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Gadolinium for neutron detection in current nuclear instrumentation research: A review

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A B S T R A C T

Natural gadolinium displays a number of remarkable physical properties: it is a rare earth element, composed of seven stable or quasi-stable isotopes, with an exceptionally high magnetization and a Curie point near room temperature. Its use in the field of nuclear instrumentation historically relates to its efficiency as a neutron poison in power reactors. Gadolinium is indeed the naturally occurring element with the highest interaction probability with neutrons at thermal energy, shared between Gd-157 (15.65%, 254000 b cross section) and Gd-155 (14.8%, 60900 b) isotopes. Considering that neutron capture results in an isotopic change, followed by a radiative rearrangement of nuclear and atomic structures, Gd may be embodied not merely as a neutron poison but as a neutron converter into a prompt photon and an electron source term. Depending on the nature and energy of the reaction products (from a few-keV Auger electrons up to 8 MeV gamma rays) that the detector aims at isolating as an indirect neutron signature, a variety of sensor media and counting methods have been introduced during the last decades. This review first draws a theoretical description of the radiative cascade following $\mathrm{Gd}(n,\gamma)$ capture. The cascade may be subdivided into regions of interest, each corresponding to dedicated detection designs and optimizations whose current status is detailed. This inventory has allowed the authors to extract and benchmark key figures of merit for the definition of a detection scheme: neutron attenuation, neutron sensitivity (cps/nv), gamma rejection, neutron detection limit in a mixed field, intrinsic or extrinsic moderation, and transportability. On this basis, the authors have identified promising paths for Gd-based neutron detection in contemporary instrumentation.

Contents

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1. Introduction

Neutron detection and counting form a crucial and multidisciplinary issue in nuclear measurement. This review will have a special, although non restrictive focus on real-time detection and dosimetry for low-level neutron monitoring in the presence of a low-level gamma background [1]. Neutron radiation is, for instance, weakly attenuated by air and is deeply penetrating, which justifies the acute attention that it receives in radioprotection. Multiple environments require specific monitoring of neutron dose rate. For nuclear industry workers, these include fuel (U-235, Pu-239) fabrication and reprocessing units, power plants, and storage and decontamination sites for spent fuel [2]. The irradiation of staff (and patients) within the proximity of medical, research and industrial accelerators, as well as users of calibration neutron sources, is also strictly regulated [3]. Neutron counting plays a major role in passive, non-destructive control of fissile isotopes all along the fuel cycle, including inventory purposes, characterization of nuclear waste packaging and criticality monitoring [4]. Eventually, neutron detection is one of the keystones of special nuclear material (SNM: U-233, U-235, Pu) detection in the frame of nuclear and radiological threat prevention [5]. Neutron radiation portal monitors (RPMs) are installed in public places, critical entry checkpoints, and national borders, to prevent nuclear material proliferation and the risk of a "dirty bomb" contaminating an extended area.

Neutron radiation, as well as electromagnetic waves, belongs to the category of neutral radiations, *i.e.* as a neutron passes through matter, it is not affected by Coulomb forces from electrons in the medium. On the contrary, free neutrons interact with matter nuclei, by recoil or by nuclear reaction, leading to the emission of secondary charged particles that ionize the sensor and allow for a charge carrier-induced signal to be collected [6]. Neutron radiation is therefore qualified as indirectly ionizing and is solely detected by the ionization signature from the charged products of neutron diffusions and reactions. Diffusions are only practically exploitable when the atomic mass of the target nucleus is close enough to that of one of the incident neutrons and when the kinetic energy T_n of the neutron falls into the fast range, usually defined between 1 and 10 MeV, essentially ^{1}H(n,n') diffusions [7]. Conversely, nuclear reactions of interest for neutron detection essentially take advantage of isotopes with interaction probability with neutron at thermal energy (i.e. E_n = 25 meV at 19 °C) [6]. These are mainly ^{3}He(n,p), ^{10}B(n,α), ^{6}Li(n,α), ^{157}Gd(n,γ), ^{155}Gd(n,γ) and ^{113}Cd(n,γ) reactions. For these reactions to be put into play, it is therefore necessary that neutron radiation has been moderated before reaching the converter, either through diffusions inside the environment (for instance, the human body in the case of personal dosimeters) or by the traversing through a dedicated moderator set around the detection medium. Table 1 summarizes the key figures of merit (equivalent microscopic surface or cross section, liberated energy or Q-value, type of interaction products) for the main neutron–nucleus interactions used in nuclear instrumentation.

Helium-3 is the most commonly deployed converter in neutron counters found on the market. The inert He-3 gas under pressure is contained inside a tube, which is itself placed within a hydrogenous moderator whose dimensions maximize the neutron capture rate inside the gas. Charged particles, produced by the reaction (protium and tritium nuclei), directly ionize the gaseous medium, generating a cloud of charge carriers that induce the detected signal. Commercial detectors display a neutron sensitivity within the order of magnitude of one count per neutron nanosievert (1 c.nSv^{-1}) against a gamma vulnerability within the order of magnitude of 10^{-3} c.nSv^{-1}, hence obtaining a n/γ discrimination ratio of 10^{-3}, which is robust up to 1 mSv.h^{-1} gamma dose rates [8]. It is thus the combination of He-3 high thermal neutron sensitivity (explained by the 5330 b cross section from Table 1) and gamma rejection of the gaseous medium that makes such detectors a reference. However, 9/11 attacks have led to the deployment of a rising quantity of SNM monitors at borders, causing the He-3 demand to skyrocket [9]. The shadow of a worldwide shortage and the strategic stakes related to the He-3 resource (United States and Russian stocks of nuclear weapons being the main providers of the isotope during the 2000 decade) have severely affected the cost of the detectors [10]. The quest for alternative technologies to He-3 proportional counters has therefore become a prominent topic of research in contemporary nuclear instrumentation.

2. Key physical features, supply data and instrumental uses of gadolinium

2.1. General physical properties of Gd

Gadolinium is naturally present as a combination of five stable and two quasi-stable isotopes, all labeled $^{A}_{64}$Gd with Z = 64 being the atomic number and A the mass number of the isotope: $^{152}_{64}$Gd $\left(T_{1/2} = 1.08 \cdot 10^{14}\ \text{a};\alpha\right)$, $^{154}_{64}$Gd, $^{155}_{64}$Gd, $^{156}_{64}$Gd, $^{157}_{64}$Gd, $^{158}_{64}$Gd and $^{160}_{64}$Gd $\left(T_{1/2} > 10^{21}\ \text{y};\beta^{-1}\beta^{-1}\right)$. Its average atomic mass equals 157.25 ± 0,03 u [11]. The electrons of a Gd atom cloud populate successive electronic layers with respect to the value of their principal quantum number n, according to the formula $(K)^2(L)^8(M)^{18}(N)^{25}(O)^9(P)^2$. Taking into account all four quantum numbers, the electronic configuration of Gd reads

$$[\text{Gd}] = 1s^2 2s^2 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} \mathbf{4f^7} 5s^2 5p^6 5d^1 6s^2 \quad (1)$$

From these considerations, it follows that Gd is part of the 6th period and 4f-subblock of the periodic table. Gadolinium is classified among the lanthanide series, comprising all elements with atomic numbers ranging from Z = 57 (La) up to Z = 71 (Lu), chemically similar to lanthanum. For a lanthanide element, including Gd, the +3 oxidation state is the most stable. As a result, Gd is found in natural form and in the majority of synthetic compounds as a Gd^{3+} trivalent cation, while it may be obtained as a Gd^{2+} divalent cation in solutions [12]. Gadolinium also finds its place among the metal group of rare earth elements, in which Sc (Z = 21) and Y (Z = 39) are added to the lanthanide series. Because of analog chemical properties, these elements are found as a mixture in a limited number of minerals, such as monazite and bastnaesite [13].

Gadolinium, when incorporated in low amounts (of the order of 1 wt%) in iron and chromium alloys, has the property to improve the workability, mechanical resistance, and high-temperature resistance to oxidation of the material [14]. Together with Fe, Co, and Ni, Gd is reported as one of the four simple ferromagnetic elements, although the notion is debated [15]: it remains that Gd presents an exceptionally high magnetization field and unusually low Curie temperature T_c, marking the transition from a state of intrinsic magnetic order to the paramagnetic state of the metal. The magnetization of Gd according to the direction of an external magnetic field then admits an inflexion point within the vicinity 289 K $\leq T_c \leq$ 293 K [16,17], so that the ferromagnetic–paramagnetic phase transition of Gd may be embodied as a room-temperature heat flux sensor [18].