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Two Pressure Cells for Quasielastic and Inelastic Neutron Scatterings

Bao Yuan[1], *Richard* Mole[2], *Chin-Wei* Wang[3], *Amy* Shumack[2], *Rachel* White[2], *Bing* Li[4], *Xin* Tong[1*], *and Dehong* Yu[2*]

[1]Spallation Neutron Source Science Center, Dongguan 523803; Institute of High Energy Physics, Chinese Academy of Sciences (CAS), Beijing 100049, China
[2]Australian Nuclear Science and Technology Organisation, Lucas Heights, NSW 2234, Australia
[3]National Synchrotron Radiation Research Center, Hsinchu 30077, Taiwan
[4]Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, 72 Wenhua Road, Shenyang 110016, China.

Abstract. Two clamp pressure cells for QENS and INS have been developed. One is a hybrid CuBe/NiCrAl cell which is for relative high pressure up to 2.5 GPa and another one is made from high strength aluminium alloy (mesolite NA723) with pressure up to 0.5 GPa. The sample volume is 0.3 mL and 1 mL, respectively. The pressure cells have been thoroughly calibrated and tested. In addition, the contribution to phonon density of states from the pressure cells has been evaluated. Measurements of the phonon density of states for two perfluorocarbon polymer liquids FOMBLIN oil and Fluorinert have indicated that they are suitable to serve as the low background pressure transmission media for high pressure INS experiments. The applications of the pressure cells for INS and QENS are demonstrated by studies of pressure-induced phase transition of plastic crystals.

1 Introduction

The application of high pressure is one of the powerful techniques, among high or low temperature, high magnetic and electrical fields, used to induce unique changes in the structure, dynamics, and the associated properties of matter. Combined with neutron scattering, high-pressure research covers a wide range of areas of science to tackle many challenging problems. This includes, but is not limited to, emergent phenomena in quantum materials; synthesis of novel materials; mimic of planetary condition for geoscientific problems and tackling problems related to biosystems such as improving food processing or enhancing protein stability.

Neutron diffraction under high pressure has become a routine technique to study material structure evolution versus pressure, however, it is still a challenging task to perform quasielastic (QENS) and inelastic neutron scattering (INS) under high pressure for material dynamics investigation. This is mainly attributed to the intrinsic several orders of magnitude lower cross section for the QENS and INS processes than elastic scattering. Consequently, a large amount of sample and minimum background contribution from the sample cell is essential for a successful high pressure QENS and INS.

There are, in general, three types of high-pressure cells widely used for neutron scattering. The well-known Paris-Edinburgh anvils cell have been routinely used for neutron diffraction up to 10 GPa pressure with limited sample volume [1]. The McWhan-type cell can accommodate a slightly larger sample volume, but with lower pressure up to around 4 GPa [2]. The piston-cylinder type cell overcomes some limitations of the McWhan cell and increases the sample volume further [3]. The piston-cylinder type of cells has seen increasing popularity in INS and QENS studies in recent years [3-4]. In terms of materials requirement, a handful of materials have been tested and used for these pressure cells. This includes CuBe, NiCrAl, TiZr alloy, NiCr alloy, and high strength Al, etc. A detailed comparison of the background contribution from these materials for high-pressure inelastic neutron scattering has been reported [5].

Two piston-cylinder type pressure cells for QENS and INS have been developed by Chinese Spallation Neutron Source (CSNS) and fully tested on the Pelican time of flight cold neutron spectrometer at Australian Nuclear Science and Technology Organisation (ANSTO) [6-7]. One is a hybrid CuBe/NiCrAl cell which is for relative high pressure up to 2.5 GPa and another one is made from high strength aluminium alloy (mesolite NA723) with pressure up to 0.5 GPa. We call the two cells as Hybrid-cell and HAS-cell respectively. The outline of the paper is as the following. The design aspect of these pressure cells will be described first. The calibration of the cells will be given followed by the evaluations on the contributions to QENS and INS from the pressure cells, and pressure transmission media. In

* Corresponding author: tongx@ihep.ac.cn;dyu@ansto.gov.au

the end, the application of the pressure cells will be demonstrated by pressure-induced phase transition in plastic crystals.

2 Design and Load Test

The design and selection of materials for the pressure cells are largely based on existing knowledge as reported previously [8-9]. The sketch, finite element analysis (FEA) and load test will be outlined here.

2.1. CuBe/NiCrAl – Hybrid Cell

The sketch, dimensions, and FEA maps are shown in Fig. 1 for the Hybrid-cell. This hybrid design allows the realisation of a maximum pressure of 2.5 GPa with a relatively large sample volume of 0.3 mL. The maximum principal stress distribution with a load of 2.6 GPa is analysed through FEA as shown on the right panel of Fig.1. A maximum stress of 2.1 GPa and deformation of 0.014 mm are found for the inside cylinder, while the corresponding values for the outside cylinder are 1.22 GPa and 0.012 mm. The sample is filled into a Teflon tube which is sealed through a CuBe insert and a carefully shaped plug ring. The Teflon tube is pushed into the inside cavity of the pressure cell. The bottom of the cell is locked with a CuBe screw with a piece of Tungsten Carbide (WC) block against the sample tube plug. On top of the sample tube is a WC piston and another CuBe block through which force is applied to the sample tube with the WC push rod. The top CuBe screw is used to lock the position of the piston and maintain the pressure.

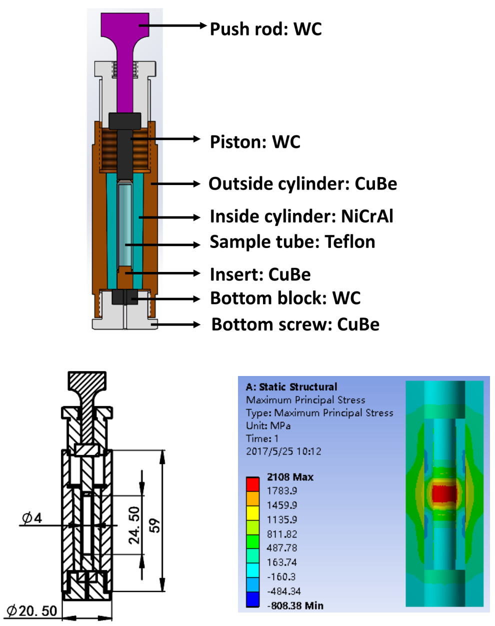

Fig. 1. The sketch, dimensions, and maximum stress distribution with FEA are shown for the Hybrid – Cell.

2.2 Mesolite NA723 – HSA Cell

As shown in Fig. 2, the same design has been applied to the HSA-Cell. Taking advantage of the high strength of the Mesolite NA723 Al alloy, this cell can deliver up to 0.5 GPa pressure with a much larger sample volume of 1 mL as compared to the Hybrid-Cell. The operation of this cell follows the same procedure as described above for the Hybrid-cell.

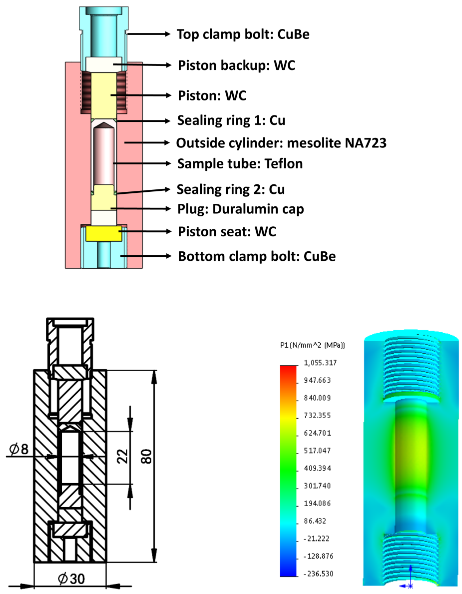

Fig. 2. The sketch, dimensions, and maximum stress distribution with FEA are shown for the HSA – Cell.

2.3 Load Test

By monitoring the resistance changes of a bismuth sensor as a function of force applied to the cell, preliminary load tests have been performed on both cells [8]. For the Hybrid-Cell, the maximum pressure of 2.55 GPa has been achieved. For safety purpose, it is recommended to keep the maximum operating pressure at 1.5 GPa. For the Mesolite NA723 Al cell, maximum safe pressure of 500 MPa has been achieved.

3 Pressure Cell Calibration

The pressure cells have been calibrated with neutron diffraction of KBr using the Wombat neutron diffractometer at ANSTO [10]. The Equation of State (EOS) for KBr [11] was used to calculate the pressure achieved corresponding to the force applied to the cells as shown in Fig. 3 and Fig. 4 below. A small amount of Fluorinert liquid (1/3 of the total sample in weight) was added to KBr powder as the pressure transmission medium. The estimated errors of about 8% are mainly from the uncertainty in the force applied.

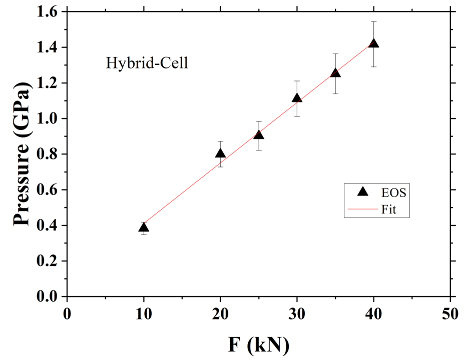

Fig. 3. The pressure calibration curve for the Hybrid-Cell.

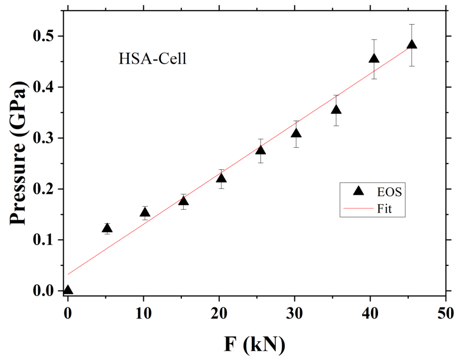

Fig. 4. The pressure calibration curve for the HSA-Cell.

4 QENS and INS contribution of the Pressure Cells

A proper evaluation of these pressure cells for quasielastic (QENS) and inelastic neutron scatterings (INS) have been performed on Pelican instrument. In addition to the empty pressure cells, the QENS and INS have been measured on two non-H contained liquids of Fomblin and Fluorinert which are frequently used as low background pressure transmission medium. Vanadium results are shown for reference. The QENS results are integrated over all Q values. The incident neutron energy is 3.7 meV corresponding to 0.13 meV energy resolution at the elastic peak. The QENS and generalised phonon density of state (GDOS), as presented in Fig. 5 and Fig. 6, respectively, are all normalised to vanadium signal. These measurements are performed at 300 K. For both Fomblin and Fluorinert, the results correspond to 0.3 g of sample weight. There are clearly QENS from these liquids. For the two empty cells, the lower QENS signal is mainly from the Teflon tube inside. For the GDOS, the contributions from the two pressure cells are much stronger (about 2 orders of magnitude) than the two pressure transmission media. These results indicate clearly that care must be taken to properly subtract backgrounds from both cells and pressure transmission medium, especially for weak scattering samples. Certainly, these pressure cells are most suitable for hydrogen-contained samples. For proper background subtraction, the following precautions need to be taken: Time for empty can measurements should be at least the same as that for sample measurements to minimise error bars due to background subtraction; The pressure cell should be masked by neutron absorption materials (Cd, for example) to ensure the same amount of cell exposed to neutrons for both sample and empty can measurements.

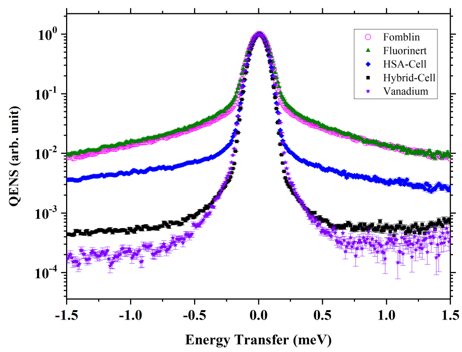

Fig. 5. QENS contributions from the pressure cells and several other materials as described in the text.

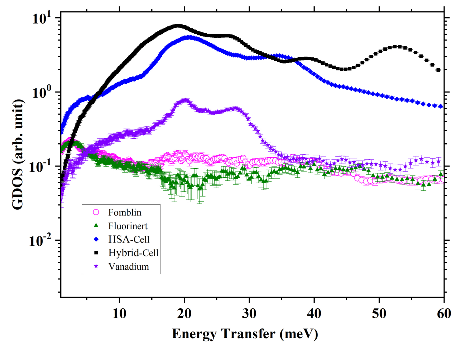

Fig. 6. GDOS from the pressure cells and several other materials as described in the text.

5 Applications

The application of the pressure cell (HSA-cell) has been demonstrated with pressure induced phase transition of NH_4I. As shown in figures 7 and 8 below, both QENS and the imaginary part of the dynamic susceptibility (χ'') have reflected the phase transition from the α-phase to β-phase upon applying 300 MPa pressure. This measurement confirms the promising application of NH_4I in solid state cooling technology through barocaloric effect [12].

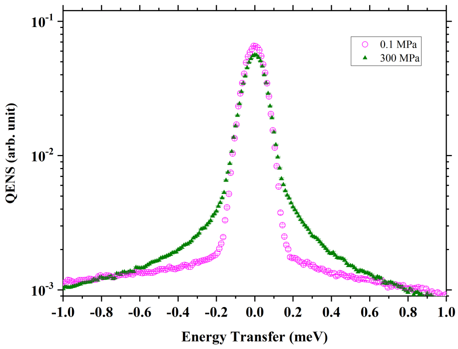

Fig. 7. QENS indicates the phase transition from the α-phase to β-phase upon applying 300 MPa pressure for NH_4I.

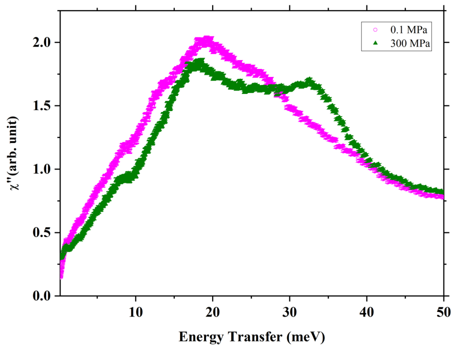

Fig. 8. The imaginary part of the dynamic susceptibility indicates the phase transition from the α-phase to β-phase upon applying 300 MPa pressure for NH_4I.

6 Summary

Two pressure cells have been developed and thoroughly tested. The contributions to quasielastic neutron scattering and phonon density of states from the pressure cells have been evaluated. A proper correction of the cell contribution must be taken especially for weak scatters. While these cells can be certainly used for diffraction experiments requiring large amount of sample for structure determination through refinement, they may be best suitable for hydrogen-contained samples for QENS and INS experiments, which has been demonstrated with pressure induced phase transition of NH_4I.

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