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The Liquid Hydrogen Moderator At The NIST Research Reactor

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ABSTRACT

In 1995, the NIST research reactor was shutdown for a number of modifications, including the replacement of the D_2O cold neutron source with a liquid hydrogen moderator. When the liquid hydrogen source began operating, the flux of cold neutrons increased by a factor of six over the D_2O source. The design and operation of the hydrogen source are described, and measurements of its performance are compared with the Monte Carlo simulations used in the design.

1. INTRODUCTION

The NIST Research Reactor located in Gaithersburg, MD is cooled and moderated by D_2O, and operates at a power of 20 MW. Its design incorporated a large thimble (540 mm D) for a cold neutron source, viewed by two ports separated by an angle of 35°. In the original (1960's) reactor planning, a large volume of D_2O ice, shielded from gamma-ray heating by a Bi tip, was foreseen as an ideal cold moderator. A 16-liter D_2O source [1] (with 7.5% H_2O) was installed in the reactor in 1987. That moderator, shown in Figure 1, was maintained at 30-40 K by cold helium gas flowing directly through the ice. The D_2O source produced a gain of 3 to 5 in the cold neutron flux, and its success lead to the construction of the Cold Neutron Research Facility [2].

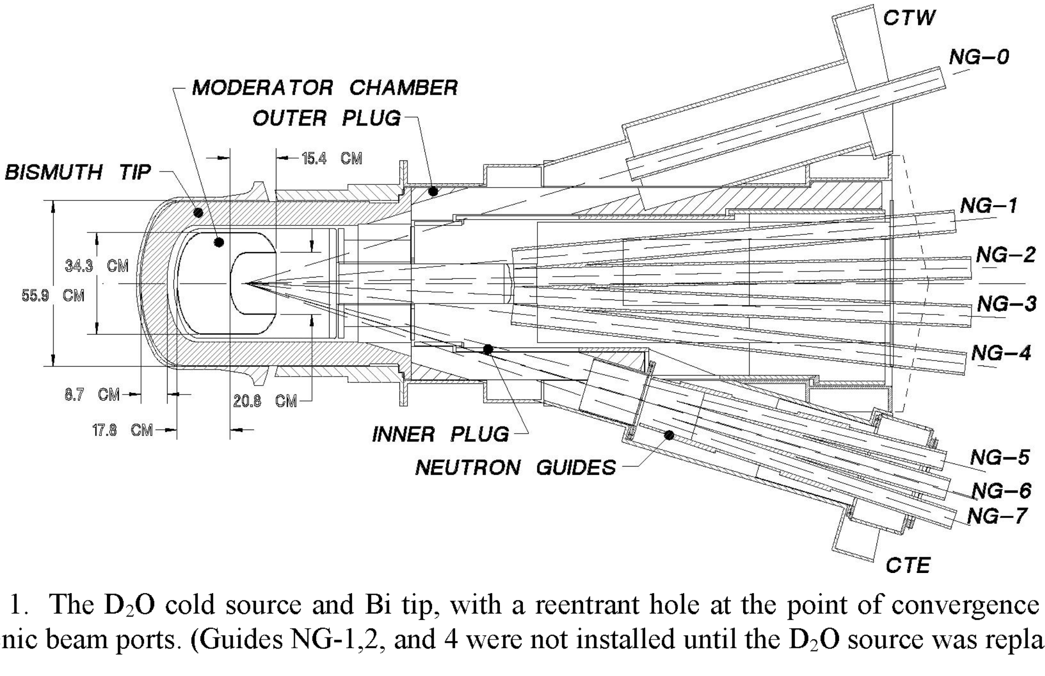

Figure 1. The D_2O cold source and Bi tip, with a reentrant hole at the point of convergence of the cryogenic beam ports. (Guides NG-1,2, and 4 were not installed until the D_2O source was replaced.)

The D_2O cold source was not without its problems, however, due primarily to the generation, buildup, and unpredictable recombination of D^+ and OD^- radicals. These "burps" could release up to one MJ of stored chemical energy, warming the source to well over 100 K, and sometimes causing a shutdown of the refrigerator. To avoid unplanned shutdowns, the source was intentionally warmed every two days, often interrupting experiments for hours. Because a 2 to 3 day reactor shutdown was required if the ice began to melt, a backup refrigerator was required to maintain the ice in the event of a refrigerator failure, the recovery from which required 8 to 10 hours. As the number of cold neutron instruments was growing annually, it became clear that the D_2O source was too labor-intensive and unreliable for the CNRF.

It was decided to begin work on a new source in order to provide better intensity and simpler operation. A liquid hydrogen source of novel design evolved through an iterative process balancing many factors, such as neutron performance, mechanical properties, thermal hydraulic behavior, and safety. This paper summarizes the design, operation and performance of the hydrogen source.

2. SOURCE DESIGN

The only realistic choices for a cold moderator in a 20-MW reactor are liquid hydrogen and liquid deuterium. While solid methane is a better moderator, the effects of radiation damage render operation difficult and complex [3], as was the case for the D_2O ice source. Liquid hydrogen was selected rather than deuterium because it required a much smaller inventory as a result of the larger scattering cross section, and it has a negligible tritium production rate. Monte Carlo simulations eventually showed that with the design adopted, in the flux gradient and geometry in the NIST reactor, the performance of hydrogen in the wavelengths of primary interest to us (2 - 10 Å) is not markedly inferior to that of deuterium.

As a result of the divergence of the existing beam ports and the height of the NIST neutron guides (150 mm), a viewed area of 300 x 300 mm^2 was essential to provide full illumination of the guides to a reasonable wavelength. We were inspired by the design of a cylindrical annulus hydrogen source for the Orphée reactor [4] to investigate annular designs, and launched an intensive simulation effort to compare various possible geometries. A sphere is an obvious choice for mechanical purposes, as it minimizes stress for given internal pressure, but a complete spherical annulus would move the viewed surface a long distance from the core, lowering intensities markedly in the large flux gradient of the NIST reactor. Eventually, we settled on the geometry shown in Figure 2, with part of the annulus removed to allow the inner surface to be viewed by the neutron guides. Intuitively, this geometry is attractive, since it more closely approaches the optimum "black body" situation. This intuition was verified by detailed simulations using MCNP [5], for which a complete model of the reactor was developed, including the exact fuel geometry, all beam ports, the thermal column, etc. Thus, the simulations, automatically included all perturbations introduced by the source. As a result of the simulations, an annulus thickness of 20 mm was chosen, which is a compromise between the weak dependence of performance on thickness (the true optimum occurred at approximately 25 - 30 mm), total hydrogen inventory, and heat load. With an outside diameter of 320 mm, the volume of the LH_2 annulus is 5 liters.

To ease fabrication and lower the heat load, the inner sphere of the system is open to the liquid, and is therefore filled with hydrogen vapor. The 300 mm of dense gas in the path of the neutron beams is certainly not desirable for neutronic performance. Beam losses were judged to be acceptable, however, based upon estimates of the ortho-para ratio being maintained at 1:4, a balance of natural conversion (para being the equilibrium form in LH_2), and ortho production (by radiative dissociation followed by recombination in the statistical ratio of 3:1). Since a predominantly para concentration is not optimal for neutronic performance in a 20 mm liquid annulus, a recirculation system was built to draw room temperature hydrogen gas over a catalyst and pump it to the condenser, to maintain a nearly normal 75% ortho content (see Figure 3). A tube separates the liquid from the gas to limit exchange of hydrogen between the two volumes to diffusion, so we expected the much lower neutron scattering cross-section of the para-rich gas would reduce the attenuation to acceptable levels.

The mechanical realization of the chosen design required optimization of many different parameters, including fabrication, heat load, licensing issues, boiling heat transfer and others. The wall thickness of the moderator vessel was selected as 1.5 mm in order to meet the requirements of the pressure vessel code (ASME Section VII). The inner sphere, however, is very thin, as it is subjected to no pressure differential. All components exposed to radiation are made from Aluminum 6061-T6 in view of the wide experience with this material in U. S. reactors, and all joints are welded. Figure 4 shows the moderator chamber inside the cryostat assembly, surrounded by its insulating vacuum, He containment, and D_2O cooling jacket.

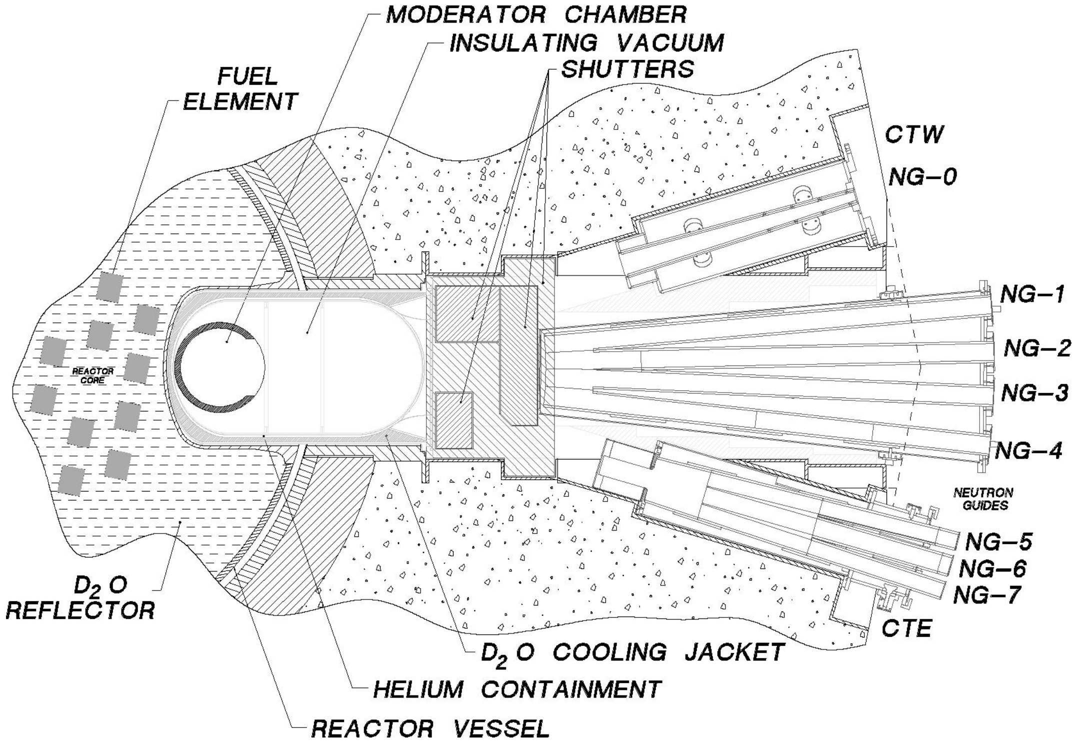

Figure 2. Plan view of the liquid hydrogen cold source, the reactor core and neutron guides.

Of the several existing choices for the mode of operation and cooling [6], a thermosiphon was chosen with a condenser mounted on the reactor face, 2 m above the cryostat beam port, and liquid hydrogen supplied by gravity. As the moderator chamber design developed into the present unconventional geometry, concerns about the stability and performance of such a system led to building a full scale mockup at the NIST laboratory in Boulder to test the system over a wide range of conditions [7]. The model, fabricated from glass spheres, contained a heater winding in the annulus to simulate the nuclear heat load, and a balance to measure the hydrogen mass. The tests demonstrated that the void fraction in the boiling LH_2 was 8 to 12% at 800 W, and that such a thermosiphon can continuously remove up to 2200 W.

Our safety philosophy for the source is quite similar to other sources - reliance on passive systems, and the use of inert gas (either helium or nitrogen) around all hydrogen containing systems. There are, however, some differences which are of sufficient interest to warrant discussion. The first is that *all* systems are surrounded by inert gas, including the insulating vacuum pumps, which discharge into closed He-filled vessels preventing the possibility of a pump failure leading to air backfilling of the space surrounding the hydrogen. The second is that there are no relief valves anywhere because the safest place for the hydrogen in the event of a rupture of any component is contained in the system, rather than vented to the atmosphere. Finally, the source is always open to the 2 m^3 ballast tank shown in Figure 3, so in the event of a refrigerator failure, the hydrogen will expand into the tank, where it resides normally when the system is shutdown. To protect the moderator chamber from overheating, an automatic reactor rundown is initiated if the hydrogen pressure increases to the point that the moderator chamber is no longer full of liquid.

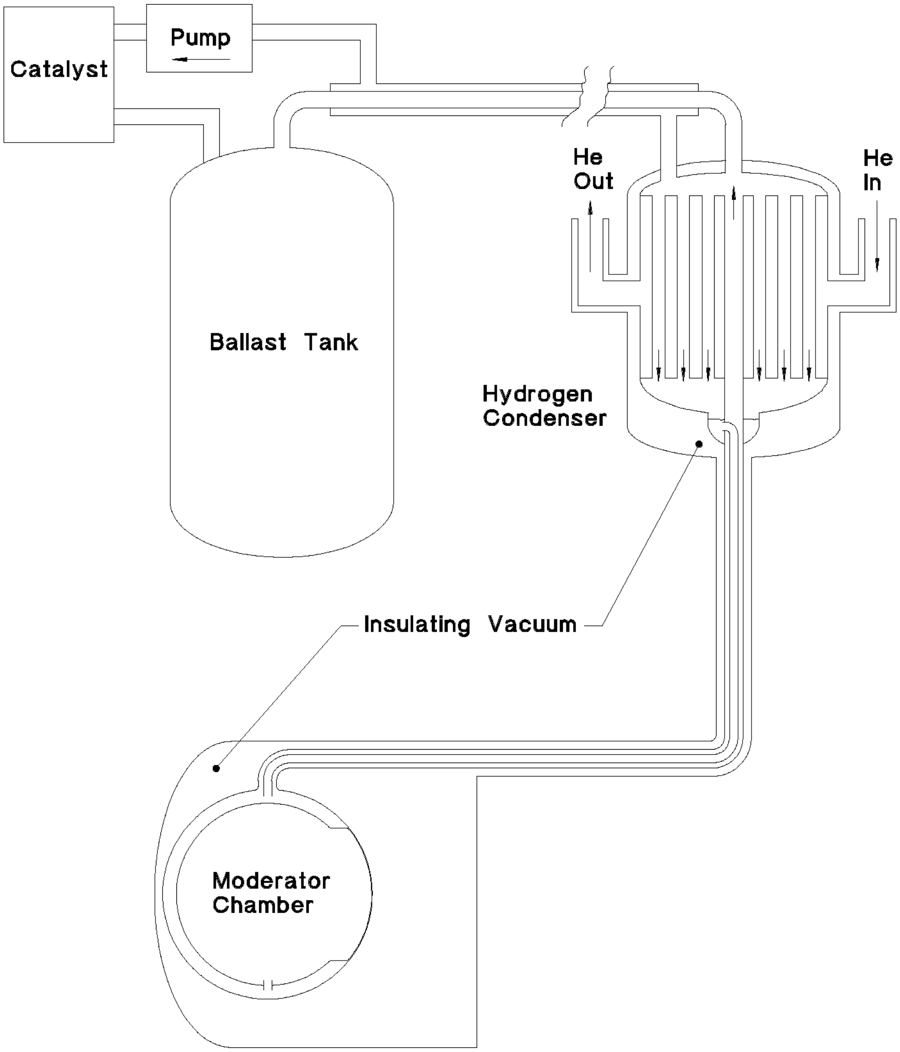

Figure 3. Schematic of the thermosiphon for the hydrogen cold source. All components are surrounded by He containments, not shown. There are two concentric hydrogen supply lines between the ballast tank and the condenser, one of which contains an ortho/para catalyst and recirculating pump at room temperature.

3. OPERATION

Even before the hydrogen source was designed, a new 3.5-kW helium refrigerator was installed for its operation. The refrigerator is PLC controlled, so it can automatically proceed through a program to its normal operational mode. When the cold source was installed, the same PLC was used to monitor the source. A single parameter, the hydrogen pressure, provides feedback to the refrigerator to modulate the flow of cold helium through the condenser as needed to maintain the hydrogen system pressure at about 105 kPa, just above one atmosphere. A load bypass line is always open, and a heater maintains the temperature of the helium returning from the load and bypass at

18 K. Thus by throttling a single valve in the refrigerator load line, varying the ratio of the flow bypassed to the flow through the condenser, the refrigerator can follow the cold source heat load as the reactor power varies from zero to 20 MW. The PLC is also programmed to restart the refrigerator after a power failure, which it can accomplish in about 2 minutes.

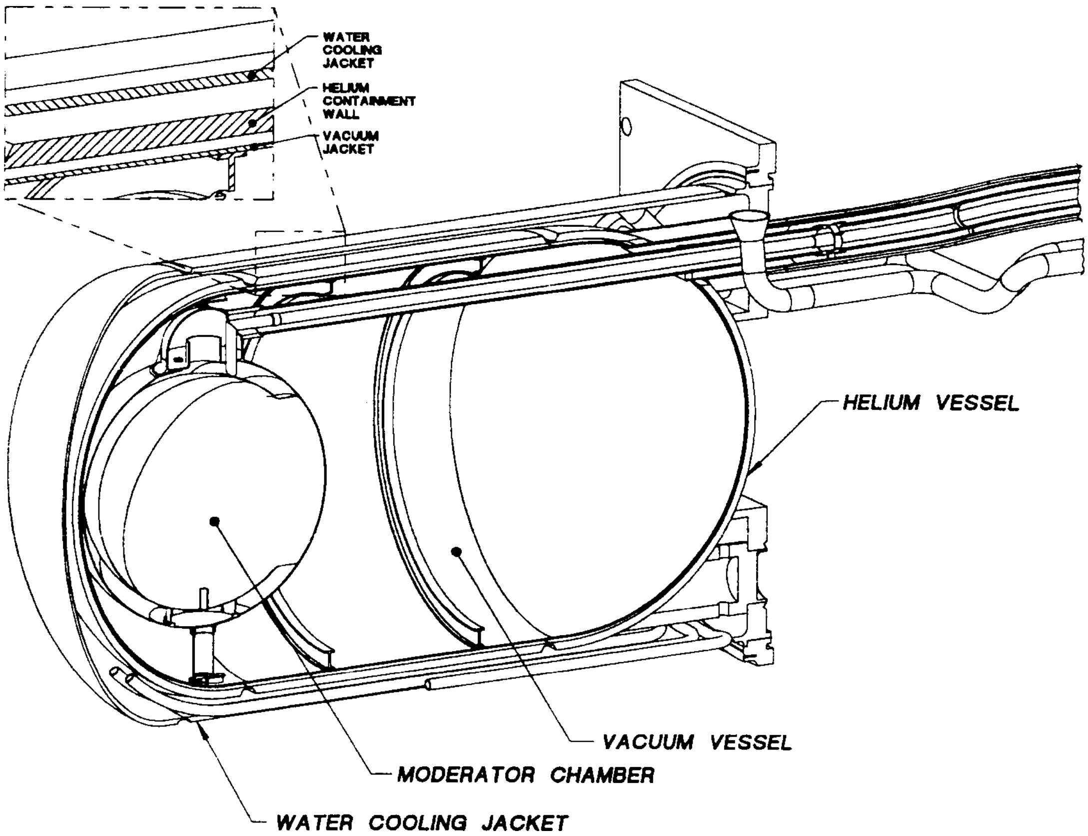

Figure 4. Side view of the cryostat assembly, which was inserted horizontally into the cryogenic beam port of the NIST Research Reactor.

By comparing the refrigerator heater power at zero and 20 MW, the nuclear heat deposited in the source has been measured as 800 ± 40 W, compared to a calculated value of 980 W (290 W in hydrogen and 690 W in the aluminum chamber) using MCNP. Photon production data for ^{235}U and ^{27}Al were modified for these calculations to include delayed gamma rays from fission products and ^{28}Al, respectively [8]. From the D_2O flow and temperature increase, the heat deposited in the cryostat assembly was also measured as 30 ± 2 kW, compared to the calculated value of 22 kW. The cryostat assembly is not thermally isolated from the reactor thermal shield, however, and is no doubt removing heat from its environment as well as the nuclear heat directly deposited in the assembly. The moderator chamber is thermally isolated by a vacuum, and one would expect better agreement.

Our original estimates of the ortho/para content of the moderator were not verified by the behavior of the source; the rate of ortho production due to radiation appears to be much higher than anticipated. Early papers on the subject [9, 10] reported that the presence of radiation stimulates the conversion from ortho to para, which according to the MCNP simulations would result in a drop in cold neutron production. No corresponding change in the intensity is observed, however, in the hours after the reactor is started. In addition, the recirculation system with the para-to-ortho catalyst, which delivers 0.1 g/s of normal hydrogen to the condenser, makes no change in intensity. We conclude that the ortho content of the liquid hydrogen remains sufficiently high, about 50%, to dominate the neutron scattering in the moderator. This conclusion is further strengthened by the observation that decreasing system pressure from 160 kPa to 105 kPa increased neutron intensities preferentially at long wavelengths, in contrast to an anticipated loss in intensity resulting from a larger void fraction. Greater attenuation at the higher density was due to cold neutrons scattering from the beam; para hydrogen would be nearly transparent to cold neutrons.

4. SOURCE PERFORMANCE

The neutron performance of the source has been evaluated in many ways, including gain over the previous D_2O ice source, gain full to empty, absolute flux, and cold neutron spectrum. The measured gains are shown in Figure 5 as a function of neutron wavelength. A time-of-flight measurement of the cold neutron spectrum was obtained at the end of NG-1, and plotted against the MCNP estimates in Figure 6.

Several comments must be made about the results in Figure 5. First, as noted in the previous section, our experience indicates that the ortho fraction in the gas in the inner sphere is at least 50%. This has two effects - a reduction in gain at long wavelength due to higher attenuation in the vapor-filled inner sphere, and an overall reduction in gain due to increased void fraction. With these two caveats in mind, the agreement with calculation is highly satisfactory, and the cold neutron gains (full/empty) are as high as those seen for any hydrogen source. Estimation of the losses due to the two effects mentioned suggest that this source geometry offers excellent cold neutron performance. The gains measured at all of the instruments are fully consistent with those shown here, which were measured at the NG-7 30-m SANS. The intensities at approximately 2.5 Å for the SPINS triple axis spectrometer are equal to or better than those at the best spectrometers at thermal beam ports at similar resolution (note that the NIST guides are not curved), providing excellent continuous measurement capability from 2.5 Å to 6 Å.

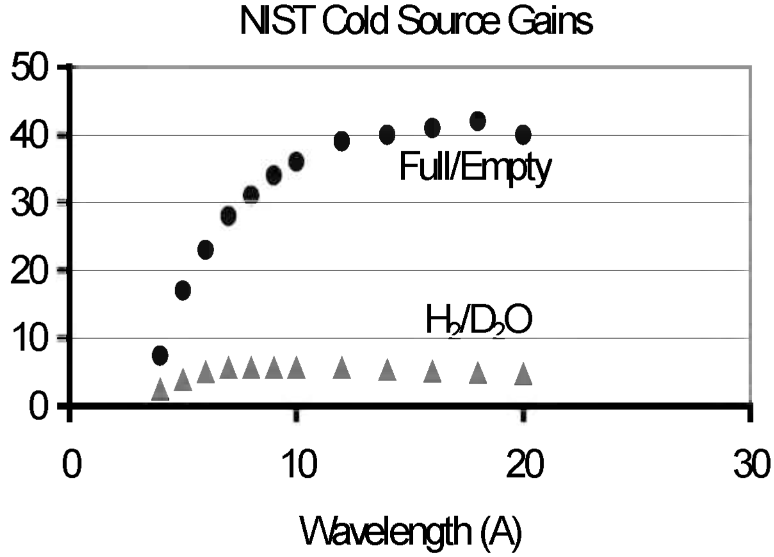

Figure 5. Neutron gain vs. wavelength. The bottom curve is the gain with respect to the old D_2O source, and the top curve is the gain with respect to the source while warm.

Another benchmark for source performance is the total capture neutron flux in the guides, an integral measurement which is most sensitive to the longest wavelengths (*e.g.* for a 45 K Maxwellian spectrum, over half of the capture flux comes from neutrons with wavelength greater than 10 Å). Hydrogen sources are certainly inferior to deuterium sources at the longest wavelengths because of absorption, and as stated earlier, the primary region of interest is below 10 Å. For the present source, the capture flux measured inside the neutron guide hall at a point 35 m from the source is in excess of 2 x 10^9 n/cm^2/s. Allowing for guide losses and for the high ortho/para ratio in the gas as discussed above, this number is in satisfactory agreement with the calculated value of 4 x 10^9 n/cm^2/s, assuming no losses.

The measured cold neutron spectrum is compared with the results of the MCNP simulations in Figure 6 The TOF data has been corrected for solid angle, guide acceptance and reflectivity, and absorption in aluminum windows, to determine the source brightness. Both the shape and intensity of the measured spectrum agree very well with MCNP calculations in which the ortho content of the LH_2 was 65% (best fit) [11]. Simulations with 100% para hydrogen predict intensities above 4 Å drop by a factor of two. All of the performance measurements are consistent with our conclusion that the liquid hydrogen is maintained with at least 50% ortho.

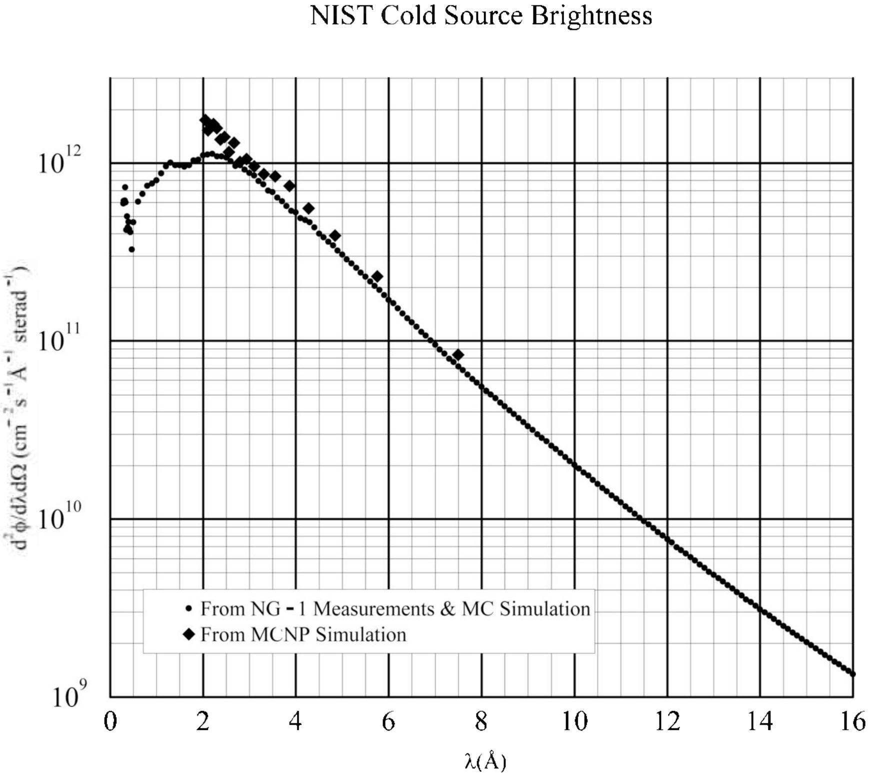

Figure 6. Calculated and measured cold neutron spectra. The measurements have been corrected for guide acceptance and reflectivity, and absorption in Al.

5. FUTURE IMPROVEMENTS

There are improvements to the present source which can be foreseen for the next version of the source, which could be installed at the next major shutdown. The most obvious, which only became obvious when the tests of the present source were complete, involves removal of the cold dense gas in the inner chamber by evacuating this volume. This will require that the total mass of the aluminum container be increased substantially (with a concomitant increase in heat load) in order to handle with the stresses imposed on the inner sphere. The resultant increase in void fraction will involve losses which could be compensated by increasing the pressure and an increase in annulus thickness, again at the expense of added heat load. The tests at Boulder indicated that the expected heat load of 1200 W can easily be handled.

A second improvement, discovered during construction of the present source, but too late to incorporate, involves reducing the reactor perturbation caused by the cold source by increasing the volume of heavy water reflector incorporated into the cryostat assembly. An ellipsoidal annulus is being studied that allows even more reflector. Since the vertical acceptance of our guides is greater than the horizontal acceptance, an ellipsoidal annulus is superior to a spherical annulus, while continuing to fully illuminate the guides. Simulations indicate that if we are able to incorporate of all of the above changes, it is possible to nearly **double** the cold neutron flux.

6. CONCLUSIONS

A new hydrogen cold source installed at the NIST reactor, which has an unusual geometry, has provided substantial gains over the previous D_2O source. The performance of the source agrees well with detailed Monte Carlo simulations performed with the MCNP code.

7. ACKNOWLEDGEMENTS

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