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Development of a ^{3}He Gas Filling Station at the China Spallation Neutron Source

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At the China Spallation Neutron Source (CSNS), we have developed a custom gas-filling station, a glassblowing workshop, and a spin-exchange optical pumping (SEOP) system for producing high-quality ^{3}He-based neutron spin filter (NSF) cells. The gas-filling station is capable of routinely filling ^{3}He cells made from GE180 glass of various dimensions, to be used as neutron polarizers and analyzers on beamlines at the CSNS. Performance tests on cells fabricated at our gas-filling station are conducted via neutron transmission and nuclear-magnetic-resonance measurements, revealing nominal filling pressures, and a saturated ^{3}He polarization in the region of 80%, with a lifetime of approximately 240 hours. These results demonstrate our ability to produce competitive NSF cells to meet the ever-increasing research needs of the polarized neutron research community.

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The China Spallation Neutron Source (CSNS) is one of the few large-scale neutron scattering facilities in the world, and the first accelerator-based pulsed neutron source in China, serving as an invaluable resource for materials and energy science research.[1] Since its commissioning in 2018, the CSNS has operated three neutron-scattering instruments in its user program (a powder diffractometer, a magnetic reflectometer, and small-angle neutron scattering), and currently has eight additional beamlines under construction.

The high-penetration and magnetic-sensitive nature of neutrons, specifically when polarized, make them an ideal probe for investigating a wide range of magnetic phenomena and material properties that other techniques, such as x-ray and electron scattering, may not be able to resolve.[2] The CSNS has therefore identified three of its beamlines as suitable for neutron polarization instrumentation: the multi-purpose reflectometer, the high-energy spectrometer, and the high-resolution diffractometer. To ensure optimized neutron polarizers and analyzers can be provided to these beamlines, the CSNS has been developing in-house facilities for the fabrication of ^{3}He neutron spin filter (NSF) cells. In comparison to other common neutron polarization and analysis methods, such as Heusler crystals,[3] or supermirrors,[4] NSFs do not alter the phase space of the neutron beam, and are able to cover a wide angular range and wavelength band simultaneously.[5–8] They are transmittance devices, whose working principle is based on the strongly spin-dependent absorption cross section of ^{3}He nuclei.[9] In addition, NSFs are cost-effective and highly customizable, allowing the diverse research needs of users to be met. The applicability of a ^{3}He NSF depends largely on the degree of nuclear polarization of the ^{3}He gas inside, and its lifetime (i.e., relaxation), which is limited by some known (dipole-dipole interaction, magnetic field inhomogeneity and interaction with the cell wall) and some ambiguous factors (X-factor).[10–13]

At the CSNS, we have developed a gas-filling station and a glass-blowing workshop for the fabrication of custom NSFs with a high degree of ^{3}He polarization, and long lifetimes, as determined via spin-exchange optical pumping (SEOP)—a method used worldwide for producing polarizing ^{3}He gas.[14–19] In this Letter, we present details of our NSF fabrication methods and cell performance. First, we discuss the design of our gas-filling station. Then, we present our fabrication methods for NSFs, including glassware preparation, and the gas-filling procedure. We investigate one of the first ^{3}He glass cells produced on this system, and discuss its performance as an NSF. Finally, we present

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our conclusions, and discuss the prospects for ^{3}He polarization capability at CSNS.

Design of the Gas-Filling Station. The ^{3}He NSF gas-filling station at CSNS consists of four major sections: gas purification, vacuum, pressure control, and a glass manifold. A schematic of the gas-filling station is shown in Fig. 1. Bottles of N_2 and ^{3}He are placed at the end of the station, with their outlets connected to corresponding gas purifiers. The gas purifiers and filling lines are baked and vented via a combination of roughing and diaphragm-backed turbo vacuum pumps, at pressures as low as 10^{-9} mbar. A residual gas analyzer (RGA) is installed next to the turbo pump to monitor gas composition. Glass manifolds can be connected to the two available outlets shown in the lower right section of Fig. 1, providing us with the ability to simultaneously fill multiple cells. Two pressure gauges are installed to monitor the pressure inside the system.

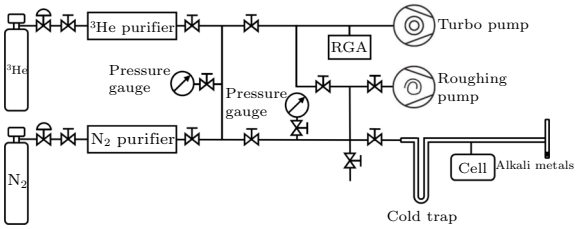

Fig. 1. Schematic of the gas-filling station at the CSNS. A depiction of a glass manifold with a U-shaped cold trap and cell are shown in the lower right section.

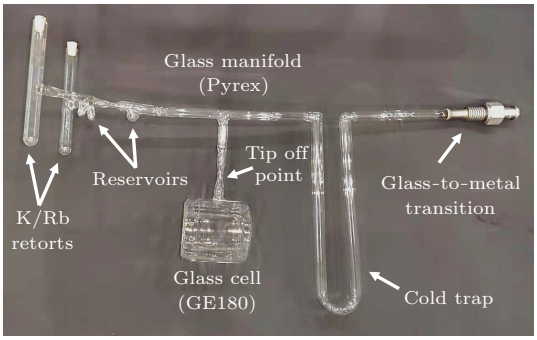

Fig. 2. Completed pyrex manifold with labeled components and GE180 glass cell assembly. The manifold is connected to the filling station by means of the VCR fitting on the glass-to-metal transition.

The glass manifold is a customized extension of the filling station, crafted in our in-house glassblowing workshop. A typical glass manifold fabricated at the CSNS is shown in Fig. 2. A completed manifold consists of one glass-to-metal transition, a U-shaped cold trap near the gas inlet to help capture impurities during filling, one cell attached for tip-off, and two alkali metal retorts for potassium and rubidium. Depending on filling requirements, modifications to the components can be made via in-house fabrication, such as additional cells and alkali metal reservoirs (see Fig. 2).

^{3}He Neutron Spin Filter Fabrication. The developed ^{3}He NSF filling station is capable of producing a custom-designed ^{3}He cell on a 2–3 weekly basis, depending on the parameters and requirements for a given cell. The ^{3}He cell-fabrication process requires three major steps in chronological order: glassware and cell preparation, gas filling, and cell tip-off.

The process begins by first selecting an empty glass cell, based on the requirements of the neutron experiment. A boron-free aluminosilicate glass (GE180) is used to make the cell, which is a desirable material for NSFs, due to its low ^{3}He permeability, excellent neutron transparency, high softening temperature, and satisfactory mechanical strength. The GE180 glass cells are cylindrical, with maximum lengths and radii measuring as much as 8 cm and 10 cm, respectively. Each cell has a hollowed stem protruding from its curved surface, which is subsequently connected to the main body of a pyrex glass manifold, and used to facilitate alkali metal transport, gas filling, and tip-off. A typical empty GE180 cell is shown in Fig. 3(a). For in-house cell fabrication, the glass must first be annealed prior to connecting it to the manifold, in order to eliminate residual stress that might result in the mechanical failure of the cell, once pressurized. The structural integrity and internal stresses of the cell and manifold are examined using a polariscope. When viewing a glass cell through a polariscope, transparent glass walls without patches or bands of color indicate regions of low-to-little stress. Figures 3(b) and 3(c) show the respective results before and after the appropriate annealing of a pyrex glass test cell. Note that a pyrex test cell is selected, rather than a GE180, for visualization purposes.

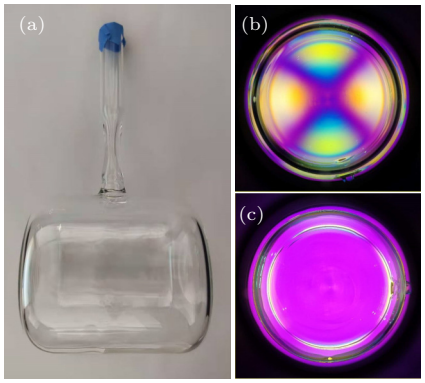

Fig. 3. (a) Typical GE180 glass cell. View from a polariscope of a test cell made of pyrex (b) before and (c) annealing. The cell diameters are approximately 6 cm.

Once the cell is connected to the manifold, the glassware is thoroughly cleaned, following the procedure described by Chen *et al.*,[20] prior to installation on the gas-filling station, and is then pressure tested to 5 bar using N_2 gas. The system is then evacuated for up to 24 hours before 1 gram ampules of >99.75% pure

K and Rb are opened, and placed into their respective retorts, under a steady stream of N_2 gas, so as to avoid oxidation. The retorts are then sealed, and the system is evacuated and baked out for approximately one week. During this time, the GE180 cell is heated to 400℃ using a custom-built oven, raised into position by an electronic lift. The oven has been designed with a large enough volume to house the GE180 cell and most of the surrounding pyrex manifold. The bake-out process is monitored by the RGA. Figure 4 shows an RGA spectrum taken from the system after a week of 400℃ bake-out, revealing a water partial pressure below 1×10^{-9} mbar, indicating a clean vacuum and cell.

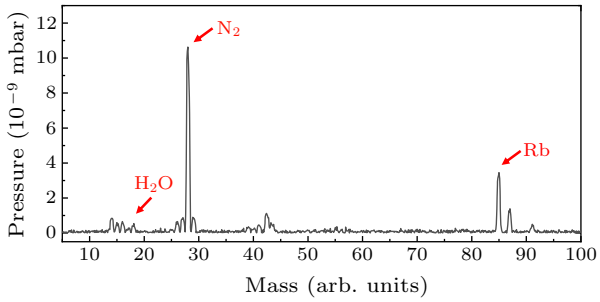

Fig. 4. Example of residual gas detection inside the system during alkali metal distillation. Gaseous components are listed according to their relative molecular mass.

Once the bake-out is complete, K and Rb alkali metals are distilled from their retorts to the GE180 cell by means of a process similar to that described by Rich *et al.*[21] The K/Rb ratio is controlled, maintaining a vapor density ratio of between 2 and 6, which is required in order to achieve high ^{3}He polarization efficiency during SEOP.[22] After the metals have been distilled, 99.99% pure N_2, and ^{3}He gases are filtered, and introduced into the system. Firstly, N_2 gas is loaded to approximately 5% of the total target pressure, and is used to quench fluorescence emitted from excited Rb atoms during SEOP. Next, ^{3}He gas is slowly introduced into the system until the target pressure is reached. Depending on the user and on instrumental requirements, we are able to fill our GE180 cells at ^{3}He pressures in the range of 0.5–3 bar.

Before the filled cell can be removed from the manifold, a procedure known as "tip-off", the pressure inside the cell should be lower than the surrounding atmosphere. If the pressure inside the cell is too high, then the softened glass will inflate or even burst, which would result in loss of precious ^{3}He gas, and a failed tip-off. Therefore, when encapsulating the gases at above 1 bar, the glass cell is partially submerged in liquid nitrogen in order to decrease the internal pressure to below 1 bar. Once pressure stabilizes, the tip-off is performed, using a torch and flame to carefully heat the narrow section of the GE180 stem until it is soft enough that the cell can be separated from the manifold, thereby sealing the gases inside. Having corrected for the contribution of N_2 gas, the pressure of the ^{3}He gas, sealed inside a cell at room temperature $p_{\rm cell}$, can be calculated using the following expression (derived from the ideal gas law):

$$p_{\rm cell} = p_{\rm i} + \frac{p_{\rm i} - p_{\rm f}}{V_{\rm c}}(V_{\rm s} + 3.87V_{\rm n}), \quad (1)$$

where $V_{\rm c}$ is the volume of the cell, $V_{\rm s}$ is the volume of the filling station gas lines and glass manifold, $V_{\rm n}$ is the volume of the cold trap that was submerged in liquid nitrogen, and $p_{\rm i}$ and $p_{\rm f}$ denote the ^{3}He pressures before and after the cell is partially submerged in liquid nitrogen, respectively. The factor 3.87 is derived from the ratio of room temperature to that of liquid nitrogen. As such, in order to accurately estimate the final pressure of the cell, the dimensions of the manifold and cell are carefully measured prior to its connection to the filling station.

Performance and Quality Testing of NSFs. NSF cells are characterized by (i) their ^{3}He filling pressure, (ii) the maximum achievable ^{3}He polarization $P_{^3{\rm He}}$, and (iii) the lifetime, T_1, of the ^{3}He polarization. Here, we present the results of neutron transmission experiments performed on our NSF cells, which confirm the nominal ^{3}He filling pressure, as well the ^{3}He polarization and lifetime measurements obtained at both the CSNS, and at Oak Ridge National Laboratory (ORNL).

The transmission of neutrons through an unpolarized ^{3}He cell is given by

$$T_0 = T_{\rm e}\exp(-n\sigma l) = T_{\rm e}\exp(-0.0732p\lambda l), \quad (2)$$

where T_0 is the total neutron transmission, $T_{\rm e}$ is the transmission of an empty GE180 cell, n is the ^{3}He number density, σ is the wavelength-dependent ^{3}He neutron absorption cross section, l is the path length inside the cell in units of cm, p is ^{3}He pressure in units of bar, and λ is the neutron wavelength in Å. The conversion in Eq. (2) with respect to ^{3}He number density and absorption cross-section to pressure is well-known.[23]

A series of unpolarized neutron transmission measurements have been performed, using the test beamline (BL–20) at CSNS. Time-of-flight (TOF) transmission spectra, with a wavelength band of $1.25\,\text{Å} < \lambda < 4.50\,\text{Å}$, were collected from unpolarized ^{3}He NSF cells using a ^{3}He detector with data accumulation times of one hour.

Here, we present data collected from our NSF, nicknamed "Salty", which possesses an axial neutron path length of $l = 8.57$ cm, as determined from its outer dimensions, plus ultrasonic measurements of the glass wall's thickness. The empty cell spectrum, $T_{\rm e}$, for our analysis was obtained from an air-filled GE180 cell with similar dimensions. Fitting the neutron

transmission data using Eq. (2) (see Fig. 5), we determine the ^{3}He pressure of Salty to be 1.52 ± 0.03 bar. This result is in excellent agreement with the room-temperature ^{3}He pressure of $p_{\text{cell}} = 1.51 \pm 0.06$ bar, calculated using Eq. (1), based on pressure and volumetric measurements during the tip-off time.

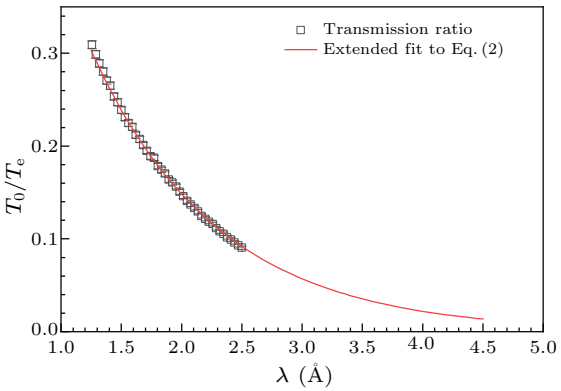

Fig. 5. Neutron transmission ratio of an unpolarized Salty cell to an empty GE180 cell (black data points). Equation (2) is used to obtain the best fit at short wavelengths (red curve), resulting in a ^{3}He pressure of 1.52 ± 0.03 bar.

Figure 5 displays an extended fit to the neutron transmission data collected in the short-wavelength region of $1.25\,\text{Å} < \lambda < 2.50\,\text{Å}$ (red curve), resulting in a 1.52 ± 0.03 bar ^{3}He pressure. We found that fitting transmission data collected in the long-wavelength region of $2.5\,\text{Å} < \lambda < 4.5\,\text{Å}$ (data not shown) deviated from the fit obtained at low wavelengths, and resulted in an elevated ^{3}He pressure of 1.62 ± 0.04 bar. This discrepancy was present in all the cells measured, and we speculate that it may originate from several factors, such as frame overlap of TOF neutrons, wavelength-dependent neutron detector efficiency, or geometric and isotopic concentration variations between the different GE180 glass cells. We acknowledge that this phenomenon between long- and short-wavelength discrepancies in the neutron transmission spectra of unpolarized ^{3}He is worthy of further study; however, due to the quality of our short-wavelength fit, and its agreement with independent pressure calculations from data obtained during the filling process, we assume the ^{3}He pressure of Salty to be approximately 1.52 ± 0.03 bar.

The highest achievable ^{3}He polarization and lifetime of the produced cell were measured using electron paramagnetic resonance (EPR), the adiabatic fast passage (AFP) method, and free induction decay (FID) on SEOP stations at both the CSNS and at ORNL. EPR polarimetry measures the frequency shift of Rb Zeeman resonance caused by polarized ^{3}He nuclei, and is therefore proportional to the degree of ^{3}He polarization.[24] Using our *ex situ* SEOP station at the CSNS, we measured a maximum shift in the nuclear magnetic resonance frequency of 17.27 kHz for Salty (inset in Fig. 6), after approximately eight hours of optical pumping. Based on the ^{3}He pressure calculated from Eq. (1), as confirmed by short-wavelength fits to the neutron transmission spectrum (see Fig. 5), and having corrected for the cell dimensions, temperature, and strength of the magnetic field during the pumping process,[25,26] we obtain a maximum saturated polarization for Salty of $P_{^3\text{He}} \approx 83.8 \pm 1.7\%$. Assuming the ^{3}He pressure of Salty to be 1.62 ± 0.04 bar, as observed based on a long-wavelength fit to the neutron transmission spectrum, the resulting ^{3}He polarization would decrease to $P_{^3\text{He}} \approx 77.7 \pm 1.9\%$.

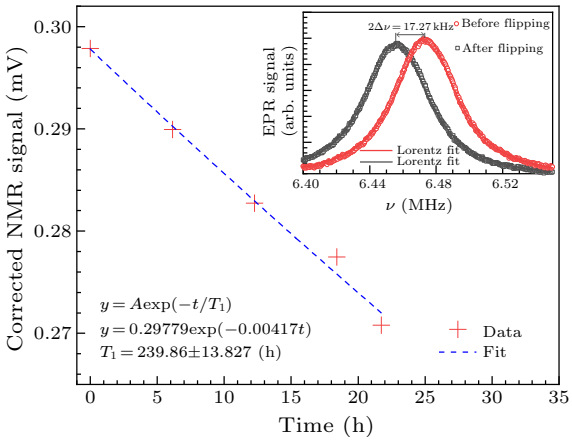

Fig. 6. Corrected nuclear-magnetic-resonance (NMR) signal obtained from Salty as a function of time. Data was fitted with an exponential decay, resulting in a ^{3}He lifetime, T_1, of 239 ± 13.8 hours. The inset shows an EPR frequency shift for Salty, measuring 17.27 kHz.

FID measurements to determine the T_1 of polarized ^{3}He in Salty were also performed on Optical Pumping Station #2 at ORNL.[19] The corrected NMR signal for polarized ^{3}He vs time is shown in Fig. 6. Here, T_1 was extracted by fitting an exponential decay (dashed blue line in Fig. 6) to the NMR data, revealing a lifetime of 239 ± 13.8 hours.

In summary, at the CSNS we have developed a gas-filling station that facilitates the production of high-quality ^{3}He-based NSFs with precise gas loading, as confirmed by neutron transmission measurements. Performance tests of our Salty NSF were conducted using EPR and FID, and reveal a maximum achievable ^{3}He polarization of up to 83%, with a lifetime of approximately 240 hours. These results demonstrate our capacity to supply customized NSFs for the purpose of polarized neutron research on various beamlines at the CSNS, whose performance characteristics are competitive with NSFs fabricated at other world-renowned facilities such as ORNL, ISIS, and J-PARC.[27,28] To expand our production capabilities, the construction of a second gas-filling station is planned. In addition, improvements are being made to both *in-situ* and *ex-situ* SEOP stations at the CSNS.

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References

[1] Chen H and Wang X L 2016 *Nat. Mater.* **15** 689
[2] Chatterji T 2006 *Neutron Scattering from Magnetic Materials* ed Chatterji T (Amsterdam: Elsevier Science) chap 1 p 1
[3] Freund A, Pynn R, Stirling W and Zeyen C 1983 *Physica B+C* **120** 86
[4] Sinclair R N and Brockhouse B N 1960 *Phys. Rev.* **120** 1638
[5] Heil W, Andersen K, Cywinski R, Humblot H, Ritter C, Roberts T and Stewart J 2002 *Nucl. Instrum. Methods Phys. Res. Sect. A* **485** 551
[6] Babcock E, Salhi Z, Gainov R, Woracek R, Soltner H, Pistel P, Beule F, Bussmann K, Heynen A, Kämmerling H, Suxdorf F, Strobl M, Russina M, Voigt J and Ioffe A 2017 *J. Phys.: Conf. Ser.* **862** 012002
[7] Boag S, Parnell S, Frost C, Andersen K and Babcock E 2007 *Physica B* **397** 179
[8] Tong X, Jiang C, Lauter V, Ambaye H, Brown D, Crow L, Gentile T, Goyette R, Lee W T, Parizzi A and Robertson J 2012 *Rev. Sci. Instrum.* **83** 075101
[9] Sears V F 1992 *Neutron News* **3** 26
[10] Fitzsimmons W A, Tankersley L L and Walters G K 1969 *Phys. Rev.* **179** 156
[11] Cates G D, Schaefer S R and Happer W 1988 *Phys. Rev. A* **37** 2877
[12] Parnell S, Babcock E, Nünighoff K, Skoda M, Boag S, Masalovich S, Chen W, Georgii R, Wild J and Frost C 2009 *Nucl. Instrum. Methods Phys. Res. Sect. A* **598** 774
[13] Babcock E, Chann B, Walker T G, Chen W C and Gentile T R 2006 *Phys. Rev. Lett.* **96** 083003
[14] Gentile T R, Nacher P J, Saam B and Walker T G 2017 *Rev. Mod. Phys.* **89** 045004
[15] Yan S, Zhang M F, Guo W C, Wang W Z, Gong J, Liang T J, Liu B Q, Peng M, Peng S M, Sun G, Tu X Q, Yan H Y, Zhang J H and Zheng H 2019 *Sci. Chin.-Phys. Mech. Astron.* **62** 102021
[16] Passell L and Schermer R I 1966 *Phys. Rev.* **150** 146
[17] Coulter K, McDonald A, Happer W, Chupp T and Wagshul M 1988 *Nucl. Instrum. Methods Phys. Res. Sect. A* **270** 90
[18] Babcock E and Ioffe A 2011 *Physica B* **406** 2448
[19] Jiang C, Tong X, Brown D, Lee W T, Ambaye H, Craig J, Crow L, Culbertson H, Goyette R, Graves-Brook M, Hagen M, Kadron B, Lauter V, McCollum L, Robertson J, Winn B and Vandegrift A 2013 *Phys. Procedia.* **42** 191
[20] Chen W, Gentile T, Fu C, Watson S, Jones G, McIver J and Rich D 2011 *J. Phys.: Conf. Ser.* **294** 012003
[21] Rich D, Gentile T, Smith T, Thompson A and Jones G 2002 *Appl. Phys. Lett.* **80** 2210
[22] Chen W C, Gentile T R, Walker T G and Babcock E 2007 *Phys. Rev. A* **75** 013416
[23] Batz M, Baeßler S, Heil W, Otten E, Rudersdorf D, Schmiedeskamp J, Sobolev Y and Wolf M 2005 *J. Research Natl. Inst. Stand. Technol.* **110** 293
[24] Chann B, Babcock E, Anderson L and Walker T 2002 *Phys. Rev. A* **66** 032703
[25] Babcock E, Nelson I A, Kadlecek S and Walker T G 2005 *Phys. Rev. A* **71** 013414
[26] Romalis M and Cates G 1998 *Phys. Rev. A* **58** 3004
[27] Okudaira T, Oku T, Ino T, Hayashida H, Kira H, Sakai K, Hiroi K, Takahashi S, Aizawa K, Endo H, Endo S, Hino M, Hirota K, Honda T, Ikeda K, Kakurai K, Kambara W, Kitaguchi M, Oda T, Ohshita H, Otomo T, Shimizu H, Shinohara T, Suzuki J and Yamamoto T 2020 *Nucl. Instrum. Methods Phys. Res. Sect. A* **977** 164301
[28] Boag S, Jiang C, Tong X and Parnell S 2014 *J. Phys.: Conf. Ser.* **528** 012019