). Though material strengths or porosities have not been quantitatively measured on all four meteorites, this observation can be explained if the regolith breccia ordinary chondrites have a higher porosity (more cracks where water can penetrate) than the non-regolith-breccia ordinary chondrites. Such a difference in porosity would be expected if Adzhi-Bogdo is a less-lithified conglomerate of different lithologies (as seen in Fig. 8) from the top surface of its parent asteroid and possibly a foreign impactor similar to L chondrites.[18]

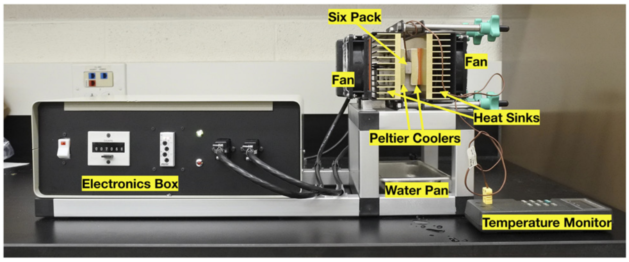

FIG. 6. The entire freeze-thaw device, with a temperature probe to monitor the uncapped vessel.

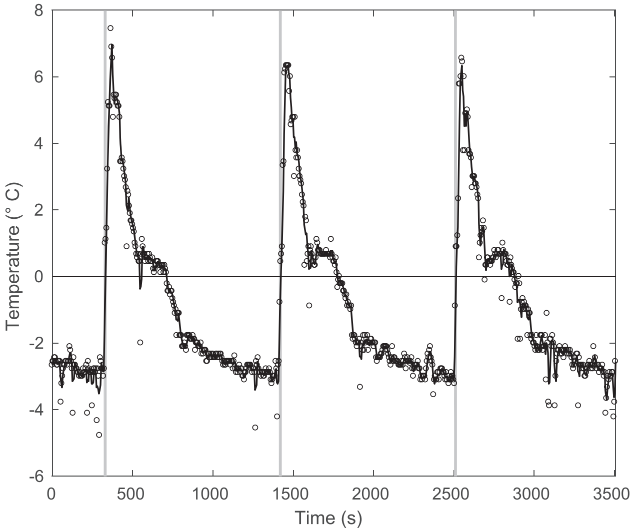

FIG. 7. Temperature as a function of time as measured inside the water-filled open vessel (Fig. 3) for 18 min of cooling time followed by ~18 s of heating time. The gray stripes indicate when the Peltier devices are heating the rest of the time they are cooling. Open symbols are data points; the solid line is the 3-point moving average.

I removed the disaggregated meteorite powder in water with a micropipette and deposited the powder and water in a welled glass slide. I evaporated off the water with a heat lamp, then picked up the fine powder with a white-bristled paintbrush, and deposited it on carbon tape mounted on a 0.5-in. scanning electron microscope (SEM) stub. Next, I coated each stub with ~2 nm of gold-palladium three separate times,

TABLE I. Description of parts used to build the freeze-thaw device.

Part label (Fig. 4)	Part description
PS1	12 V, 30 A power supply
PELTEC 102 TIMER	Peltec 102 assymetric cycler
COUNTER	Reddington P9-4096 counter
PELTIER	Marlow Industries thermal cycler XLT2422-01S + Teca thermoelectric air-cooled aluminum cold plate 12/24VDC AHP-150CP USG
FAN1	Sunon MA-1062HVL (electronics cooling fan)
FAN2	Sunon PMD1206PMB1-A, 12 V/10.6 W (defrost fan)
R1	Potter and Brumfield KRPA-11DN-12 relay
K1	Contactor, McMaster Carr part #7384K43, 12 V DPDT with contacts 28VDC @ 30A
S1+L1	McMaster Carr part #7395K252, DPST-NO 110VAC/20A
Diodes	1N4007
GREEN LED	McMaster Carr part #2779K121, 12VDC
RED LED	McMaster Carr part #2779K132, 12VDC
F1	Fuse 1: 5A @ 250VAC
F2	Fuse 2, 3: 10A @ 250VAC

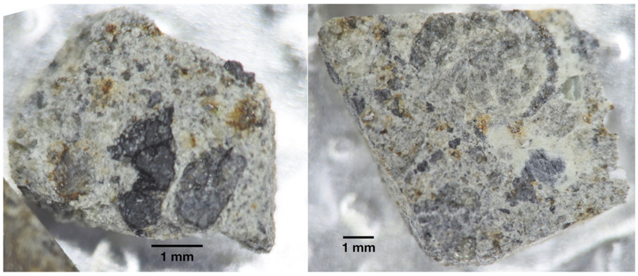

FIG. 8. Adzhi-Bogdo samples before disaggregation.

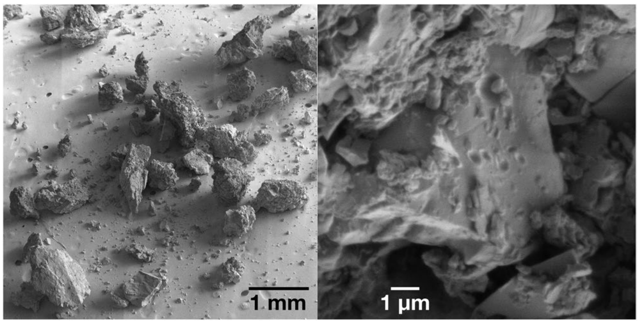

FIG. 9. (Left) Secondary electron images (1 kV accelerating voltage) of disaggregated samples of the Adzhi-Bogdo meteorite showing typical sizes of disaggregated grains (10 μm–1 mm). (Right) High-magnification secondary electron image (1 kV accelerating voltage) of crater-like impact features on an Adzhi-Bogdo grain.

each time with the stub laying at different angles in the sputter coater. This was to ensure electrical conductivity by minimizing shadowing of the conductive coat.

The disaggregated Adzhi-Bogdo samples were then imaged with a Tescan MIRA-3 field-emission scanning electron microscope. I acquired secondary electron images at low accelerating voltage (1–2 kV) for the highest surface sensitivity (Fig. 9). The images were acquired in the "depth mode" which uses an intermediate lens to provide enhanced depth of field at the expense of some spatial resolution. Automated image acquisition via the Tescan "Image Snapper" package with auto-focus, auto-brightness, and auto-contrast allowed for the collection of ~90 000 images of the Adzhi-Bogdo disaggregated grains, each with a 50 μm field of view and 1536 × 1536 pixels.

These images were manually searched for interesting features that could be related to micrometeoroid bombardment: impact craters, splash melt, and other impact residue. Several promising candidates for impact craters were identified (Fig. 9). These features will be removed by focused ion beam lift-out and analyzed by transmission electron microscopy. Eventually, as more samples are imaged and the number of images to be searched becomes very large, this process will be crowd-sourced: the images will be searched by volunteers online using the open-source Pybossa crowd-sourcing framework. Initial imaging studies of the non-regolith-breccia ordinary chondrite powders have not shown any impact features like those in Fig. 9, implying that these features did not form during the disaggregation process.

IV. DISCUSSION

A modification that could be made to this system is to pair temperature monitoring with the freeze-thaw cycle. A temperature probe could be inserted into the water of the empty 6th vessel, and the value could be read electronically. When the temperature falls below −1 °C, a timer would start for ~8 min. When the timer finishes, the polarity of the voltage to the Peltier device would reverse, and the sample would be warmed. Since the required warming time is so short, the warming time would be best set by a timer. At the end of this timer, the cooling cycle would start again. Active temperature monitoring could reduce the time duration for one freeze-thaw cycle and make the system more efficient. It would also automatically handle the decaying efficiency of the Peltier devices. However, active temperature control would also complicate the system and create another source of failure. For example, the temperature probe could fail or the water in the vessel that it is monitoring could evaporate.

V. CONCLUSIONS

I have described a Peltier-based freeze-thaw device that can be used to disaggregate meteorites with relatively low porosities (6.1% for the L chondrite Bath) over thousands of freeze-thaw cycles. The described device can be built and maintained cheaply and requires little supervision. I have employed this device to gently disaggregate regolith breccia meteorites. Identification and further analyses of surface-exposed regolith grains from these samples will allow for a more sophisticated understanding of space-weathering effects on other airless bodies in the Solar System.

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