A LIQUID DEUTERIUM COLD NEUTRON SOURCE FOR THE NIST RESEARCH REACTOR – CONCEPTUAL DESIGN

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ABSTRACT

The NBSR is a 20 MW research reactor operated by the NIST Center for Neutron Research (NCNR) as a neutron source providing beams of thermal and cold neutrons for research in materials science, fundamental physics and nuclear chemistry. A large, 550 mm diameter beam port was included in the design for the installation of a cold neutron source, and the NCNR has been steadily improving its cold neutron facilities for more than 25 years. Monte Carlo Simulations have shown that a liquid deuterium (LD_2) source will provide a gain of 1.5 to 2 for neutron wavelengths between 4 Å and 10 Å with respect to the existing liquid hydrogen cold source. The conceptual design for the LD_2 source will be presented.

To achieve these gains, a large volume (35 litres) of LD_2 is required. The expected nuclear heat load in this moderator and vessel is 4000 W. A new, 7 kW helium refrigerator is being built to provide the necessary cooling capacity; it will be completely installed and tested early in 2014. The source will operate as a naturally circulating thermosiphon, very similar to the horizontal cold source in the High Flux Reactor at the Institut Laue-Langevin (ILL) in Grenoble. A condenser will be mounted on the reactor face about 2 m above the source providing the gravitational head to supply the source with LD_2. The system will always be open to a 16 m^3 ballast tank to store the deuterium at 500 kPa when the refrigerator is not operating, and providing a passively safe response to a refrigerator trip. It is expected the source will operate at 23 K, the boiling point of LD_2 at 100 kPa. All components will be surrounded by a blanket of helium to prevent the possibility of creating a flammable mixture of deuterium and air. A design for the cryostat assembly, consisting of the moderator chamber, vacuum jacket, helium containment and a heavy water cooling water jacket, has been completed and sent to procurement to solicit bids. It is expected that installation of the LD_2 cold source will begin in April of 2016.

Funding for the refrigerator and the cold source upgrade has been granted by the National Nuclear Security Administration of the Department of Energy as a mitigation strategy to offset the anticipated 10% loss in neutron flux when the NBSR is converted to low-enriched uranium (LEU) fuel.

1. Introduction

The NBSR is a 20 MW research reactor operated by the National Institute of Standards and Technology (NIST) Center for Neutron Research (NCNR), primarily for neutron scattering instruments to study the properties of materials, but also for research in Fundamental Physics and Nuclear Chemistry. With completion of the first guide hall in 1990, and the installation of the first liquid hydrogen (LH_2) cold neutron source (CNS) in 1995, the NCNR has become one of the world's premier centres for cold neutron research. In 2012, the NCNR completed another major expansion project with the addition of a new guide hall and 5 new guides. As shown in Figure 1, about 20 instruments will be using cold neutrons when the entire suite of instrumentation is completed in 2015. Along with the expansion of the facility, the cold neutron sources have been improved and expanded. The original LH_2 source was replaced with the Advanced LH_2 Cold Source (Unit 2) in 2002, doubling the flux of cold neutrons to all the instruments [1,2]. In addition, a second LH_2 source was installed in the thermal neutron beam port, BT-9, solely for the Multi-Axis Crystal Spectrometer which was relocated to BT-9 to accommodate the 5 new guides [3]. The only way to improve upon the highly optimized Unit 2 is to replace it with a large volume, liquid deuterium (LD_2) source.

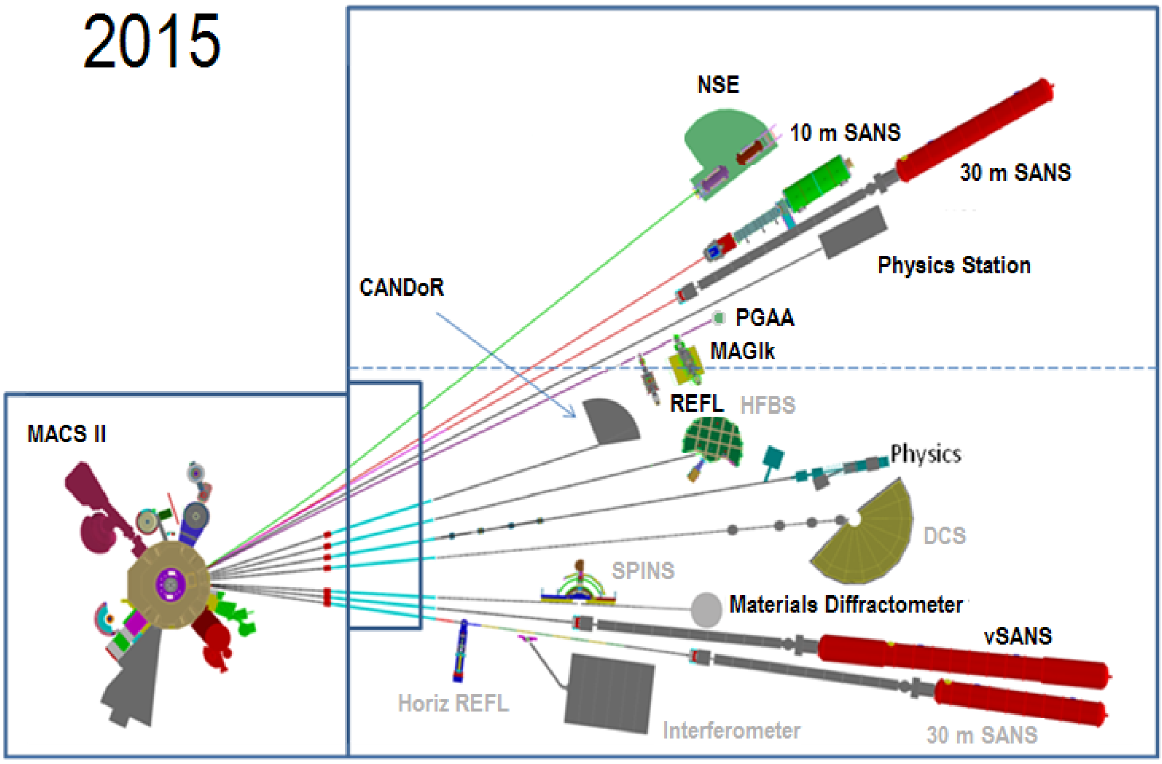

Figure 1. Anticipated layout of cold neutron instruments at the NCNR upon completion of the Expansion Project funded by the national American Competitiveness Initiative. The new guide hall, above the dotted line, nearly doubles the area of the cold neutron facility, and increases the number of guides from 7 to 12.

2. LD_2 Cold Source Performance Calculations

Monte Carlo Simulations using MCNP were performed to optimize the gain in cold neutron brightness of the LD_2 with respect to Unit 2, and to calculate the anticipated heat load of the LD_2 source. Reference 1 provides a description of the methods used for these calculations, which will be briefly summarized here. The NCNR maintains a very detailed MCNP model of the NBSR that has been used for relicensing of the reactor, LEU conversion studies, and cold source development. A powerful variance reduction tool of MCNP, the DXTRAN feature, is used to achieve good statistics for rare events, namely the tallies of cold neutron currents into narrow energy and angle bins at the entrance of neutron guides, far from the source. (At every collision, a pseudo particle is generated and directed toward the DXTRAN sphere [4] around the tally surface. The probability that it will be scattered and transported to the DXTRAN sphere is calculated, and its weight adjusted accordingly. Inside the sphere, pseudo particles are transported normally, contributing to the tallies. If the neutron actually reaches the sphere it is killed so as not to be counted twice.)

A two-step process is used, starting with the generation of a surface source file around the region of the cold source from a criticality calculation. The surface source provides the starting particles for subsequent calculations to study the effects of ***minor changes*** in the source geometry, LD_2 density, ortho-para fraction, etc. using the DXTRAN sphere. A minor change is one that does not affect the NBSR power distribution; major changes require separate surface source calculations. Recently, MCNP6.1 was released [4], so the gain and heat load calculations are being repeated using the latest version of the code and the ENDF/B-VII.1 cross section data [5] released with it.

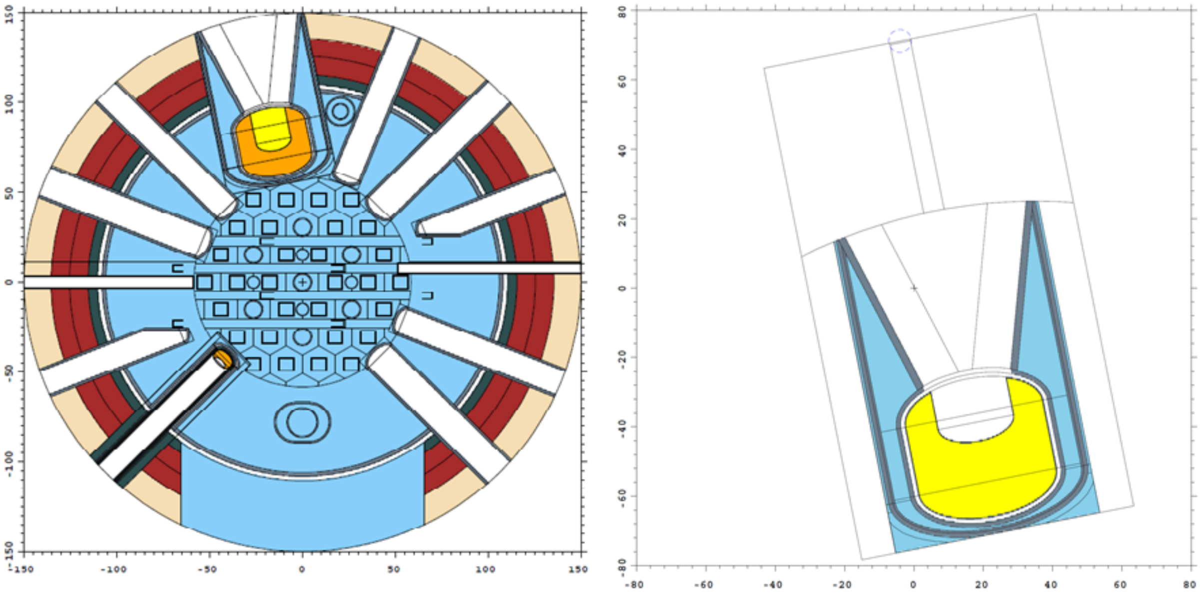

Figure 2. MCNP models of the NBSR core (left) and the LD_2 cold source (right). The surface source generated from the core calculation provides the starting neutrons for the CNS performance calculations (note the DXTRAN sphere, top right in the center).

2.1 Gain Calculations

The gain is defined as the ratio of the brightness of the LD_2 source to that of Unit 2 at a particular wavelength, or in general, for neutrons with wavelengths greater than 4 Å. The brightness in units of n/cm^2-s-ster-Å is obtained from the current tallies across a surface within the DXTRAN sphere. Simulations of cold neutron production and transport depend heavily on the scattering kernels (cross sections for low energy neutrons, or S(α,β) data) of the cold moderators. Kernels for ortho and para liquid hydrogen and liquid deuterium, and for liquid and solid methane are provided in the nuclear data released by the Los Alamos National Laboratory (LANL) with MCNP. The recently released ENDF/B-VII.1 data include continuous energy and angle S(α,β) data [6] and MCNP6.1 has improved interpolation routines that eliminate non-physical peaks and valleys in the current tallies with small energy and angle bins.

Because the LANL kernels for LD_2 were evaluated at 19 K but its boiling point at 100 kPa is 23.6 K, an alternate set of S(α,β) data was obtained [7]. These data, prepared by the Institut fur Kernenergetik und Energiesysteme (IKE), University of Stuttgart, included kernels for ortho and para LH_2 and LD_2 at a few temperatures. Introducing the IKE kernels for LH_2, however, resulted in quite a difference (15% to 35%) from the Unit 2 performance with the LANL kernels. The IKE ortho-LH_2 kernel has lower final energies than the LANL kernel for the same initial energies, so an "IKE model" of Unit 2 has substantially higher brightness than a "LANL model", all other parameters being equal. The actual ortho-LH_2 content of Unit 2 is unknown, but the shape of the spectrum indicates that the scattering is dominated by ortho, which has a much higher cross section.

The modeling discrepancy led to a series of time of flight measurements on one of the new guides, NG-Bu, to benchmark the MCNP model of Unit 2 against the brightness inferred from the measurements. The LH_2 average void fraction for the Unit 2 benchmark came directly from data collected from the original mockup tests conducted at NIST Boulder [8]. Using the ENDF/B-VII.1 kernels from LANL, the best agreement with the measurements was obtained with an ortho-LH_2 content of 25%, considerably lower than previously thought. At this writing, the benchmark is incomplete because MCNP6 has only been available for a short time. Using the brightness measurements, however, the gains calculated for LD_2 are with respect to the best possible TOF data available. The IKE kernels were used only to obtain an average correction factor to account for the flux decrease at 23.6 K versus 19 K, a factor of 0.88 over the range of 4 Å to 10 Å.

There are many other factors that affect the performance of the LD_2 source, such as the size of the vessel, the depth and diameter of the reentrant hole, the void fraction, and the ortho/para fraction. In general, the performance increases with the volume of the source, but not as fast as the heat load increases. We chose a 400 mm diameter, 400 mm long cylinder with a 220 mm diameter reentrant hole, 180 mm deep (see Figure 2). The resulting volume is 35 liters, about twice the size of any other LD_2 source. Our choice was a vessel that is as big as possible, but allowing ample room for the required vacuum, helium containment and heavy water cooling jackets. The cryostat assembly will be installed horizontally into a 550 mm ID thimble. The diameter of the reentrant hole was fixed by the requirement that the neutron guides extending ± 16° from the axis be fully illuminated to at least 10 Å. The reentrant hole depth represents a compromise between the highest gains at the longest wavelengths (~2) versus the losses at 2.5 Å (0.6). Most of the instruments in the guide hall use wavelengths between 4 Å and 9 Å.

To estimate the void fraction, the Kazimi/Chen Correlation for pool boiling [9] was compared to measured values for three cold source thermal-hydraulic mockups, the LH_2 mockup at NIST-Boulder [8], the R-134 mockup of the small BT-9 source at NIST [3], and the mockup of the horizontal LD_2 source [10] at the ILL. In all cases the correlation over-estimates the void fraction by about a factor of two. These mockups (and all three cold sources) vary significantly from pool boiling in that (1) there is no liquid/vapor interface (they are all flooded with a two phase mixture return flow) and (2) there is a continuous supply of liquid from the condenser. Therefore, for these situations the correlation is multiplied by 0.5:

$$\alpha' = 0.5 * \{ 1 + [0.645*(V_s/V_\infty)^{0.35}] / \ln (1 - 0.645*(V_s/V_\infty)^{0.35}) \}$$

where,

α' = CNS void fraction
V_s = superficial gas velocity = (volume of gas per second) /(flow area)
$V_\infty = [\sigma*g*(\rho_L-\rho_V)/ \rho_L^2]^{0.25}$
σ = surface tension
g = acceleration due to gravity
ρ_L = density of liquid, and
ρ_V = density of gas.

Applying the modified correlation the estimated average void fraction is 13%.

Deuterium, like hydrogen, exists in two states, ortho and para, owing to the nuclear exchange symmetry of the deuterons in the molecule. Unlike hydrogen, however, there is just a small difference in the scattering cross section between ortho and para, with the ortho state favoring the production of lower energy neutrons. At room temperature the ortho-LD_2 fraction, governed by quantum statistics, is 2/3. The liquid will reach an equilibrium ortho concentration of 0.955 at 25 K in the absence of radiation. In a cold neutron source, the molecular dissociation favors recombination in the 2/3 quantum ratio, so the ortho fraction will equilibrate at some point between those two limits. Raman spectroscopy measurements of the LD_2 source at SINQ (the spallation neutron source at the Paul Scherrer Institut in Switzerland) determined that the ortho content is 0.762 at a power density of 220 mW/g [11]. As seen in Section 2.2 below, the direct heat deposition in the NIST LD_2 source is calculated to be 290 mW/g, so the ortho fraction is expected to be about 75%. Using all the parameters above in the MCNP6 model, and using current tallies at the entrance of guide NG-B, the ratio of the calculated brightness of the LD_2 source to the measured brightness of Unit 2 is plotted in Figure 3. The average gain for cold neutrons, with wavelengths greater than 4 Å, is 1.5, with a gain of a factor of 2 at the longest wavelengths.

2.2 Nuclear Heat Load

MCNP6 was also used to calculate the expected heat deposition rate in the LD_2 and the moderator vessel. Aluminum alloy 6061 will be used for the vessel; the wall thickness will be 3.2 mm. The LD_2 source is much more massive than Unit 2 and the nuclear heat load will be about 3600 W (see Table 1). Because the heat load will triple with respect to Unit 2, and because the boiling and freezing points of LD_2 are about 3 K higher than those of LH_2, a new 7 kW cryogenic helium refrigerator is being installed. The apparent overcapacity is needed for the LH_2 cold source in BT-9 (operating at 200k Pa) and a contingency against unavoidable heat leaks and for future expansion. The refrigerator is scheduled to be fully operational in April 2014. The cryostat assembly, with a total mass of about 150 kg, is cooled by the D_2O Experimental Cooling System.

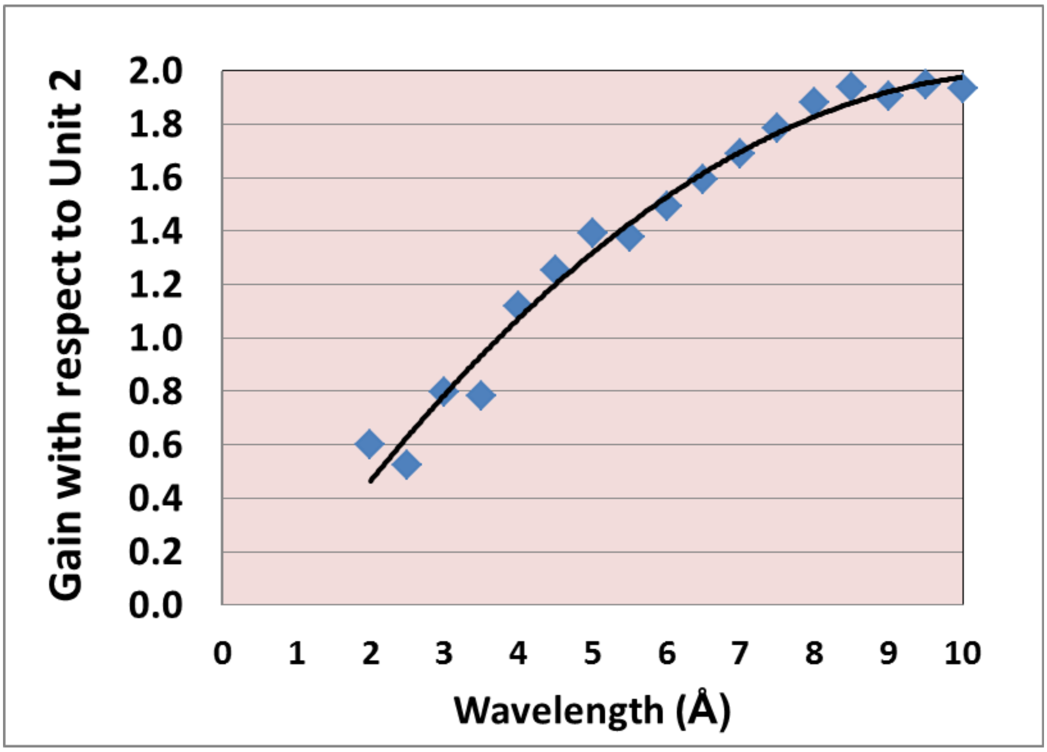

Figure 3. Anticipated gains of the LD_2 cold source with respect to the existing LH_2 source.

	Deuterium 5160 g		Aluminum 7155 g	
Radiation Source	**Rate (W/g)**	**Heat (W)**	**Rate (W/g)**	**Heat (W)**
Neutrons	0.0851	440	0.0008	6
Beta Particles	-	-	0.0793	567
Gamma Rays	0.204	1053	0.215	1538
Subtotal		1493		2111
TOTAL Cryogenic Heat Load = 3604 W				

Table 1. Heat Load in the LD_2 Moderator and Vessel.

3. Thermal-Hydraulics of the LD_2 Thermosiphon

The NIST LD_2 cold source must be installed horizontally into the cryogenic beam port, and the condenser installed no more than 2 m above the beam port (its height is limited by a radial crane). A naturally circulating thermosiphon is the simplest way to operate the source. Liquid from the condenser flows by gravity to the moderator vessel and the vapor produced rises back to the condenser with no need for a pump. There is one other horizontal LD_2 source, operating successfully in the High Flux Reactor at ILL since 1988. Extensive tests of a mockup of the ILL source were conducted to measure its thermal-hydraulic parameters to ensure that the thermosiphon would operate at the expected heat load, 3 kW, with an acceptable void fraction at 150 kPa [10]. The data from Reference 10 have been analyzed and scaled to model the NIST source, and this model was used to determine the size of the of the LD_2 supply line and the vapor/liquid return line for operation at 1 bar and a heat load of 4 kW [12]. The internal diameters of the supply and return lines will be 22 mm and 31 mm, respectively. The flow areas need to be greater than the ILL lines because that source has 3 m of head available, and operates at a higher pressure. To ensure the vessel remains completely filled with liquid, the portion of the return line extending into the moderator vessel will have two rows of small holes in the top, replicating the "piccolo" in the ILL source (and in the small LH_2 source at NIST).

4. The Deuterium System

The existing LH_2 sources at NIST are connected to ballast tanks providing a low pressure (less than 500 kPa) expansion volume in the event of a refrigerator failure. A very large 16 m^3 ballast tank is being fabricated for the LD_2 source to store the entire inventory at a pressure not to exceed 500 kPa. It is expected that the tank will be charged to an initial pressure of 400 kPa and the source operated at 100 kPa, but it is sized to operate at a higher pressure if desired. The tank will have a helium containment surrounding it and all of the connecting valves and pipes. It will be located outdoors along the west wall of the new guide hall, and a small enclosure, about 10 m^2, will be built to house instrumentation and the charging manifold, and to allow the cold source team to pump out the system and load it with deuterium.

Another consequence of the added heat load of the LD_2 source is the need to replace the existing 3.5 kW LH_2 condenser. A 6 kW, brazed aluminum, plate-and-fin type deuterium condenser is being fabricated and will be installed on the reactor face above the beam port. New LD_2 supply and return lines are required also. Vacuum jackets and helium containments will surround these components, as they do on the LH_2 sources.

A vacuum pump skid will be located on top of the guide shields to provide the insulating vacuums for all of the cold components. The vacuum system will be securely fixed to the shields and the vacuum lines protected, precluding a guillotine break in the lines and uncontrolled air ingress. The pumps will operate in a sealed, helium containment so in the event of a leak into the vacuum, the D_2 will mix with an inert gas. The LH_2 source in BT-9 will have its own vacuum pump skid (under construction) completely isolating the LD_2 vacuum system.

A conceptual design of the cryostat assembly, consisting of the moderator vessel, vacuum jacket, He containment, and D_2O cooling water jacket, is now complete. NIST has issued a request for proposals to fabricate the entire assembly and provide a complete quality assurance program to document the materials used (Al-6061), radiographs of all the welds, and the results of all leak and pressure tests. A contract for this work will be awarded in the next few months.

5. Safety

Many of the safety features of the LD_2 source have been described above. The underlying philosophy at NIST is that the cold source be simple, reliable and safe. This is assured by providing at least two barriers between the deuterium and air, minimizing gas handling, rigorous quality assurance standards, protecting components from physical hazards and by a passively safe design. All deuterium-filled components are surrounded by monitored helium containments maintained above one atmosphere to signal if a barrier is compromised. The system is loaded with D_2 and then sealed, hopefully for many years. The D_2 system is completely welded and checked for leaks via helium mass spectroscopy so that the leak rate is less than 10^{-9} STD cm^3/s (no detectable leaks). System components are also surrounded by protective shields, preventing an accident with the crane or a fork truck. Inside the confinement building, the piping to the ballast tank is located in a totally inaccessible floor trench. Thus a massive release of deuterium into the building with the reactor operating is not credible. In the event of a refrigerator failure, the LD_2 boils and the gas flows back to the ballast tank where it was loaded initially, requiring no active components or relief valves in the D_2 system. The reactor must be shutdown, however, until the refrigerator recovers to avoid overheating the moderator vessel.

An accident analysis is being prepared for the Engineering Change Notice, the standard internal review process required by the Nuclear Regulatory Commission to ensure compliance with 10 CFR 50.59 (the chapter in the Code of Federal Regulations concerning nuclear facility changes). The helium containment vessel surrounding the moderator chamber is designed to withstand the maximum pressure generated in the cryostat assembly by any credible leak of LD_2 into the vacuum system, or leak of air into the vacuum system. The Maximum Hypothetical Accident (MHA) assumes that the vacuum pump containment is inadvertently left filled with air instead of helium, followed by a pump failure allowing the air to flow into the vacuum and freeze on the surface of the moderator vessel. It further assumes that as the mass of oxygen reaches its maximum, the vessel fails and there is a LD_2 – solid O_2 detonation. Pressure measurements of such detonations were made by Ward et al [13] and the results can be scaled to predict the peak pressure. The peak pressure generated in the MHA would be 2.2 MPa, well below the design pressure of the helium containment which is greater than 6 MPa. There are already five other LD_2 cold moderators at other nuclear facilities, incorporating similar safety standards, accumulating decades of accident free operation.

6. Conclusion

The NCNR, continuing its commitment to expand its cold neutron research capabilities, is planning to replace the existing cold source with a LD_2 source that will provide an average gain of 1.5 for cold neutrons, and a factor of 2 for the longest wavelengths. The source is scheduled to be installed by the end of 2016.

7. Acknowledgements

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8. References

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