In situ deformation apparatus for time-of-flight neutron diffraction: Texture development of polycrystalline ice I_h

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This article documents a new *in situ* deformation apparatus built for neutron diffraction investigations of polycrystalline materials in low-temperature environments and the first experiment in which it was used. We performed texture analysis of fine-grained polycrystalline D_2O ice I_h deformed uniaxially between 230 and 240 K using time-of-flight neutron diffraction on the high-pressure preferred orientation diffractometer at the Manuel Lujan, Jr. Neutron Scattering Center at Los Alamos National Laboratory. The new deformation apparatus operates at 1 atm of ambient pressure and over temperatures in the range of $77\ \mathrm{K} < T < 298\ \mathrm{K}$, and accommodates up to 667 N of uniaxially applied load. It is suitable for diffraction studies of any bulk polycrystalline material, ideally cylindrical in shape, and is adaptable to multiple neutron spectrometers, including those at other polychromatic and monochromatic neutron facilities. The first experiment on a hexagonal ice sample demonstrates development of fiber texture in the direction of the applied load. The equipment has many applications to earth science, glaciology, and ice engineering. © *2006 American Institute of Physics.* [DOI: 10.1063/1.2349603]

INTRODUCTION

The crystallographic structures and unique physical behaviors of ice have been studied for over a century, not only to understand the evolution of the Earth and glacial flow, but also to use ice as an analog for both materials and ceramics sciences. There is a need to measure natural and experimental ice samples at the atomic level during deformation under *in situ* or real-time conditions. The form of ice (I_h) found on the Earth has a hexagonal crystal structure that leads to a wealth of unique physical properties also found in natural rocks and some metals. Here and throughout the article we refer to hexagonal ice I_h as ice.

To investigate the crystallographic behavior of polycrystalline fine-grained ($2-10\ \mu$m) ice under real-time load conditions, we built an *in situ* low-temperature, deformation apparatus for neutron spectrometry. In the past, laboratory investigations of deformation regimes in polycrystalline ice have been restricted to investigations where the ice sample is deformed, removed from the deformation apparatus, and then measured for crystallographic information.[1–5] As a result, only the final texture of a sample can be measured; the stages of texture development during the experiment are lost.

We designed and built a low-temperature deformation apparatus to deform ice *in situ* using time-of-flight (TOF) neutron diffraction on the unique high-pressure-preferred orientation (HIPPO) diffractometer[6] housed at the Manuel Lujan, Jr. Neutron Scattering Center (Lujan Center) at LANL in Los Alamos, New Mexico. The deformation apparatus is suitable for virtually any polycrystalline sample of appropriate size with a length and width up to $2.54 \times 2.54\ \mathrm{cm}^2$ in size. The new apparatus is suitable for most spectrometers at the Lujan Center and may be adapted for use at other neutron source facilities.

Our polycrystalline ice sample was made with D_2O. In neutron diffraction studies, it is common to substitute deuterium (D) for hydrogen (H).[7] Because neutrons scatter hydrogen incoherently, replacing H with D decreases the absorption cross section by a factor of approximately 1000. The result is that D is a stronger scatterer of neutrons and contributes less noise to the neutron signal than H.

With this apparatus, we measured deformation and crystallographic strain directly and simultaneously, including texture (preferred crystallographic orientation) development. Neutrons are ideal probes for bulk-material analyses and are well suited to investigation with special environments. As relatively weak scatterers, they are also nearly transparent to many materials that are used to build environmental cells,

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and large bulk samples containing light and heavy elements and coarse grains can be analyzed quantitatively.

To achieve our scientific goal of measuring the texture evolution in a polycrystalline ice sample during deformation and at glacial temperatures, we built a deformation rig with a sliding-piston cylinder. This article presents the first experimental results using the new deformation rig; we have measured texture evolution *in situ* of laboratory-produced, polycrystalline ice under load.

EXPERIMENTAL DESIGN

Our goal was to build an apparatus to apply a constant load to a D_2O polycrystalline sample, suitable for neutron-diffraction experiments at low temperatures. Our scientific goal was to relate texture evolution to strain in ice. For this task we chose to design a rig for the new TOF neutron diffractometer HIPPO. HIPPO sits on Flight Path 4 at the Lujan Center, with a time-averaged flux on a sample of $\sim 1 \times 10^7$ neutrons cm^{-2} s^{-1}. The diffractometer has 1393 He^3 gas tube detectors that surround the large (30 cm diameter, 183 cm deep) sample chamber, at five different scattering angles in 360° rings. To detect the diffracted neutrons, this instrumental setup is ideal for efficient quantitative *in situ* measurement with environmental cells; the multiple scattering angles and detectors combined with TOF diffraction analyses allow for simultaneous multiple peak analysis without rotating a sample or sample environment. On HIPPO, a full diffraction pattern ($0.12\ \text{Å} < d < 47.5\ \text{Å}$, where d is the spacing between diffraction planes) may be collected almost instantaneously. Refinements of the TOF patterns allow for not only a detailed quantitative texture analysis, but also analysis of complex crystal structures, phase proportions, grain shapes, residual stresses, and crystallographic textures, simultaneously. Various Rietveld methods provide standard analyses for HIPPO.[6,8–11]

Five design objectives were identified for the apparatus to achieve *in situ* deformation and characterization of polycrystalline ice using neutron diffraction. The first requirement was to accommodate large samples for bulk measurement in the HIPPO chamber. Second, the sample chamber of the deformation apparatus needs to be isolated from the surrounding ambient atmosphere so that a controlled environment could be maintained around the sample to protect the integrity of the sample and its crystal structure. The third requirement was to have low-temperature capability and adequate temperature control to keep the sample at a chosen temperature or to warm or cool the sample quickly. Fourth, easy access to the platform had to exist for supporting the weight of the load material (or the dead-load stage) for quick, convenient load increase or decrease. Finally, due to the cumbersome hoses normally attached to the apparatus for temperature control, we did not want to rotate the entire setup in order to achieve adequate pole-figure coverage.

To address these design objectives, first we designed an aluminum deformation apparatus to fit into the HIPPO chamber and to accommodate a sample size of approximately 2.54 cm length by 2.54 cm width. The deformation rig holds a sample at the end of a cold finger, and perpendicular to the applied load (Figure 1). Figure 2 shows an engineering sketch of the apparatus sitting in the HIPPO chamber. A load plate sits perpendicular to the sample cold finger and bears multiple circular weight plates to increase or decrease the

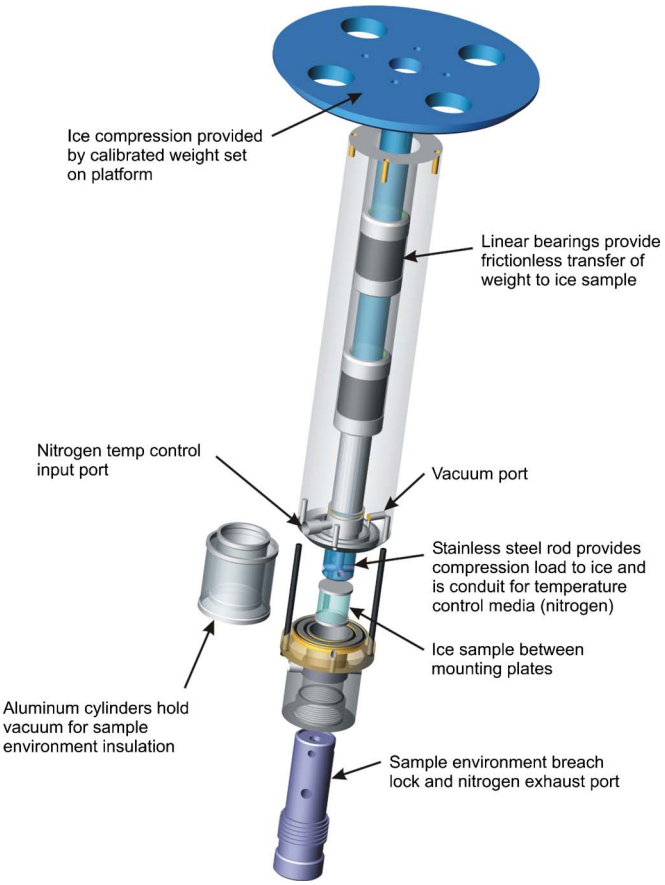

FIG. 1. (Color online) A close-up schematic diagram of the deformation apparatus. The ice sample is visible in the deformation chamber. The top of the ice sample is flushed with the piston (bearing load plates) and the bottom of the sample is held in place by the end plug. Four cooling channels sit at 90° intervals above the sample; only two are visible in this image; one is labeled. Schematic courtesy of Mark Taylor and Danny Gallant.

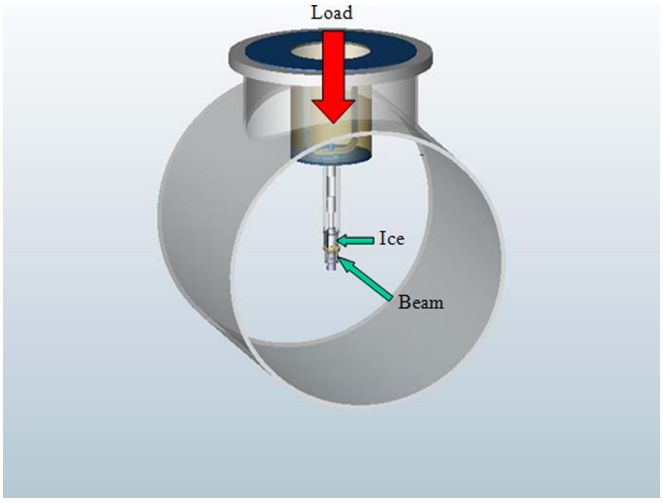

FIG. 2. (Color online) The deformation apparatus lowered into the HIPPO chamber. The weights are placed on top of the apparatus on a plate (not pictured here) directly below the red arrow marking the load direction. The direction of the neutron beam is shown. Schematic courtesy of Mark Taylor.

load. Our sample was cylindrical in shape for texture measurement to avoid absorption corrections; however, samples of different sizes and shapes could be used. A sample may be smaller in size, but cannot be larger or the apparatus would not have a tight seal, and temperature control would be compromised.

Second, to achieve temperature control in the environment around the sample, we built a vacuum chamber around the sample using 0.0580-cm-thick aluminum "windows" (Figs. 1 and 2). This aluminum is thin enough to allow easy penetration by neutrons. Aluminum is a weak coherent and incoherent scatterer of neutrons and is commonly used in sample environments.[12]

Third, we used a temperature controller to regulate the flow of liquid nitrogen to the sample chamber. To protect the sample from direct contact with the liquid-nitrogen coolant, it was wrapped in thin aluminum foil (again nearly transparent to neutrons). Thermocouples attached to the base of the sample chamber measured temperatures at the base of the sample. Low-temperature o-rings provided seals to the sample chamber for both constant temperature and vacuum maintenance.

Fourth, the rig is top loading with a series of weighted plates to minimize time to change applied loads, and can accommodate a maximum load of 667 N. We transferred the sample into the cooled sample chamber of the deformation apparatus outside the HIPPO chamber to minimize time between sample changes. The load plate can be accessed through the diffractometer "top hat," so that the rig and sample remain isolated in the chamber while load is increased or decreased.

Last, there was no need to rotate the press for texture analyses for two reasons: the advantage of a TOF analysis, and the unique design of HIPPO. Traditionally, texture was measured using monochromatic radiation (a nuclear-reactor source). Usually, diffractometers at a reactor source have detectors that move in order to collect data at different d spacings in the sample. This results in a necessary sample rotation, which is difficult with environmental cells. The diffraction pattern for the sample is collected in parts, as the detector moves, which can require a collection time on the order of many days.

In a neutron TOF measurement, neutrons with multiple wavelengths are collected simultaneously. In other words, an entire diffraction pattern is collected at once and in a matter of hours. The design of diffractometers can also be used to exploit the multiple scattering intensities available from TOF neutrons. We chose to use HIPPO because it is comprised of 1393 detector tubes grouped into 59 detector banks and arranged in a circular array. This allows texture measurements to be done on a very few sample orientations. Coverage of pole figures by HIPPO detectors [Fig. 3(a)] for this study is shown in Fig. 3(b). We did not use all available detectors in our analysis because the deformation rig blocked some of the detector panels. This coverage is adequate for a calculation of the orientation distribution function (ODF) when cylindrical symmetry can be assumed based on the applied load.

The main body of the press was built almost entirely of aluminum, which is strong enough to sustain a load, is nearly

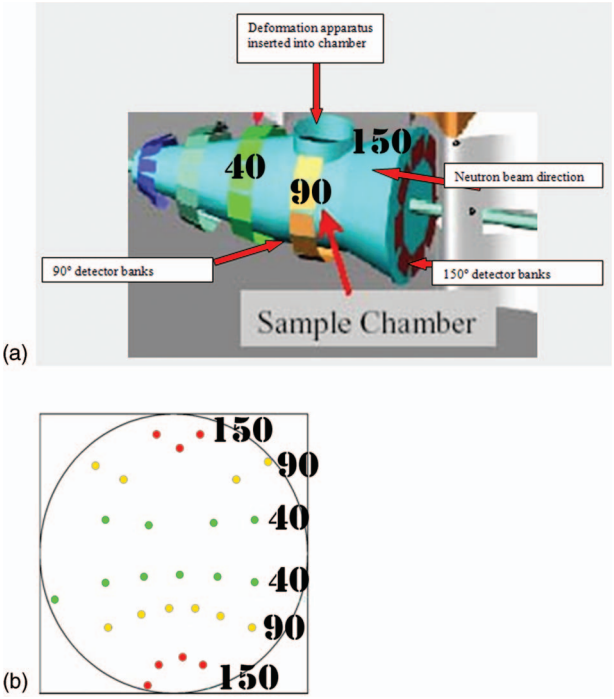

FIG. 3. (Color) (a) HIPPO detector banks and sample chamber. The detector banks are marked by different colors. The numbers of 40, 90, and 150 (deg) correspond to the angle of the detector banks in HIPPO. The circular array of these banks, which is possible with TOF neutron measurements, increases pole-figure coverage and minimizes the need for rotation of the apparatus. The neutron beam (marked with an arrow) comes into the HIPPO chamber from the right. The deformation apparatus is inserted through the top of the chamber so that the sample sits at the level of the beam. (b) Pole-figure coverage of HIPPO detector banks produced in MAUD. The numbers of 40, 90, and 150 (deg) correspond to the angle of the detector banks in HIPPO. The center of the pole figure is the compression axis. The neutron beam is going into the page, perpendicular to the pole figure.

invisible to neutrons, and can be fitted with various "top hats" and therefore adapted to several diffractometers in the Lujan Center. The piston and the ball bearings in the shaft of the cylinder that make the piston slide unhindered were built of steel. In a collected diffraction pattern, the aluminum peak from the deformation apparatus is small and does not affect peak fitting during Rietveld refinement.

EXPERIMENT AND RESULTS

Figure 4 shows the deformation apparatus with the ice sample mounted on the displex just before loading the rig into the HIPPO sample chamber. The main cylindrical shaft of the press sits perpendicular to the top hat. The sliding steel piston is attached perpendicular to a flat plate that fits inside the top hat and holds the loading plates. The piston is positioned inside the shaft of the deformation apparatus and rests against the top of the sample. The sample is inserted through the bottom of the rig and a stationary holder is screwed in flush against the bottom of the sample to hold it in place.

For the first experiment, we loaded a fine-grained (2–10 μm) D_2O polycrystalline laboratory-fabricated[3] ice sample (2.23 cm length $\times$ 2.54 cm diameter) into the deformation apparatus. The piston was flushed against the upper surface of the sample, which was shaved flat during sample preparation and loaded into the sample chamber. The defor-

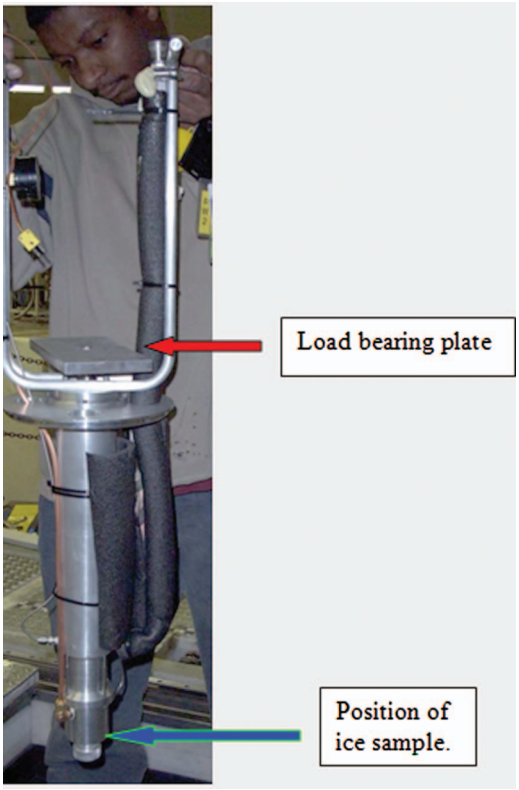

FIG. 4. (Color) The deformation rig is positioned above the HIPPO chamber, ready for insertion into the chamber. HIPPO instrument scientist Darrick Williams is holding the rig. The arrow shows location of ice sample in the deformation chamber. The load plate is visible between the U-shaped handles. Weights are placed on the plate after the deformation rig is inside the HIPPO chamber.

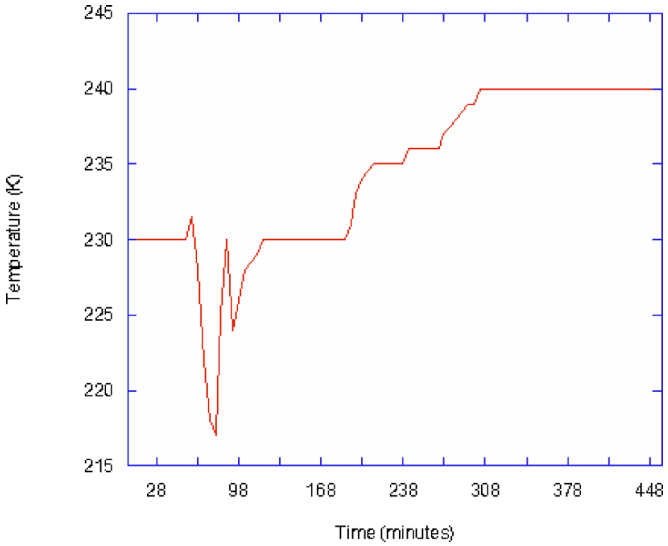

FIG. 5. (Color online) Temperatures and time spent at each temperature. Temperature fluctuated as the level of the liquid nitrogen in the Dewar decreased. The cooling lines from the Dewar to the deformation rig were long (4.6 m) which also caused a problem for maintaining a steady temperature. Future experiments on the rig will use a cooling medium that sits within or on top of the HIPPO chamber, creating shorter travel routes for the cooling medium. This will help us stabilize the temperature during an experiment.

mation rig was placed into the HIPPO chamber and a load of 20 kg was placed on the ice sample parallel to the sample axis. Using a constant load, the sample was shortened a total of 0.2667 cm in length for a total strain of 0.210. Temperature was varied between 230 and 240 K over a period of 7.5 h (Fig. 5).

Diffraction patterns were collected in 5 min intervals over 7.5 h while maintaining the constant load. Figure 6 shows the ice diffraction patterns as a function of orientation after 110 min of deformation with an axial stress of 3.8 kPa and the principal diffraction peaks for hexagonal ice (10-10), (0002), (10-11), and (10-12). Each color in the figure represents a different detector bank in the circular array of high angle (150°) banks for HIPPO, and therefore a separate orientation. If no texture were present, the individual diffraction patterns would have similar peak-height ratios throughout the patterns with respect to orientation. The difference in intensity indicates that a texture has developed in this sample.

We collected the TOF neutron data in 5 minute real-time intervals on all 1393 HIPPO detectors, and examined textures as a function of increasing temperature, using Rietveld analysis and the direct inversion method material analysis using diffraction (MAUD) texture package. [10] MAUD uses a Marquardt least-squares method to fit diffraction powder peaks and to obtain the ODF for a texture analysis. We used the extended-Williams-Imhof-Matthies-Vinel (E-WIMV) algorithm direct method to compute our spectra collected on HIPPO.[13] We used the program to calculate the pole figures for the relevant ice diffraction peaks.

Figures 7(a) and 7(b) show the (10-10), (0002), (10-11), and (10-12) pole figures for the ice sample twice during the 7.5 h of total deformation. Pole figures are plotted in equal-area projection in multiples of random distribution (m.r.d.) with the direction of stress in the center of the pole figure. Figure 7(a) shows pole figures measured at $T=230$ K after 148 min of total deformation time (out of 450 min). Figure 7(b) shows pole figures at $T=240$ K after 448 min of deformation.

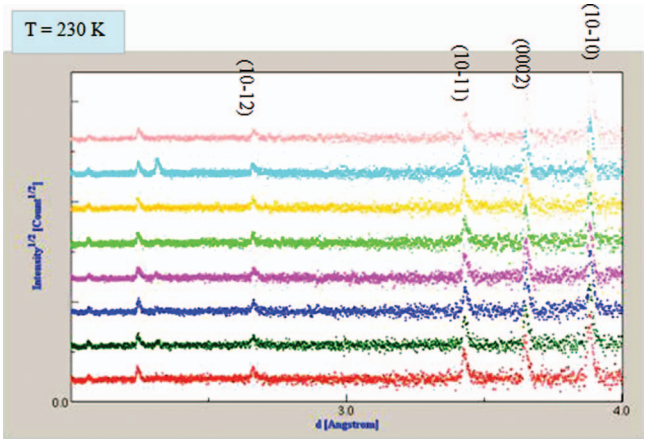

FIG. 6. (Color) Ice diffraction pattern in HIPPO of high-angle (150°) detector banks (background subtracted) after 148 min of deformation at a temperature of 230 K. Each color represents a different detector bank in the circular array of high angle banks for HIPPO. The vertical axis shows intensity scaled by counts, or the number of counts of neutrons reflected off different crystallographic planes. The four prominent peaks for hexagonal ice are labeled with their Miller indices (*hkil*).

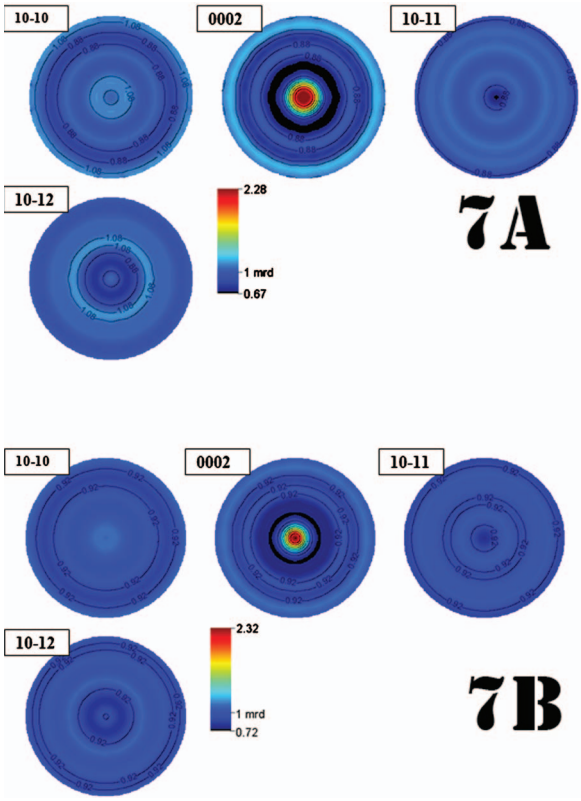

FIG. 7. (Color) (a) Equal-area projection of pole-density distribution in ice after 148 min of deformation with temperature at 230 K. (b) Equal-area projection of pole-density distribution in ice after 448 min of deformation. Temperature was 240 K.

Texture analyses of these two time intervals reveal a similar pole distribution for the sample as time increases, with twice as many (0002) poles parallel to the compression axis than as normal to it. The sample has 2.28 m.r.d. at 148 min and 2.32 m.r.d. at 448 min.

DISCUSSION

This work demonstrates a first successful measurement of texture in polycrystalline ice I_h during *in situ* low-temperature deformation under constant load. First results compare well with previous results on texture in ice I_h. Natural glacial samples show this development of *c* axes oriented parallel to the compression direction arising from slip on the basal plane.[14,15] Models also demonstrate this dominance of the basal plane.[16] Results show that there are approximately twice as many (0002) poles parallel to, than perpendicular to, the axis of compression. This result is expected at this temperature regime where laboratory experiments have shown that easy basal slip occurs in ice at these temperatures, allowing crystallographic *c* axes to rotate and align with the compression direction.[17,18]

We could improve these pole figures by (1) increasing the number of neutron counts and (2) rotating the apparatus in the HIPPO chamber. Collecting for longer count intervals would give sharper diffraction peaks, making Rietveld refinement more accurate. Also, rotating the deformation apparatus inside the HIPPO chamber would achieve better pole figure coverage. With ice, which in this case has fiber texture, rotation was not necessary for an adequate ODF calculation. However, with a sample of lower symmetry, rotation is necessary.

Overall, the apparatus design proved successful. Temperature control could be improved by placing a cold source such as liquid nitrogen or liquid hydrogen in a Dewar located inside the HIPPO chamber or directly above the HIPPO door. Also, placing the cooling medium in the HIPPO chamber would allow for rotation of the deformation rig; one rotation of 90° or 180° would improve the coverage of pole figures.

The texture results are expected. With increasing strain and warming temperatures, we anticipated seeing the texture increase. Yet the texture of the sample does not significantly increase with time. We present two possible explanations for our results: the texture could be inherited from the initial sample or a balance has arisen between strain-causing-fabric development and recrystallization. Since we were unable to measure the initial texture of the sample with this deformation apparatus, we do not know if the ice had a completely random initial texture or if some texture was imparted in the making of the initial material. The sample receives an immediate load from the weight-bearing platform at the top of the deformation rig upon placement in the apparatus. Future experiments should be performed on a material whose initial texture is known.

Future work in ice deformation and texture analyses includes *in situ* characterization of texture development at various temperatures and ambient pressure conditions as a means of exploring dominant deformation mechanisms in ice. Geologists, materials scientists, and chemists could use this rig to explore any low-temperature or ambient temperature material parameters that would respond to the load applicable by this apparatus. Also, research in liquid-chemistry fields where solids may form under varying temperature conditions could also be carried out with this rig.

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