COLD NEUTRONS AT NIST

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Received July 1, 2005

The design process for the series of cold neutron sources installed at NIST is presented, with particular emphasis on the reason for the decisions and choices made. These developments are used to illustrate some of the general principles of CNS design.

KEYWORDS : HANARO2005, Abstracts, CNS Design, Hydrogen, Deuterium, NIST, NCNR

1. INTRODUCTION

Over the past 20 years, the NIST Center for Neutron Research has been developing improved capabilities for the production of neutrons with long wavelength. Starting in 1985 with a commitment to develop a D_2O ice cold neutron source, installation of a first generation liquid hydrogen source in 1994, a second generation source in 2002, and continuing to the present with design of a liquid deuterium source, the emphasis has been on cost-effective upgrades to better serve the U.S. scientific community. At the same time, continuous beam delivery and instrument installation and upgrade has steadily improved the experimental capabilities. The result has been a steady growth in facility use and output, and a steady stream of new science.

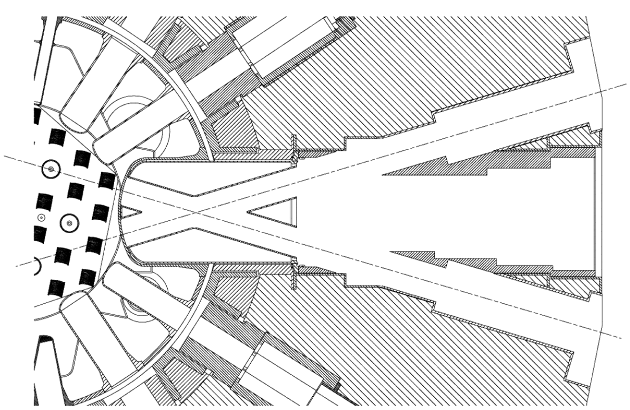

Fig. 1. Cold Source Cavity (54 cm D) and Beam Tubes in NIST Reactor

While both source and instrumentation are critical to this development, the present manuscript will focus on the development of the cold neutron sources with emphasis on the factors leading to particular design choices.

2. D_2O ICE SOURCE

From the very first days of the reactor design [1], a D_2O cold neutron source was planned, but neither funds nor staff effort were available to design, construct and install it. The only components provided were a large volume cavity in the reflector viewed by two 16 cm D beam ports, as shown in Figure 1, and a helium refrigerator. Beginning in the early 1980s(the reactor went into full operation in 1969 at 10 MW, and power was increased to 20 MW in 1985), design studies [2] of a CNS were begun at a low level. It was quickly apparent that it would be impossible to remove the nuclear heating from the ice if it were allowed to be irradiated with the full γ and fast neutron flux, primarily because of the low thermal conductivity of the ice and limited capacity of the refrigerator. Therefore, a lead-bismuth shield was designed and fabricated to be inserted into the cavity to reduce the γ heating to 0.025 W/g (see Fig. 2). This shield was water cooled, and generated several tens of kilowatts of heat.

At the same time, design efforts began on a cryostat to contain approximately 16 liters of D_2O ice and cool it by means of coils of tubing embedded in the ice through which cold helium flowed. By 1987, sufficient resources were available to fabricate and install this source [3], which was viewed by a small angle scattering (SANS) instrument and a crystal monochromator time-of-flight spectrometer

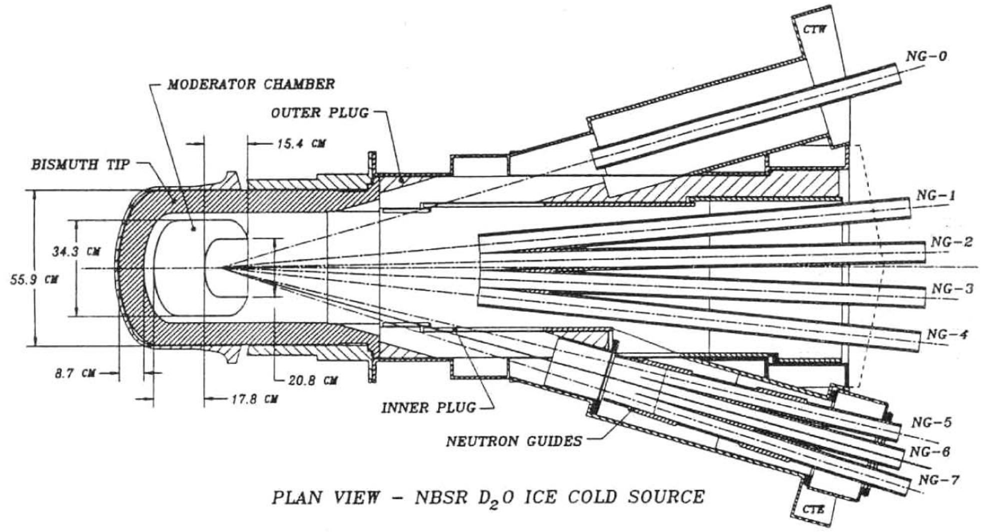

Fig. 2. Layout of Lead/bismuth Shield, D_2O Ice Source and Guides in NIST Reactor

inside the reactor hall. The overall gain was modest (approximately a factor of 3 above 5 Å), but it was sufficient to launch the cold neutron guide hall and instrumentation initiative in that same year. This cold source supplied neutrons to the first instruments in the neutron guide hall from 2001-2003, with the guides as shown in Fig. 2.

Why D_2O ice? Even when the source was being inserted, we knew that hydrogen would be better. We also knew that radiation damage would be a problem, although we believed that we could handle it. The one surprise was the so-called burping effect – under prolonged irradiation at low temperature (<30 K), the source temperature would suddenly and rapidly rise in temperature to well above 100 K, then recover. These excursions were not easily predictable as to frequency or initiation, but were disruptive. The essential problem is that the radiation causes dissociation, and the ions are immobilized in the lattice at low temperature preventing recombination. When the density of ion pairs reached a critical level, which depends on many parameters, the stored chemical energy would be released spontaneously, as the release warmed up the ice, increasing mobility, in a feedback mechanism that caused the instantaneous power to rise quickly. This problem was solved by warming the ice every two days, so that there was never a critical level of chemical energy stored. (Similar problems occur with methane, e.g. at IPNS, and are dealt with in the exact same way). However, we went with the original source design for three reasons. First, since it had been planned from the first, and the refrigerator was available, it was faster, easier and less expensive. Second, we had no experience with liquid hydrogen, and no funds to buy it. Third, because it was envisioned in the original Safety Analysis Report, we believed that it would be easier to obtain regulatory approval.

However, even as we were doing the first tests in 1987-88, and beginning the guide hall project, we also started to investigate liquid hydrogen sources. Our first efforts were MCNP simulation studies and conceptual engineering designs, until we were able to begin detailed design and construction in the early 1990s.

3. THE FIRST LIQUID HYDROGEN SOURCE

The first choice that we made during the design was to use a closed natural circulation loop to remove the radiation heating. In this system, the liquid hydrogen is allowed to boil under saturation conditions, and liquid is constantly replenished by gravity flow from a condenser located above the source. The evaporated gas is allowed to flow up to the liquefier, where it is re-condensed and allowed to flow back down to the source. This system is connected to a large room temperature tank that can contain the entire inventory (at 400 kPa) when the whole system is at room temperature. As the source is filled, the pressure drops to approximately 100 kPa under normal conditions at NIST. As the hydrogen system is a closed natural circulation loop, there is no need to connect to the venting system; that is, the moderator cell and liquefier are always connected to the storage tank during operation, with no valves of any kind. Since this system is designed to hold all hydrogen-inventory, there is never a need to vent it. In a deflagration or detonation, the venting process would be so slow with any vent line of conceivable diameter that it would be of no use in mitigating the pressure rise (which has a rise time measured in ms). When maintenance is needed, the gas can be removed by absorption in a metal hydride system. One other unique feature of the NIST system is that all connections are welded (all welds are radiographed and leak tested as performed), and there are no non-welded connections in the hydrogen system. When the source is removed the lines are cut, and then re-welded. With this arrangement, we have never had a leak in 10 years of operation.

The most difficult constraint in designing the liquid hydrogen cold source was imposed by the geometry shown in Fig. 1. The original design of the beam ports that view the reactor had them crossing each other approximately 36 cm from the end of the cavity. Since, for a hydrogen source the optimum thickness is of the order of 4 cm and the optimum position is in the highest flux region at the end of the cavity, the source would have to be very large in the horizontal and vertical dimensions for good neutron optics (full illumination of the planned 15 cm high guides). Working from the past experience of others, our first design choice was for an oblate ellipsoid with major axis 32 cm and minor axis 4 cm. However, this is a highly unsatisfactory solution, as the very large eccentricity leads to failure by buckling at quite modest pressures, and in order to avoid this, either a very heavy vessel is required (leading to unacceptable heat loads) or the eccentricity must be reduced (leading to bad cold neutron economy).

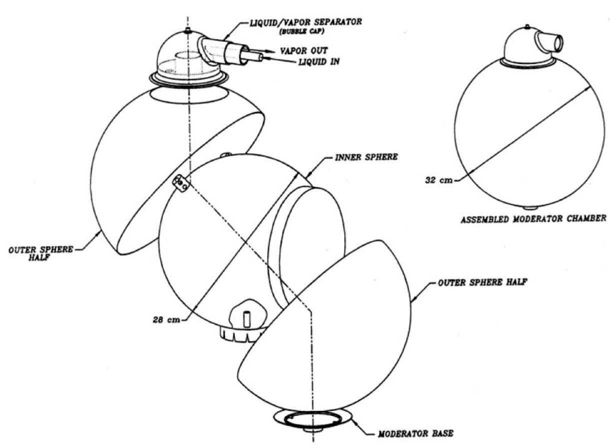

Fig. 3. The Arrangement of the First Liquid Hydrogen Source

After considering other modifications of this basic design (*e.g.* metal ties from one face to the other), and different slab geometries (*e.g.* an overlapping array of vertical tubes), we settled on the construction shown in Fig. 3 as the best combination of mechanical properties with high cold neutron flux. The liquid hydrogen is contained in a spherical shell formed by two concentric spheres of diameter 32 and 28 cm. In the region of the shell that is viewed by the neutron beam tubes and guides, there is a 20 cm D cylindrical viewing hole formed by a "bump" on the inner sphere, as shown in Fig. 3. Note that the inner sphere is connected to the shell only at the bottom. As a result, when the metal of the inner sphere is heated by radiation, any liquid in the inner sphere boils off, and this volume contains only hydrogen gas. To the best of our knowledge, the use of the cylindrical annulus moderator-cell in which the outer shell contains liquid and the inner shell only vapor was first proposed by Paul Ageron for a hydrogen CNS at Saclay [4]. One additional feature shown in the figure is a "bubble cap" at the top of the sphere, which serves as a phase separator.

Since the unusual geometry of the proposed source made computation of the thermal hydraulic performance very difficult, a full scale mockup was built at the Boulder site of NIST. Because no refrigerator of suitable characteristics was readily available, the test was done by adding liquid hydrogen from a storage dewar, and venting the vapor. The system was made of glass, to allow visualization of the flow characteristics, and incorporated a mass scale to allow direct measurement of the mass of hydrogen in the source (and hence the void fraction). As a result of these tests, several changes were made to the design, of which the most important was a decision to operate with two phase flow in the return line, as is done with the horizontal deuterium source at the ILL. In this mode, the arrangement was extraordinarily stable, responding quickly and predictably to rapidly changed conditions such as sudden loss, change or onset of heating.

It should be noted that this design process was not linear, nor as straightforward as stated here. It consisted of many iterations of neutron flux calculations (now using MCNP), mechanical design calculations including finite element analysis of stresses, and safety analyses to arrive at the final design installed. Many passive safety features were added to the design, including:

- Absence of non-welded joints
- Helium blanket surrounding *all* hydrogen containing components and volumes where hydrogen might leak to (*e.g.* not only vacuum spaces, but also vacuum pumps)
- All components protected from inadvertent damage by steel, concrete or other barriers
- Absolute minimum of gas handling

A pictorial view of this source, along with its successor, is shown in Fig. 4. This hydrogen source was installed [6] in 1994, and operated with no problems until it was replaced by the second hydrogen source in 2002. The gain in flux that this source gave over the D_2O ice source is shown in Figure 5.

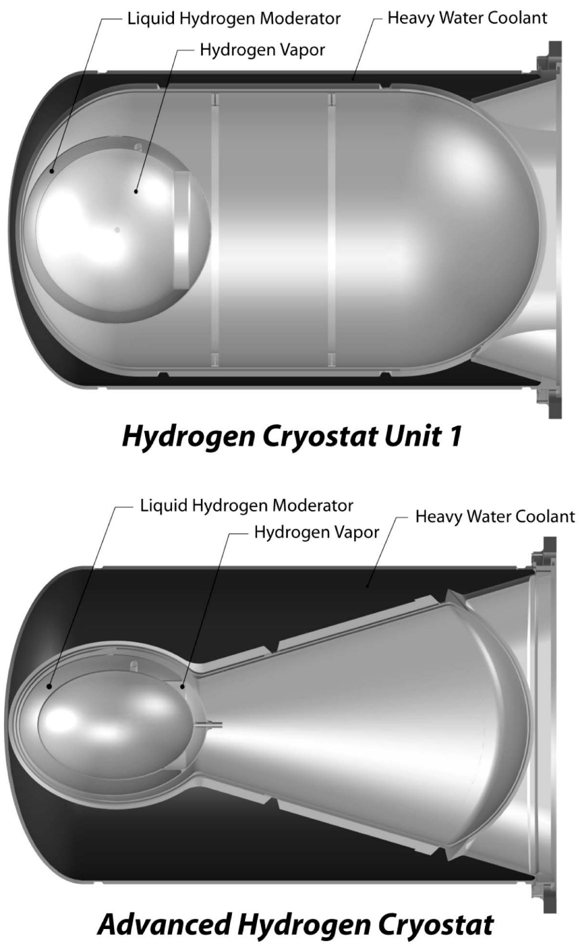

Fig. 4. Pictorial Views of the First and Second NIST Hydrogen Cold Sources. The Larger Amount of D_2O Surrounding the Source is Readily Observed

4. THE SECOND HYDROGEN SOURCE

Once again as we were installing the first source in 1994, we had begun design on an improved version, based on the extensive MCNP simulations that were continuing. The simulations showed that a major gain could be realized by increasing the amount of room temperature D_2O surrounding the hydrogen, and smaller gains were possible by, for example, putting more hydrogen near the reactor, and by evacuating the inner chamber. The result of these changes was the design shown in Figure 4, where a pictorial view is compared to the first source.

The components of this source were much harder to manufacture, and in most cases, the starting point was a solid block of aluminum, that was machined to final shape on a high speed 5-axis milling machine. Because of the less favorable shape, the second source has more aluminum, and hence a higher heat load, but the new measured heat load of 1100 watts is 20% below calculations, and well within the refrigerator capacity. This source was installed [7] in 2002, and has operated without problems since. In fact, there have been no shutdowns of the cold source when the reactor was ready to run over the entire period. This is our answer to the problem of impact on reactor availability – the design is robust, there are spare components for all parts of the system (including a spare compressor), and preventive maintenance is performed regularly. As a result, even during the initial commissioning of the first hydrogen source, the cold source reliability never fell below 95%, and this was adequate for our requirements. The gain in intensity from this source is shown in Fig. 6.

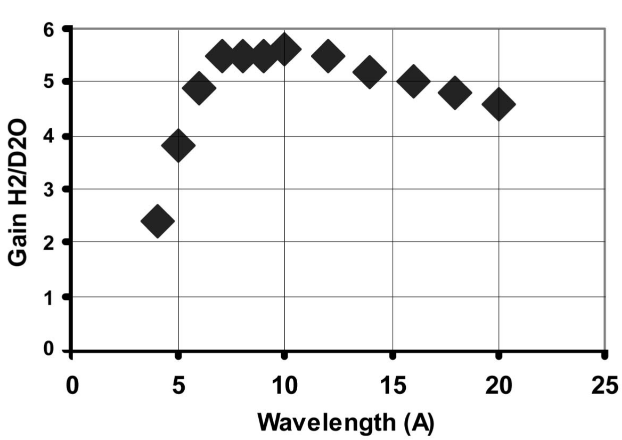

Fig. 5. Gain in Cold Neutron Flux, First Hydrogen Source Over D_2O Ice

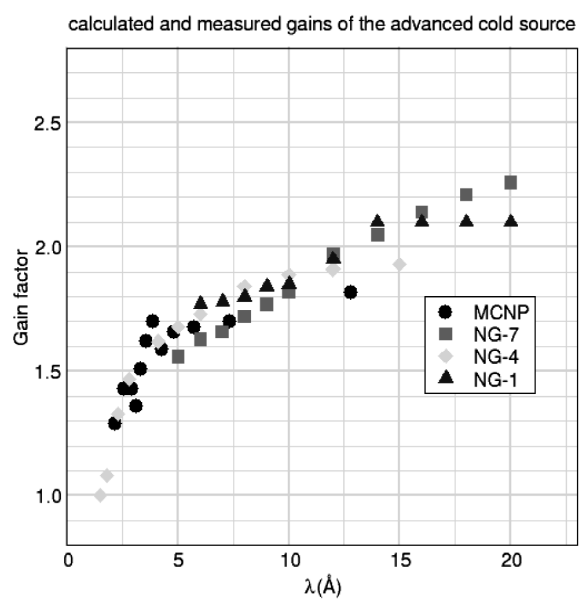

Fig. 6. Gain of Second Hydrogen Source to First, as Measured at Three Separate Instruments, and as Calculated by MCNP

5. IMPACT OF COLD NEUTRONS

The impact of these developments in cold neutron sources on NIST and on U.S. science has been dramatic. The development of the CNRF, which encompassed a cold neutron guide hall and 15 cold neutron instruments was funded in 1987, and the first neutrons were in the guides in 1990. The guide hall is now almost completely filled, as shown in Fig. 7, and the time is approaching to replace existing instruments, either with improved versions, or with entirely new types of instrument. For example, a new instrument funded by the National Institutes of Health was commissioned last year, taking the last available space. This year, a new cold neutron triple axis spectrometer is being installed at the port that had served Depth Profiling, while that facility is moving into the guide hall to share a beam with an improved version of Prompt Gamma Activation Analysis.

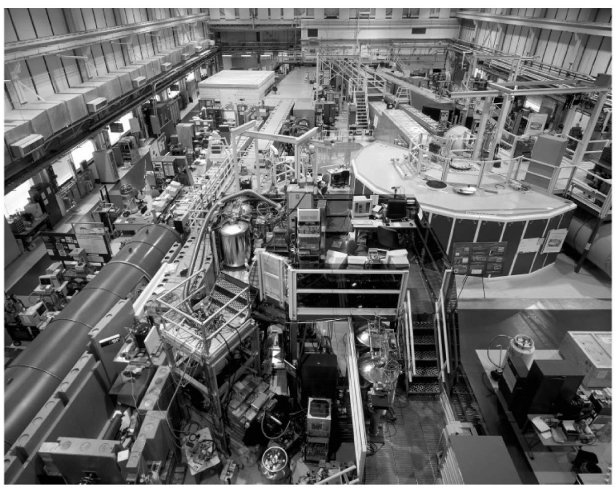

Fig. 7. NCNR Guide Hall in 2003, Prior to Installation of Membrane Diffraction Instrument

As these capabilities were developed, usage increased dramatically, as shown in Fig. 8. Even more important, the quality of the work being done improved, and new users, many of whom had never considered neutron techniques, were attracted to use the facilities. As a result, both the National Science Foundation and the National Institutes of Health have become major supporters of instrument development and operation. As a measure of scientific impact, it is notable that at least 5 major scientific prizes, not neutron prizes, were awarded at least in part based upon work done at the NCNR. These included the High Polymer Prize and the Young Investigator Prize of the American Physical Society. The benefits were not restricted to cold neutron research, as the success of the cold neutron part of the NCNR helped to bring benefits to the thermal neutron program, in which two new triple axis spectrometers are now being installed in the reactor room.

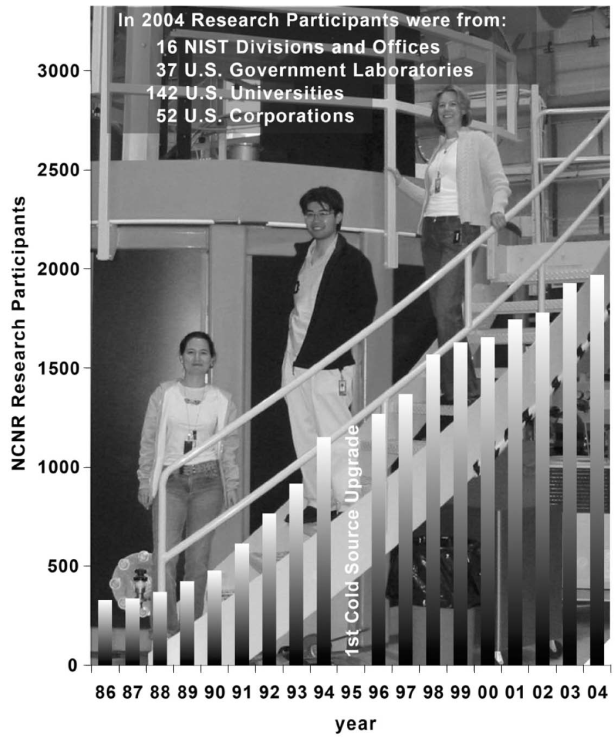

Fig. 8. Growth of NCNR Usage as Capabilities Improved

6. PROSPECTS AND CONCLUSIONS

At this point, it is hardly necessary to say that we have started design on yet another source – this one to be a large (approximately 30 l) volume of deuterium. We expect to be able to achieve further gains comparable to those obtained in going from the first to second hydrogen source [8]. If we are successful, we expect to see the result shown in Fig. 7 for the overall gain since the story of cold neutrons at NIST began. We expect to install the D_2 source in 4 years, at the next regularly scheduled long shutdown for reactor maintenance.

Once again, one can ask why we didn't do this the last time, to which the answer would be very similar to before – the second hydrogen source was straightforward, could be done quickly, and resources were limited. The main point is that one should never quit trying to improve a source, *but* get a source in, even if a better one can be envisioned. The other aspect of this problem is that the same must happen with instrumentation. In spite of the large gains envisioned in Fig. 7, equally large gains come from improvements in instrumentation, as for example at the High Flux Backscattering Spectrometer at NIST, in which every known improvement was made to existing designs to achieve an overall gain of at least 7 over existing instruments. Also, the NIST facility was one of the first to go beyond the usual guide construction, using ^{58}Ni coatings for a 30 % gain; now, new facilities are using supermirrors wherever possible, for further gains of 4-10. Thus, although we have concentrated on the CNS in this paper, an equally important paper could be written on what happens after the CNS – the guides and instruments.

The final statement is now as it was at my last visit here – the NIST Research Reactor and NCNR are well positioned for the next 20 years. In 2004, an application for a renewal of the reactor operating license was submitted to the Nuclear Regulatory Commission. Ongoing work on the reactor is aimed at maintaining it at the highest standards, including new control room instrumentation, improved electrical systems, and other projects. In 2005, the NCNR received a $6M increase in operating funds (in a very tight budget year) to improve service to users and continue developing new instrumentation. I expect that the next twenty years will look just as exciting as the past twenty have been.

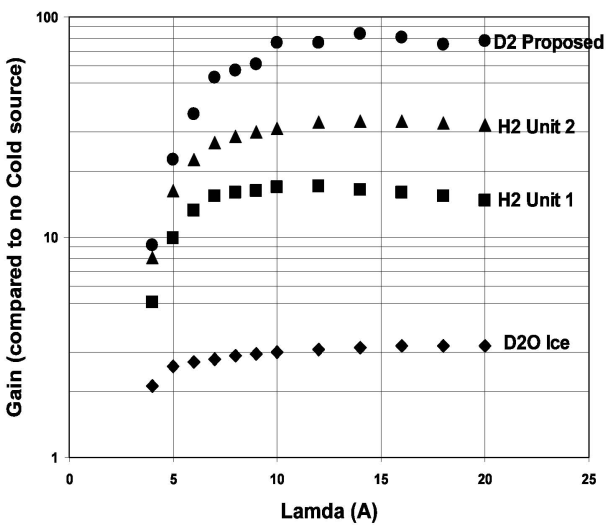

Fig. 9. Gains from NIST CNS – D2O Ice, H_2 Unit 1, H_2 Unit 2, D_2 (proposed, MCNP calculation)

ACKNOWLEDGEMENTS

The authors acknowledge the support and contributions of all of their colleagues at the NCNR who helped with the source construction and installation, and with the guides and instruments that perform the science and serve the users, which is the justification for the effort.

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