

Neutron measurement at the thermal column of the Malaysian Triga Mark II reactor using gold foil activation method and TLD

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Abstract. In order to design facilities for boron neutron capture therapy (BNCT), the neutron measurement must be considered to obtain the optimal design of BNCT facility such as collimator and shielding. The previous feasibility study showed that the thermal column could generate higher thermal neutrons yield for BNCT application at the TRIGA MARK II reactor. Currently, the facility for BNCT are planned to be developed at thermal column. Thus, the main objective was focused on the thermal neutron and epithermal neutron flux measurement at the thermal column. In this measurement, pure gold and cadmium were used as a filter to obtain the thermal and epithermal neutron fluxes from inside and outside of the thermal column door of the 200kW reactor power using a gold foil activation method. The results were compared with neutron fluxes using TLD 600 and TLD 700. The outcome of this work will become the benchmark for the design of BNCT collimator and the shielding

1. Introduction

Cancer is one of the leading causes of death by 42% in the developing country [1]. Even though there were improvements in the common cancer treatments such as surgery, radiotherapy and chemotherapy, the patient of glioblastoma still have a very short overall survival period, which less than 18 months [2]. Thus, boron neutron capture therapy (BNCT) is looked as a promising method was studied around the world because the conventional treatment are not efficient enough to cure cancer [3] like melanoma and glioblastoma multiforme. According to [4], the patient will be irradiated with slow energy of neutron (thermal neutron or epithermal neutron) to reach the ratio of high concentration of boron compound in the tumour cell injected with a Boron-10 compound. The irradiation with slow neutron will caused nucleus of Boron-10 to capture the slow neutron to form



Boron -11 which then decays to Li-7 from the emission of an alpha particle He-4 [5]. BNCT was called binary treatment for cancer based on its nuclear reaction of the slow neutron bombarded with the non-toxic and non-radioactive compound of Boron-10.

In general, most of the BNCT studies use neutron source from the research reactors. This research is aimed at developing a BNCT facility for the cancer treatment studies that is safe and controlled from radiation over exposure and practically uses slow neutrons emitted from the Malaysian TRIGA MARK II reactor. Thus, the first step in order to achieve the long-term goal of this project is to establish the suitability of neutron fluxes produced from the research reactor. Commonly, most of the TRIGA reactor has a thermal column facility, which is comprises of graphite moderator designed to produce epithermal neutrons and thermal neutrons which can be utilised in BNCT facility. Therefore, the thermal column of Malaysian TRIGA MARK II reactor was identified for BNCT study as it has thermal and epithermal neutron source for this research. Previous researches indicated that the thermal column in the Malaysian TRIGA MARK II reactor has a sufficient a quality neutron beam to develop BNCT facility [5]. In order to develop a BNCT facility at the thermal column at this reactor, neutron measurement needs to be carried out comprehensively in the thermal column (graphite), thermal column door and outside of the thermal column to produce benchmark for material to be used for collimator and shielding this research.

2. Methodology

2.1. Experimental Arrangement

In this research, the experimental arrangement for the thermal column was divided into three phases as shown in figure 1.

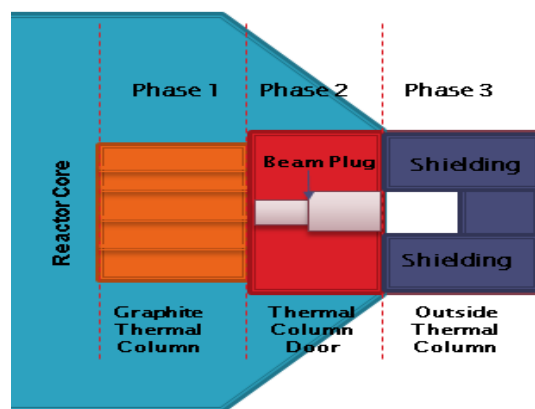


Figure 1. Three phases of thermal column of Malaysia TRIGA MARK II reactor.

Phase 1 consists of two parts; the inner part of the graphite which is right at the peripheral of the graphite reflector, and the second part which is the outer part right behind the door of the thermal column. The outer part is a pile of graphite stringers stacked in crisscross layers along the thermal column. There are 7 graphite stringers which were labelled according to their positions. Most of the graphite stringers have a 127cm length and $10.16 \times 10.16 \text{ cm}^2$ area, except for the lowermost and the uppermost graphite stringers which have areas about $10.16 \times 3.18 \text{ cm}^2$ with the same length. All of the stringers except a central stringer are provided with the six holes with 2 cm diameter and 0.7 cm depth with 24.5cm distance between the holes as a purpose for the sample irradiation in the thermal column. The central stringers were labelled as G7 consists of 13 holes and the 10cm distance between the holes. The central stringers were different compared to others stringers because of the neutron flux passing through these stringers are the highest because the location of central stringers is directly

pointing from the reactor core. Figure 2 shows the drawing of removable graphite stringers labelled and their position at the thermal column.

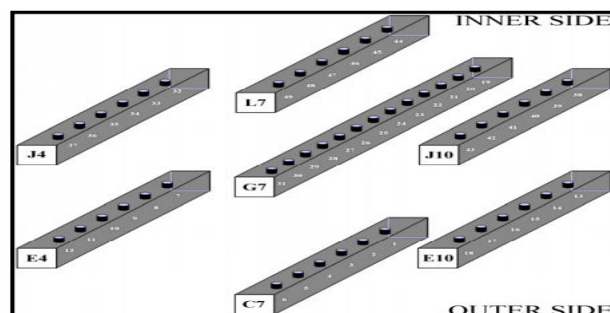


Figure 2. Schematic diagram of removable graphite stringers labeled and their position in the thermal column [6].

Phase 2 consists of thermal column door which have one beam pug across the beam line and G7 stringers. The beam pug divided in two sections which is internal and external beam pug. The internal beam pug has small diameter compare to external beam pug. Currently, both beam pugs are made of fully solid stainless steel and function as a beam stopper. During the experiment, both beam pug will be pulled out and replace with 8 cm diameter of temporary collimator made from polyethylene and filled with paraffin. The beam pug is potentially being developed for BNCT collimator.

Phase 3 are referring to the outside of the thermal column, which is covered about 100 cm from the exit of the thermal column door (Phase 2). The measurement of neutron in this region is important to estimate the neutron and gamma flux across the beam line and the surrounding of the thermal column for the safety and precautions prepared for the future BNCT facility.

2.2. Gold foil activation method

The neutron flux measurement is determined by using gold foil activation method that utilizes ^{197}Au reaction with neutrons to produce ^{198}Au . In this experiment, there are two types of samples being prepared which is pure 99.99% gold foil and Cadmium (Cd). The gold foil is used in this experiment because the gold foil will decay and emitting single gamma of well-known energy 412keV and the half-life of 2.81 days. The cadmium is well known as the thermal neutron filter that will be used to cover gold foil for to separate the thermal and epithermal neutron fluxes. There are 20 samples of bare gold foils with less than 1 mm in length and 20 gold foil covered cadmium samples with less than 1.3mm length were prepared for irradiation in Phase 1. All the samples were put in the irradiation vials which were labelled and that make the samples easier to handle and reduce contamination from irradiation of materials and for safety precautions. The positions of irradiated samples loaded into the stringers are shown as table 1 below:

The irradiation starts and continues for exactly 6 hours before getting the reactor shutdown. The time of reactor shutdown is also recorded for the purpose of activity calculations. The thermal column is opened again in the following day for unloading samples. In the Phase 2 and Phase 3, the position of the samples is shown as table 2 and figure 3:

Table 1. Stringers and holes use in experiments

Stringers	Holes
E4	E07
	E09
	E12
E10	E13
	E16
	E18
G7	G19
	G24
	G26
	G28
	G31
J4	J32
	J35
	J37
J10	J38
	J40
	J43
L7	L44
	L46
	L47

Table 2. The sample of bare gold foil and cadmium-covered gold foil for the Phase 2 and Phase 3 with the distance from G7 stringer

Phase	Sample	Distance from Graphite Thermal Column (cm)
2	A1	142
	A2	182.7
	A3	208
	A4	233.3
3	A5	258.6
	A6	283.9

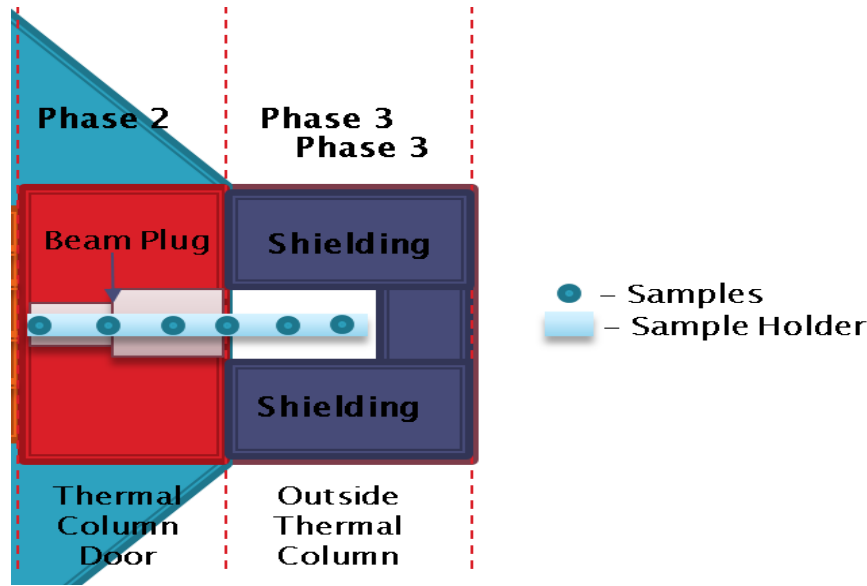


Figure 3. Illustration of sample position for the Phase 2 and Phase 3.

After irradiation, all of the samples in Phase 1, Phase 2 and Phase 3 were counted by a detector. The detector uses a germanium-detector gamma ray spectrum acquisition and analysis with 4.59×10^{-3} of detector efficiency. The count rate of radionuclide took more than two weeks to fully complete as the sample must have less than 25% of dead time or to repeat and, each sample took more than 30 minutes to be counted. The data then analysed by using the specific equation for neutron activation analysis to obtain thermal neutron flux and epithermal neutron flux [7]. At first, the count rate was converted into absolute foil activity by incorporating the detector efficiency by using the relation in equation (2.1):

$$A_{abs} = \frac{C_r}{\eta_d} \quad (2.1)$$

where A_{abs} absolute foil activity (counts/s), C_r = observed count rate (count/s), η_d = detector relative efficiency

By finding the activity of bare gold foil and the activity of cadmium covered gold foil, the thermal neutron flux was obtained by using equation (2.2):

$$\phi_{th} = \frac{\rho(A_{abs} - A_{cd})}{m\Sigma_a(1 - e^{-\lambda t_i})e^{-\lambda t_c}} \quad (2.2)$$

where A_{cd} = activity of cadmium covered gold foil, ρ = density of the foil material (gm/cm^3), m = weight of the foil (gm), Σ_a = macroscopic thermal absorption cross-section (cm^{-1}), λ = decay constant of radioactive isotope formed during radiation (s^{-1}), t_c = cooling time (s), t_i = irradiation time (s)

For gold, the response region that can absorb thermal neutron and epithermal neutron is in between 0.0015eV to 5.8eV and the cut-off energy of cadmium of 0.55eV was used to mark both thermal and epithermal neutron region. Therefore, the difference between the activity of bare gold foil and the cadmium covered gold foil, if both were irradiated in the same neutron flux under the same circumstances, is the activity caused by thermal neutron flux. The ratio of thermal to epithermal fluxes was determined by cadmium ratio, which is defined by the equation (2.3):

$$\text{Cadmium ratio} = \frac{\text{Activity of bare gold foil}}{\text{Activity of Cd covered gold foil}} \quad (2.3)$$

By using equations 2.2 and 2.3, the epithermal neutron flux can be calculated by the equations given below.

$$R_{cd} - 1 = \frac{\phi_{th} \cdot \sigma_{th}}{0.1526 \text{ barn}} \quad (2.4)$$

and,

$$\phi_{epi} = \theta \text{Log} \left(\frac{E_2}{E_1} \right) \quad (2.5)$$

where ϕ_{th} = thermal neutron cross-section (barns), θ = intermediate neutron flux density per unit lethargy, ϕ_{epi} = epithermal neutron flux (neutrons $\text{cm}^{-2}\text{s}^{-1}$), $E_1 = 0.5$ eV, $E_2 = 2$ MeV, I_r = resonance integral (1526 barns).

2.3 Thermoluminescent dosimeter

In this experiment, two types of TLD was used which is TLD-600 and TLD-700 for measuring gamma and neutron dose rate. The advantage of using TLD in this research is that a TLD has a long-term storage of information data and have high sensitivity towards radiation. TLD-600 could detect and is sensitive to both neutron and photon such as gamma while TLD-700 is sensitive to detect and measure photon only [8]. On the other hands, the use of TLD detector is convenient because the atomic density of LiF is definitely close to the human body. TLD detector is also cheap and affordable compare to another detector use in the recent world market. The experiment carried out using TLD-600 and TLD-700 only for the Phase 3 (A5 and A6), because of the time limitation with only 60 seconds of irradiation time and the safety issues as the Phase 1 and Phase 2 are highly exposed to radiation of gamma and neutron. Figure 3 and table 2 above illustrates the position of the TLD- 600 and TLD-700 for sample A5 and A6.

3. Results and discussion

3.1. Gold foil activation measurement

Based on experimental results shown in table 3, the thermal neutron flux from the phase 1 proved that stringer G7 provide highest thermal neutron flux compared to other stringers. Thermal neutron is defined as slow neutrons that are approximately in thermal equilibrium with a moderator. The highest thermal neutron flux came from hole G19 from stringer G7 with $7.42 \times 10^{11} \text{ n.cm}^{-2}\text{s}^{-1}$ while the lowest is $1.35 \times 10^9 \text{ n.cm}^{-2}\text{s}^{-1}$ from hole E18 of stringer E10. Most of the samples loaded in stringer G7 have higher thermal neutron flux due to the position of the stringer that located closest to the reactor core. As the distance of the stringer is close to the reactor core, more neutron fluxes would be produced. Meanwhile, the position of stringer E10 (which is located above the reactor core) gave the lowest thermal neutron flux due to the high volume of shielding material there. Besides that, the epithermal neutron flux graph indicated that the highest result of epithermal neutron come from hole G19 with $1.16 \times 10^{11} \text{ n.cm}^{-2}\text{s}^{-1}$ as compared to the lowest result from stringers L7. Epithermal neutron is defined as neutron having energy in the range immediately above the thermal range, roughly between 0.02 and 100 eV. This result also occurs due to the position of the stringer with stringer G7 located near to the reactor core. Meanwhile, the position of stringers L7 is above of G7 and located near to the moderator. The interaction of the neutrons in irradiated samples and coolant from thermalisation effect has produced less epithermal neutron flux. This indicates that both thermal and epithermal neutron flux in G7 at Phase 1 are applicable for BNCT purpose.

Table 3. Thermal and epithermal neutron flux measured using the gold foil activation method for Phase 1

Stringers	Holes	Thermal Neutron Flux (neutron.cm ⁻² s ⁻¹)	Epithermal Neutron Flux (neutron.cm ⁻² s ⁻¹)
E4	E07	7.05×10^{10}	1.11×10^{10}
	E09	2.34×10^{10}	3.10×10^8
	E12	2.24×10^{10}	2.18×10^9
E10	E13	5.75×10^{10}	2.64×10^9
	E16	1.04×10^{10}	1.33×10^9
	E18	1.35×10^9	6.61×10^7
G7	G19	7.42×10^{11}	1.16×10^{11}
	G24	2.81×10^{11}	2.30×10^9
	G26	2.41×10^{10}	1.32×10^8
	G28	1.47×10^{10}	2.03×10^8
J4	G31	1.85×10^{10}	1.73×10^8
	J32	5.39×10^{10}	2.94×10^9
	J35	8.31×10^{10}	2.12×10^8
	J37	7.31×10^{10}	1.81×10^8
J10	J38	6.78×10^{10}	2.50×10^9
	J40	1.63×10^{10}	2.07×10^8
	J43	9.79×10^9	1.13×10^8
L7	L44	2.10×10^{10}	1.24×10^9
	L46	1.48×10^{10}	6.65×10^7
	L47	1.20×10^{10}	3.98×10^7

Table 4 shows the result of thermal and epithermal neutron flux measurement at Phase 2 and Phase 3. From table 4, the highest thermal neutron flux is $3.62 \times 10^7 \text{ n.cm}^{-2}\text{s}^{-1}$ at the distance of 142 cm from the stringer G7. The thermal neutron flux decreased linearly with the distance at Phase 2. At the Phase 3, the thermal neutron flux at position A5 decreased about 25% from the $3.53 \times 10^6 \text{ n.cm}^{-2}\text{s}^{-1}$ and increase back to $128 \times 10^6 \text{ n.cm}^{-2}\text{s}^{-1}$ when the distance measured at 283.9 cm from the thermal column. The surrounding factor become the main factor of this fluctuate of thermal neutron from the Phase 2 to the Phase 3 as the Phase 3 is outside of the thermal column and exposed to the air.

The epithermal at the Phase 2 and Phase 3 proved that the neutron flux decrease linearly towards the distance from the graphite thermal column. The sample A1 at the distance 142 cm was the highest epithermal neutron flux with $5.91 \times 10^6 \text{ n.cm}^{-2}\text{s}^{-1}$ while the sample A6 at the 283.9 cm was measured the lowest with $3.89 \times 10^4 \text{ n.cm}^{-2}\text{s}^{-1}$. Technically, the epithermal neutron flux reduced to about 99.34% from Phase 2 to the Phase 3.

3.2. Thermal and epithermal neutron flux across the beam line

The result of neutron measurement from the Phase 1, Phase 2 and Phase 3 was combined to show the thermal and epithermal neutron flux across the beam line with 283.9 cm from the reactor core of TRIGA MARK II. The measurement start with the stringers G7 as a beam line for the Phase 1 and continued by Phase 2 and Phase 3 as shown in the Table 5 and Figure 4.

Table 4. Thermal neutron and epithermal neutron measured using the gold foil activation method for Phase 2 and Phase 3.

Phase	Sample	Distance from Graphite Thermal Column (cm)	Thermal Neutron Flux (neutron. cm ⁻² s ⁻¹)	Epithermal Neutron Flux (neutron. cm ⁻² s ⁻¹)
2	A1	142	3.62 x 10 ⁷	5.91 x 10 ⁶
	A2	182.7	5.51 x 10 ⁶	3.98 x 10 ⁶
	A3	208	5.27 x 10 ⁶	1.90 x 10 ⁶
	A4	233.3	3.53 x 10 ⁶	8.90 x 10 ⁵
3	A5	258.6	8.89 x 10 ⁵	2.78 x 10 ⁵
	A6	283.9	1.28 x 10 ⁶	3.89 x 10 ⁴

According to the table 5 and figure 4, both neutron thermal and epithermal neutron flux decreased linearly with the distance from 142 cm to 283.9cm from graphite thermal column as the flux generally is exponentially decreasing with distance[9]. From the Phase 1 to the Phase 2, the thermal and epithermal neutron flux are significantly reduced to about 99.8% for thermal neutron flux and 96.5% for epithermal neutron flux. The use of graphite as a neutron moderator for the research reactor in Phase 1 become the main factor for the tremendously decreases of flux from the Phase 1 to the Phase 2. The property of graphite that have higher thermal conductivity result in the increase the measured thermal and epithermal neutron fluxes at Phase 1. Across the beam line, the thermal neutron flux was higher than epithermal neutron flux with 1.00×10^2 n.cm⁻²s⁻¹ difference. The aim of this measurement to clearly see the thermal and epithermal neutron across the beam line for the optimization purpose for collimator. The result also shows that the thermal and the epithermal neutron flux at the thermal column of the Malaysian Triga Mark II reactor can be used for BNCT research purpose according to the IAEA standard[10].

Table 5. Thermal neutron and epithermal neutron measured using the gold foil activation method across the beam line for Phase 1,Phase 2 and Phase 3

Distance (cm)	Thermal Neutron Flux (neutron. cm ⁻² s ⁻¹)	Epithermal Neutron Flux (neutron. cm ⁻² s ⁻¹)
0	7.42 x 10 ¹¹	1.16 x 10 ¹¹
28.4	2.81 x 10 ¹¹	2.30 x 10 ⁹
56.8	2.41 x 10 ¹⁰	1.32 x 10 ⁸
85.2	1.47 x 10 ¹⁰	2.03 x 10 ⁸
113.6	1.85 x 10 ¹⁰	1.73 x 10 ⁸
142	3.62 x 10 ⁷	5.91 x 10 ⁶
182.7	5.51 x 10 ⁶	3.98 x 10 ⁶
208	5.27 x 10 ⁶	1.90 x 10 ⁶
233.3	3.53 x 10 ⁶	8.90 x 10 ⁵
258.6	8.89 x 10 ⁵	2.78 x 10 ⁵
283.9	1.28 x 10 ⁶	3.89 x 10 ⁴

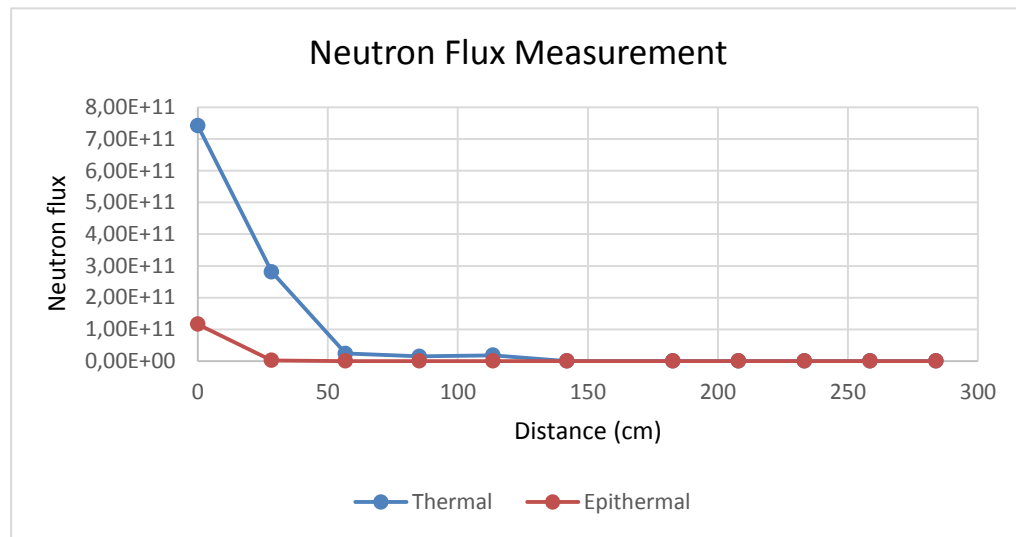


Figure 4. The neutron flux measurement across the beam line for the Phase1, Phase 2 and Phase 3.

3.3. TLD-600 and TLD-700 measurements

Table 6 shows the result of neutron and gamma at the Phase 3 obtained from TLD-600 and TLD-700. Only Phases 3 was available to use TLD as the TLD required shorten time to measure dose and to prevent any safety issues regarding to over exposure of radiation. The previous results revealed that the outside of the thermal column contained higher neutron than gamma. Therefore, at 200 kW experiment, the outside part of the thermal column is safe with low gamma radiation[11]. By comparing both gold foil activation and TLD method, the neutron (mSv) at sample A5 is higher with TLD. As can be showed on table 6, the gold foil activation measurement are significantly not in sync with TLD experiment measurement due to different count rate. The TLD measurement was counted right after the experiment been conducted while for the gold foil activation analysis, the sample was counted when the activity of the sample are low. The gap of the time of sample counted for both experiments effected the measurements [11].

Table 6. The comparison between the measurement of neutron and gamma using TLD and gold foil activation method

Sample	Gold foil activation measurement		TLD-600 and TLD-700 measurement	
	Neutron (mSv)	Gamma (mSv)	Neutron (mSv)	Gamma (mSv)
A5	3.00	-	4.20	0.25
A6	4.20	-	2.38	0.27

4. Conclusion

The thermal and epithermal neutron fluxes at thermal column of Malaysian TRIGA MARK II reactor were successfully measured using two methods; gold foil activation and TLD. The purpose of this measurement is to explore the feasibility of the neutron source from the thermal column to be used for BNCT application. From the experiments conducted, the thermal column produced significantly higher thermal and epithermal flux at the stringers G7. Besides that, the data also proved that the

thermal and epithermal fluxes decreased linearly with distance as the experiment are set up across the beam line on the Phase 1, Phase 2 and Phase 3. This experimental data would be further used in designing collimator and shielding for BNCT facility.

5. References

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