# FINAL REPORT (Phase I)

# Top Loading Helium Cryostat Integrated with High-Pressure Cell with Fast Remote Pressure Control

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Grant # DE-SC0019678

## ABSTRACT

***Purpose of the research:*** A large number of physical phenomena, such as superconductivity and quantum critical phenomena, often appear only at very low temperatures below 5 K. There is an immense interest to investigate these phenomena at high pressure as a means of tuning interatomic distances, and thus the interaction parameters controlling these phenomena, in a continuous and controlled fashion. The current P-T condition for neutron scattering experiments are limited to either relatively low pressures of about 2 GPa at temperatures below 5 K, or to relatively high temperatures at pressures or tens of GPa. The purpose of this research is to develop neutron sample environment instrumentation for reaching 10-20 GPa at 1-2K with rapid and reliable online pressure and temperature control (i.e. without having to interrupt the experiment).

***Description of the research:*** The ultimate goal of the project is to design an integrated fast-cooling low-temperature sample environment cryogenic system compatible with state of the art neutron diamond anvil cells suitable for single-crystal neutron scattering experiments for temperatures down to 2K and pressures of several tens of GPa. The integrated system will consist of top-loading Helium flow cryostat with in-situ sample alignment mechanisms, large-volume diamond anvil cells (DAC) made from novel superalloy Pascalloy and optimized for fast cooling and heating, and a compact remote pressure control mechanism for the DAC based on a novel concept of inflatable bellows integrated with a lever-arm based force amplifier.

***Results of the research:*** In Phase I, we have designed, manufactured, and tested prototypes of the novel compact force-amplified pneumatic pressure control mechanism for Neutron Diamond Anvil Cell (nDAC), which allows to use pneumatic bellows system for smooth remote pressure control in the nDAC inside a top-loading cryostat with bore size of 70 mm or larger. This allows significant minimum temperature decrease in remotely controlled nDAC from 5-10 K down to 2 K. We also studied mechanical properties of a novel non-magnetic superalloy Pascalloy with different heat-treatment conditions and preliminary results indicate that up to date this is probably the strongest and most suitable material for making more compact and lightweight cryogenic nDACs for neuron scattering experiments at extreme conditions, allowing much faster cooling and heating.

***Potential applications:*** The new developments will allow to create a wide range of compact Diamond Anvil Cells for neutron diffraction which can reach several tens of GPA pressure at 2-4 K while preserving accurate remote pressure control capabilities. The new concepts can be used for developing sample environment instrumentation outside the neutron scattering field. The new development have very strong potential for expanding experimental capabilities in materials sciences and have high commercialization potential. The proposed new cryogenic high-pressure system or any of its individual components will be in high demand not only in neutron scattering facilities, but also at synchrotron beamlines and other high-pressure research facilities around the world.

# 1. Overview

The DOE 2019 SBIR Phase I Program Solicitation requested grant applications under INSTRUMENTATION AND TOOLS FOR MATERIALS RESEARCH USING NEUTRON SCATTERING Topic 17a: "Advanced Sample Environments" to develop faster cooling furnaces and cryostats. DAC Tools LLC, in collaboration with the Neutron scattering facilities at the Oak Ridge National Laboratory, has submitted the proposal to develop a top loading Helium cryostat integrated with high-pressure cell with remote pneumo-mechanical pressure control.

A large number of physical phenomena, such as superconductivity, magnetic ordering, and quantum critical instabilities, often appear only at very low temperatures below 5 K. There is an immense interest to investigate these phenomena at high pressure as a means of tuning interatomic distances, and thus the interaction parameters controlling these phenomena, in a continuous and controlled fashion. Typically high pressure experiments in tens of GPa range are more suited for synchrotron techniques, where the x-ray beams can be readily focused to (sub) micron sizes and samples with only several micrometers in size can be studied. Nevertheless, neutron diffraction and scattering offers studying properties and phenomena than cannot be distinguished by x-rays, such as magnetic structure determination and crystallographic measurements on many important systems containing light elements such as H (and D), Li, Be, B, C. Another strength of neutrons lies in single crystal diffraction which allows the exploration of nuclear and magnetic structures as a function of pressure, temperature and magnetic field and gives particular insights into diffuse scattering and hydrogen within the lattice.

The simultaneous generation of high pressure and ultra-low temperatures is a well-known problem for neutron scattering, in particular at multi-GPa pressures where the pressure cells are rather massive and therefore need more complex cryogenic and pressure control solutions. Thus, the current P-T conditions for neutron scattering experiments are limited to either relatively low pressures of about 2 GPa at temperatures below 5 K, or to relatively high temperatures at pressures of tens of GPa [4]. The ability to reach 10-20 GPa at 1-2K and having the ability to control pressure and temperature rapidly and reliably online (i.e. without having to interrupt the experiment and while minimizing setup and sample alignment time) is critical for understanding the behavior of quantum and other multifunctional materials at such extreme conditions.

DAC Tools has proposed to design an integrated fast-cooling low-temperature sample environment cryogenic system compatible with state of the art diamond anvil cells (DAC) suitable for single-crystal neutron scattering experiments for temperatures down to 2K and pressures of several tens of GPa. The integrated system would consist of top-loading Helium flow cryostat with in-situ sample alignment mechanisms, large-volume DACs optimized for fast cooling and heating by using novel non-magnetic materials, and a compact pressure control mechanism for the DAC based on a novel concept of inflatable membranes (or high-pressure bellows) integrated with a lever-arm based force amplifier. For Phase 1 of the SBIR grant the goal of DAC Tools was to develop such instrumentation that allows to remotely control and measure pressure in "large-volume" neutron scattering compatible DACs in an accurate and consistent manner. Such remote pressure change should be done without the need for time-consuming thermal cycling and avoiding the drawbacks associated with the radiological constraints of handling the irradiated high pressure assembly, which are crucial for effective operation of modern neutron facilities and efficient utilization of the valuable neutron beam time for experiments.

The proposed developments can be potentially utilized at dozens of neutron instruments all over the world, including existing and future beamlines at ORNL (HFIR, SNS and STS), NIST, ILL ISIS, ESS, etc. Moreover, individual components of the proposed development can be used separately from the rest of the system. For example, the new more compact non-magnetic DACs made from a non-magnetic superalloy Pascalloy (a novel domestic version of 40HNU-VI or "Russian alloy") can find application in tens, if not hundreds, of laboratories in the worlds dealing with superconductivity and electric/magnetic properties of materials at extreme conditions. The compact pneumo-mechanical device for remote and accurate pressure control in opposite / diamond anvil cells can become very popular across the disciplines using high pressure techniques, especially when the size and properties (e.g. magnetic) of the high-pressure device are of a significant concern (e.g. in cryostats and high magnetic field environments).

In the course of performing the proposed research DAC Tools was maintaining close collaboration with the staff from Oak Ridge National Laboratory, Spallation Neutron Source and High Flux Isotope Reactor: António Moreira dos Santos, Clarina Dela Cruz, Bianca Haberl, and Reinhard Boehler. This collaboration will be continued in the future independently of the further involvement of DAC Tools into the relevant SBIR program.

# 2. Phase I technical objectives

The ultimate goal of the project is to develop a multi-component high-pressure cryogenic system for neutron diffraction and scattering. The target system should be capable of reaching pressure in excess of 10 GPa with large sample volume sufficient for high-quality neutron diffraction and scattering at modern neutron facilities while reaching temperatures below 5K, with the immediate goal of 2 K. The cell should be made of optimized materials having highest strength to weight ratio to minimize the size of the Diamond cell thus allowing fast temperature change on cooling and heating. The diamond cell should have a compact pressure control mechanist which can generate remotely controllable load in the order of 10-12 tons and provide smooth pressure change at base temperatures without the need of significant thermal cycling of the system.

The Work plan for Phase I was developed to specifically address the primary goals listed above and included the following tasks:

1. Make a preliminary design of the compact remote pressure control DAC attachment based on pneumatic drive with lever arm force amplification mechanism.
2. Perform FEA analysis and optimize the design for different practical loads and displacement.
3. Manufacture the pneumo-mechanical device / attachment for remote pressure control and load testing frame.
4. Perform in-house tests and analyze the data.
5. Test the device with currently operating diamond cells at ORNL at cryogenic conditions.
6. Heat treat samples of Pascalloy to different temper conditions and arrange strength/toughness tests in the outside facilities at liquid helium temperatures.
7. Based on the results of #5 make a preliminary design of the top-loading "wet" cryostat (if schedule permits).
8. Final report preparation. Formulation of the proposal for the Phase II program.

In the execution of Phase I, we have completed all the proposed tasks except for tentative #7. The preliminary design was supposed to be done in collaboration with a major US cryogenic company, such as Janis Research, but due limited time and technical issues on Janis Research side the execution of #7 was postponed.

Key performance metrics that were obtained during Phase I work are the successful design and test of the compact force-amplified pressure control mechanism and preliminary tests of the strength and toughness of Pascalloy with different tempers at cryogenic conditions. These metrics would determine the further directions of the development, i.e. type and size of the cryostat, alternative pressure control mechanisms below 5K, as well as other issues identified in the course of the development in case DAC Tools will continue pursuing this development in the future. The compact force-amplified pressure control mechanism have been manufactures and tested and now it can be used at neutron facilities either as is or new modifications for different nDACs can be readily made based on the new design.

# 3. Results of the research: Remote pressure control system

## 3.1. Overview

Pneumatic way or remote pressure control in cryogenic diamond anvil cells was introduced many decades ago right after the invention of the Diamond Anvil Cell [5] and is by far the most common and convenient way or accurate remote pressure control, especially at non-ambient temperatures [2, 6-10]. Thus bellows and diaphragms/membranes remain very popular, specifically at cryogenic conditions, because of their simplicity and efficiency. Typically helium gas (which has lowest freezing temperature) inflates either a bellows [9], double-diaphragm [2] or single-sheet membrane [11] which produces axial displacement and applies force to the diamond anvils, thus generating pressure in a sample chamber squeezed between two diamonds.

In diamond anvil cells the pressure is generated by applying some load on the frame which drives the diamonds closed and squeezes / pressurizes the sample. Pressure is merely force divided by area (P=F/A). 1 bar approximately corresponds to a load of 1 kg over the surface area of 1 $cm^2$. Therefore for DACs with a typical diamond culet of 350 µm (~0.1 $mm^2$) 10 kN (~1 ton) of force will translate into approximately 1 megabar (100 GPa) pressure. Typical DAC membranes with 40-50 mm OD (and area of 10-15 $cm^2$) are capable of generating loads of 1-2 tons with 2200 psi (150 bar) gas pressure [6], and this is sufficient for reaching megabar pressure in majority of conventional DACs, covering the complete pressure range in >95% of experiments.

Unlike regular diamond anvil cells, in neutron DACs with culet sizes 1-3 mm [3, 12] the sample size is orders of magnitude larger than in regular DACs, and order of magnitude larger forces/loads are required. Increasing membrane gas pressure above the standard 2200 PSI is a possible option which is unfortunately a serious safety concern and technical challenge. Another simple solution - to scale up the membrane to generate a required load of 11-12 tons - was proposed by the PI and realized by the SNS/CIW team several years ago [3] (Figure 1). The scaled-up double leaf membrane is with 100 mm OD is capable of generating the load of 12 tones when properly clamped [12]. Such load is normally sufficient for high pressure work high at neutron facilities.

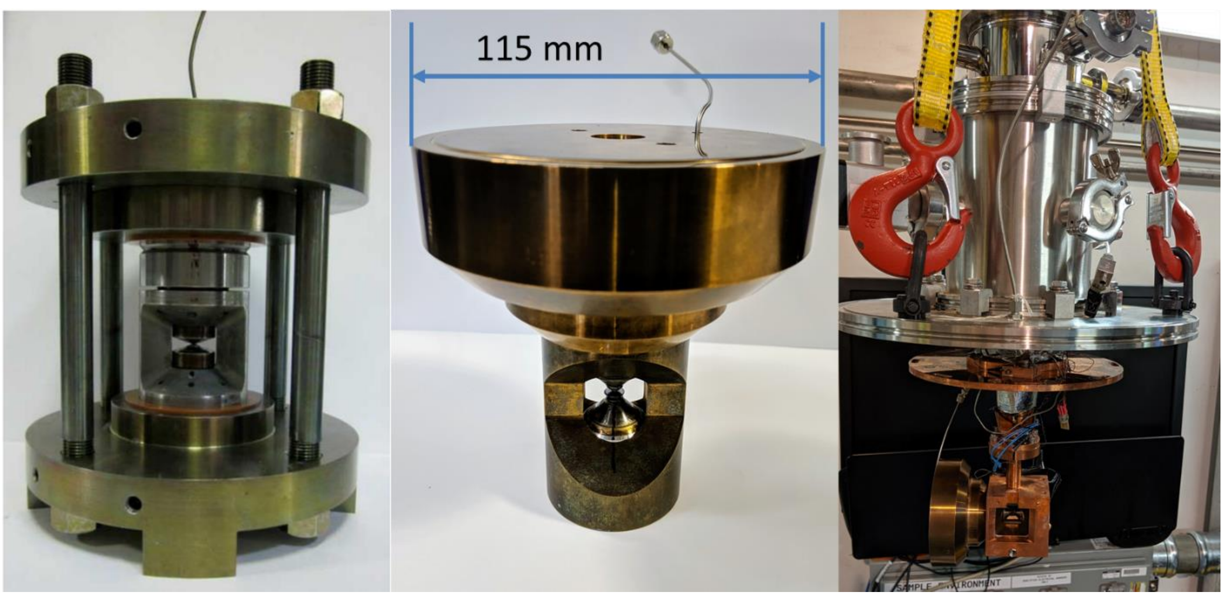


*Figure 1. Left: Tie-rod pressurizing frame for 100 mm OD double-diaphragm[2] membrane and neutron DAC (early versions [3]); Center: New membrane attachment for neutron DAC and 100 mm OD membrane; Right: the neutron DAC – membrane assembly in bottom-loading cold finder cryostat at SNAP / SNS / ORNL.*

While the concept was proven to be successful for many different experiments, being currently a common pressure drive for neutron DACs at SNAP/SNS, there are still a lot of inconveniences and restrictions associated with such large membrane. In addition to huge thermal mass of the existing device and significant (more than 5 hours) amount of time required to cool down the nDAC to a working temperature of 8-9K.

The other major problem is the compatibility of the membrane drive with other neutron instrumentation. The majority of top-loading cryostats at SNS and HFIR at ORNL have a sample bore of 100 mm or smaller. The enclosure for the 100 mm membrane has an OD of ~115 mm – too large to fit into standard cryostat bores. It is possible to scale down the membrane to make the drive compatible with the cryostat, but this will decrease membrane efficiency and generated pressure, and the thermal weight would still be very significant if not prohibitive for "sample-in-exchange gas" cryostats. Thus one of the most important objectives of this work was to design a pneumatic system attachable to the neutron DAC (e.g. [12]) which has a relatively small size (e.g OD < 70 mm to fit into mid-sized top-loading cryostat) and capable of generating 12 tons of useful load at liquid helium temperatures.

## 3.2. Design goals

Unlike regular Diamond Anvil Cells, the neutron DACs have a number of non-obvious specifics not typical for regular DACs. For example, in order to pressurize the sample to several tens of GPa the distance between diamond culets needs to be decreased by less than 100-200 micrometers. Nevertheless the load in the order of 10-12 tons causes serious mostly elastic deformation (elongation) of the DAC body, and therefore the effective stroke of the pressurizing device should be in the order of 0.5 – 1.0 mm rather than 0.1-0.2 mm. This effective stroke of 1 mm (after mechanical amplification) with a load capacity of 12 tons was set a design requirement.

Another design goal was the overall Outside Diameter of the pressurizing attachment. Even though 100 mm bore top loading cryostats do exist and are available at neutron facilities, the top loading cryostats with 70 mm bores are more common and more versatile and efficient. Thus another design goal was to constrain the OD of the pressurizing attachment not to exceed 69 mm.

Other design goals included full compatibility with common neutron DACs used at SNAP, ease of use (assembly and disassembly), ease of adapting to other DACs, and ability to bench test the device without using actual DACs. In addition, the compatibility of the assembly with sample sticks for top loading cryostats was mandatory, but technically did not present any serious design problem.

## 3.3. Conceptual design

The conceptual design of pneumo-mechanical device with <70 mm outside diameter capable of generating of 12 tons of useful load from bellows with 2200 PSI gas pressure (with up to 5X lever force multiplier) was given in the original proposal (Figure 2). While it would effectively work for small loads, the initial design turned out to be

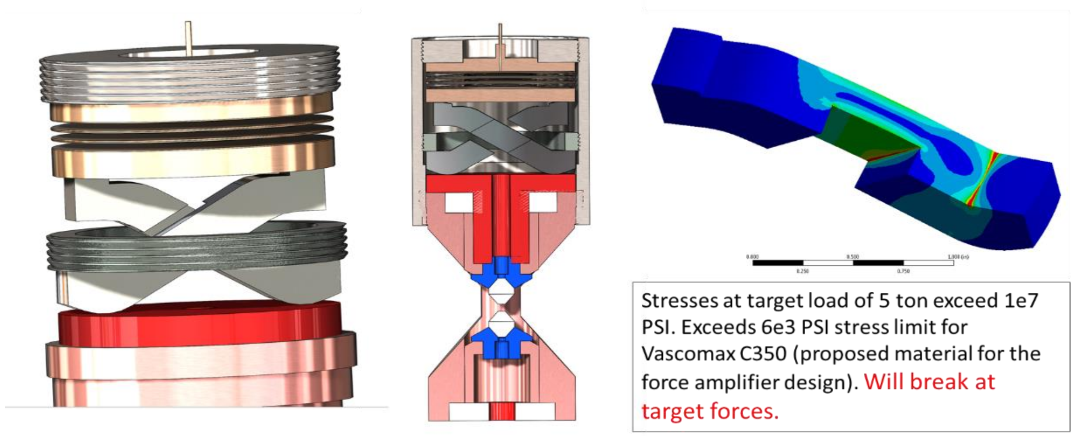


*Figure 2. Initial conceptual design of the compact membrane / bellows lever-based force multiplier.*

too weak and the levers would break far before reaching the target load of 6 tons each (12/2). Therefore we were forced to look for a different lever geometry and possibly to a different configuration and number of levers.

### 3.4. Finite element optimization of lever arm mechanism

After it became clear that the lever shape suggested in the conceptual design is not capable of handling 12 tons of load (or proportional fraction of it), we put a very significant effort into searching for other lever shapes and arrangements. Figure 4 shows a variety of proposed lever shapes for two-level configuration. All of them were carefully analyzed and neither shape and/or arrangement was found suitable for generating 12 tons of load with 1 mm payload displacement (or at least 1 to 3 force amplification ratio).

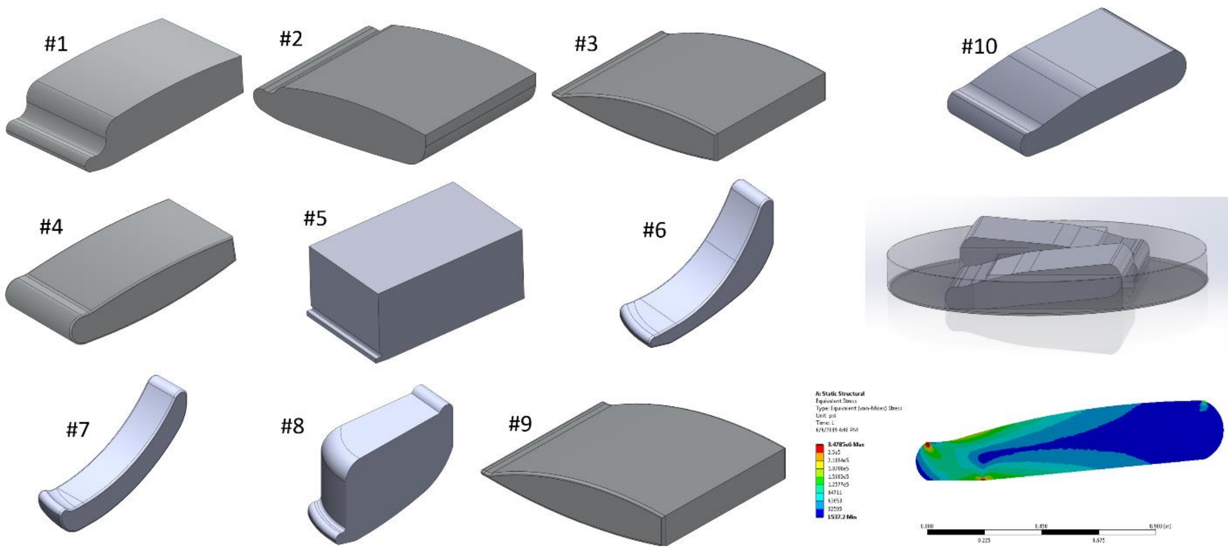


*Figure 4. Different shapes of levers considered and analyzed for this proposal. Neither of these shapes found satisfactory to satisfy all requirements.*

### 3.5. Working design

After extensive search for the design of the lever arms and multiple FEA iteration on each design we have converged on a design with three shorter non-crossing levers (Figure 3).

Even though the levers are much shorter than original, they are fully capable of providing the working displacement of 1 mm with force amplification of ~3 (due to geometry the force amplification ratio changes as the pushing piston is getting displaced). For cryogenic use all parts of the assembly are made of maraging steel Vascomax C-300 heat treated to HRc 54-56, and the levers themselves are made of Vascomax C-350 hardened and tempered to the hardness of HRc 58-60. Special care was taken to manufacture the levers in such a way that they do not have any notches or other places of

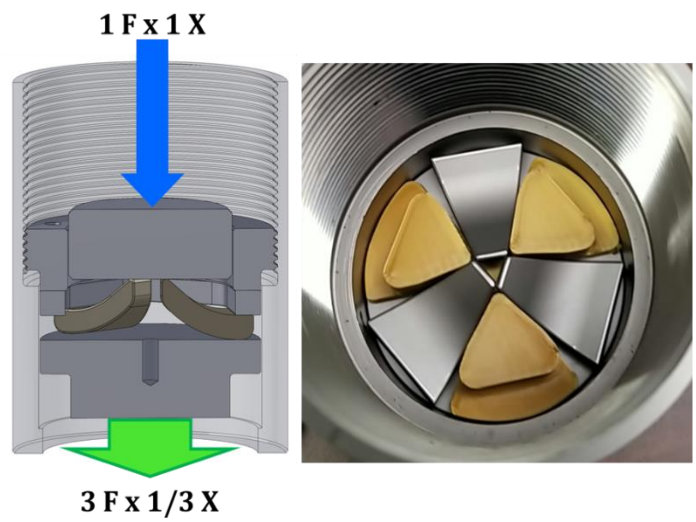


*Figure 3. Left: The design of Bellows Force Amplifier with three levers. In current configuration the displacement is decreased and the force is increased by about 3 times. Right: Top view of the assembly; three M11 levers are in the middle separated with yellow 3D printed PLA spacers.*

stress concentration, which can result in premature breakage. The FEA calculations show that if the levers are manufactured and heat treated correctly – they can withstand required loads and provide safety factor exceeding 1.8 (Figure 5).

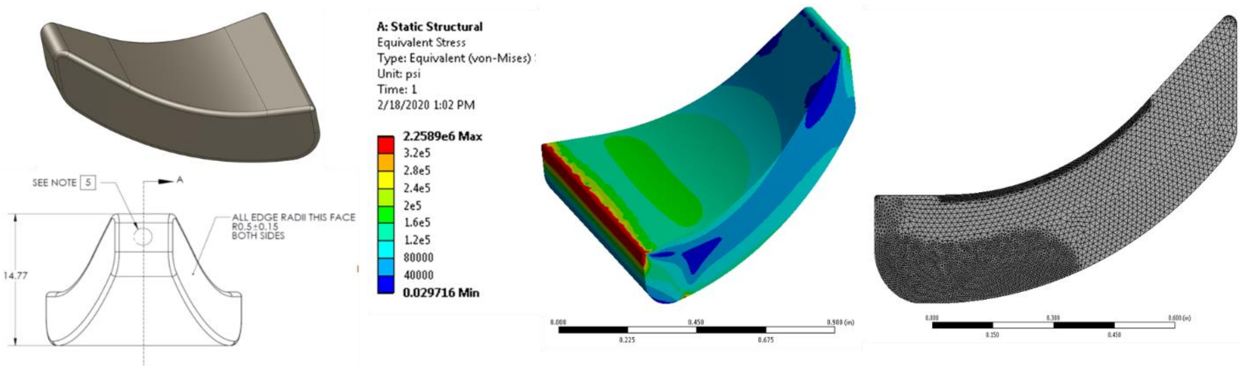


*Figure 5. FEA calculations for the M11 level. For Vascomax C-350 with yield strength of 320 ksi and load of 4 tons (12/3) the maximum calculated stress is ~172 ksi, corresponding to a safety factor of ~1.86.*

## 3.6. Bellows design and manufacturing

The design goals for the bellows included the following specifications:

- Diameter of about 60 mm to maximize the generated force but still be compatible with 70 mm core cryostat;
- Rigid (~4-6 mm thick) top and bottom;
- Made of Series 300 (304, 316) stainless steel which does not become brittle at cryogenic conditions.
- Flexible 1/32” - 1/16” gas capillary;
- Up to 3 mm displacement;
- Operating at pressures up to 2,500 PSI in constrained confinement.

None of the commercial companies were able to quote such bellows because the typical laser edge welded bellows are operating at significantly lower pressure. Finally we managed to convince a domestic company Metal Flex Welded Bellows Inc. to make several custom bellows for our purposes on the promise that because this is a

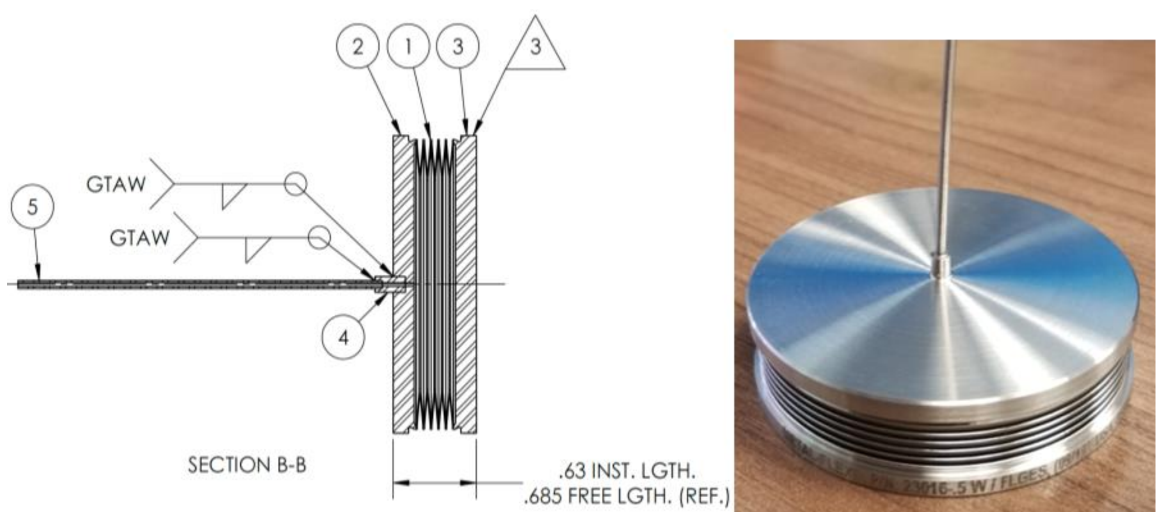


*Figure 6. 2.3” bellows custom designed and made for this project.*

research and development project - we would not hold them responsible for bellows failure as we would be taking them far outside of standard operating conditions.

The final design and a photograph of the manufactured bellows is shown on Figure 6. The bellows are custom machines and welded from SS316 (convolutions) and SS304 (the rest).

The effective OD of the bellows is 2.3" (58.4 mm), which corresponds to an area of 26.8 $cm^2$ or 4.14 $in^2$. Therefore with gas pressure of 2,200 PSI the bare bellows should be able to generate a pushing force of ~9140 pounds or ~4150 kg. This was later confirmed during bench top tests.

The bellows of this size with this number of convolutions (5) are rated for normal displacement of ±3 mm (total 6 mm). This range was adequate since we did not anticipate extending the bellows by more than 1.5-2.0 mm in normal working conditions.

The thickness of the convolution leaflets was 0.006" / 0.15 mm, and was determined by the capabilities of punching and shaping equipment at bellows manufacturing facility. The bellows with such thin walls are more prone to plastic deformation which decreases their high-pressure service life and a number of cycles. For the future we should try to order the bellows with thicker walls and larger number of convolutions.

One of unwelcome surprised was the fact that the bellows can leak through very thick (about 5 mm) stainless steel rigid plates (Figure 7). One of four custom made bellows had a pinhole in the middle of the thick plate which was leaking air at high pressure. The rate of pressure loss was measurable bur somewhat acceptable for bench tests with pressurized air. It would not have been acceptable for actual experiment with pressurized helium. Perhaps this king of defect in the stainless steel stock is extremely rare, but nevertheless it is worth checking the bellows for such kings of unexpected leaks before actual experiments.

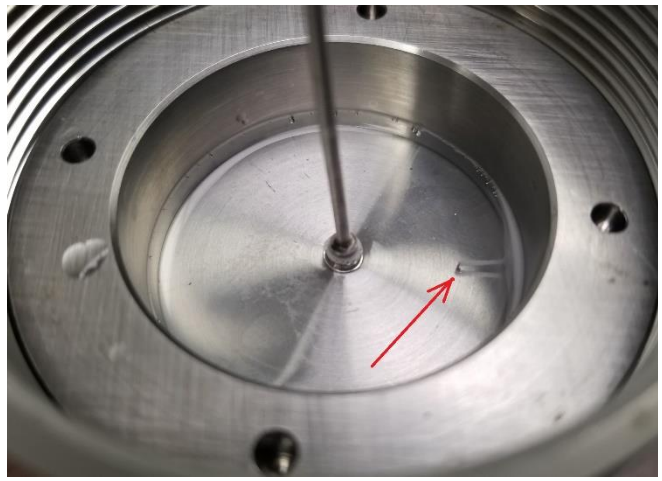

*Figure 7. Pinhole the bellows top plate.*

### 3.7. Bench tests at DAC Tools LLC

After the force amplifier assembly and bellows were manufactured, we have performed a series of benchtop test of the device both without levers and with lever arm amplification. For benchtop tests we used configuration shown on Figure 8 with a special transducer container and a load cell LCMWD-100KN capable of handling loads up to 200 kN (~20 tons). The numbers correspond to the following elements: 1-Force Amplifier Container; 2-Lever M11; 3-Load Plate; 4-Mushroom Piston vSS440; 5-Transducer Container; 6-Dummy Bellows; 7-Lower Retaining Plate; 8-Piston; 9-Bellows Retaining Ring; 11-Load cell Omega LCMWD-100KN; 11-Set Screw.

The actual setup for benchtop measurements is shown on Figure 9.

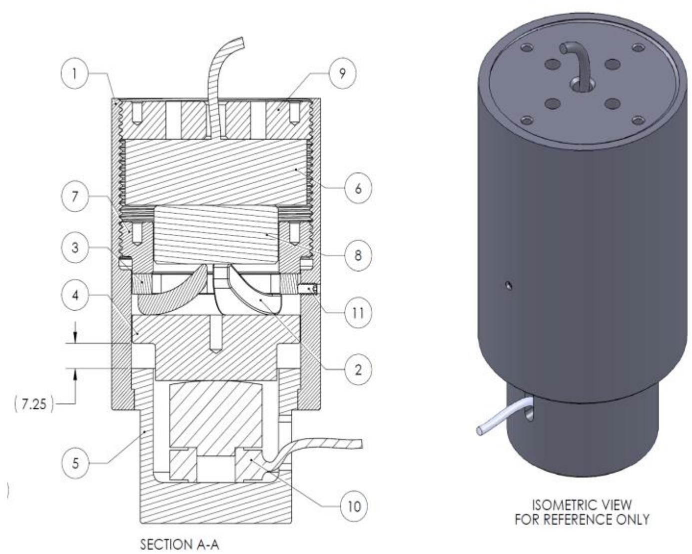


*Figure 8. Bellows force intensifier prototype – Force amplifier assembly M11.*

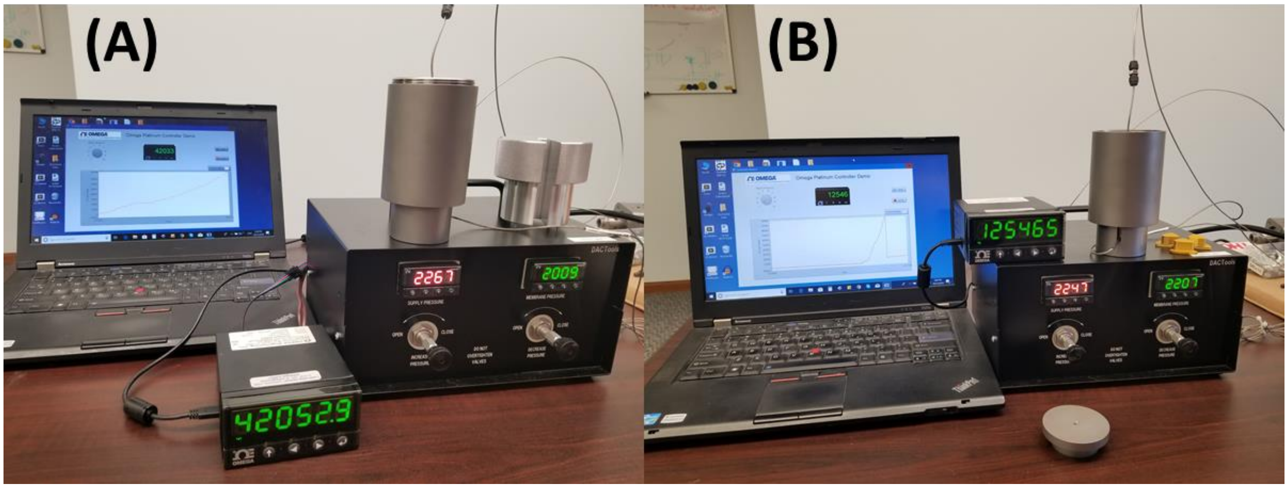


*Figure 9. Benchtop setup at DAC Tools LLC facility for testing the Bellows Force Amplified assembly and measuring Gas pressure – Generated force relations. The left picture (A) shows the test of bare bellows without lever amplification (force of 42 kN at 2,009 PSI of gas pressure). The right figure (B) shows a useful load of 125 kN (>12 tons) at 2,207 PSI of gas pressure.*

The measurements were performed in two modes – just bare bellows without any lever amplification (4 runs) and bellows with lever amplification (3 runs) (see Figure 10). The bellows were pressurized to a maximum gas pressure of ~2,300 PSI. All runs with and without lever amplification showed consistent results and generated predictable pressures. Thus in all four runs without lever amplification we reached at least 40 kN (~4 tons) with bare bellows. While using the lever amplification we have reached about 3 times of this load (at least 120 kN or ~12 tons) in all runs. Note that due to the shape of the levers the amplification ratio varies in the range of ~3-4.

Note that when the lever force amplifiers are used the gas pressure – generated load curves show significant hysteresis in unloading curves. This is most probably related to plastic deformation of the thin stainless steel material in bellows convolutions. When the load is removed – pressure – force curve pretty much follows the same path as in earlier run. Some hysteresis is also observed in base of bare bellows, but because during those test the bellows were more constrained and did not experience the same amount of plastic deformation – the hysteresis curve was significantly smaller.

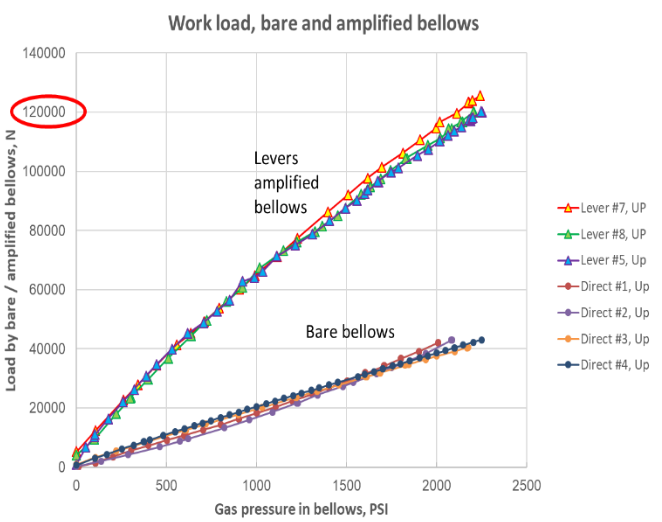


*Figure 11. Bench top test of the Bellows Force Amplifier attachment with (triangles) and without (circles) levers.*

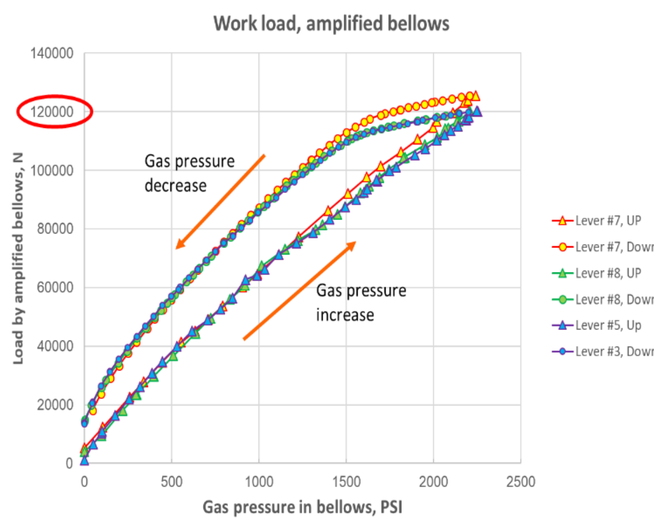


*Figure 10. Bench top test of the Bellows Force Amplifier attachment with levers. The hysteresis in loading unloading curves due to plastic deformation of the bellows is clearly seen.*

### 3.8. Bellows displacement limit - rupture tests

In any work which involves pneumatic membranes and bellows it is critical to know the extension limits of the devices. The normal working range of such bellows is ±3 mm (total 6 mm), and this should be sufficient for compressing Diamond Anvil Cell (which requires maximum bellows extension not more than 2-3 mm). Nevertheless such bellows, manufactured by MetalFlex Welded Bellows are rated to very small pressure of only a few Bars while we need up to 140 Bar for successful operation. To check the expansion limit of the bellows at maximum working pressure we performed the following tests: the bellows were placed into the force amplifier container and the maximum allowed extension was varied in 2 mm increments (2, 4, 6, 8, 10 mm). At each constrained extension the bellows were pressurized to 2500 PSI (~170 bar). At each extension length the bellows were holding the pressure with out any signs of leaking, and at 100 mm extension the bellows burst and one weld (between the bellows rigid base and the flexible leaflets) was destroyed. With normal bellows extension of 1-2 mm this 10 mm extension at burst represents the safety factor of 5-10 and is considered to be safe for use in the National Laboratory.

*Figure 12. Broken bellows – the bellows ruptured at ~10 mm displacement at 2500 PSI.*

Before conducting tests in SNS / ORNL we have conducted safety assessment with SNS/ORNL team to make sure that even if the bellows burst at – this would not hurt any people or result in serious damage to the equipment. The estimates show that with bellows area of ~28 cm$^2$, at 2 mm displacement the bellows would hold ~7 cm$^3$ of compressed gas. If the bellows would rupture at 150 bar – it will result in producing / releasing approximately one liter or gas at 1 bar. If the bellows burst in a cryostat – which typically has a volume of several tens of liters, this would result in a local pressure increase of several hundredth (0.01-0.05) of a Bar. Since the bellows are constrained in a very strong container – the gas will escape the container rather slowly thru threads and service holes, and any burst will not result in a catastrophic event. As a matter of fact – during the first test at SNAL / SNS / ORNL, the bellows ruptured at ~68 Bar and lost pressure. This was accompanied by a muffled pop sound, but nothing has moved since as a safety precautions the DAC with the bellows assembly was placed into a protective sleeve and the pressurizing air has escaped the device through multiple paths (Figure 13, right picture).

### 3.9. Room temperature tests SNAP, SNS, ORNL

The first test with an actual diamond anvil cell have been performed at SNAP beamline of SNS / ORNL on November 11-12, 2019 together with SNS crew (António Moreira dos Santos, Jamie Molaison, Bianca Haberl, and Reinhard Boehler). Unfortunately during these test there was no way of measuring pressure inside the Diamond Anvil Cell as the diamond used were polycrystalline non-transparent and could not be used with optical ruby fluorescence pressure measurement system, while measuring pressure with neutron beam was not available at that time.

The "Standard" SNAP Neutron Diamond Anvil Cell with attached Bellow Force Amplifier is shown on Figure 13 (left). The left figure also shows the original membrane canister/attachment for this DAC. It has significantly larger outside diameter (~115 mm) than the new attachment (~68 mm) and cannot be used in top-loading cryostats (maximum bore size of 70 and 100 mm) capable of cooling the cell to 2-4 K. In addition, the thermal mass of the new device is several times smaller, which allows to cool and warm the diamond cell with the attachment at least two times faster.

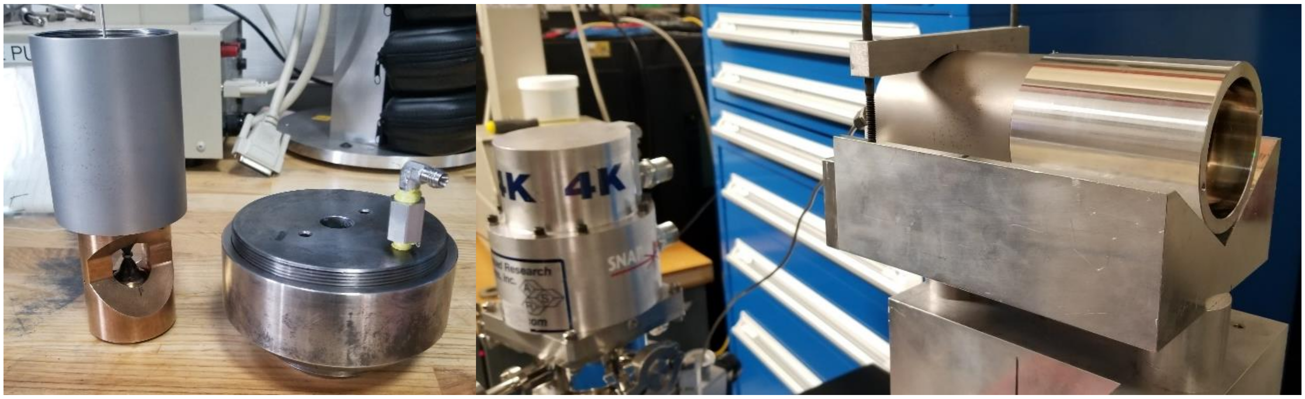

*Figure 13. Left: "Standard" SNAP Neutron Diamond Anvil Cell with attached Bellows Force Amplifier (68 mm OD), on the right there is an original 100 mm membrane attachment (~115 mm OD). Both pneumatic attachments are capable of providing 12 ton push on the diamonds in the Diamond Cell. Right. The Neutron Diamond Cell with Bellows Force Amplifier during the first bellows high-pressure test.*

During this visit we have tested the Bellows Force Amplifier attachment. The Diamond Cell was kept inside SNAP experimental station but outside the cryostat or any other containment. As a safety measure the body of the diamond cell was covered with aluminum tube to prevent diamonds from flying in case of diamond breakage. For that test we used bellows which have been extended before (with measurable plastic deformation) and recompressed to the original size. During this test we were able to reach only ~68 Bar (~9,860 PSI) – at that pressure the bellows ruptured and gas pressure was lost. This premature failure of the bellows can be explained by previous pressure cycling and accumulated plastic deformation and fatigue. It is clear that such bellows can be considered consumables (at a price of less than $1K per custom bellows), but nevertheless they can be used multiple times (as typical high-pressure membranes) if treated properly. Therefore it is important to understand the usable life cycle of these bellows – how many times can they be used, what level of extension and plastic deformation is acceptable, what are the signs of fatigue indicating that this particular bellows can not be used anymore, and so on. This work will eb continued in the future.

### 3.10. Cryogenic tests at SNAP, SNS, ORNL

The final tests of the bellows / lever arm assembly were performed at SNAP beamline of Spallation Neutron Source at Oak Ridge National Laboratory by ORNL staff – Jamie Molaison and Antonio Moreira Dos Santos at the end of December 2019. The assembly was attached to the real Neutron Diamond Anvil Cell with polycrystalline diamonds and steel gasket. The lever/bellows/DAC combo was mounted in a bottom-loading cold-finger style cryostat and cooled down to 9.5K and kept at this temperature during the tests (Figure 14). The pressure was applied in 15 bar increments up to 90 Bar (corresponding to ~8 tons of load on DAC). At that pressure / load the diamonds broke (perhaps due to small angular

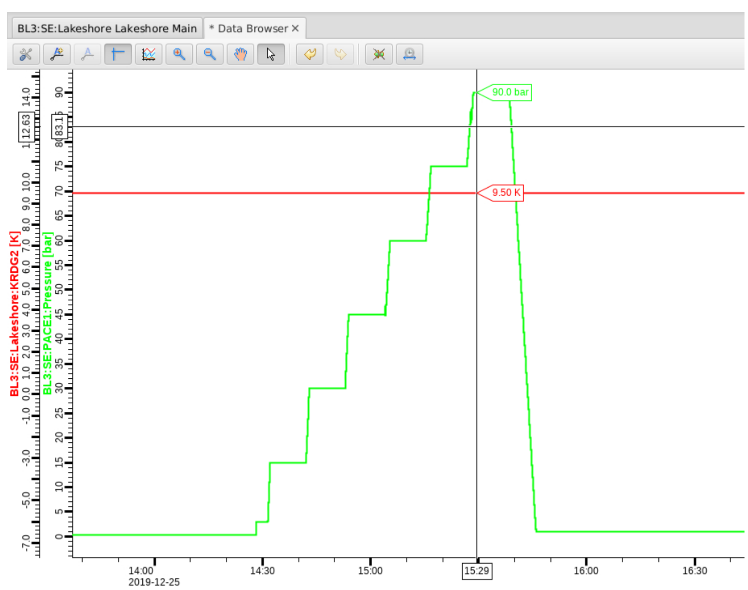


*Figure 14. Test of bellows / lever arm assembly at SNAP, SNS, ORNL.*

misalignment) and the gasket was destroyed. Nevertheless after a thorough inspection the bellows appear to be intact and fully functional. This proves that such bellows can be successfully used to remotely control pressure in neutron Diamond Anvil Cells at cryogenic conditions providing that the cell is assembled and prepared correctly and all elements of the DAC – Bellows Force Amplifier assembly are in good order and are perfectly matching.

### *3.11. Path forward*

It is obvious that in its current condition the new Bellows Force Amplifier Assembly can be routinely used with Neutron Diamond Anvil Cells "as is" at room temperature and cryogenic conditions, including top loading cryostats with bores down to 70 mm. Moreover, this design can be adapted to other large and small diamond anvil cells and used for accurate remote pressure control in a variety of Diamond Cells (neutron, synchrotron, laboratory) in sample environments with size / bore restrictions (cryostats, magnetic fields, etc.).

*Figure 15. Damage / indentation at the tip of one of the levers resulting from touching the second lever during pressurization.*

Nevertheless there are several issues which needs to be addressed before the system can be widely used.

The first is optimization of the design to make it more stable and user friendly. For example, the design of some components of the assembly can be supplemented by additional service holes simplify the assembling and handling the system.

The second is checking the detailed design for possible interference of components. For example, the lever made from Vascomax C-350 hardened to HRc 58-60, was interfering with the other lever during pressurization and thus was slightly damaged (indented, see Figure 15). Such interference and indentation ant cryogenic conditions can result in premature failure of the levers and possible failure of the sample. Thus we need to optimize the design of the levers and spacers to avoid such interference and damage in the future.

The third, and perhaps most important, is understanding the behavior, limits, life cycle and life expectancy of the welded bellows and optimizing their design. Perhaps increasing the number of convolutions in the bellows will result in smaller relative plastic deformation of thin metal convolutions, less stress and fatigue, and longer service life.

# 4. Results of the research: Pascalloy

### *4.1. Overview*

Except for some special cases such as flexural DACs [13] or 3-4 pin DACs [14], the majority of opposite anvil / diamond anvil cells are based on piston / cylinder principle where a piston is sliding inside a "cylinder"([4]). In this case the piston is in compression mode and the "cylinder" (which serves as a loading frame) is under tension. For proper performance of the opposite anvil cell it is critical that the cylindrical parts have an adequate tensile strength and toughness (to minimize elongation and avoid failure) while the piston should be have high elastic modulus to minimize bulging and locking the cell at high loads (several tons) required for pressurizing the millimeter-sized samples.

Traditionally the material of choice for diamond anvil cells for cryogenic conditions was (and remains) Beryllium-Copper alloy with about 2% of beryllium. This alloy is heat treatable, has high yield strength (up to ~ 200 MPa), toughness, stiffness, low temperature ductility, and excellent thermal conductivity (although it becomes progressively more difficult to find a manufacturer of BeCu DACs due to Beryllium toxicity and environmental regulations). While high thermal conductivity is critical for "cold-finger type" cryostats (e.g. Closed Cycle Refrigerators, CCR) with sample in vacuum, for "wet" cryostats (sample in exchange gas or "bath" cryostats) thermal conductivity is not that critical because all parts of the high-pressure cell are directly cooled with a cold gas. In this case the thermal mass of the system becomes a more important consideration and it is desirable to use DAC (tensile/loading frame) materials with maximum reasonable tensile strength and toughness, thus reducing the thermal mass of the system and allowing faster cooling and heating. For example, increasing the strength of DAC frame material by a factor of two will allow to reduce the tensile load bearing cross-sections of the DACs by a factor of two and either decrease the thermal mass by 2-3 times or allow larger opening and more flexibility in DAC design.

Unfortunately there are only a handful of high-strength materials suitable for application at liquid helium temperatures [15]. Normally the high-strength alloys have body-centered cubic (*bcc)* crystal structure and typically undergo ductile-to-brittle transformation at low temperatures and become extremely brittle. Maraging steel (e.g. C-300 grade) is often a steel of choice for DACs used at room and high temperature, even neutron DACs [12], but it becomes very brittle and unreliable at liquid helium temperatures, especially with frequent thermal cycling. The metals with a face centered cubic (*fcc*) crystal structure (such as 300 series stainless steels, Ni alloys such as Monel and Inconel) are exhibiting good ductility and toughness at LH temperatures, but have significantly lower yield strength.

One of the possible candidates for making DACs for LH temperatures is Ni-Cr-Al alloy, commonly referred to as "Russian alloy" or 40HNU-VI. It has mostly *fcc* crystal structure and remains ductile and tough at LH temperature (with proper heat treatment). Until recently it was not available commercially in the US (except for small amounts of borated material from Japan[15]). Nevertheless a domestic Illinois-based company Tevonic (www.tevonic.com) has recently started manufacturing alloy under a trade name of Pascalloy [16]. Pascalloy has very high yield strength (320 ksi), comparable to the strength of maraging steels (Vascomax) C-300 and C-350 (~290 and 350 ksi) and almost double the strength of hardened BeCu alloy. Pascalloy is heat treatable to a variety of conditions to ensure the best mechanical properties for particular applications and relative easiness in machining DAC parts (Figure 16). It also has excellent corrosion resistance and extremely low magnetic susceptibility (practically non-magnetic) which significantly expands the range of its application. Thus DAC Tools has proposed to start using this material for making compact, tough, high-strength neutron DACs out of Pascalloy.

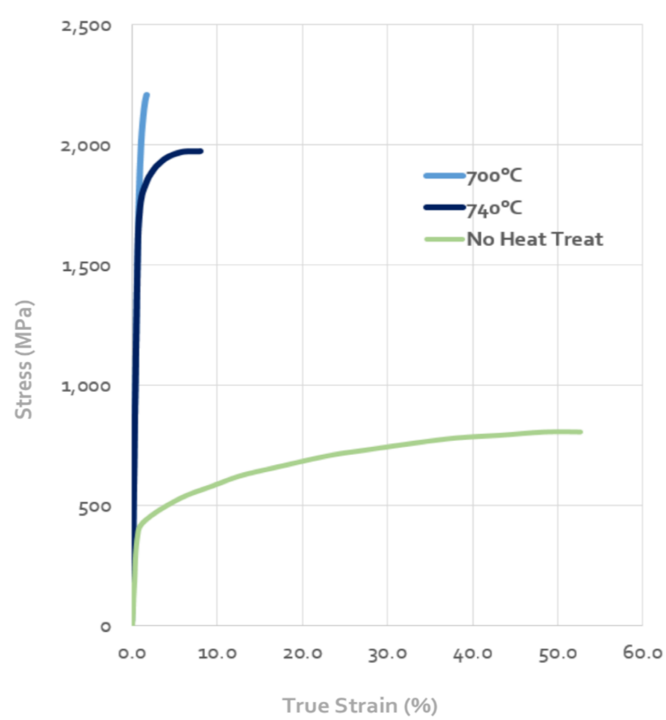


*Figure 16. The room temperature stress-strain plots for Pascalloy aged at 700° and 740° compared to annealed material (From [1]).*

The Business manager of Tevonic, Justin Carmichael, is collaborating with DAC Tools on different projects, in part involving Pascalloy, and during execution of the first phase of the SBIR proposal he has served as a consultant to DAC Tools LLC. In part he has prepared the samples of Pascalloy with different heat treatment conditions and thermal history and helped to arrange tests of strength and toughness of Pascalloy in Stanley Tozer's lab in National High Magnetic Field Laboratory for testing of mechanical properties at LH temperatures. The goal of DAC Tools in this part of the project was to check if Pascalloy is indeed superior high strength high toughness alloy at LH temperatures and if possible - find the proper temper of this material that best fits the purpose of designing and manufacturing compact large-opening neutron DACs which are capable of operating at 2 K in fully elastic regime with the load of 10-12 tones.

## 4.2. Tensile strength measurements at National High Magnetic Field Laboratory – preliminary results

A number of tensile strength tests have been performed in National High Magnetic Field laboratory (NHMFL). The tests have been performed by R.P. Walsh and K.J. Radclif under supervision of Dr. Stanley Tozer during 2019.

Due to limited number of samples and limited time as not enough data was taken for some temperatures to get statistical data, thus the presented data are **preliminary results** and should be treated as such. All the details are given in the Engineering Memorandum / cryo-mechanical report MD&C EM 19-012 of the NHMFL. Below are extracts from the report.

### 4.2.1. Introduction:

Two different diameter rods (0.5 in and 1.0 in) were submitted to the machine shop for tensile specimen machining. The 0.5 in. rod was supplied in two pieces that were identified to have 2 different conditions (annealed and age hardened). The 1.0 in. rod was identified to be in the annealed condition. There was a marginal amount of 1.0 in material so smaller tensile specimens were made in order to get a total of (4) specimens for testing. For the 0.5 in material, (4) tensile specimens of each condition were made. 2 or 3 tensile tests have been performed for each material condition at 4 K and 1 each at 295 K.

### 4.2.2. Material:

Stan Tozer obtained Pascalloy (NiCrAl:B) from Justin Carmichael (Tevonic, LLC, Lisle, Illinois 60532). The properties of the alloy are described elsewhere [1].

Pascalloy (NiCrAl:B) is a high entropy alloy with composition 40%Cr, 3.5%Al, 0.01%B, ballance Ni. It is based on the Russian alloy 40HKN. After machining, the Rockwell hardness of each tensile specimen was measured and is shown along with the tensile test results in **Error! Reference source not found.**.

### 4.2.3. Tensile test results

The Pascalloy samples for tensile tests are shown on Figure 17. The tensile test results are shown in **Error! Reference source not found.**. Figure 18 shows pictures of the fracture surface on Specimens #10 and #11, from the age hardened 0.5 in dia. material. Specimen #10 exhibited a yield point and NHMFL staff were able to determine the 0.2% yield strength while Specimen #11 failed in the linear elastic region and did not exhibit yielding or plastic deformation before fracture. There are some obvious defects on the fracture surface of Specimen #11 as can be seen in Figure 18 that were probably responsible for the premature failure. The measurements on Specimen #12 have failed because the threads on the gripped have broken while the Specimen #12 did not fracture of was not affected otherwise.

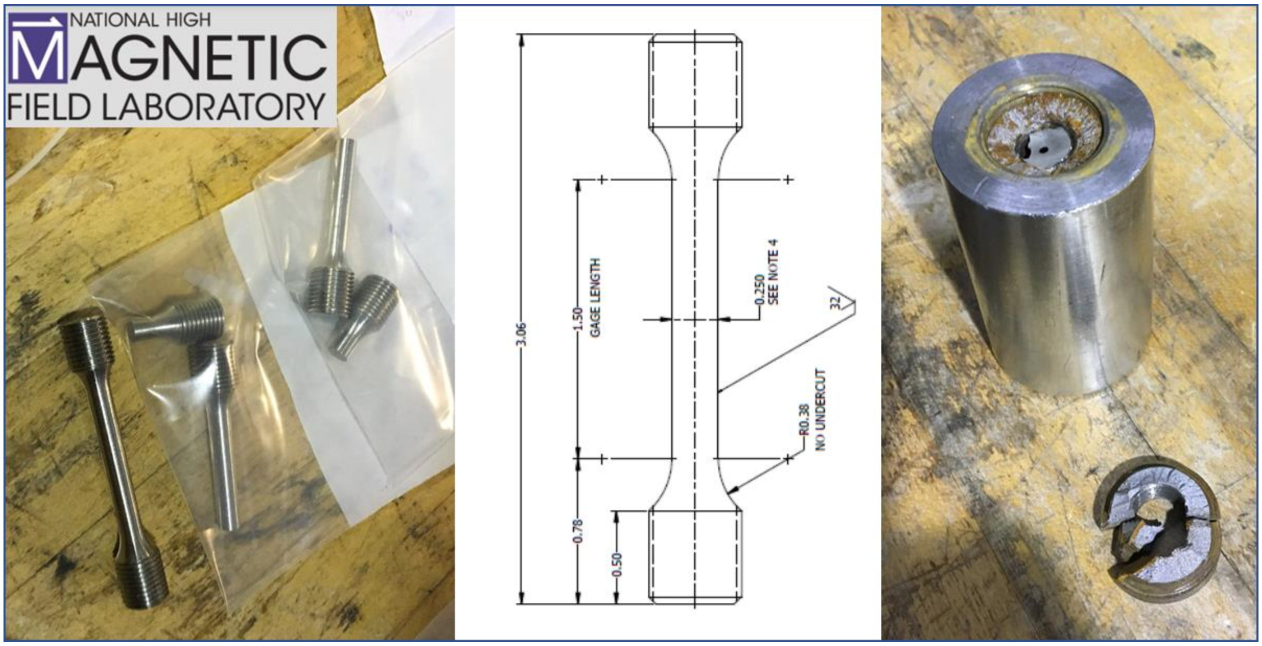


*Figure 17. Left: Specimens for tensile tests made of Pascalloy. Center: Dimensions of the tensile specimen used for 0.5" rod material. Right: Tensile test gripper broken during test of specimen #12.*

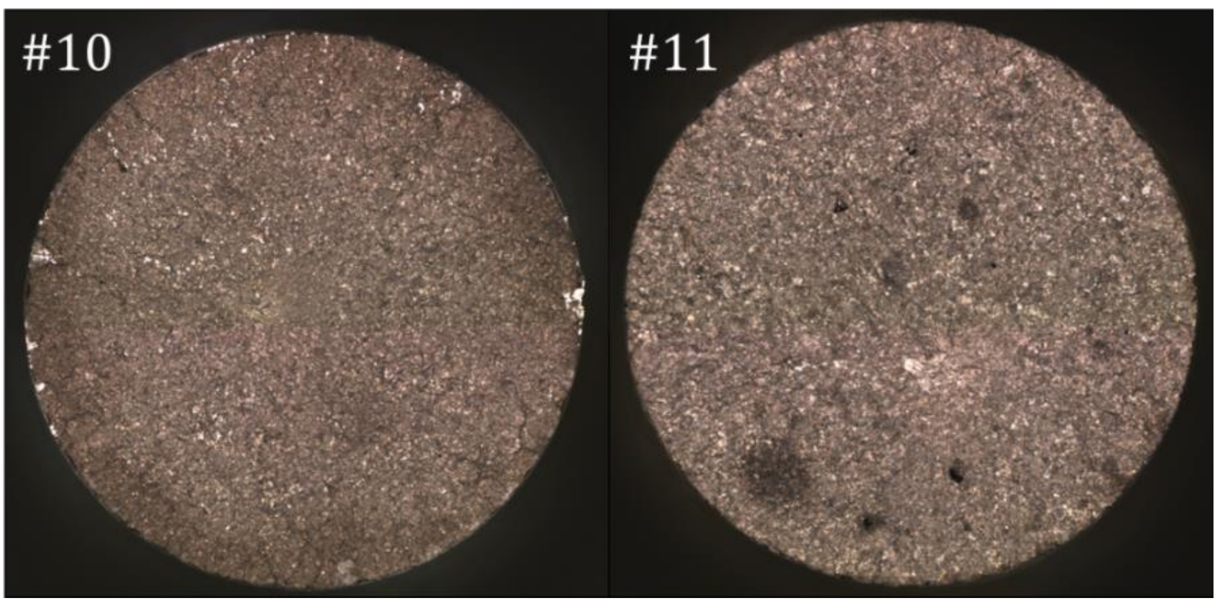


*Figure 18 Left: Fracture surface of specimen # 10 that exhibited a yield point during the 4 K tensile test. Right: Fracture surface of specimen # 11 that failed in the linear elastic portion of the 4K tensile test shows some black colored defects on the fracture surface.*

| Material ID | Specimen ID | Test Temp Temp K | Tensile Modulus GPa | Yield Strength MPa | Tensile Strength MPa | Elong. % | Reduct'n of Area % | Rockwell Hardness HRC | Comments |
|---|---|---|---|---|---|---|---|---|---|
| Annealed 1" | 1 | 295 | 216 | 689 | 1027 | 32 | 31 | 30 | |
| | 2 | 4 | 213 | 1026 | 1569 | 40 | 32 | 30 | |
| | 3 | 4 | 235 | 1044 | 1576 | 26 | 30 | 31 | |
| Annealed 1/2" | 5 | 295 | 209 | 803 | 1247 | 1.80 | 2 | 46 | |
| | 6 | 4 | 228 | 1406 | 1617 | < 1 | < 1 | 45 | Broke at scribe marks |
| | 7 | 4 | 222 | 1675 | 1783 | < 1 | < 1 | 49 | Broke at scribe marks |
| | 8 | 4 | 220 | 1350 | 1552 | < 1 | < 1 | 48 | |
| Age Harden 1/2" | 9 | 295 | 229 | 1833 | 1866 | <1 | <1 | 56 | |
| | 10 | 4 | 240 | 2432 | 2605 | < 1 | < 1 | 59 | |
| | 11 | 4 | 229 | N/A | 1865 | < 1 | < 1 | 58 | Broke before yield, evidence of defects on fracture surface |
| | 12 | 4 | 230 | N/A | >2162 | N/A | N/A | 59 | Threads broke grip, specimen did not fractured |

*Table 1. The tensile test results for Pascalloy in different annealing / heat treatment state at room temperature and 4K.*

### 4.3. Conclusions from tensile properties measurements on Pascalloy

The material most commonly used for making cryogenic Dimond Anvil Cells for neutron scattering is Beryllium Copper C17200. In heat treated state (e.g. hardest temper TH04) it has an ultimate tensile strength of 1310-1480 MPa and Yield strength of 1140-1415 MPa[17, 18]. These numbers are significantly lower than those for Pascalloy which, in proper heat treated condition shows the ultimate strength of ~2,210 MPa[16] and Yield strength of about 2,000 MPa at room temperature. **Preliminary results** of the tests performed in the National High Magnetic Field Laboratory at LH temperature (4K) indicate that the Ultimate Yield and 0.2% Tensile strength increase to ~2600 and 2400 MPa respectively and with proper heat treatment Pascalloy, unlike maraging steels, remains relatively ductile (not brittle). Room temperature and preliminary cryogenic strength measurements show that Pascalloy is significantly stronger than strongest modifications of Beryllium copper and making Diamond Anvil Cells from properly treated Pascalloy will allow to decrease mass of the Diamond Cells by more than two times. Given comparable heat capacity of Pascalloy and Beryllium copper – this will decrease the cooling / warming time of the cells made of Pascalloy by a factor of two when used in "wet" cryostats.

Despite the obvious advantage of Pascalloy, there are still issues which need to be resolved before it can be used in mass-productions of cryogenic high pressure cells. One of this issues is still finding the proper heat treatment conditions to receive the alloy with optimal strength and toughness at cryogenic conditions. Another issue is homogeneity of the alloy – inhomogeneities and defects in the material can make the overall product quite brittle and result in premature failure, thus finding the way of producing very homogeneous and defect-free bulk material is paramount for success of Pascalloy as a material of choice in manufacturing cryogenic high-pressure devices.

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