Study of neutron radiation effect in $LaBr_3$ scintillator *

LU Yi(卢毅)1) SONG Zhao-Hui(宋朝晖) LI Gang(李刚) TAN Xin-Jian(谭新建)
ZHAO Xiao-Chuan(赵晓川) ZHANG Kan(张侃)

Northwest Institution of Nuclear Technology, Xi'an 710024, China

Abstract: The resistance to neutron irradiation of $LaBr_3$ scintillator was studied in this work. The change of background counting rate, light output and energy resolution of the $LaBr_3$ scintillator were analyzed to determine whether the scintillator was damaged under different neutron flux rates induced by ^{241}Am-Be, D-T neutron generator, and reactor neutron source. The results show that the neutron radiation damage in $LaBr_3$ scintillator is mainly affected by neutron flux rate. Under low flux rate, the properties of the scintillator were hardly changing, while under high flux rate, there is obvious deterioration in the background spectra and in the energy resolution because of the neutron activation. After a period, the neutron radiation damage will spontaneously recover.

Key words: neutron resistance, $LaBr_3$ scintillator, neutron flux

PACS: 29.30.Hs, 29.40.Me **DOI:** 10.1088/1674-1137/39/10/106003

1 Introduction

The recently discovered cerium-doped $LaBr_3$ scintillator owns superior properties such as high energy resolution, fast response time, small nonproportionality, and high scintillation yield. The $LaBr_3$ detector is proved to be suitable for the gamma spectrum detection especially for high-energy gamma rays and has been widely applied in different research areas. Recent research shows that the $LaBr_3$ scintillator is also fit for neutron detection [1, 2], and the $LaBr_3$ scintillator has favorable resistance to proton and γ irradiation [3–5]; however, few studies about the effect of neutron radiation in $LaBr_3$ scintillator has been reported.

In this work, we intend to study the neutron radiation damage or the resistance to neutron of $LaBr_3$ scintillator under different neutron flux rates. The change of self-radioactive background in the scintillator, light output, and the energy resolution were measurement and compared to analyze whether the $LaBr_3$ scintillator was damaged by neutron irradiation. Three neutron sources of ^{241}Am-Be, D-T neutron generator and reactor were used to supply different neutron flux rates.

2 Experiment

In this experiment, a 5% cerium-doped scintillator $LaBr_3$ (size, ϕ50×10 mm) supplied by CTI was used. A 10^6 n/s ^{241}Am-Be neutron source was used to provide low neutron flux. The calculated neutron flux rate in the position of $LaBr_3$ scintillator is about 2×10^2n/(cm^2·s) according to the intensity of the source. Because the ^{241}Am-Be neutron source can offer a continuous and stable neutron flux, the irradiation experiment lasted several weeks. After a short period of irradiation, the background spectra were measured and recorded.

The background spectra include self-radioactivity in $LaBr_3$ and the surrounding radiation, as shown in Fig. 1.

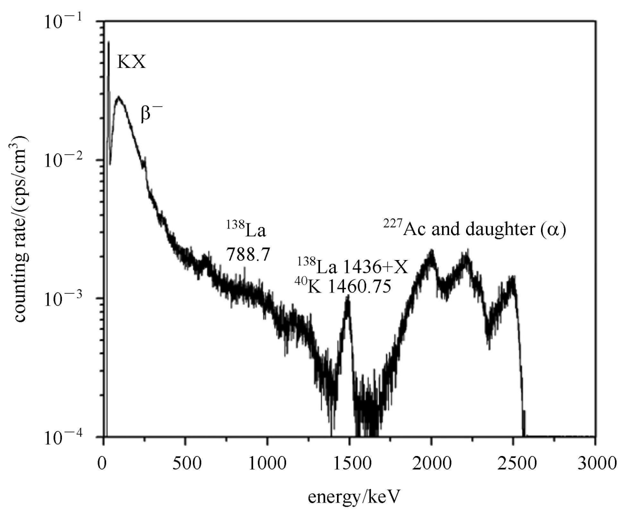

Fig. 1. Background spectra measured by $LaBr_3$ scintillation detector.

Received 28 February 2015
∗ Supported by National Natural Science Foundation of China (11275151, 11205122)
1) E-mail: luyi@nint.ac.cn

The self-radioactivity of $LaBr_3$ scintillator is mainly caused by the unstable isotope ^{138}La and the contamination of ^{227}Ac. ^{138}La decays into ^{138}Ba by electron capture with a branching ratio of 66.4% emitting a 1436 keV γ ray in coincidence with a 32 keV Ba X-ray and into ^{138}Ce by β^- emission in the other 33.6% with an end-point energy of 255 keV. The background in the energy region between 1500 and 2750 keV is due to the daughter nucleus of ^{227}Ac, such as ^{227}Th, ^{223}Ra, ^{219}Rn, ^{215}Po, and ^{211}Bi [6].

Then, the $LaBr_3$ scintillator was irradiated under a higher neutron flux supplied by the D-T neutron generator at the China Institute of Atomic Energy. The neutron generated from the source is about 6.7×10^8 n/s average in 4π direction. Because the stability of the neutron beam is not easy to maintain, an average calculated neutron flux with the value of 3.2×10^3 n/(cm^2·s) is adopted, and the total net irradiating time is about 60 min. Finally, an experiment with a neutron flux rate of nearly 10^5 n/(cm^2·s) was taken on the 1# radial beam port at Xi'an Pulsed Reactor. The background spectra and energy resolution before and after the experiment were measured and compared to show whether the scintillator was damaged by neutron irradiation.

3 Results and discussion

3.1 ^{241}Am-Be neutron source experiment

The counting rate of background and surrounding gamma rays, as well as self-radioactivity, was recorded under increasing neutron fluence (total neutron number injected in the scintillator) as shown in Fig. 2.

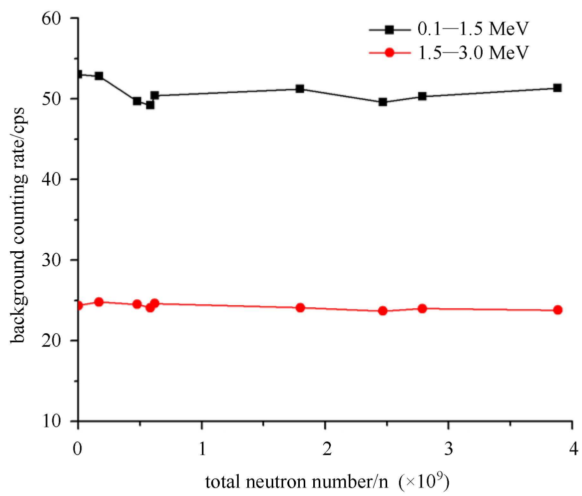

Fig. 2. (color online) Change of background counting rate of the 19.63 cm^3 $LaBr_3$ scintillator.

The contribution of the surrounding natural isotopes was considered to be unchangeable because the background spectra were measured under the same condition. The counting rate of the energy region from 0.1 to 1.5 MeV (indicating ^{138}La decay) and from 1.5 to 3.0 MeV (indicating ^{227}Ac decay) changes slightly while the neutron fluence increases. Although the total neutron fluence in the scintillator reached about 3.88×10^9 neutrons, there was almost no change in the background counting rate of the scintillator.

The spectra of the background, ^{137}Cs and ^{60}Co measurement by the $LaBr_3$ detector before and after the irradiation of ^{241}Am-Be source are in Figs. 3 and 4. In Figs. 3 and 4, the background counting rate and energy resolution (3.8%@662 keV, 2.9%@1173 keV, and 2.6%@1332 keV) of the $LaBr_3$ scintillator almost remained the same after neutron irradiation, but the light output decreased by approximately 5% according to the left shift of the channels. (The difference between the counting rate before and after irradiation is because of the changing of the gamma source position.)

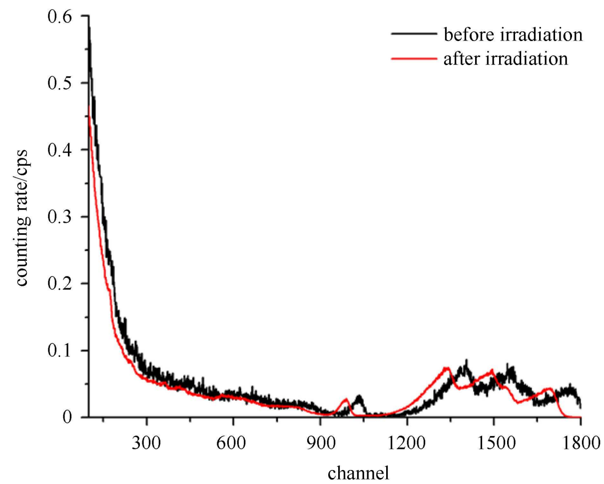

Fig. 3. (color online) Background spectra acquired by the $LaBr_3$ detector before and after the irradiation of ^{241}Am-Be source.

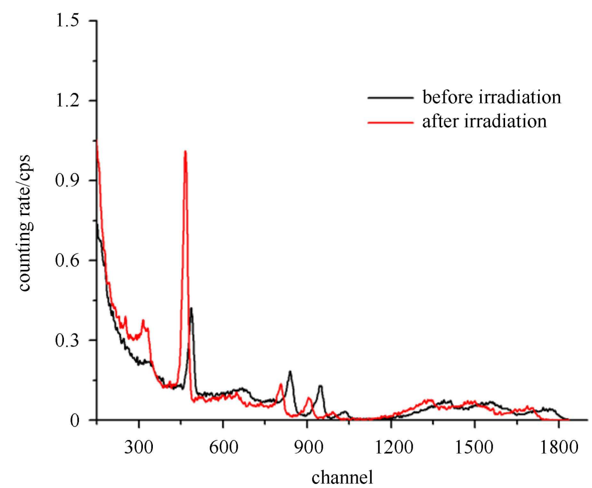

Fig. 4. (color online) ^{137}Cs and a ^{60}Co spectra acquired by the $LaBr_3$ detector before and after the irradiation of ^{241}Am-Be source.

3.2 D-T neutron generator experiment

The neutron flux of the D-T neutron generator is about 3.2×10^3 n/(cm^2·s) in the $LaBr_3$ scintillator, and the total neutron fluence is about 2.3×10^8 n. The background counting rate of the detector and the energy resolution before and after the irradiation was compared.

As shown in Fig. 5, the background counting rate of the scintillator greatly increased especially in the low energy region after the irradiation.

There was also a decline in the energy resolution of the scintillator. The energy resolution at 661.6 keV gamma ray of ^{137}Cs dropped from 3.8% to 5.4%, as shown in Fig. 6.

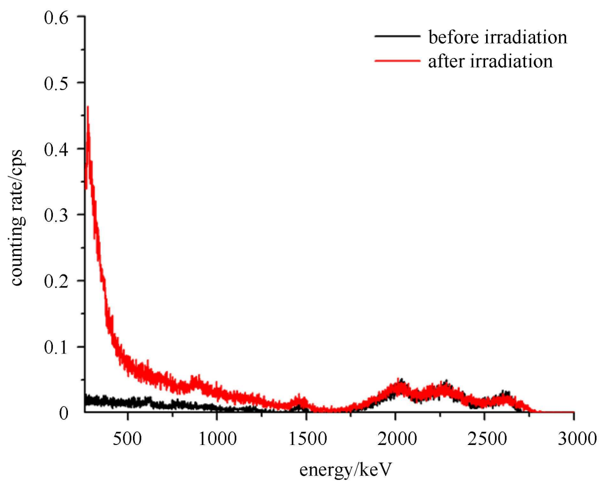

Fig. 5. (color online) Background spectra acquired by the $LaBr_3$ detector before and after the irradiation of the D-T source.

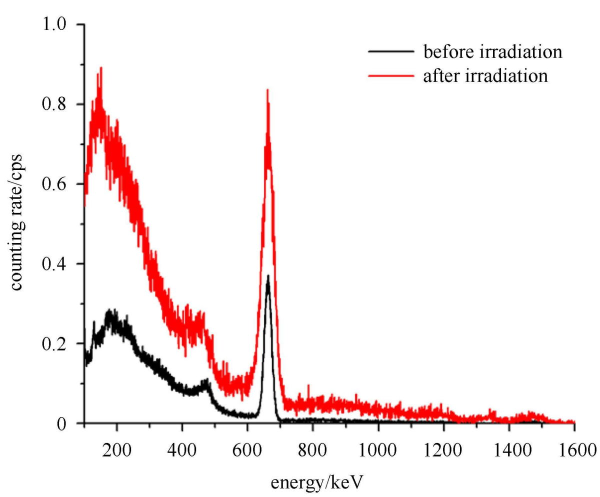

Fig. 6. (color online) ^{137}Cs spectra acquired by the $LaBr_3$ detector before and after the irradiation of D-T source.

As observed in Figs. 5 and 6, the counting rate of the energy region from 0.1 to 1.5 MeV was increased, which comes from ^{138}La decay, but the counting rate of the energy region from 1.5 to 3.0 MeV did not. It means that the radioactivity of ^{138}La may increase because of the irradiation of neutrons, and the neutron activation through (n, 2n) reaction [7] may lead to a worse energy resolution.

3.3 Reactor experiment

The 1# radial beam port at Xi'an Pulsed Reactor supplied neutron flux of nearly 10^5 n/(cm^2·s) in the scintillator, and the total neutron fluence was about 3.5×10^9 after a 30-minute irradiation.

After the irradiation, the background counting rate of the 0.1–1.5 MeV energy region increased to 114.4 cps compared with the ^{241}Am-Be source experiment (49.8 cps average), whereas the energy resolution at 661.6 keV gamma ray of ^{137}Cs dropped from 3.8% to 4.5%. A possible explanation is the activation of La in the scintillation by neutron reaction. However, the light output increased by 7.6% according to the right shift of the channels, which is contradictory to the result of the ^{241}Am-Be experiment. After about three weeks, the background counting rate of 0.1–1.5 MeV energy region dropped to about 80 cps, and the energy resolution of 661.6 keV gamma ray recovered to 4.0%.

The background counting rate may be mainly affected by the neutron flux rate. In these three experiments, the total number of neutrons injected in the scintillator of the three sources are all above 10^8 neutrons, but their neutron flux rates differ from 10^2 to 10^5 n/(cm^2·s). As a result, under low flux rate (^{241}Am-Be source), the background spectra and the energy resolution of the scintillator hardly changed, but under high flux rate (D-T and reactor source), there are obvious changes in the background spectra counting rate and energy resolution, which come from the short-lived activation induced by the neutrons. The ^{241}Am-Be source emits both thermal neutrons and fast neutrons while the D-T source just emits fast neutrons and the reactor source mainly emits thermal neutrons. Short-lived activation could be produced by thermal neutron capture reaction and by the (n,2n) reaction with fast neutrons. Although the mechanism is different, the performances are alike. So the background counting rate of the scintillator seems to get more influence from the neutron flux rate than neutron energy. It is hard to discern the relationship between the energy resolution and the neutron energy, because even though the flux rate of the reactor neutron is much higher than the D-T neutron source, the energy resolution does not get even worse. The light output may be affected by the neutron energy, because the changes were observed in both the ^{241}Am-Be source and reactor source experiment and what is more, the light output of the scintillator seems to have dropped after the irradiation of ^{241}Am-Be, whereas the result of the reactor experiment is opposite. In this work, the neutron radiation damage finally recovers after a period, regardless of whether the $LaBr_3$ scintillator acquires irrecoverable damage when the neutron flux rate increases even more. To deal with these problems, further research is needed.

4 Conclusion

The resistance to neutron irradiation of $LaBr_3$ scintillator was studied in this work by the measured change of background counting rate, light output, and energy resolution of the $LaBr_3$ scintillator under the irradiation of ^{241}Am-Be, D-T accelerator, and reactor neutron. The total number of neutrons injected in the scintillator of the three sources are all above 10^8 neutrons, but their neutron flux rates differ from 10^2 to 10^5 n/(cm^2·s). As a result, under low flux rate, the properties of the scintillator hardly changes, there is little radiation damage in the scintillator. On the other hand, under high flux rate, there are obvious changes in the background spectra and in the energy resolution. Thus, the neutron radiation damage in $LaBr_3$ scintillator is mainly affected by neutron flux rate and, after a period, the scintillator will spontaneously recover. It is hard to explain the effect of neutron energy on energy resolution and light output based on the present work; further studies and detailed work should be carried out in the future.

References

1 Oberstedt A, Billnert R, Oberstedt S. Nuclear Instruments and Methods in Physics Research A, 2013, **708**: 7–14
2 Cazzaniga C, Croci G, Giacomelli L et al. Nuclear Instruments and Methods in Physics Research A, 2013, **732**: 384–387
3 Owens A, Bos A J J, Brandenburg S et al. Nuclear Instruments and Methods in Physics Research A, 2007, **572**: 785–793
4 Drozdowski W, Dorenbos P, Bos A J J et al. IEEE Transactions on Nuclear Science, 2007, **54**(4): 1387–1391
5 Normand S, Iltis A, Bernard F et al. Nuclear Instruments and Methods in Physics Research A, 2007, **572**: 754–759
6 Quarati F G A, Khodyuk I V, Van Eijk C W E et al. Nuclear Instruments and Methods in Physics Research A, 2012, **683**: 46–32
7 Cazzaniga C, Nocente M, Tardocchi M et al. Nuclear Instruments and Methods in Physics Research A, 2015, **778**: 20–25