

Conceptual Study for the Moderator Selection of the Cold Neutron Source Facility for HANARO

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Abstract

Basic concept of a cold neutron source for a 30 MW heavy water moderated reactor (HANARO) is developed. The source is a cold bottle located in a vertical hole near the reactor core. Since the bottle does not have sufficient volume for cooling, the optimum liquid mixture ratio is studied between liquid hydrogen and liquid deuterium. We also studied the variation of the gain depending on the volume of the bottle. The calculation is performed by a coupled MCNP model and by a semi-analytic approach. For the current geometry, 80% liquid deuterium mixture with liquid hydrogen gives the highest gain at 10 Å neutron wave.

1. Introduction

The development of high intensity cold neutron sources is one of the important matter in condensed matter neutron research and in fundamental neutron physics[1]. The KAERI(Korea Atomic Energy Research Institute) has a plan to build a new cold neutron source for the new test reactor HANARO. HANARO is a 30 MWth reactor operating since February 1995. The reactor core is loaded with 28 cluster fuel assemblies and 4 booster fuel assembly positions. The fuel is 20 percent enriched uranium silicates. They are moderated by heavy water and cooled by light water. There are 7 horizontal beam tubes. One of the beam tubes is reserved for the cold neutron source(CN). The CN is coupled with vertical thimble(CNS) of diameter 16 cm. The CNS is filled with light water currently.

From the physics aspect, the most important

thing to decide is the moderator. The objective of moderator selection is to get maximum cold neutron flux and to have minimum heat generation. There are several candidate materials such as the liquid hydrogen, the liquid deuterium[2], and the solid methane[3]. We have considered mixture of liquid hydrogen and liquid deuterium.

2. Calculation Model

2.1. Property of Material

The liquid hydrogen has small mass number, large scattering cross-section, and low evaporation temperature. However it has relatively large absorption cross-section of 0.016 to the total cross-section. The liquid deuterium has twice the mass number of the hydrogen, and has 0.15 times of the hydrogen scattering cross-section and

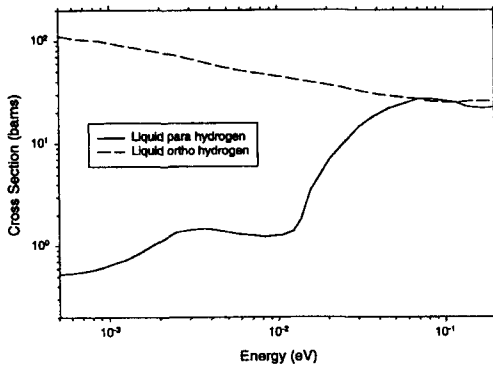


Fig. 1. Scattering Cross-section of Liquid Hydrogen

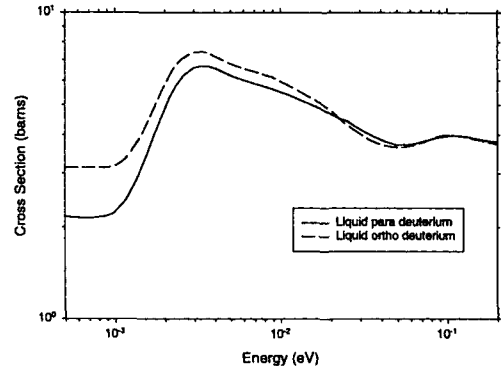


Fig. 2. Scattering Cross-section of Liquid Deuterium

has similar evaporation temperature comparing with hydrogen. However, the absorption cross-section is just 0.00016 to the total cross-section. When neutrons are sufficiently cooled down, the temperature of neutron will be reach that of medium. So, when the moderator volume is sufficiently large, we can expect the liquid deuterium, which has less absorption to scattering ratio, will result in more cold neutrons than liquid hydrogen. However, when the volume is limited, the neutron in the liquid hydrogen will reach near the medium temperature faster.

By the spin of proton, the hydrogen molecule has two species of para- and ortho-hydrogen. At room temperature, there are 1/4 of para-hydrogen and 3/4 of ortho-hydrogen. But at the normal boiling point of liquid hydrogen (20.4°K), equilibrium shifts to almost 100 % para-hydrogen during modern liquidification process[4]. For deuterium molecule, the room temperature equilibrium is 2/3 ortho-deuterium and 1/3 para-deuterium. Near the boiling point almost 98 % is ortho-deuterium.

The cross-section for liquid ortho-hydrogen is about 100 barns in low energy and that of para-hydrogen is less than 0.5 barns as shown in Fig. 1. The sharp drop below 50 meV is due to spin

coherence, and the second drop below 3 meV is due to intermolecular interference. For ortho-deuterium, the cross-section is about 3 barns and that of para-deuterium is about 2 barns as shown in Fig. 2. The drop in the cross-sections below 3 meV is due to intermolecular interference.

2.2. MCNP Model

The continuous energy Monte Carlo code MCNP-4A[5] is used as the calculation tool. Fig. 3 shows a cross-sectional view of HANARO core. It is a very time consuming calculation to model detailed geometry including core and fuel parts. We have employed a coupled MCNP model[6] by separating core geometry and source location geometry as displayed in Fig. 4. The core calculation is performed in absence of cold bottle. The location of cold bottle is filled with light water. From this calculation, a surface source for CNS is obtained. The cold neutron bottle is modelled as a cylinder of the radius 6.5 cm and of the height 17 cm. The radius of surface source is 15 cm and the separation of 8.5 cm between the surfaces of bottle and source is about twice the mean free path in heavy water. Since the Monte Carlo simulation of a problem is same as solving a

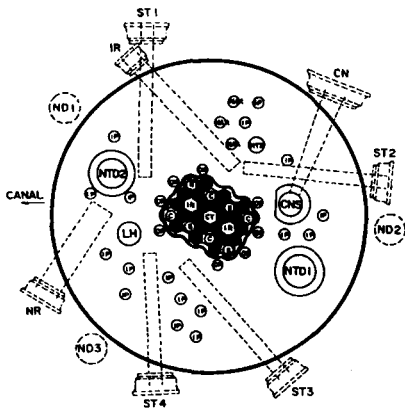
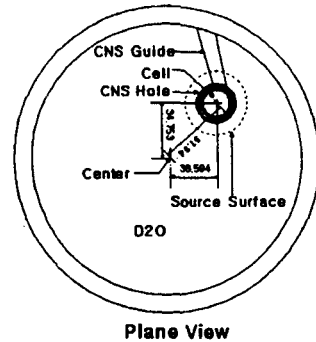


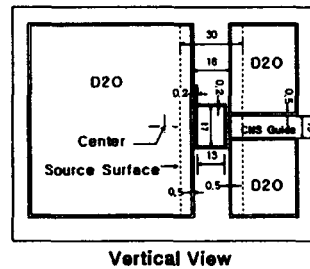
Fig. 3. HANARO Core

second kind Fredholm integral, the solution inside of a convex region with given boundary condition (or boundary source) is identical to the solution obtained by the original whole region solution (detailed core model). The source location geometry model employed in this study includes not only inside of boundary source, but also outside of boundary source. In this instance, the flux emerging from inside of boundary source will interact with neighbouring material. However as long as the distance from the source is far in terms of mean free path of neutron, the interference of fluxes from the inside and outside of boundary source will be small as verified in the study of Lee et al. [6]. During the calculation of source location geometry, various mixtures of hydrogen and deuterium are studied.

The double differential scattering cross-section for thermal neutrons is represented by the scattering law for the incident neutron energy less than 4 eV[7]. The hydrogen or deuterium molecule requires the quantum mechanical treatment to account for the spins of two atoms in the same molecule. This problem was considered by Young and Koppel[7]. The liquid hydrogen or deuterium is pictured as a cluster of about 20 molecules and undergoing vibrations similar to those in a solid. The actual scattering law is



Plane View



Vertical View

Fig. 4. MCNP Model of CNS

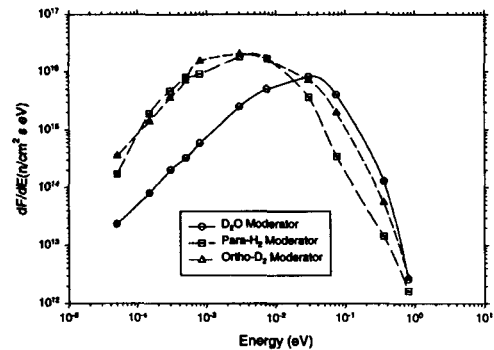


Fig. 5. Neutron Spectrum with Various Moderator

presented by LANL in numerical form[8].

To achieve less than 2 % statistics in the relative error of the energy dependent flux, we use 4,000,000 histories for MCNP run. The calculational result of neutron spectrum is displayed in Fig. 5. The spectrum for the normal heavy water is near to the Maxwell-Boltzmann distribution. However, the spectrum for cold source is apart from the Maxwell-Boltzmann

distribution for the transition region of the energy dependent cross section.

2.3. Semi-analytic Method

The neutron in equilibrium under infinite medium without absorption will have the Maxwell-Boltzmann distribution[9].

$$\phi(E) = \frac{2\pi N}{(\pi kT)^{3/2}} \sqrt{\frac{2}{M}} E e^{-E/kT} \quad (1)$$

where $\phi(E)$ is the energy dependent neutron flux, M is the mass of neutron, k is the Maxwell-Boltzmann constant, T is the neutron temperature, and N is the neutron density. From above equation, we know that the peak of neutron flux occurs at the temperature for which $E = kT$. From the calculated neutron spectrum, we can find the neutron temperature. However the flatness of the peak hampers the decision of the peak energy (or the temperature). So, we can define a suitable region near peak of the spectrum, and derive following formula to determine the peak energy.

$$kT = \frac{\int_{E_1}^{E_2} \phi(E) dE / \phi_{th}}{e [e^{-E_1/kT} (1 + \frac{E_1}{kT}) - e^{-E_2/kT} (1 + \frac{E_2}{kT})]} \quad (2)$$

where ϕ_{th} is the peak value of flux. One can find the neutron temperature after several iterations with Eqn. (2).

For low energy neutron, it is usual to express the energy in the wavelength unit. The relation between De Broglie wave length of neutron with the energy is as following formula.

$$\lambda \text{ (in \AA)} = \frac{9.04}{\sqrt{E \text{ (in meV)}}} \quad (3)$$

One defines the cold neutron gain as the ratio of neutron flux of the case of a cold source is

installed to that of the case when there is only heavy water. From Eqn.(1), one can derive the gain factor as follows:

$$G(E) = \frac{N}{N^0} \left(\frac{T^0}{T}\right)^{3/2} e^{-E(\frac{1}{kT} - \frac{1}{kT^0})} \quad (4)$$

where superscript 0 denotes the case of heavy water moderator. The trend of the gain factor is displayed in Fig. 6. In the zero energy limit, one gets

$$G(\lambda = \infty) = \frac{N}{N^0} \left(\frac{T^0}{T}\right)^{3/2} \quad (5)$$

For the Maxwellian limit, the theoretical gain for a 20°K cold moderator against a 312°K heavy water is 62, if there is no drop of scattering cross section around 2 meV.

3. Results

3.1. Mixture

With the coupled MCNP calculation model, the neutron temperature and number density are calculated using the semi-analytic approach as displayed in Table 1. For the heavy water moderator, it is nearly in equilibrium. And for the cryogenic moderators, the neutron temperature is still high above the liquid moderator temperature, in other words, it is not sufficiently cooled.

As a figure of merit for comparative study of cold bottle, we have used the neutron gain. The result for mixture of para-hydrogen and ortho-deuterium is displayed in Fig. 7. It is noticeable that the ortho

Table 1. Neutron Temperature and Number Density

Case	Neutron Temperature	Number Density
D ₂ O	306°K	2.5 × 10 ⁹ #/cc
para-H ₂	62°K	2.9 × 10 ⁹ #/cc
ortho-D ₂	87°K	4.2 × 10 ⁹ #/cc

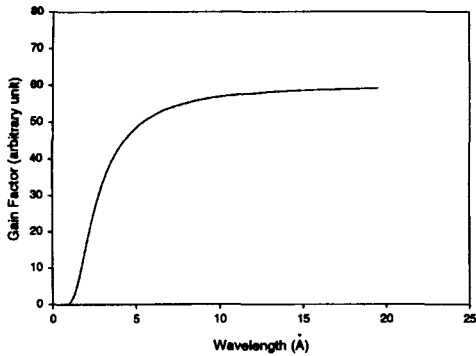


Fig. 6. Gain Factor - Semianalytic Method

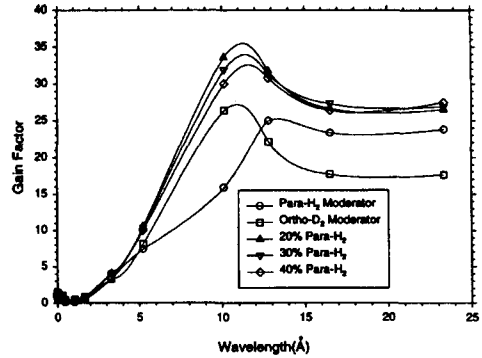


Fig. 7. Gain Factor with Various Mixture

Table 2. Flux and Heat Generation in the CNS Liquid

Radiation	Energy	para-H ₂		ortho-H ₂		ortho-D ₂	
		flux	heat	flux	heat	flux	heat
neutron	upper limit						
	0.625 eV	33.64	0.003	39.91	0.03	58.94	0.0002
	0.18 MeV	0.89	0.04	0.88	0.04	1.72	0.01
	0.82 MeV	0.05	0.06	0.05	0.06	0.07	0.02
	20 MeV	0.10	0.32	0.10	0.30	0.10	0.13
	Sum	34.67	0.42	40.93	0.44	60.83	0.16
photon	0.1 MeV	0.14	0.0004	0.13	0.0004	0.14	0.002
	0.5 MeV	2.71	0.06	2.79	0.06	2.31	0.03
	0.6 MeV	0.39	0.02	0.40	0.02	0.36	0.01
	1.3 MeV	1.03	0.08	1.03	0.08	1.00	0.04
	3.0 MeV	5.95	0.93	7.38	1.16	0.98	0.07
	7.5 MeV	0.55	0.13	0.52	0.12	0.57	0.07
	14 MeV	0.15	0.05	0.12	0.04	0.15	0.02
	Sum	10.92	1.27	12.38	1.49	5.51	0.24
Total heat			1.69		1.93		0.40

* unit: flux = 10¹³/cm²-sec, heat = W/gr

deuterium moderator case gives very high gain around 10 Å, which confirms the molecular binding effect of liquid deuterium as the measurement of Saclay[2] suspected. Due to the low cross-section of liquid deuterium in comparison with liquid hydrogen, the gain behaves in smoother pattern for increasing liquid hydrogen fraction. Fig. 8 shows variation of gain with deuterium mixture near 10 Å. The gain is

maximized when 80 %(volume) of deuterium is mixed with para-hydrogen.

3.2. Source Volume

For liquid hydrogen, it is well known that there is an optimum volume of cold bottle because of the competition between cooling and absorption. In this study, the radius of bottle is fixed as 6.5 cm

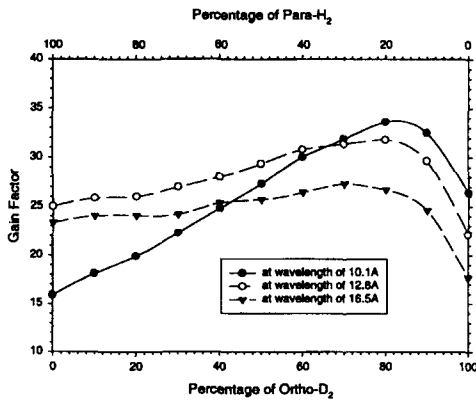


Fig. 8. Gain Factor at Certain Wave Length

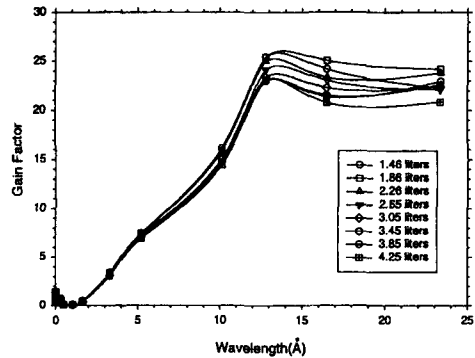


Fig. 9. Gain Factor for Para-hydrogen

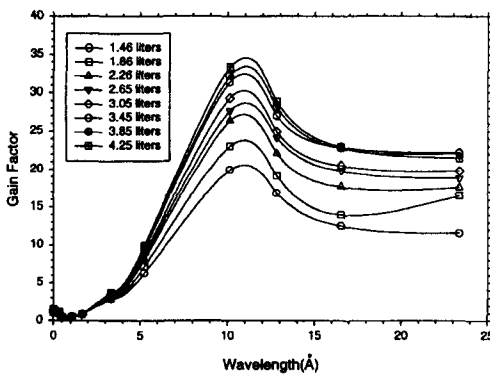


Fig. 10. Gain Factor for Ortho-deuterium

and the height is varied from 11 cm to 32 cm. The result for liquid para-hydrogen is displayed in Fig. 9 and that for liquid deuterium is in Fig. 10.

For liquid para-hydrogen, it is evident that the optimum height is 11 cm. There is decrease in the gain slightly for the large or small volume. However for the liquid deuterium, the gain increases as the volume increases since the absorption is ignorable.

3.3. Heat Generation

The heat generation estimation for the cold neutron source is very important factor to design a

Table 3. Heat Generation in CNS

Material	Radiation	para-H ₂	ortho-H ₂	ortho-D ₂
liquid hydrogen	neutron	0.42	0.44	0.16
	photon	1.27	1.49	0.24
	sum	1.69	1.93	0.40
aluminum	neutron	0.0005	0.0004	0.0005
	photon	0.56	0.60	0.31
	beta	0.53	0.53	0.53
sum		0.99	1.13	0.84

* unit: heat = W/gr

cryogenic system. There are three kinds of heat sources for the CNS cell. Those are:

- neutron interaction,
- photon interaction,
- beta decay of Al-28 after capture from Al-27.

Most heating is from the fast neutrons with energy higher than 1 MeV. So, we have not considered the variation of the mixture, and considered only the pure liquids. The results are summarized at Table 2.

One may find that the 2.225 MeV photon generated by the capture of hydrogen nuclide, contributes mainly for a hydrogen source. For ortho-hydrogen, this value is larger than that of para-hydrogen. The total heat generation by a hydrogen source for same mass is about 4 times

that of a deuterium source. However, this fact means that it is about 2 times for same volume.

For aluminum tube surrounding the bottle, the heat generation by neutron is very small. However, the main source of heat for the aluminum tube is due to the beta contribution. It is not possible to calculate directly the beta contribution by MCNP code. We have employed semi-analytic method to estimate the beta contribution. The neutron flux distribution inside of the aluminum tube is assumed to be the Maxwellian distribution again. And the absorption cross-section of aluminum for low energy neutron (less than 100 eV) follows $1/v$ law:

$$\sigma(E) = \frac{\sigma_0 (= 0.03695)}{\sqrt{E \text{ (in eV)}}} \text{ (barn)} . \quad (6)$$

We can compute the total absorption rate with assumption of the Maxwell-Boltzmann neutron spectrum as follows:

$$\begin{aligned} I &= \int dE \phi(E) \Sigma(E) \\ &= \int dE \frac{2\pi N}{(\pi kT)^{3/2}} \sqrt{\frac{2}{M}} N_{Al} \sigma_0 \sqrt{E} e^{-E/kT} , \quad (7) \\ &= n N_{Al} \sqrt{\frac{2}{M}} \sigma_0 \end{aligned}$$

where N_{Al} is the number density of aluminum. Eqn. (7) says that the absorption rate is independent of the neutron temperature for the Maxwell-Boltzmann distribution. The Al-28 at 4.642 MeV excitation level by capture of neutron, will make beta decay to Si-28 at the 1.779 MeV level. Then it decays with photon emission. The effective beta decay energy for this process is 1.247 MeV[10]. The total heat generation by beta decay is then:

$$\text{Heat (W/gr)} = 1.247 \times 1.602 \times 10^{-13} \times I / \rho,$$

where ρ is the density of aluminum. The energy carried by the neutrino is ignored since it is

distributed far away.

The total heat generation including moderator and aluminum tube is summarized at Table 3. The total volume of the bottle is 2.26 liters and the mass of tube is 531 grams. So total heat generation for para-hydrogen is 796 watts, where 270 watts are from the liquid and 526 watts from the tube. For the case of ortho deuterium, the total heat generation is 599 watts where that of the moderator is 153 watts and that of the tube is 446 watts.

4. Conclusions

We have studied basic properties of cold neutron source. The gain is about 25 for long wave range. This number is limited by the size of the bottle in comparison with other sources. At 1 meV (9 Å), we have 10^{16} n/(cm² s eV) neutron flux. The gain is maximized when 80 %(volume) of liquid deuterium is mixed with liquid para-hydrogen.

The heat generation of the system has been determined by the neutron and gamma spectrum above 1 MeV range. And the effect of moderator is rather small except the number densities of liquid. The total heat generation ranges from 599 watts to 796 watts depending on the moderator selection.

It needs further studies for the case of using the "enriched" ortho-hydrogen and extending the response to the cold neutron guide tube. And it is also needed to study the influence of core fuel and control rod movements.

Acknowledgement

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