SCINTILLATORS FOR PGNAA IN MINERAL EXPLORATION

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SUMMARY

Current gamma-ray detectors, based upon scintillation, are not likely to perform well in narrow diameter logging-while-drilling (LWD), such as NQ diamond drilling. The normally used halide scintillators of NaI, CsI are considered to not have sufficient resolution to compensate for a lack of stopping power in PGNAA applications. Whereas, BGO, a robust oxide scintillator performs so poorly with respect to resolution, light output, and temperature sensitivity that the higher density and stopping power does not compensate fully for its limitations.

We have evaluated several novel halide and oxide scintillators that might improve the viability PGNAA in small diameter LWD application. Specifically, we have looked at CsI, SrI, BGO, CWO, CeBr, GAGG, LaBr, YAP scintillators, and find that CeBr and CWO may be a useful alternative to LaBr in PGNAA applications. Our modelling and analysis indicates that very dense scintillators such as CWO, with moderate resolution capability, or moderate density with high resolution, such as CeBr and LaBr, are the best candidates for measuring line spectra from PGNAA with small, less than 50 mm diameter, detectors. However, none of the newer scintillators are a panacea as the penetrative nature of the PGNAA process means that a high background of scattered gamma rays will obscure many weak elemental peaks even with a "perfect" detector.

Key words: Scintillator, PGNAA, Gamma Spectrometer.

INTRODUCTION

There is increasing interest in applying LWD technologies to slim hole drilling in mineral exploration. The acquisition of real time lithological and mineralogical information can lead to significant cost savings for an exploration program. LWD is standard practice in the petroleum industry but the challenge of the miniaturisation of existing sensing technologies has inhibited the development of suitable tools (tools for mineral exploration have smaller working diameters, 50 mm or less, than petroleum tools). The use of natural-gamma and spectral gamma wireline tools is not uncommon in mineral exploration so a natural question to ask is whether these existing sensors technologies can be adapted for LWD applications and, further, are they suitable for PGNAA applications? Specifically, the most fundamental (and important) component of any gamma spectrometer is the scintillator, which converts gamma-rays to light.

Traditionally, the principal technical characteristics for a scintillator are its stopping power and resolution. Stopping power is a measure of the scintillator's ability to completely absorb the energy of a gamma-ray, minimising the amount of scattered energy present in the resulting spectrum. Resolution is a measure of the degree to which gamma-rays of different energy can be accurately discriminated. There are other properties that we should take in practical consideration, especially in an LWD environment where robustness and temperature stability are important. For this study, we concentrate on the principal quantities and consider other factors in light of the results.

The quality and characteristics of scintillating materials has improved in recent years, with many modern scintillators now available in sizes up to 50 mm in diameter (Lecoq 2016). In this research, we have compared some of the normally used scintillators with more novel ones to see what benefits newer scintillators might offer to PGNAA and to what degree. Newer halide scintillators have been used in new generation wireline tools for iron ore (FastGrade tool using pulsed a pulsed neutron source (Sodern)) and Uranium (Penney et al. 2012) grade control, LaBr and CeBr respectively. These new halide scintillators, which include SrI:Eu, (i.e. similar in physical properties to NaI:Tl and CsI:Na and CsI:Tl) offer many advantages in terms of resolution and have slightly higher density whereas the newer oxide scintillators have even higher density, but often lower resolution. So what really matters?

METHOD AND RESULTS

We have used the Geant4 Monte Carlo software (Agostinelli et al. 2003) to model the expected performance of each scintillator under consideration. The model consisted of a cylindrically symmetric borehole 10 cm in diameter (approximately HQ in size), in a 2m diameter x 1m high formation, whose composition was chosen to reflect typical elemental crustal abundance and density (Fleischer 1953). A 5cm diameter x 10 cm length scintillator crystal is located in the centre of the borehole. In all, eight simulations were run for the eight different scintillators shown in Table1, which summarizes their relevant physical properties. The energy resolution of crystals is based on our laboratory measurements (Carson and Kepic 2016; Kepic et al. 2015), personal communication with suppliers, and

published data. Note that the resolution and light collection efficiency can vary significantly from manufacturer-to-manufacturer. Additionally, most published resolution figures tend to be "optimistic", the best result achieved with a small sample. Larger scintillators tend to have lower resolution than published results because of greater difficulties growing highly uniform and transparent crystals.

Scintillator	Density (g/cc)	Resolution at 662KeV	Comment	Published data
CsI:TI	4.5	6.5	Low density	(Moszynski et al. 1998)
SrI:Eu	4.5	4.5	Low density, few suppliers	(Sturm et al. 2010)
BGO	7.1	16	Poor resolution. Light yield strongly temperature dependent.	(Gierlik et al. 2006)
$CdWO_4$	7.9	8	$_{48}Cd$ is a strong neutron absorber. Slow scintillation time.	(Kim et al. 2008)
$CeBr_3$:Ce	5.2	4.2	Expensive, one supplier	(Quarati et al. 2013)
GAGG:Ce	6.6	6.5	$_{64}Gd$ is a strong neutron absorber.	(Iwanowska et al. 2013)
$LaBr_3$:Ce	5.1	3.5	Expensive, one supplier	(Quarati et al. 2011)
YAP:Ce	5.3	5	Expensive, few suppliers	(Kapusta et al. 1999)

Table 1: Physical properties of the scintillator crystals studied for PGNAA.

Note that we are not using an exact model of a PGNAA tool (because neutron transport modelling is not performed), but rather concentrate on examining the 'ideal' performance of each scintillator in a realistic medium after the neutron induced prompt gamma conversion. Rather than model the diffusion and subsequent capture of source neutrons, we have generated the capture gamma-ray spectrum explicitly and assumed that the thermal neutrons have diffused fairly uniformly in the model volume (as the neutrons are very penetrative even relative to gamma rays). Nor are fast neutron reactions modelled. A weighted neutron capture cross-section for each isotope in the formation model is calculated from the IAEA PGNAA database (IAEA) to calculate the line spectra for each isotope.

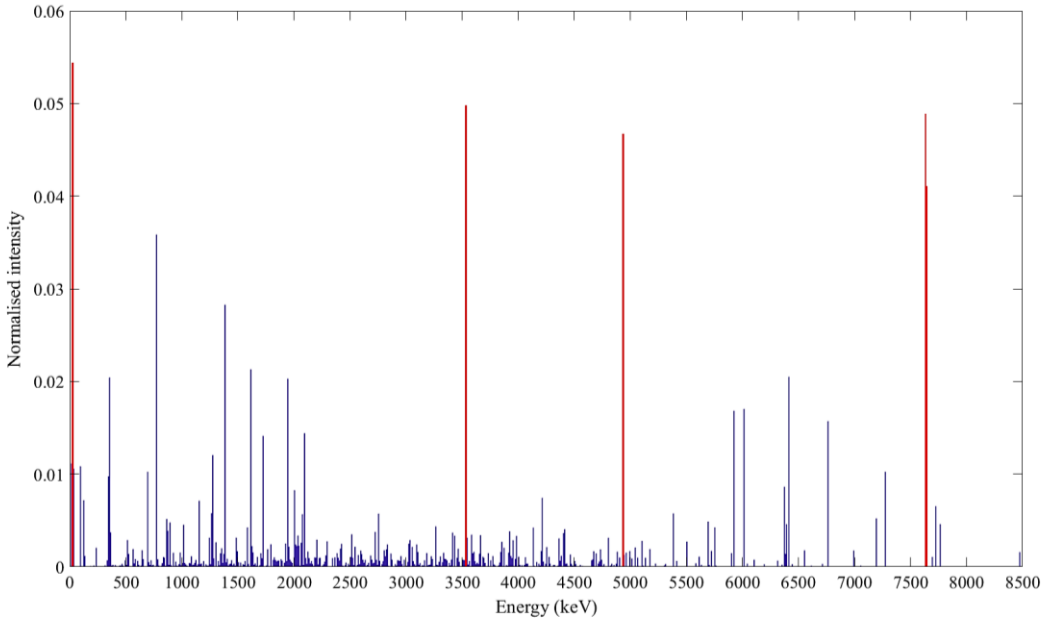

Figure 2: Line spectrum used as an input for GEANT4 simulation. The five most intense lines in the spectrum are coloured red.

Figure 1 shows the relative gamma ray intensities of over 1000 separate gamma-ray lines that result from our thermal neutron capture model of a "typical" crystalline rock. This spectrum forms the input to the GEANT4 computer simulation. Immediately we can see the full complexity of the initial spectrum and the fact that many spectral lines are not well separated or indeed, overlap even with 10 keV resolution in the plot. Given that all of the scintillators have a far worse energy resolution that in figure 1, it is clear that the degree of inter-element interference in each detector will be an important factor in determining their efficacy for PGNAA. The model was set up to simulate gamma-rays with energies randomly drawn from this input spectrum (which is the weighting function of the random variable – starting gamma ray energy) at each location (again randomly chosen in the Monte-Carlo process) in the formation. Gamma-rays are allowed to scatter and absorb throughout the volume and any subsequent energy deposition in the scintillator recorded. All relevant electromagnetic processes were simulated including Compton scattering, photoelectric absorption and pair production, the three most important processes. The Monte Carlo simulation ends when at least 100,000 gamma rays have deposited energy into the scintillator, requiring about 60M starting gamma rays. This provides a captured gamma spectrum in the scintillator that is reasonably statistically robust for analysis.

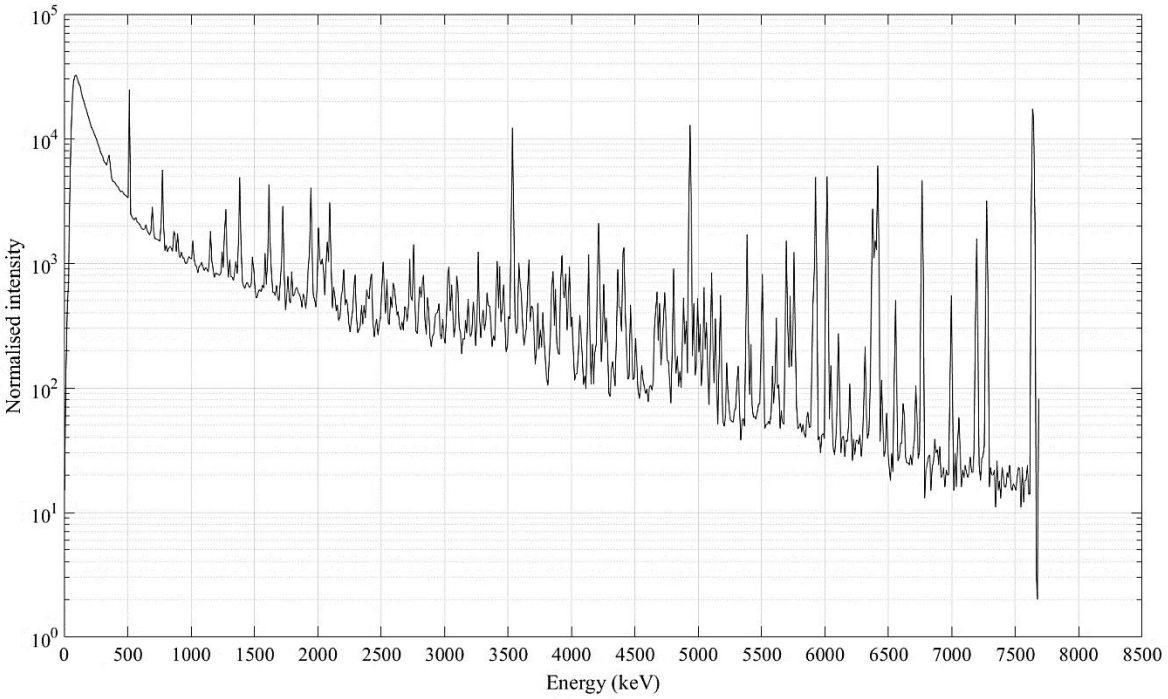

Figure 2: Spectrum of gamma-rays prior interaction with the scintillator.

The gamma spectrum resulting from the neutron gamma ray interactions with the rock prior to entering the scintillator is displayed in Figure 2. Figure 2 highlights another important aspect of the problem or elemental analysis via PGNAA: spectral peaks of the elements are still visible, but now a significant background is present. This background comes from source gamma-rays scattering and losing energy within the formation, and creates a challenge in low levels of detection, where the peaks will be often less than the background. Removing the background continuum by is possible; however, there will be a random residual due to statistical fluctuations in the scattering processes that add "noise" to any estimate of elemental concentration. Lower resolution detectors smear the peak signal over a larger range of energies, necessitating estimates with higher background fluctuation contribution. From figure 2 we would predict that if all other matters being equal (such as stopping power) a detector with higher resolution will produce more precise elemental abundance estimates.

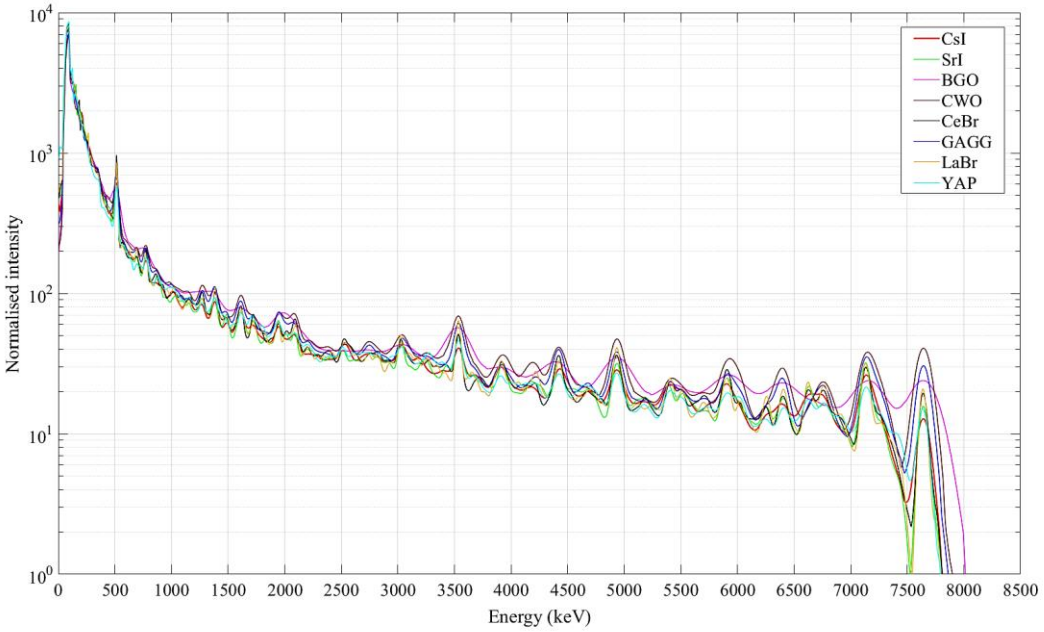

Figure 3: Typical PGNAA induced gamma-ray spectra from the simulated scintillators.

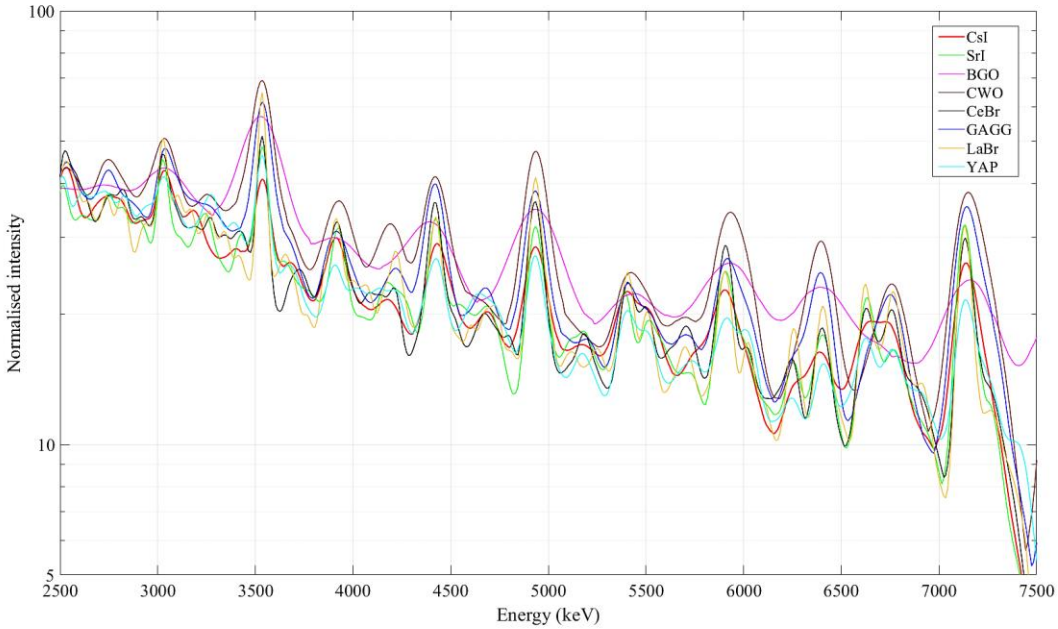

Figure 4: Gamma-ray spectra from simulated scintillators (enlarged image of area of interest).

Figure 3 shows the "recorded" energy spectra for each of the eight scintillators. Note that the spectra have been convolved with detector resolution functions constructed from the energy resolutions (calibrated at 662 keV) shown in Table 1. Over the range of 0.1 to 8 MeV the relative resolution of a scintillator is proportional to $1/\sqrt{(\text{Energy})}$. The same spectra are shown in Figure 4, with energy range limited from 2500keV to 7500keV, to show the differences better between scintillators.

Since there is no single measure that enables us to evaluate scintillation materials, we have developed a couple of different figure of merits (FoM) to evaluate and rank the scintillators. All calculations are made for the middle portion of the spectrum, from 2 to 5MeV, which we believe to the prime region for estimating elemental abundances, and presented in Table 2 (scintillators are presented in alphabetical order). The spectrum of gamma-rays prior interaction with the scintillator, shown on Figure 2, is the spectrum that would be obtained from a "perfect" scintillator. The perfect detector is then compared to the simulated and convolved detector responses in our analyses. Numbers in column one show how close (in percentage) the scintillation crystals are from the "ideal" scintillator in capturing and registering spectral peaks. The second column calculates a relative signal to noise (S/N) FoM by dividing the capture efficiency by the square root of the scintillators' resolution to provide an indication of the ability to estimate peak area accurately. The third column (Peak/Bckg FoM) is a notionally a peak to background ratio that also looks at the ability to estimate the peak area with a different methodology: areas of the spectrum are determined using calculations presented in Lépy, Pearce and Sima (2015) by analysing the three strongest, most isolated, peaks. Given the importance of inter-element interference for PGNAA application, we developed a FoM that estimates the liklihood of a an additional element peak located in a part of the spectrum free of interference (the last column in the table). While we cannot reliably identify for certain any peaks that are less than 3x standard deviation of count variations, a peak identification algorithm will report a result if there is at least 1x standard deviation from count fluctuations. Thus, the last column is the fraction of spectrum that is less than background plus one standard deviation of background count variations. In all cases a larger number indicates better performance.

Scintillator	Capture efficiency (%)	S/N FoM	Peak/Bckg FoM	Inter element interference (IEI) FoM
BGO	35.6	4.90	11.94	0.36
CeBr	15.3	8.01	9.65	0.74
CsI	13.7	4.64	6.57	0.69
CWO	28.7	7.89	14.68	0.51
GAGG	18.2	6.18	10.88	0.64
LaBr	12.7	7.97	10.60	0.75
SrI	12.6	6.14	8.12	0.76
YAP	5.6	2.46	5.75	0.83

Table 2: Figure of merits for scintillation crystals.

What table 2 shows is that for a scintillator with 50mm diameter density matters in capturing the gamma rays. Both BGO and CWO are clear leaders and YAP with relatively low density and low Z poorest (NaI is similar to YAP in this regard – terrible). The measures of peak detection ability with respect to S/N or peak-to-background (columns 2 and 3) are mixed: a few clear leaders with some differences in order due to one FoM estimate favouring density and the other resolution. Neither of these measures produce a clear winner and CsI and BGO are not terrible in comparison. Examination of figure 4 corroborates the numbers in that there are a number of scintillators that produce similar spectra recording reasonably similar detail. The last column is perhaps the most pertinent in the selection of "best", the ones with highest resolution. The FoM measures in table 2 are designed to provide a linear progression of good-better-best; thus, there is at best a three-to-one advantage in any one category, which would not make up an order of magnitude difference in elemental detection levels. So newer scintillators are better, with LaBr and CeBr appearing to have the best balance of resolution, density and lack of other issues associated with PGNAA. However, using conventional CsI in larger detectors should not degrade performance too badly.

CONCLUSIONS

None of the newer scintillators will make dramatic improvements in the detectability of most elements as there is a high background of scattered gamma rays in PGNAA. PGNAA is not a method that generally lends itself to trace elemental analysis. However, choosing a newer high resolution scintillator will greatly assist in resolving peak interference issues. Stopping power matters, but no combination of stopping power and resolution of the available materials produces a clear winner. Our modelling and analysis indicate that LaBr and CeBr are good candidates, and theoretically CWO is good for small diameters and lower count rate application. However, other issues such as pulse pile-up with a slower responding scintillators such as CWO and SrI mean that if price is not a factor then LaBr and CeBr are the best choices. If cost is especially important then one of the conventional scintillators such as CsI or high quality BGO would offer reasonable performance at much less cost.

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