# Gadolinium-Neutron Capture

Gadolinium exhibits interesting characteristics particularly desirable in the field of neutron detection. Upon thermal-neutron interaction, it displays a **significant reaction probability** and high Q-value. The gadolinium-neutron capture yields particles with a characteristic energy distribution. *Each particle contributes to a unique component of the total energy spectrum. It is this spectrum that makes neutron observations possible.*

It is imperative that users of gadolinium-based neutron detectors understand the origin of the spectrum components. This chapter starts with an introduction to gadolinium and its response to thermal neutrons, followed by a more comprehensive report on the resulting energy spectrum and its components

## Gadolinium

Gadolinium (Gd) is a chemical element with atomic number 64. It is a metal and appears as a solid under standard pressure and room temperature. In nature, Gd occurs as a composition of seven isotopes; the most abundant being Gd-158 (24.84\%), followed by Gd-160 (21.86\%), Gd-156 (20.47\%), Gd-157 (15.67\%) and Gd-157 (14.80\%).

%Introduction to neutron detection???

## Cross-section

Neutron capture cross-section of natural Gd (48800±150 barns) is given by the weighted sum of isotopic cross-sections. The relative abundance of gadolinium isotopes and their neutron capture cross-section are listed in table 1. Of all known natural occurring nuclei, Gd-157 has the highest thermal-neutron absorption cross-section (\*). In natural gadolinium, Isotope Gd-157 and Gd-155 collectively contribute 99.99\% of the capture probability. Gd may therefor be simplified as a “two-absorbing isotope system” consisting of the isotopes Gd155 and Gd157 [Dumazert, 2018].

## Reaction Equation

*Since nat-Gd interaction with neutrons can be ascribed* to isotopes Gd-157 and Gd-155, it is worth studying their corresponding nuclear reaction equation.

\begin{equation}

\_{64}^{155}Gd \rightarrow \_{64}^{156}Gd^\* \rightarrow \_{64}^{156}Gd + \gamma + ICe^- (Q=8.5 MeV)

\end{equation}

\begin{equation}

\_{64}^{157}Gd \rightarrow \_{64}^{158}Gd^\* \rightarrow \_{64}^{158}Gd + \gamma + ICe^- (Q=7.9 MeV)

\end{equation}

After a gadolinium nucleus absorbs a neutron it is excited to a higher energy state (neutron-capture state) from which it decays by gamma-transition. The decay primarily results in emission of gamma-rays ($\gamma$-ray) and internal conversion (IC) electrons. Byproducts are Auger and Coster-Kronig (ACK) electrons and X-rays, prompted by vacancies left by the IC electrons.

The Q-value ($Q$) is defines as the difference in mass before and after a nuclear reaction. It represents the net energy released when the nucleus has completely decayed. The reaction energy is distributed as kinetic energy among reaction products. Due to the Gd nuclei’s large mass, compared to a photon (massless) and an electron, its recoil energy is neglectable ( Modern Nuclear Chemistry, page 219\*) and most of the Q-value is transferred to the gamma-rays and IC electrons.

## Reaction Energy Spectrum

From Gd(n,gamma) capture, the excitation energy is distributed among reaction products; roughly 99\% is carried by prompt gamma-rays and the rest by low energy electrons [Sakurai et al.]. The energy spectrum ranges from 0 MeV to the Q-value. Prompt gamma-rays lie all over the spectrum, from low to high, while energies of IC electrons and their biproducts are mainly located at the lower end, below 0.2 MeV.

The resulting spectrum is a superposition of two basic components. The first, a continuous spectrum representative of energetic prompt gamma-rays. The second, a discrete set of lines who correspond with energies of the low-energy prompt gamma-rays, IC electrons, Auger electrons and X-rays. The prompt gamma-ray emission adds to both components, while the remaining reaction products only supply the discrete.

\begin{figure}[h]

\centering

\includegraphics[width=7cm]{fig/lvl.png}

\caption{Nuclear levels of an arbitrary nucleus}

\label{fig:1}

\end{figure}

The spectrums form is closely related to the nuclear structure of gadolinium. Figure ? illustrates excited states of an arbitrary nucleus. A low-lying nuclear level has less excitation energy than a high lying nuclear level. Low-lying levels are discrete and easily distinguishable, each with a known spin and parity. The nuclear level density increases with excitation energy. Eventually, high lying levels become indistinguishable and resemble a continuum. In fig. ?, the quasicontinuum domain of energy states is represented by a gradient, where the energy level density increases as the gradient darkens. Energy levels within the quasicontinuum are marked by dotted lines and levels within the discrete domain are marked with uninterrupted lines. No clear boundary separates the domains; the transition between the two is smooth and gradual. The highest energy level represents neutron capture state and the lowest level ground state, both are indicated by a bold uninterrupted line. A transition from one level to another is indicated by and arrow.

### Prompt Gamma-Rays

A nucleus may transition once or several times before it reaches ground state.

Transitions can occur (1) between states in the continuous domain, (2) between states in the discrete domain or (3) between states from one of each.

In the continuous domain, there are nearly an endless amount of states a nucleus can decay from. Transitions from these (slightly) different energy states is what brings about about the spectrum’s apparent continuity.

As the nucleus de-excites and gets closer to ground state, transitions start to take place between states in the discrete domain occurs. Here, *the amount of levels are limited, and the number of possible gamma-emission energies are finite. This results in discrete peaks near the lowest part of the energy spectrum.*

\begin{figure}

\centering

\includegraphics[width=7cm]{fig/Gd\_ExcitedStates.png}

\caption{Excitation levels of Gd-155\* and Gd-157\* and corrosponding gamma-transitions}

\label{fig:1}

\end{figure}

In the spectrum, gamma-rays take the form of a broad peak structure along with a couple of prominent spikes. The distribution stretches from one end of the spectrum to the other. The broad peak favors energies in the bottom half of the spectrum, its apex between 0 and 2 MeV, and tapers off slowly as it approaches maximum energy release.

Fig. \ref{fig:Gd\_ExcitedStates} illustrates gamma-transistions associated with excited gadolinium nuclei. Two discrete peaks, in the lower end, dominate the spectrum. In a purely isotopic material of either Gd-156\* or Gd-158\* a set of two energy pairs is emitted, {88.97 keV, 199.22 keV} and {79.51 keV, 181.94 keV}, respectively. The lowest energy of each set is characteristic for the transition between the first excited state with spin-parity ( J^π) 2$^+$ and the ground state 0$^+$. The larger energy is characteristic for transitions between the second excited state with spin-parity 4$^+$ state and the first excited state 2$^+$[C.W. Reich, Nuclear Data Sheets for A = 156, Nucl. Data Sheets 113 (11) (2012) 2537–2840.]. The gamma-ray emission rate probability (per neutron capture) of Gd-157 and Gd-155 are listed in table ?? [Gräfe et. al]

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### Internal Conversion Electrons

Alternatively, the atom can de-excite by means of internal conversion (IC), the direct emission of an orbital electron. In gadolinium excited by neutron capture, IC is most probable for transitions from first state to ground state $(2^+ \rightarrow 0^+)$ and second state to first state $(4^+ \rightarrow 2^+)$. These transitions are responsible for 96.7\% of the energy carried by IC electrons [??!]. They also happen to be the same transitions from which the discrete gamma-ray duplets, mentioned previously, are produced.

IC and gamma emission are competing decay modes. The ratio of IC decay rate $\lambda\_{ICe^-}$ to gamma decay rate $\lambda\_{\gamma}$ can be described by the internal conversion coefficient (ICC) $\alpha$:

\begin{equation}

\alpha = \frac{\lambda\_{ICe^-}}{\lambda\_{\gamma}}

\end{equation}

In cases where gamma decay is preferred the coefficient is small, perhaps even negligible, and differently when IC is preferred the coefficient is large.

The probability of IC depends on the electron shell (K,L, M, …)and shells therefor have respective coefficients ($\alpha\_K$,$\alpha\_L$,$\alpha\_M$, …).

Inner shell electrons, such as those from the K shell, are more likely to interact directly with the nucleus, since its wavefunction has finite probability of penetrating the nucleus. The probability of IC in a shell becomes less likely the further away it lies from the nucleus. In other words, internal conversion depends heavily on the atomic electron density inside the nucleus. (studie of the probability of IC from shells [ref?], table?). Consequently, odds of nuclear interaction with the K-shell is more likely than with the L-shell, than the M-shell and so on.

The total ICC is the ratio of total number of IC electrons to gamma-rays emitted by a nucleus and it can be expressed as a sum of shell coefficients:

\begin{equation}

\alpha\_{TOT} = \sum\_i \alpha\_i \ , \ i = K, L, M, ...

\end{equation}

Transition levels of lower energy favor internal conversion. As calculated by [A.AHarms, Table 4], transitions of Gd-157\* from the second lowest transition level, L-shell of the first excited state, are 3 times more likely of mediation by IC than gamma-emission. While de-excitation from higher states are less prone to IC. Already at the third excitation state, Gd-157\* exhibits a low coefficient of 0.1, meaning IC electron rates are 10 times lower than gamma rates.

The energy of an IC electron is determined by the available transition energy and the binding energy of a shell.

\begin{equation}

E\_{ICe^- }=E\_T-E\_{bi,i},\ , \ i = K, L, M, ...

\end{equation}

IC electrons contribute to discrete lines in the 20-200 keV range. Eminent is the intensity at which specifically 71 keV electrons are produced, nearly 0.27 nc$^-1$ in natural gadolinium [T.Aoyama].

### X-rays and Auger Electrons

When a conversion electron is expelled from subshell m a vacancy is left behind. Still in an excited state, the atom undergoes further de-excitation. An electron from a higher subshell p (>n) descends and fills the vacancy, releasing atomic excitation energy by either emission of x-rays or Auger electrons.

X-ray emission occurs when the transition energy goes into electromagnetic radiation. Difference in binding energy makes up the x-ray energy.

\begin{equation}

E\_{x-ray} = E\_T = E\_{bi,p}-E\_{bi,m}

\end{equation}

On the other hand, the transition energy may go into freeing an electron of an intermediate subshell n where n lies between shell m and p. The so-called Auger electron has energy equal to the difference in transition energy and binding energy.

\begin{equation}

E\_{Ae^-} = E\_T-E\_{bi,n}

\end{equation}

X-rays and Auger electrons, from L- and K-shell, of gadolinium-neutron capture are radiated in the 0-50 keV range. The most prominent x-ray has energy of 43 keV and an emission rate of 0.47 nc$^-1$ (per neutron capture). Auger electrons worth noting are of energy 35 keV (0.08 nc$^-$) and 5 keV (0.21 nc$^-1$). In comparison to x-rays, they are less frequent and of lower energies. [Dumartez]

Rest

As efficient neutron absorbers, gadolinium plays an important role in neutron shielding alloys for nuclear reactor safety and storage (\*). An additional use of great gadolinium-neutron capture is as Gd-based neutron poison. For instance, Gd(III) nitrate is used in moderator systems to regulate power generation and shutting-down Heavy Water Nuclear Reactors (\* page 31). In addition to a notable reaction probability, gadolinium-neutron capture also has a comparably **large Q-value**. These features are desirable, and makes gadolinium highly suitable for the usage, in the field of neutron detection. Reaction products of Gd(n,) yield a characteristic energy spectrum and it is this spectrum which makes neutron observations possible.

This chapter therefore provides

* starts with a brief introduction to gadolinium as an element,
* a moderate explanation of gadoliniums behavior upon neutron interaction and
* a comprehensive report on the resulting energy spectrum.