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Measurements of the change of neutronic performance of a hydrogen moderator at Manuel Lujan Neutron Scattering Center due to conversion from ortho to para-hydrogen state

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Abstract
The ortho/para hydrogen ratio is a key parameter for the neutronic performance of a liquid hydrogen moderator. In order to get a better understanding of the influence that the ortho/para-hydrogen ratio has on the performance of such a moderator, we measured the neutronic performance of the partially coupled liquid-hydrogen moderator at the LANSCE Manuel Lujan Jr. Neutron Scattering Center, Los Alamos, USA, as a function of time after condensation. This was done by measuring the energy spectra and pulse shapes (neutron emission time distributions) of this moderator. It was found that the neutronic characteristic of the moderator changes with the elapsed time after condensation, and that the changes were so significant that for any scattering experiment made on such a moderator, the effect should be carefully evaluated.

Keyword: Pulsed neutron source, Hydrogen moderator, ortho-para conversion, energy spectrum, emission time distribution

1. Introduction

At 1 MW class high power spallation neutron sources, liquid hydrogen is at present a unique realistic candidate for a cold moderator material. Therefore, liquid hydrogen moderators have been adopted in the new high-power spallation neutron sources, JSNS (Japan Spallation Neutron Source) in Japan and SNS (Spallation Neutron Source) in the USA.

Simulation calculations indicate that the neutronic performance of a liquid hydrogen moderator depends on the para-hydrogen concentration.[1-3] Also, it is shown in the experimental result that the neutron pulse peak intensities increase with the higher para hydrogen concentrations.[4] However, there is no experimental data of time dependent neutronic performance from a hydrogen moderator in the high radiation field. Such data would be of great benefit not only to new spallation neutron sources but also to existing neutron facilities.

The Manuel Lujan Jr. Neutron Scattering Center has four water moderators and two liquid hydrogen moderators but there is no ortho-para conversion catalyst in the hydrogen loop. Therefore, it is expected that the para-hydrogen concentration in the loop will increase over time after liquefaction. The para hydrogen concentration in the state of equilibrium for hydrogen at T=20 K is 99.8%, whereas at T=300 K the equilibrium is 25% para hydrogen. Since the liquefaction process is significantly faster than the ortho-to-para conversion time without a catalyst at T=20K, the hydrogen loop at the Lujan Center always contains 25% para and 75% ortho-hydrogen right after a fresh filling. However, due to the different points of equilibrium for hydrogen at T=20K and at room temperature, a conversion from ortho to para hydrogen will start immediately. Even though the time constant for this process is fairly long, the ortho/para concentration and with it the neutronic performance will change during a typical period of operation (~10 days) at the Lujan Center. We have therefore measured the neutronic performance of the lower tier partially coupled liquid hydrogen moderator at the Lujan Center continuously for 132 hours to get a better understanding of how the neutronic performance changes over time as a function of the ortho/para ratio.

2. Experimental Setup

The measurements were performed at the LANSCE Manuel Lujan Jr. Neutron Scattering Center. The proton energy was 800MeV and the current was about 100μA, the power was about 80kW. In particular, this experiment was conducted on the SPEAR instrument (flight path 9). This instrument is viewing the lower tier partially coupled hydrogen moderator. Moderator dimensions are 13 x 13 x 5cm^3. A layout of the Lujan Mark-II target systems including its moderators is shown in figure 1. [5, 6]. During the period of operation, the temperature of hydrogen in the moderator was estimated to be at 21.6 K [7]. In the experiment, the regular SPEAR neutron beam was collimated down to 5 x 10 mm^2 using a B$_4$C collimator. For the energy spectrum measurement, a low efficiency He-3 detector was used. This detector has an efficiency of 0.01% at 25meV and the 1/v law of the absorption cross section was used to evaluate the energy dependence of the detector efficiency.

For the time distribution measurement, the neutron beam was monochromated by Bragg scattering with a mica crystal (d=9.96Å) with a Bragg angle of 85 degree, and detected by three He-3 detectors. These measurements were performed continuously for 132 hours starting right after the liquefaction/filling process was completed.

3. Observed change in energy spectrum

As mentioned above, the measurements were conducted over a period of 132 hours. In order to be able to compare the intensities of the various measurements with each other, it was essential to normalize the measured count-rates by the proton current on the target during the time each measurement. We therefore requested the proton current records for the 1L target from the central control room. We normalized our counts rates with the proton current data given. Unfortunately, this had only limited success. As can be seen in figure 2, the neutron intensity, which integrated below 25meV, normalized with proton current changes twice significantly over the duration of the experiment. We found out the following fact after the experiment. The final part of the SPEAR collimation consisted of two B_4C slits, and these slits could be adjusted depending on the requirements of the experiment. Occasionally the controllers for these slits did not function correctly, which could cause fluctuation of the slit width, namely the neutron intensity. While this affected the absolute measured neutron intensity, it had no influence on the spectrum shapes or on the time distributions, since the viewed area of the moderator was almost fixed and the area of the detector was only changed. However, in order to normalize our data we had to look for an alternative method. An experiment conducted by Ooi et al [4] at Hokkaido University showed that for a range of para-hydrogen concentration from 35 to 99% , the absolute neutron intensity at E=15 meV did not change within the uncertainty of the measurement. (This is also true up to about 70% in the calculated results shown in figure 4 indicating the intensity ratios which were normalized at para 25% for LANSCE TMRA.) We therefore normalized all our data assuming that the flux intensities at E=15 meV had the same value.

Figure 3 shows the spectral intensities and intensity ratios normalized at 15meV. It can be seen that in the energy range below 15 meV, the neutron intensity increases. In order to get information on the ortho/para ratios of our measurements, we calculated the change of the neutron intensity of this moderator as a function of the ortho/para ratio by using the most commonly used Monte Carlo transport code MCNPX [8]. Hydrogen kernel was evaluated by R. E. MacFarlane[9][10]. The results of the calculations are shown in Figure 4. There are some significant differences between the experimental and the calculated results. For example, the calculated results show a peak around 10 meV whereas the experimental results do not. This observation coincided with the findings of Muhrer et al. [11], who showed that the scattering kernel presently used in MCNPX does not always reproduce the measured data within the required accuracy. However, the calculations reproduce the overall trend of experimental results fairly well. The calculations indicate that the intensity from 15 to 100 meV decreases monotonically with the increase of the para-hydrogen concentration. Below

10 meV, the calculations indicate that the intensity increases monotonically until the para-hydrogen concentration reaches 60%, and decreases with over 70% of the para-hydrogen concentration. On the other hands, the experimental data shows that the intensity increases monotonically over time in this energy range although the intensity at 132hours seems to be approaching maximum since the difference between the intensities at 88hours and 132hours is small, which is very similar to the trend of the calculations at the para-hydrogen concentration up to 60%. The ortho/para ratio expected by natural conversion [12] appears in Figure 5, which indicates about 65% para-hydrogen after 132 hours, so we concluded that the effect of the radiation on the ortho-to-para conversion is not very large at the Lujan center. If we assume the natural conversion, the para ratios at each time are about 26% at 2.5h, 47% at 48h, 57% at 88h and 65% at 132h. To confirm the ortho/para ratio more precisely, it would be very useful to perform the experiment beyond 132 hours, so that we might observe the intensity decrease below 15 meV. However, we were not able to do this due to operational constrains.

4. Observed changes in pulse shapes
In addition to the neutron spectrum, we also simultaneously measured the neutron pulse shapes (emission time distributions) for various energies as described in chapter 2. During the analyzing process we made the following three corrections to the experimental data:
1) background subtraction,
2) broadening of the measured data due to the mosaic spread and geometry of the collimation system
3) absolute intensity normalization.
 The background was evaluated from the front part and the end part of the individual pulse peak by fitting analytical functions, in which the polynomial, the exponential or the logarithmic function were used. Figure 6 shows an example of background evaluation.

If the neutron scattering angle with analyzer crystal is small, the setup configuration without time-focusing can cause the significant broadening of the measured pulse shape. [13] However, even though our setup was not in time focusing geometry, the correction due to this effect was small enough to neglect even at the longest wavelength, since our geometry was almost backscattering, Bragg angle of 85 degrees, to obtain good angular resolution, and the solid angles of the moderator to the analyzer and the analyzer to the detector are small enough (under 1 degree). Therefore, we were only left with the task to correct for the so-called mosaic spread, which is due to the imperfection of the crystal. In our case, we assumed that the mosaicness of the mica crystal followed a Gaussian distribution with a FWHM of 0.8 degrees and corrected the experimental data for it. For the 5.19 meV pulse shape, for example, this mosaicness corresponds to an instrument resolution function with FWHM of 2 microseconds.

The absolute neutron intensities were obtained using the intensities of the energy spectra normalized at 15 meV as presented in the previous chapter. The corrected pulse shapes at the

energies of 1.87 meV, 5.19 meV, 20.8 meV and 40.7 meV are shown in Fig 7. And also calculated pulse shapes are shown in Fig 8. It can be seen in this figure that in the cold energy region (E < 5 meV), the pulse peak intensity increased over time (see left figures of Fig 7), and there is no noticeable change in the tail of the pulse (see right figures of Fig 7). In the thermal energy region (around 20 meV), the peak intensity also increased over the duration of the experiment; however, unlike in the cold energy region, the tail of the pulse does decrease over time. Therefore, the decrease of the spectral intensity above 15 meV is due to the decrease of the pulse tail. In the energy region above 20 meV there are no changes noticeable either in the peak flux or in the pulse tail, which is due to the fact that in this energy region there is almost no difference in the neutron total cross section between ortho and para hydrogen. In the comparison with experimental pulse shape and calculated one, trend with para concentration (time) about pulse peak intensity and the change of the tail part (see 20meV) are almost same. But pulse decay in calculated results is slow (compare the intensities at 800 microseconds). It means calculation simulate the effect of para hydrogen concentration well in these study and para concentration after 132 hours is estimated to be around 60%.

Figure 9 shows pulse peak intensities as a function of the neutron energy. As shown in this figure, the pulse peak intensity increases in the energy region less than 30 meV. In the experimental results (left figure), the biggest gain can be seen at about 7 meV, with approximately 30% after 130 hours. In the calculated results (right figure), the biggest gain appear around 10meV and pulse peak gain around 1meV is smaller than that of experimental result.

Figure 10 shows the FWHMs of these pulses. It can be observed from this plot that the FWHM decreases over time in the energy region less than 20 meV by up to 20%, whereas there are no changes being observed above 25 meV as one would expect. Calculated pulse width ratio is also shown in Fig 10. We expect 130 hour as a para 65%. So, relative para hydrogen effect on the pulse width is over estimated in the calculation.

5. Conclusion

The neutron energy spectra and pulse shapes at various energies of the partially coupled lower tier hydrogen moderator at the Lujan center were measured continuously for 132 hours. Since there was no ortho-to-para-hydrogen conversion catalyst in the hydrogen loop, the ortho/para-hydrogen ratio changed over time. If one assumes a natural conversion process, the para-hydrogen concentration would be about 60% at the end of the experiment after 132 hours, while the concentration was 25% at the beginning of the experiment. Corresponding to the change of the ortho/para-hydrogen ratio, we observed a change of the neutronic performance of the moderator. The spectral intensity increased below about 15meV and decreased above this energy. The pulse peak intensity increased and the FWHM decreased for neutrons with energies below 20meV. While an increase in flux with a decrease of the

FWHM is a preferable effect for neutron scattering experiments, an ever changing spectrum is not. From the comparison of the change of spectral intensity and the pulse shape with the calculated ones, we estimated the para concentration is around 60% after 132 hours operation at Lujan center. This value is almost the same as that of natural conversion, but the value is not so correct due to the uncertainty in the calculation results. We, however, would say that the conversion occurred almost naturally. To determine the coefficient of radiation effect, it would be required to measure the para concentration by more direct methods.

Anyway, we have concluded that for future moderator designs, in order to optimize the performance of a liquid hydrogen moderator and keep its performance stable, an ortho-to-para converter or an ortho/para ratio stabilizer is indispensable.

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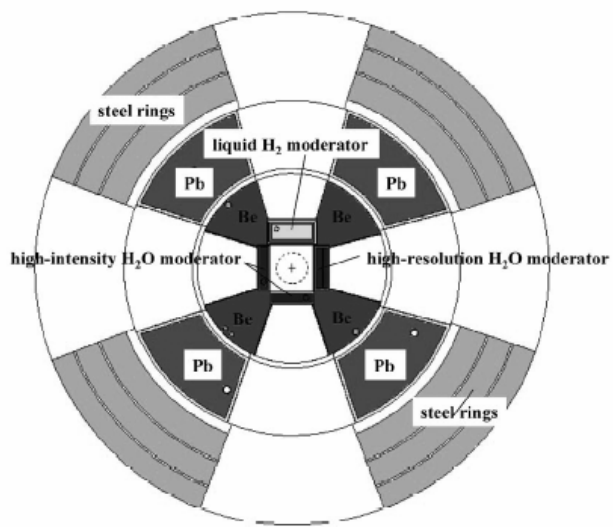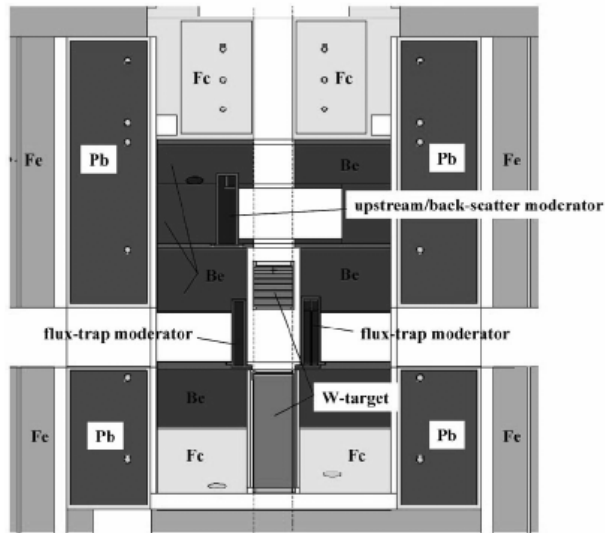
Figure 1. Target moderator reflector system of Lujan center.
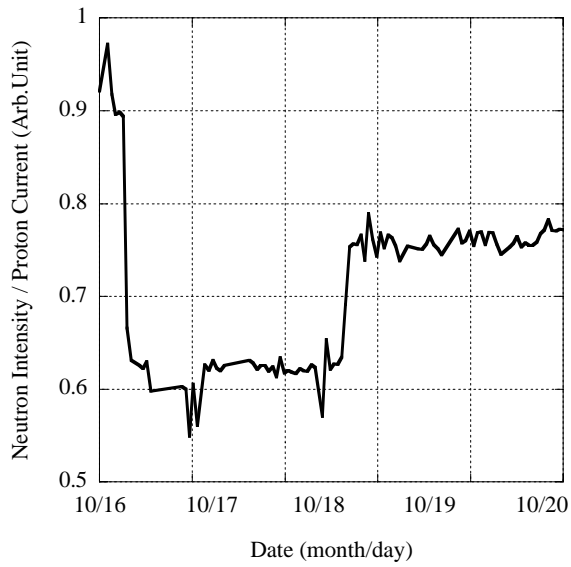
Figure 2. Log of proton current and neutron intensity from a hydrogen moderator normalized by proton beam current. Horizontal axis indicates the date of the experiment. The neutron intensity was in arbitrary unit only to indicate the trend of the intensity change.
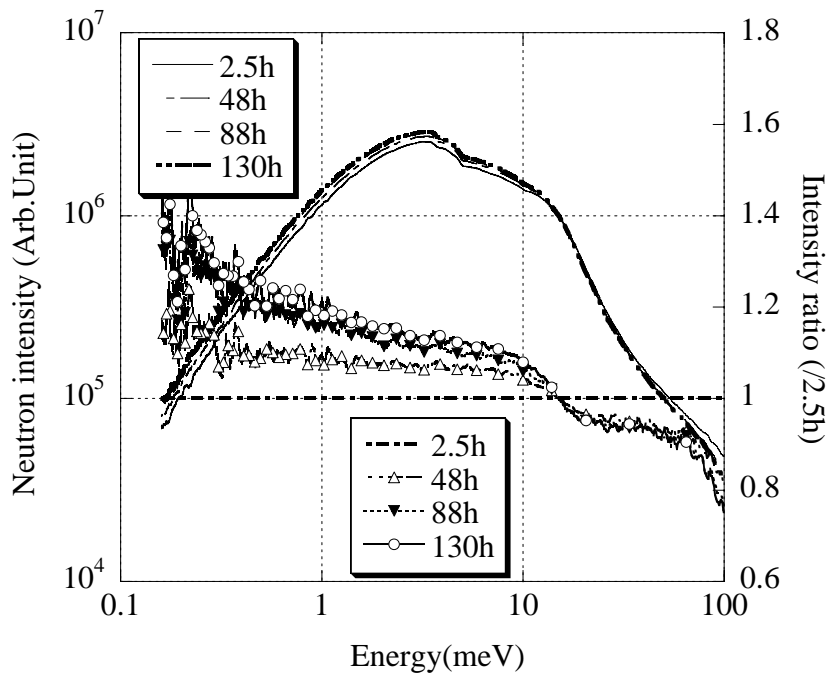
Figure 3. Neutron energy spectra and intensity ratios at various hours after condensation.
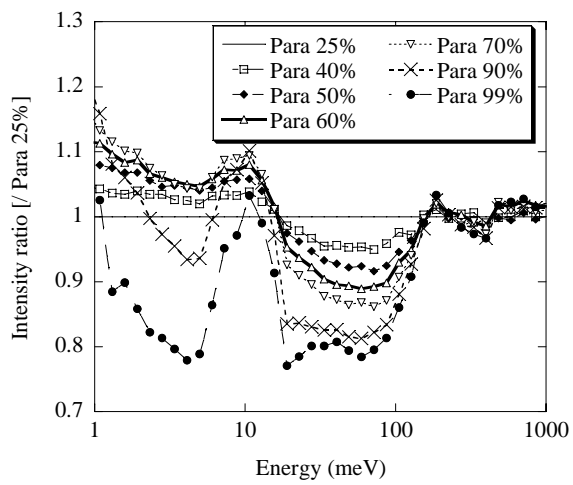
Fig. 4 Simulation calculation results of intensity ratios at various ortho/para ratios.

Natural Hydrogen Relaxation

132 hours, 65% para-hydrogen
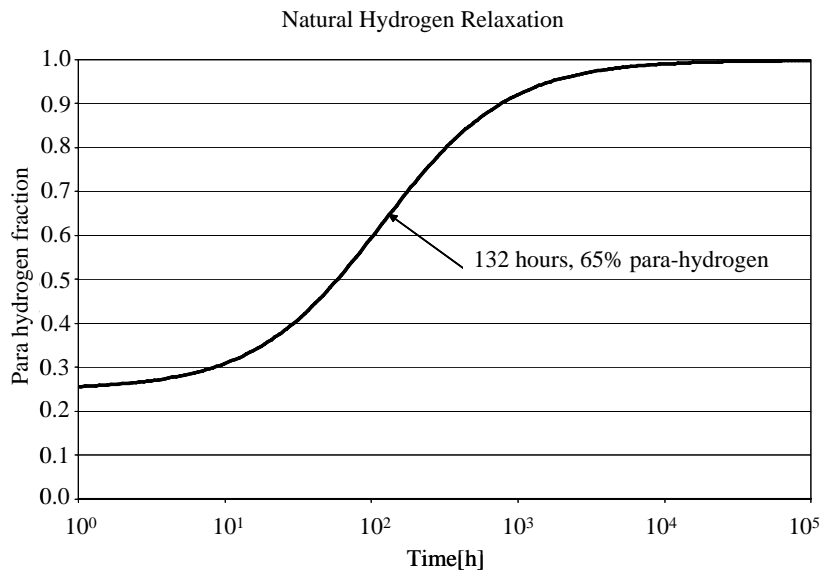
Figure 5. Natural hydrogen conversion from 25% to 99.8% para-hydrogen.
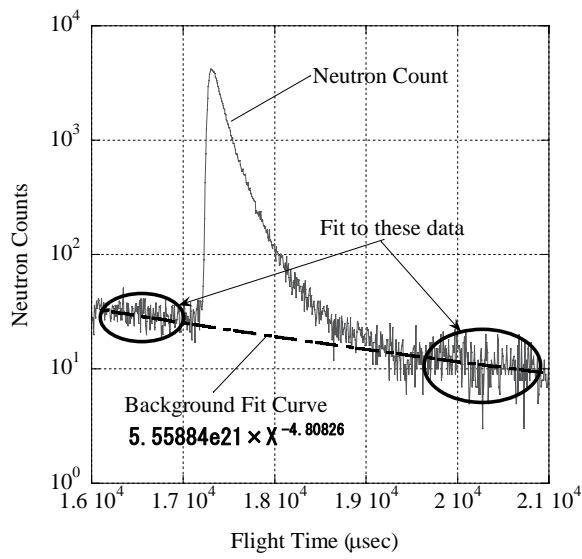
Figure 6. Example of background curve fit for pulse measurement.
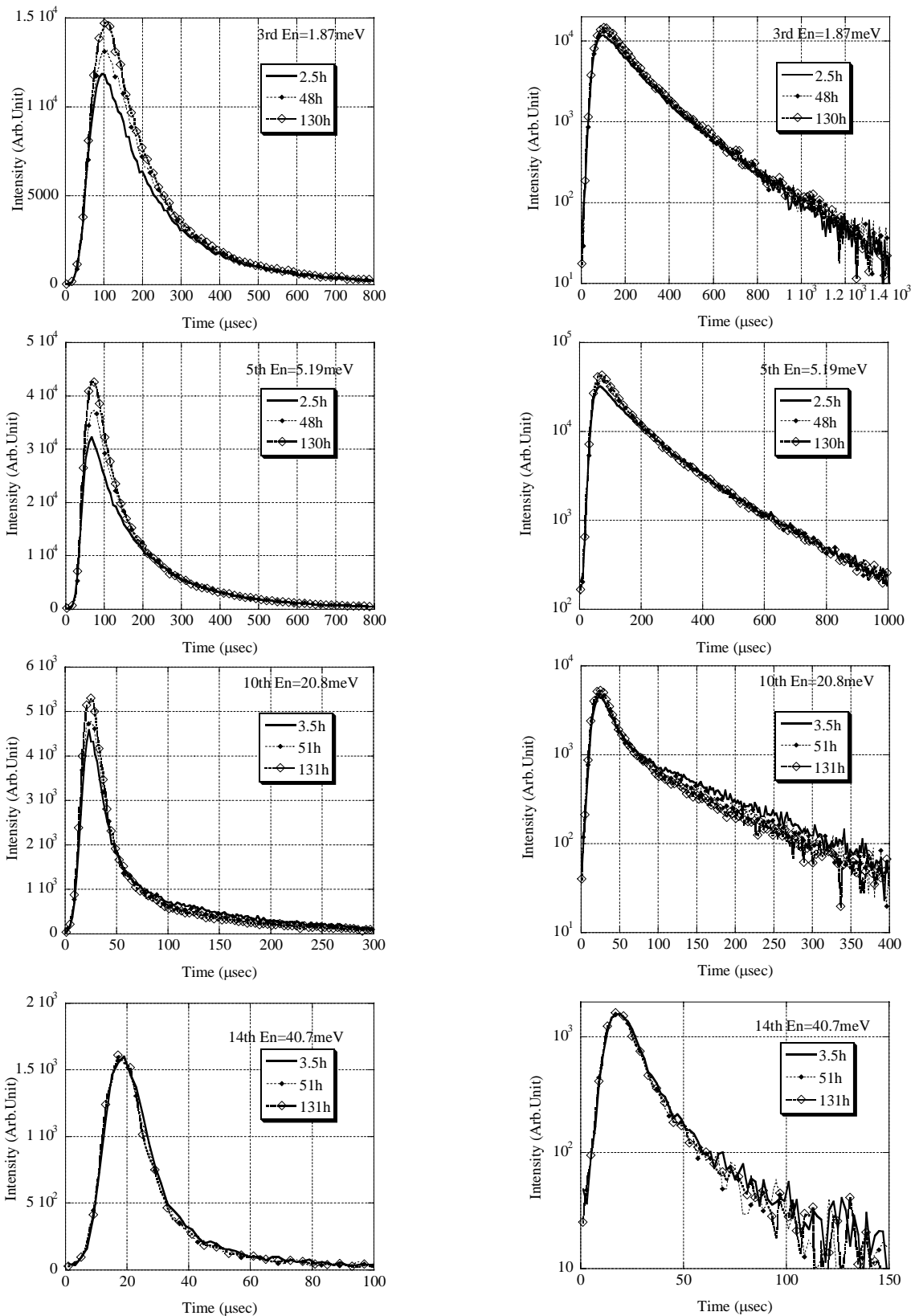
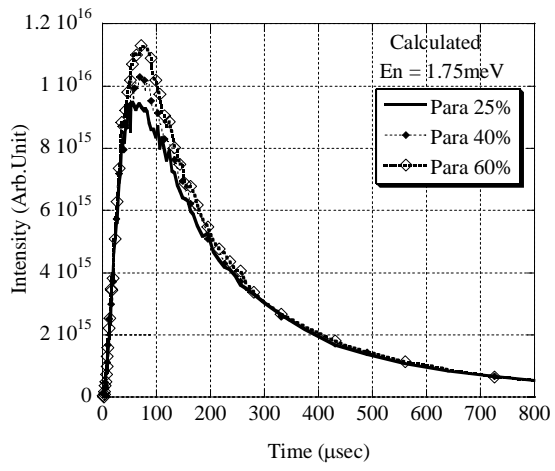Figure 7. Neutron pulse shapes. Left figures are linear plot and right semi-logarithmic plot.
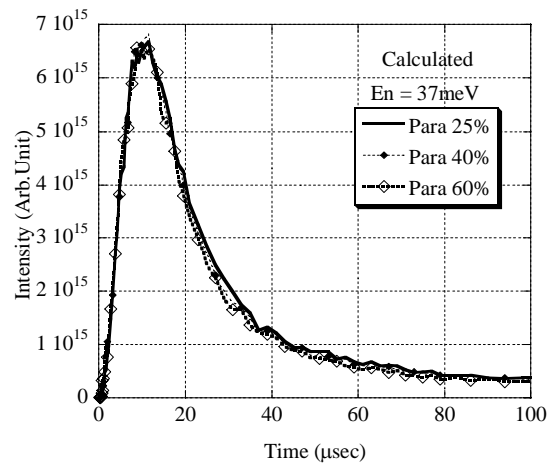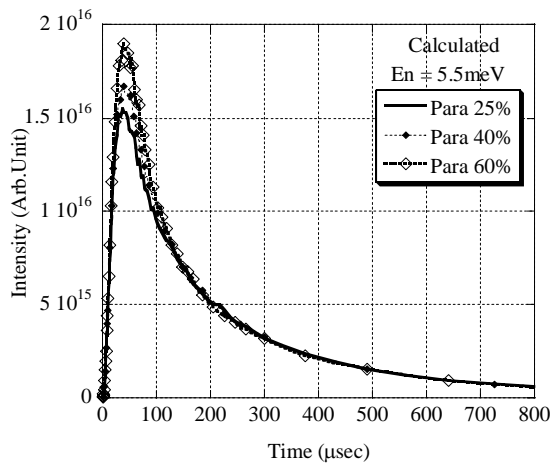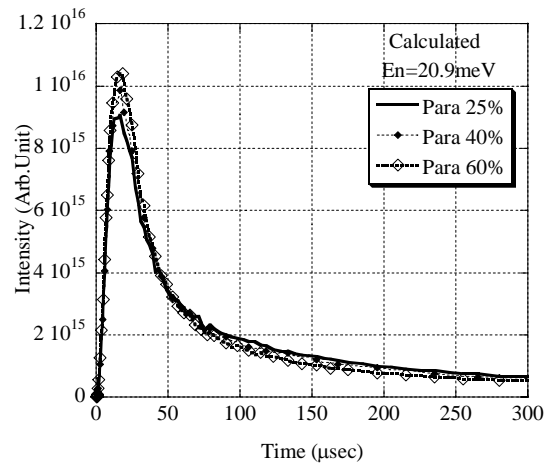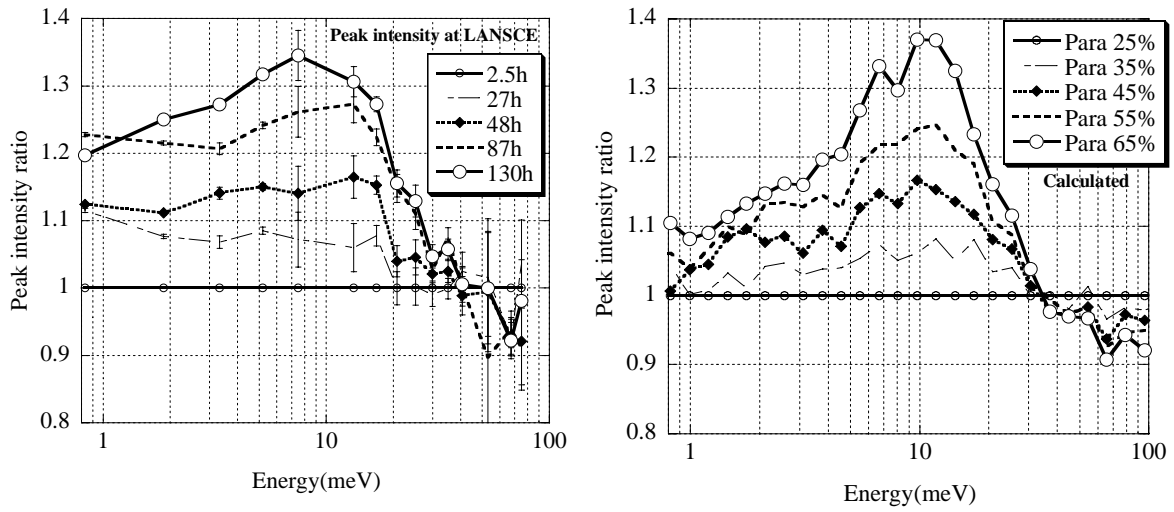
Figure 8. Calculated neutron pulse shapes.

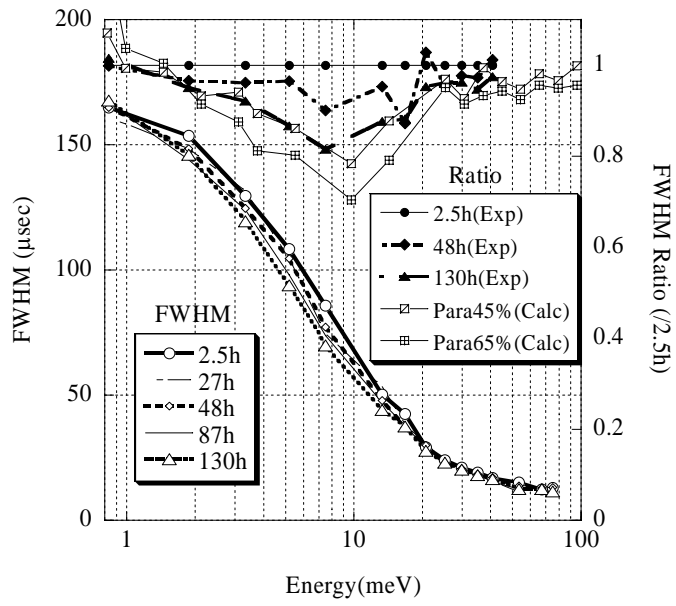Figure 9. Pulse peak intensity ratios depending on the time(Left). And calculated pulse peak intensity ratio (Right).

Figure 10. Time dependence of FWHM of pulse shapes.