Lujan Neutron Scattering Center's TMRS Mark II and Mark III Operational Successes and Lessons Learned

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Abstract. The Los Alamos Neutron Science Center (LANSCE) Lujan Neutron Scattering Center (Lujan Center) is a User Facility that provides neutrons for a broad spectrum of science. The neutron source is a single piece Target-Moderator-Reflector System (TMRS). After eight run cycles of operation, TMRS Mark II was replaced with TMRS Mark III in April 2010. Several scientific and operational upgrades were designed into Mark III including cladding of the tungsten targets with tantalum, replacement material for the hydrogen transfer lines, elimination of the hydrogen transfer line bellows assemblies, and inclusion of a cold beryllium filter for one of the cryogenic moderators. Operational upgrades were intended to improve reliability of the TMRS and enhance maintainability of supporting systems.

1. Introduction

The Target-Moderator-Reflector System (TMRS) is a short pulse neutron source at the LANSCE accelerator that generates moderated neutrons from an 800-MeV proton linear accelerator. The target assembly is designed for operations at 200 uA and 20Hz. TMRS Mark I operated between 1998 and 2001 and received 500 mA-hrs of integrated beam current, TMRS Mark II operated from 2002 to 2009 and received 2400 mA-hrs of integrated beam current, and TMRS Mark III was installed in 2010 and has so far received about 700 mA-hrs of integrated beam current.

The TMRS Mark I was replaced early after only 500 mA of integrated beam current due to a known cooling deficiency in the lower lead reflector and a suspected cooling shunt of the upper target. An increase in the flow coefficient through the upper target assembly led system experts to believe that the water was being bypassed through a weld failure in the plenum common to both the upper target's inlet and outlet cooling lines.

TMRS Mark II featured a new design of the lower reflector utilizing stainless steel and beryllium and a redesign of the upper target. The new reflector corrected the Mark I cooling problem and also eliminated further generation of mixed waste (lead is considered hazardous and is regulated as a RCRA metal and once it becomes radioactive it becomes very costly to dispose of). The upper target's supply and return manifold was redesigned with independent inlet and outlet lines so that cooling water could never be shunted. Operation of Mark II for nine run cycles proved to be very challenging from a reliability and personnel safety standpoint. Because so much was learned during this time, the focus of this paper will be a discussion of lessons learned during the lifetime of Mark II and what was changed for Mark III.

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Mark III includes tungsten targets clad with tantalum, Invar cryogenic transfer lines, and a redesigned hydrogen moderator. All three major changes have proven to be a success so far.

2. Water Cooling Systems

The TMRS is cooled by three pressurized closed loop water systems and a liquid hydrogen system. The three water systems are the Target Water System, the Reflector Water System, and the Moderator Water System (MWS). Each water system contains two water pumps, a heat exchanger, a water purification loop, a surge tank, water make-up and drain valves, flow control valves, flow, pressure, temperature, and water chemistry instrumentation, and control system.

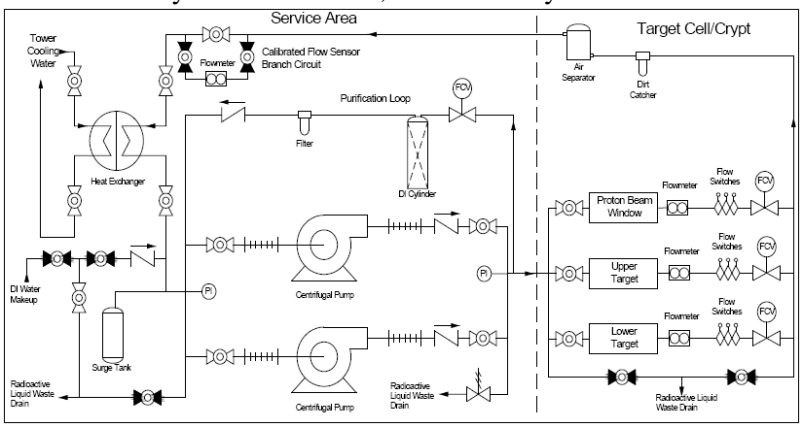

Figure 1

The target water purification loop is a slip steam design that shuts approximately 10% of system flow at a flow rate of about 11 l/min (3 gpm) to minimize soluble and insoluble materials that become activated from interaction with the proton beam and secondary particles. The target water system services all components where the proton beam can pass directly through the cooling water resulting in the most complicated chemistry of the three water systems. Resultantly, many of the lessons learned have originated from difficulties in operating and maintaining the target water system.

2.1. Target Water History and Operational Issues

During the first four years of TMRS Mark II operation several target water system failures resulted in a loss of beam to target. The primary culprit was target safety equipment such as spring loaded flow switches and paddlewheel flow sensors. Component failures were a result of cuprous oxide deposit on these moving components. Although nearly all of the copper containing components had been removed prior to installation of Mark II, the one remaining component was a braze pack heat exchanger which contained stainless steel plates brazed together with copper. The amount of copper was thought to be small and was not appreciated until the heat exchanger was replaced with a nickel-brazed heat exchanger. After this, component failures were nearly eliminated.

The replacement of the heat exchanger was preceded by a major corrosion excursion in the entire target water system. The purification loop, at the time, contained an oxygen resin bed followed by an anion/cation mixed resin bed followed by 10 micron and 1 micron cartridge filters. The loop also contained inlet and outlet water sample lines to allow periodic water sampling. The pH of the process water was not controlled but water sample indicated a fairly steady pH of 6 to 6.5 for most of Mark II's lifetime. The by-products of beam interaction with cooling water such as H_2O_2 and O_2 were realized but probably under appreciated. With beam off target the oxygen scavenging resin could maintain oxygen levels at less than 10 ppb but with beam on target the oxygen levels typically saturated to approximately 50 ppm. These beam cycles and subsequent swings in oxygen concentration from one extreme to the other subjected the heat exchanger's copper brazes to the high corrosion conditions, figure 2 [1].

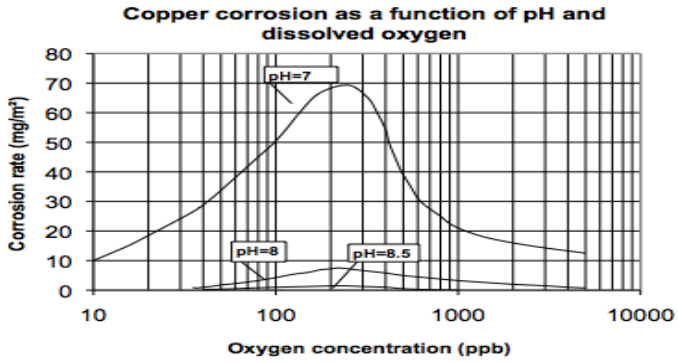

Figure 2

Corrosion of the copper brazes as well as erosion/corrosion of the bare tungsten targets typically saturated the mixed bed in about three months of operation. For personnel radiation exposure concerns, the DI bottles were not replaced during the run cycles. The service equipment room had a limited allowable floor loading which made it impossible to increase DI loop lifetime by adding extra bottles. The 60-300 R/hr bottles required a four inch thick steel cask to help reduce personnel exposure during annual DI bottle replacement and to help reduce general area radiation levels. General area radiation levels increased drastically if the target water system was operated without the purification loop in service, even if the beds were expended. For the first four years of Mark II operation, the philosophy was to leave the purification loop in service after depletion of the DI bottles to retain the insoluble material filtering capability using the installed filter(s) and even the DI resin as a mechanical filter. This practice resulted in negative consequences during the 2005 run cycle. Post run cycle water analysis revealed a steady lowering of the water pH from 6.8 (January 2005) to 4.6 (October 2005) with a steady increase in the concentration of metal cations (mostly Cu^{2+}) and anions such as chlorides and sulfates which resulted in decomposition of the ion exchange resins and serious corrosion damage throughout the water system. All target water system piping and components were replaced during the 2006 accelerator outage at which time the copper brazed heat exchanger was replaced with a nickel brazed heat exchanger. The oxygen-scavenging cylinder was not reinstalled but was replaced with a second mixed bed DI cylinder in an attempt to double the operational lifetime of the purification loop. During the final four run cycles of TMRS Mark II, the average DI cylinder lifetime was 8-10 weeks each and once the resin beds were expended the bottles were isolated but cartridge filters were left in service.

During the 2006 run cycle, with approximately 1300 mA-hrs of integrated beam current on the target, the beam window formed a leak, which was most likely due to the previous years corrosion excursion. Although post mortem analysis has not been performed, it is suspected that the leak was at an electronic beam weld joint for a thermocouple pad on the Inconel 718 window face. The leak continued intermittently for the remaining operational lifetime of the target (1100mA-hrs) and at its peak leaked 3 gallons of water per day into the crypt vacuum space. The crypt was evacuated with an oil-lubricated pump until the onset of this leak. Water vapor emulsified the oil rendering this pump useless, so dry a vacuum pump was utilized for the remainder of Mark II and continues for Mark III.

3. Cryogenic System Operational Issues

The TMRS Mark I to Mark III target assemblies have all incorporated two liquid hydrogen moderators operated at 20 Kelvin. Mark I and Mark II target assemblies utilized a formed bellows on the otherwise rigid stainless steel insulating vacuum jacket to relieve stress created when the inner stainless steel transfer line contracted at cryogenic temperatures. Three run cycles of vacuum jacket bellows cycling (<100 cycles) resulted in leaks in two of the bellows. The bellows were not replaceable or easily repairable so a silicone sealant patch was applied around the bellows to

temporarily plug the leaks. Due to radiation damage of the silicone, it was peeled off and replaced twice over the next four run cycles.

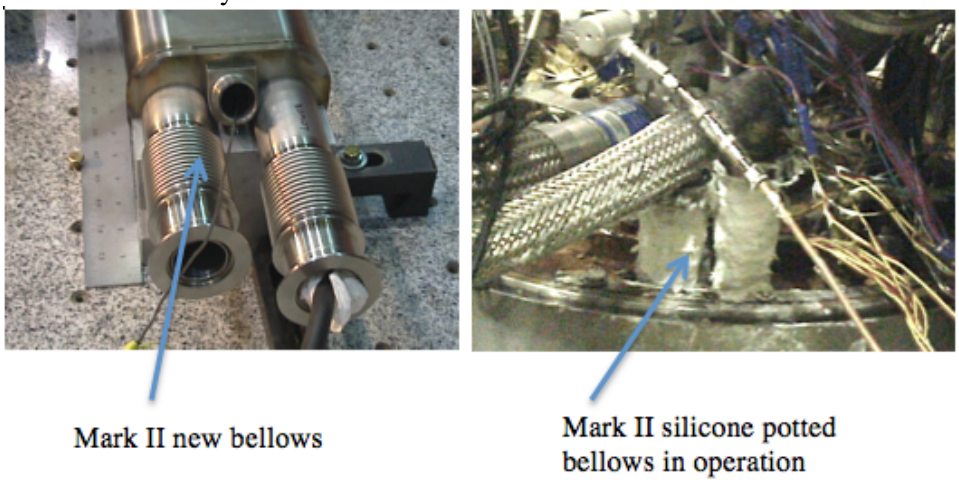

Figure 3

4. TMRS Mark III Design Improvements and Results

Many lesson were learned during the operation of TMRS Mark II and several of these lessons were addressed during the design of TMRS Mark III.

4.1. Target Design Improvements and Results

Tungsten has many favourable properties as material for a spallation neutron source but also has some drawbacks including poor corrosion and erosion resistance when subjected to a proton beam and associated cooling water. One approach to avoid these issues is to clad the tungsten with a material such as tantalum or stainless steel. Favourable clad material properties along with previous cladding successes at both KENS (spallation neutron source at KEK) and ISIS [2] led us to choose tantalum as the cladding material. A customized HIP'ing (hot isostatic press) process was perfected and used to clad the tungsten target with .010" (.25 mm) tantalum.

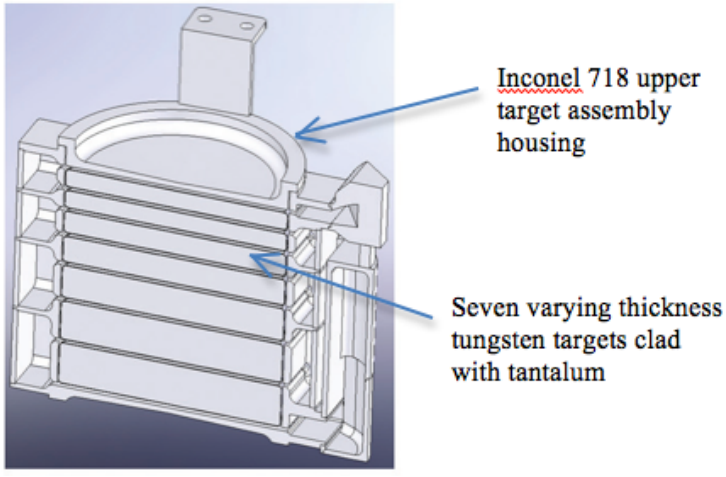

Figure 4

Mark III has been in operation for two run cycles (700 mA-hrs) and the results of the cladding have been positive. General area radiation levels have decreased by a factor of 10 compared to MK II levels and the lifetime of a single target water DI cylinder is approximately 7 months. Only one set of

DI cylinders has been removed since installation of Mark III and the dose rate was 15 R/hr. This compares favourably with the first bottles removed from Mark II, which were above 100 R/hr on contact.

The first six months of water samples showed only trace amounts of stainless activation products and no spallation products. After six months (250 mA-hrs) of operation, area radiation levels started increasing slightly due to the first presence of tantalum spallation products in the cooling water. From this point on, water samples have been gradually increasing in activity and the first sign of tungsten (W-187) were observed after 8 months of operation (330 mA-hrs).

In addition to gamma spectroscopy, water samples are analyzed for metals using Inductively Coupled Plasma (ICP) mass spectroscopy. Of primary interest is tungsten and tantalum, neither of which has increased over new target baseline values. If tantalum were to have eroded to expose tungsten, the presence of tungsten would increase in time, which is not the case. Calculating the flow coefficient through the upper target is a useful diagnostic for determining if flow resistance is changing. An increasing flow coefficient would indicate a decreasing resistance and thus possible erosion. As is evidenced by figure 5, the during the first two run cycles of Mark III the upper target flow coefficient has remained steady whereas during the first two run cycles of Mark II the flow coefficient increased. Mark II's flow coefficient continued to increase throughout its lifetime indicating possible erosion of the tungsten target.

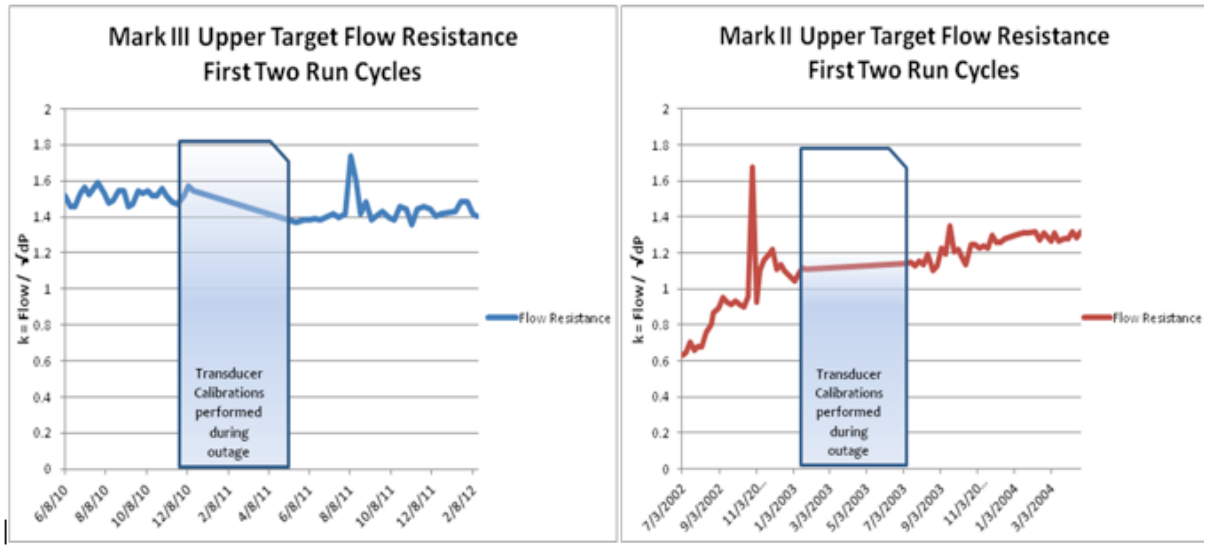

Figure 5

4.2. Cryogenic Design Improvements and Results

The bellows assemblies were eliminated from the liquid hydrogen moderator's vacuum jacket. Thermal contraction gradient stress between the cold inner hydrogen lines and the warm vacuum jackets were eliminated by the use of Invar metal for the cold inner lines. Invar has very low thermal contraction when compared to 304 or 316 stainless steel.

The TMRS Mark III lower hydrogen moderator was redesigned to include a beryllium pre-filter as shown in figure 6. This pre-filter is located downstream of the hydrogen moderator and also provides mechanical support for the adjacent moderator wall. It is cooled by contact with the cold moderator. The primary goal of the pre-filter was to cause a factor of two increase in the low-energy part of the neutron spectrum in the flight paths that view this hydrogen moderator, while not increasing the heat load on the hydrogen moderator from contact with the beryllium over that seen in TMRS Mark II.

Initial results indicate that the desired improvement in the low-energy neutron spectrum has been achieved, with improvements of up to a factor of 2.4 seen. The heat load on the hydrogen moderator

has increased significantly, however. Beam-related heat loads in both Mark I and Mark II were measured to be about 2 watts per microampere of beam on target. The same measurement on Mark III yields 4.5 watts per microampere; the excess evidently being due to the beryllium. The parasitic heat load with Mark III is not significantly different from that of either Mark I or Mark II.

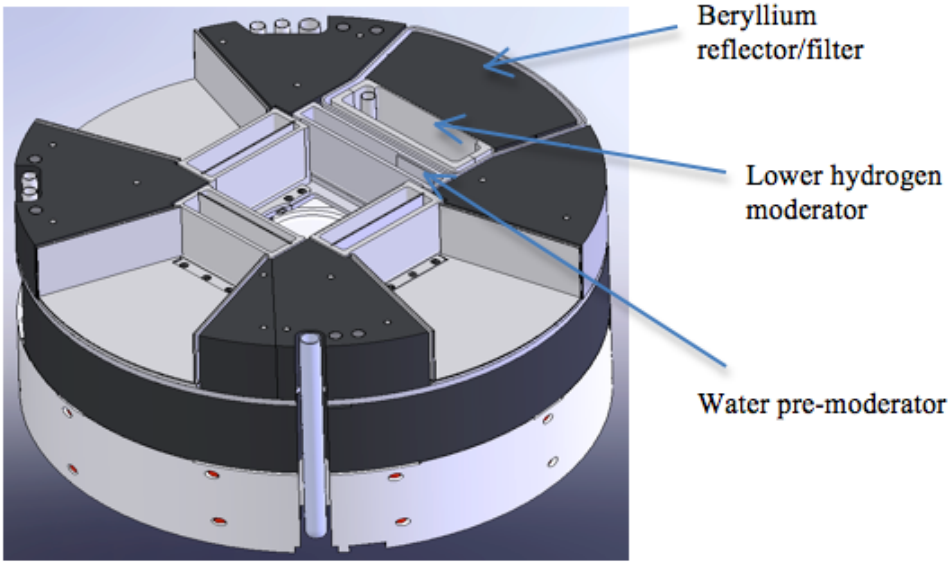

Figure 6

4.3. Window Design Improvements and Results

During the design of Mark III it was determined that the window thermocouples located on the face of the window were under utilized as a diagnostic for target operations or beam steering and so were removed from the Mark III design. This eliminated the possibility for future weld corrosion leaks at on the window face.

5. **Conclusion**

Despite a few design shortcomings of TMRS Mark II, it proved to be very robust and reliable during its eight run cycles of operation as there were no catastrophic failures after 2400 mA-hrs of operation. Several operational challenges presented themselves in the form of a window leak, hydrogen transfer line bellows leak, erosion of the tungsten targets and severe corrosion of the target water cooling system.

The most significant change to the cooling systems was removal of all copper components from the target water system, which increased target reliability and decreased radiation exposure for the operations and maintenance personnel. Many improved practices have been learned from the target water chemistry control standpoint but we still have more to learn.

Cladding of the tungsten targets is by far the greatest operational improvement for TMRS Mark III. It has reduced area radiation levels by a factor of ten and increased DI resin bed lifetime resulting in ability to complete a run cycle without expending both resin bottles. After two years of operation, trace amounts of tungsten have appeared in the water but it is not believed to be from erosion but possibly from a hairline crack in the tantalum. We will continue to monitor for the presence of tungsten and the overall performance of the target.

6. **References**

[1] Kawai M, et al. 2003 R&D of a MW-class solid-target for a spallation neutron source 2003 *Journal of Nuclear Materials* **318** 38-55