Technical Physics Letters, 2023, Vol. 49, No. 8

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Optical and chemical stability of the liquid scintillator of the iDREAM detector at the Kalinin nuclear power plant

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Received May 16, 2023
Revised May 30, 2023
Accepted May 31, 2023

Optical and chemical stability studies were performed for a Gd-doped liquid scintillator synthesized for using in a prototype of industrial reactor antineutrino detector iDREAM. It is shown that the scintillator solution 1.1 m^3 in volume remains stable for two years of observations provided the temperature is kept below $\sim 20^\circ$C and contact with air is prevented.

Keywords: neutrino detector, liquid scintillator, reactor antineutrino.

DOI: 10.61011/TPL.2023.08.56682.19627

Long-term stability of properties of gadolinium-doped liquid organic scintillators (Gd-LOS) is necessary to ensure long-term collection of neutrino interaction statistics. Gadolinium possesses a large cross-section of thermal neutron capture and is introduced into the scintillator in order to increase the neutrino detectors efficiency in detecting a neutron arising during the antineutrino-proton interaction according to the reverse beta-decay reaction: $\bar{\nu}_e + p \rightarrow e^+ + n$. Beginning from the first studies performed at the Rovno NPP neutrino laboratory in the 1980s, in a great number of experiments researchers failed to prepare large-volume (1 m^3 and larger) Gd-LOSs able to retain for a long time the optical and chemical stability [1–3].

By now, the international practice has provided data on the Gd-LOS stability for scintillation compositions based on white-spirit [4], phenylxylylethane [5] and linear alkylbenzene (LAB) [6,7]. In all cases, the gadolinium concentration did not exceed 1 g/l. Earlier, in the framework of developing and preparing for testing the neutrino detector iDREAM (industrial Detector of Reactor Antineutrinos for Monitoring), we have studied samples of LAB-based scintillation compositions [8], synthesized Gd-LOS samples up to 30 l in volume, and investigated their stability [9]. In 2021, a prepared Gd-LOS sample 1.1 m^3 in volume was poured into detector iDREAM installed in the neutrino flux at the Kalinin NPP (KNPP). The results of observing the optical and chemical stability of that Gd-LOS are presented below.

The iDREAM detector has been designed for monitoring the nuclear reactor via the neutrino emission from the active zone. The detector is located in the subpile room at level 0.0 of the KNPP Unit 3 at the distance of 20 m from the reactor VVER-1000 active zone (thermal power $P_{th} = 3000$ MW). As the neutrino target, a LAB-based 1.1 m^3 Gd-LOS was used, to which metal gadolinium included in gadolinium 3,5,5-trimethyl-hexanoate was added. The gadolinium concentration was 1 g/l. To prevent Gd-LOS against contact with air, an excess nitrogen pressure of $0.5-1.5$ kPa was maintained under the tightly closed detector cover. The detector was initially filled with scintillator free of gadolinium; then a concentrated solution of gadolinium 3,5,5-trimethyl-hexanoate in LAB (Gd-complex) was added. Detailed description of the detector and procedure for preparing Gd-LOS is given in [10].

The measurement conditions in the industrial reactor subpile room stipulate the neutrino detector operation at temperatures above 30°C. It is known that this can result in degradation of the LAB-based Gd-LOS optical characteristics [11]. During the measurements at KNPP, the Gd-LOS temperature is maintained in the range of $\sim 12-20^\circ$C by air conditioning inside the detector protection. Since the protection surrounds the detector on all sides, its cooling proceeds in a confined volume. This results in minimizing the effect of seasonal variations in environmental temperature upon both the Gd-LOS and photomultiplier tubes (PMTs) whose quantum efficiency depends on temperature. Each PMT is controlled by voltage and current consumption accurately to 0.2% [10]. Stability of output signals of adders-discriminators summing-up all

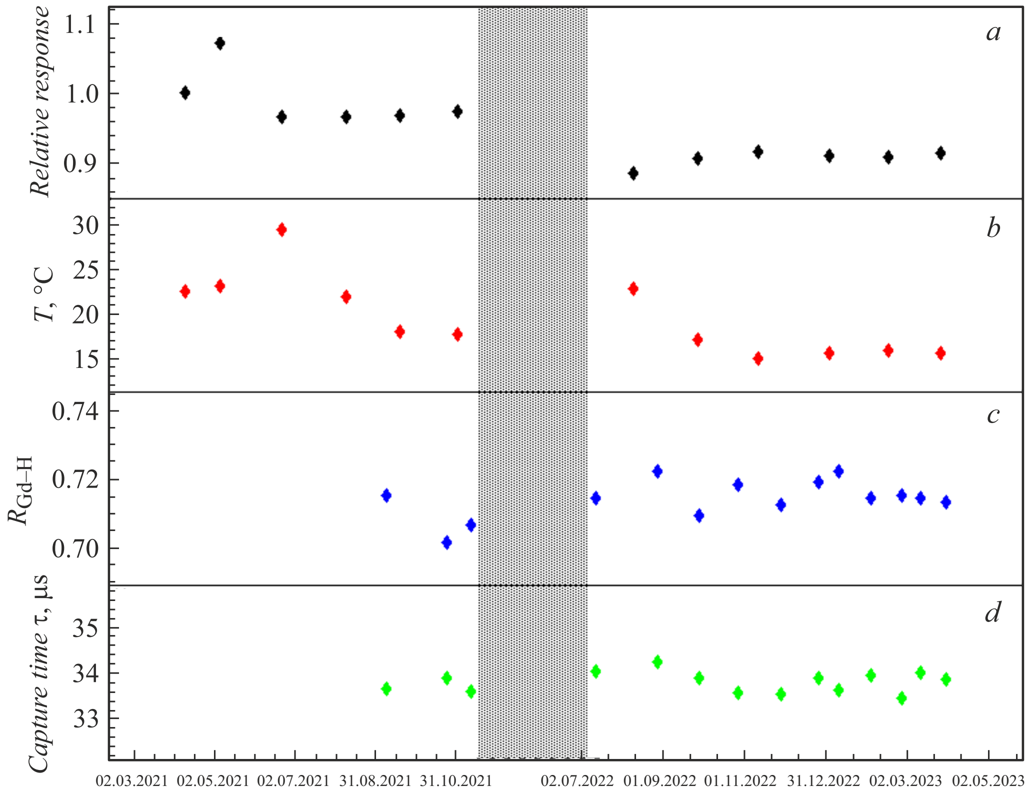

Figure 1. Relative response (*a*), Gd-LOS temperature (*b*), $R_{\text{Gd-H}}$ (*c*), and neutron capture time τ in Gd-LOS of the iDREAM detector (*d*). During the detector shutdown, no measurements were performed .

the PMT signals is controlled using a generator calibration pulse.

Degradation of the scintillator optical properties may result in loss in its transparency and/or light output, which is expected to reduce the light collection and, hence, to decrease the detector response. Optical properties of Gd-LOS were monitored by observing variations in the detector light collection. There was observed relative variation in the position of the complete absorption peak of γ-photons from the ^{60}Co ($E_\gamma = 1.17 + 1.33\,\text{MeV}$) source in the center of detector. The peak position was determined via its normal-distribution approximation. As a unit, there was taken the peak position measured in the first day after adding to scintillator the Gd-complex and stirring it by purging with nitrogen. Notice that intense foaming made it necessary to supply nitrogen under low pressure ($\sim 10\,\text{kPa}$), due to which stirring was inefficient.

Fig. 1, *a* demonstrates relative variation in the detector response (the values are averaged over 1.5 months). In the first one or two months, a 8% increase in light collection was noted, which might be caused by a gradual increase in transparency of Gd-LOS in the process of stirring by diffusive convection. The subsequent decrease in response to ~ 0.97 may be associated with an increase in temperature of Gd-LOS that was not being cooled at that time. The Gd-LOS temperature was increased to 33°C (the temperature data averaged over 1.5 months are shown in Fig. 1, *b*). Right afterwards, forced air conditioning was performed inside the detector protection.

In the period from December 2021 to July 2022, the detector was off. The excess nitrogen pressure was not maintained at that time, and cooling of Gd-LOS was not performed. When data collection was recommenced, a 10% reduction in the response to ~ 0.88 was observed (Fig. 1, *a*). Gd-LOS was purged with nitrogen, the excess nitrogen atmosphere under the cover was restored, and cooling was recommenced. After that (during 2022), a small ($\sim 3\%$) response increase was observed, which may be explained by a slow transparency growth caused by settling of dust particles and floating-up of microbubbles of dissolved gases formed during purging. The same effect was observed in the Double Chooz near detector [5].

Chemical stability of Gd-LOS is characterized by variation in the Gd-complex solubility and/or nonuniformity of distribution of the Gd-complex included in Gd-LOS. As the measure of the Gd-LOS chemical stability, the ratio between the numbers of neutron captures in gadolinium and hydrogen is typically considered; this ratio is defined as

$$\frac{N_{\text{Gd}}}{N_{\text{H}}} = \frac{\sigma_{\text{Gd}}}{\sigma_{\text{H}}}\frac{\rho_{\text{Gd}}}{\rho_{\text{H}}},$$

where σ_{Gd} and σ_{H} are cross-sections of the thermal neutron capture in gadolinium and hydrogen, respectively, while

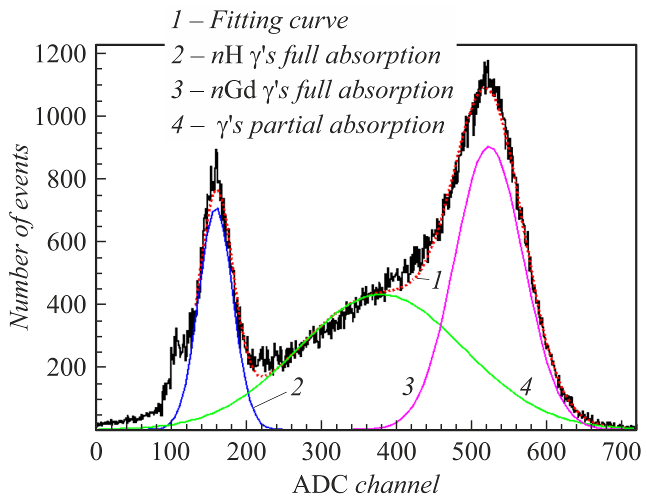

Figure 2. Charge spectrum of the ^{252}Cf neutron captures in the center of detector iDREAM and the best-fit curve (line *1*). *2* and *3* are the complete absorption spectra of γ-photons from neutron captures by hydrogen and gadolinium, respectively, *4* is the partial absorption spectrum of γ-photons.

ρ_{Gd} and ρ_{H} are the gadolinium and hydrogen densities in Gd-LOS.

The chemical stability was monitored beginning from September 2021 by using fast-neutron source ^{252}Cf in the center of detector. The events of neutron capture in Gd-LOS were selected by the delayed coincidence method: first the detector registers instantaneous γ-photons from spontaneous ^{252}Cf fission (prompt event), and then γ-photons from capturing the neutrons slowed down to thermal neutron energies by nuclei of ^{157}Gd ($\sigma = 2.5 \cdot 10^5$ b, $E_\gamma = 7.9\,\mathrm{MeV}$) and ^{155}Gd ($\sigma = 5.6 \cdot 10^4$ b, $E_\gamma = 8.5\,\mathrm{MeV}$) or of hydrogen ($\sigma = 0.332\,\mathrm{b}$, $E_\gamma = 2.23\,\mathrm{MeV}$). Neutron capture time τ is determined by the time of neutron deceleration and diffusion in Gd-LOS and matches the time between the prompt and delayed events. Fig. 2 presents the charge spectrum of ^{252}Cf neutron capture. The peak in the region of ~ 160 channels of the analog-digital converter (ADC) relates to the neutron capture by hydrogen, while the peak in the region of ~ 520 ADC channels corresponds to gadolinium.

In the case of small-size iDREAM detectors it is rather difficult to determine the total numbers of the N_{Gd} and N_{H} neutrons captures. This is because the ^{252}Cf fast neutrons get scattered over the detector and captured, among other regions, at the Gd-LOS periphery. For such events, partial energy absorption of emitted γ-photons due to the edge effect is detected, and the total energy deposit may be below the detector threshold. To characterize the chemical stability, quantity $R_{\mathrm{Gd-H}} = N'_{\mathrm{Gd}}/(N'_{\mathrm{H}} + N'_{\mathrm{Gd}})$ was chosen, where N'_{Gd} and N'_{H} were defined as integrals in the peaks of complete γ-photons absorption as a result of neutron capture by gadolinium and hydrogen, respectively (Fig. 2, curves *3* and *2*). Therefore, $R_{\mathrm{Gd-H}}$ means the share of neutron captures by gadolinium in the total number of captures by gadolinium and hydrogen for the case when the edge effect is ignored. The $R_{\mathrm{Gd-H}}$ measurements are presented in Fig. 1, *c*. The $R_{\mathrm{Gd-H}}$ value remains stable accurately to $\pm 2.4\% (\pm 3\sigma)$.

Fig. 1, *d* presents the measurements of neutron capture time τ that can vary because of degradation of the Gd-LOS chemical properties. As the figure shows, neutron capture time τ remains stable accurately to $\pm 2\% (\pm 3\sigma)$.

Thus, the paper presents the results of studying the optical and chemical stability of Gd-LOS of the iDREAM detector. The observation period was two years long. The detector response remained stable while the Gd-LOS temperature was maintained below 20°C. In the absence of cooling and excess nitrogen atmosphere under the detector cover, a drop in light collection by $\sim 10\%$ was observed; the light collection got partially restored after purging and switching to normal cooling. No signs of a decrease in the Gd-LOS chemical stability were found.

The experience of the iDREAM detector shows that, under the conditions of constant temperature (below 20°C) and minimization of contact with air, the prepared Gd-LOS may be used in long-term measurements in the framework of solving fundamental and applied problems of reactor antineutrino physics.

Financial support

The study was supported by the Russian Science Foundation (project № 22-12-00219).

Conflict of interests

The authors declare that they have no conflict of interests.

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Translated by Solonitsyna Anna