

Elemental analysis of Kimberlite and associated Country Rock

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Abstract. The elemental analysis of Kimberlite and Country Rock was performed as part of an ongoing study into the Mineral-PET online rock sorting technique. This is a technology proposed to locate high densities of carbon in kimberlite and country rock using the very well known medical physics technique called Positron Emission Tomography (PET). Carbon in kimberlite is not a natural positron emitter; one has to convert the naturally occurring Carbon into a positron-emitting isotope. In this case it is done through the photo-nuclear transmutation reaction $^{12}\text{C}(\gamma, n)^{11}\text{C}$. We accomplish this reaction by irradiating the host rock using gamma rays with energies in the Giant Dipole Resonance (GDR) region. It is then important to determine the full inventory of radioisotopes produced in this process. The irradiation of Kimberlite was performed using the 100 MeV electron microtron at Aarhus University in Denmark. In this paper, we first describe the experiment, and then perform an analysis of the data. This provides quantitative identification of the prompt, short, mid-term and even long-term radioactivity of irradiated Kimberlite. This is necessary in order to assess more efficiently the radiation safety of the equipment and people working in the facility. A unique feature of this analysis is the unambiguous identification and quantification of each radioisotope formed by the two dimensional spectroscopy of the energy and the emission time of the signature gamma lines using time differential spectroscopy.

1. Introduction

The extraction of diamonds from excavated rocks has so far been a wasteful process using huge equipment requiring lot of energy. In a typical process, the rocks are first reduced into pieces of about 10 to 15 cm diameter and then ultimately crushed into pieces of some few millimetres. It is assumed the diamonds will be liberated or exposed at the surface. Different techniques are then used to extract them [1]. Only a small fraction of the rock contains any diamond making this method very inefficient. It would be very interesting if after the first crush, only diamond bearing rock could be processed further and this is where the Mineral-PET comes in. This method has its root in a the well known medical Positron Emission Tomography (PET) but rather than introducing the positron emitter in the patient, it is produced within the host rock by irradiating it with a photon beam with energies in the Giant Dipole Resonance (GDR) region [2, 3]. It is important to determine the full inventory of radioisotopes produced in this process. The irradiation was performed using the 100 MeV electron microtron at Aarhus University in Denmark. Time differential gamma-ray spectroscopy was used to identify the peaks of the spectrum. The possible primary reactions in the host rock are respectively the (γ, n) , (γ, p)

and (γ, α) . These products can interact with the host rock in many other secondaries. It is then very important to assess all the nuclides involved in the activation of kimberlite as the viability of the mineral-PET technique relies on this assessment. The time differential gamma-ray spectroscopy offers the advantage of allowing one to identify active isotopes based on both lifetime and energy, rather than only the energy, which is the only information available in normal gamma ray spectroscopy.

2. Experiments

The irradiation system was the Aarhus 100 MeV electron Microtron. This facility is equipped with an irradiation stage for the samples. The beam energy can be modified by the use of degraders. The electron current and irradiation time are measured. The electron and photon irradiation spectrum distribution is modelled with Monte Carlo transport calculations. The irradiation dose details can therefore be controlled, measured and modelled. In the example below the irradiation condition was 100MeV electrons incident on a 2 mm stainless steel exit port.

We irradiated a piece of Kimberlite rock, keeping the sample in position in front of the beam exit point. The irradiation time was 1000s for a dose of $5.25 * 10^{13} \text{electrons/cm}^2$. Note that this is about 17 times the MinPET dose[5]. After irradiation, a gamma spectroscopy system based on event-by-event acquisition implemented on a VME standard records a time-stamped energy data for each event. The data is then analyzed offline in a time differential manner, to provide both the gamma-line energy and lifetime. One detection run therefore yields complete information about both the energies and lifetimes of all isotopes present, allowing for unique isotope identification.

3. Data analysis and results

3.1. Method

The time-stamped energy data is collected for each event and a calibrated energy histogram is then plotted. In figure 1, one can easily distinguish the different peaks corresponding to diverse energy lines present in the spectrum. The graph in figure 1 presents many peaks with the most dominant ones being the 511 keV, 846 keV, 1271 keV, 1365 keV, 1432 keV, 1460 keV, 1730 keV, 2164 keV, 2241 keV and 2754 KeV. A time differential energy spectrum of the irradiated kimberlite shows that the height of the peaks varies with time. Figure 2 shows the dominance of the 511 peak, and also some very short-lived peaks vanishing before 1000s. One can also distinguish very long-lived peaks with the height remaining almost constant for few hours. The data is time sliced with different time bin sizes depending on the number of events needed to have a good histogram representing the different peaks. We then fit the peaks with an asymmetric gaussian and calculate the area of the peak for every time slice[4]. This gives us the peak height (area) as a function of the time. From the peak height for different time slices, one can then extract the lifetime of each energy line as a function of time. We fit these new data with an exponential decay. In some cases with interfering peaks, two components in the decay of the line needed to be included. The energy of the line and the half-life then gives an unambiguous isotope identification using a database of nuclide level schemes[6].

Figure 3 presents for different time-slice the histograms together with the asymmetric gaussian fit for the 511 keV peak. The histograms and the fits are one on the top of the other, remaining exactly at the position 511 keV . In figure 5, the peak tends to be moving which suggest the 845.8 KeV might be a combination of two very close lines with the centres at 844 keV and 846 keV.

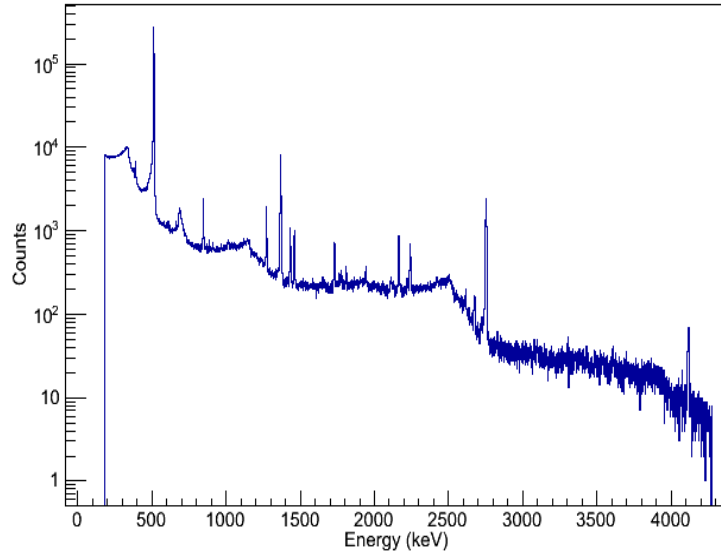


Figure 1. Calibrated energy spectrum with the dominance of the 511 KeV PET line

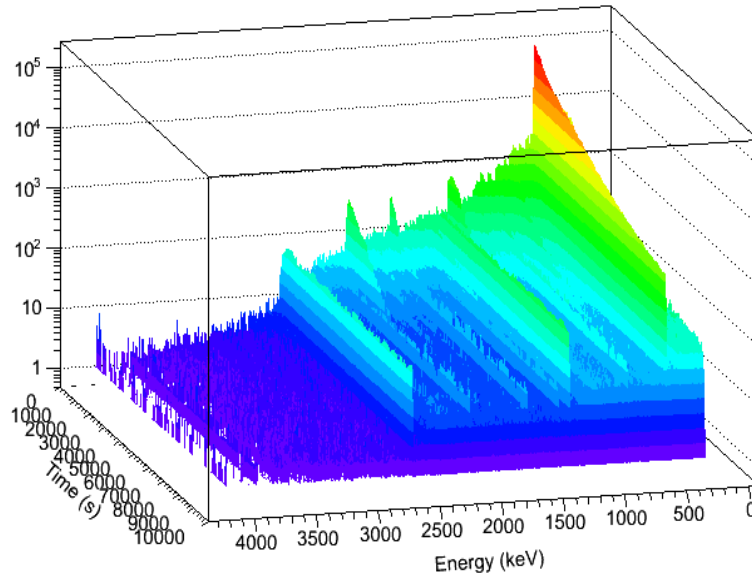


Figure 2. Time differential gamma spectroscopy for life-time determination for each gamma-line

3.2. Results and discussion

The single decay fit of the 845.8 keV line shown in figure 5 gives a lifetime of 62 minutes with some discrepancies in the early and the latter part of the spectrum. This suggest a two component fit of the data. For the long-lived component of the fit, we get candidates such as ^{87}Kr with lifetime of 76.3min 9s and ^{56}Mn with a lifetime of 2.58h. ^{87}Kr could come from neutron absorption on the natural ^{86}Kr but from the kimberlite composition, there is no ways Krypton could be present in the environment[7]. ^{56}Mn could either come from neutron capture on ^{55}Mn (abundance 100%) which is present in kimberlite or from (γ, p) reaction on ^{56}Fe (abundance 91.75%) which is also present in kimberlite. For the short-lived component, we fixed the long-lived one to ^{56}Mn and

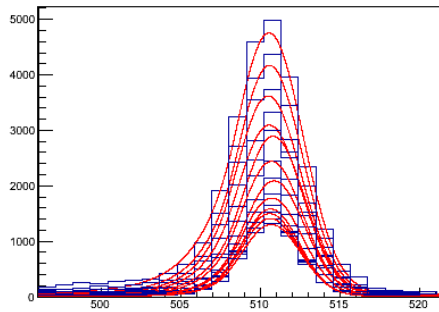


Figure 3. Single spectrum for the 511 KeV gamma-line for consecutive time slices.

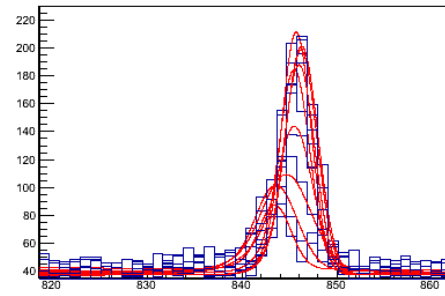


Figure 4. Single spectrum for the 845.8 KeV gamma-line for consecutive time slices.

extracted a lifetime of 10 min for the unknown short-lived isotope. The database points to ^{27}Mg which has a half-life 9.46 min. ^{27}Mg could come from neutron capture on stable ^{26}Mg or (n,p) reaction on ^{27}Al .

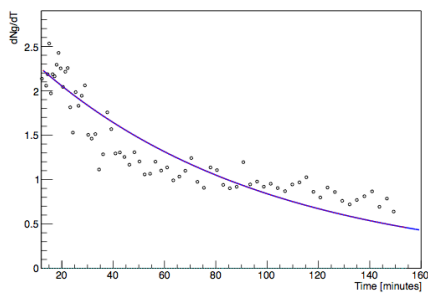


Figure 5. One decay best fit of the 845.8 KeV signal with the fit missing the data after 100 min.

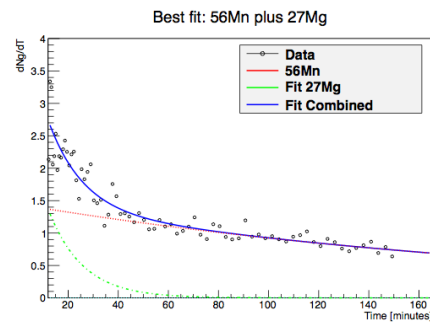


Figure 6. Lifetime spectrum of the 845.8 signal which is a sum of 2 decays: Magnesium and Manganese.

the graph on figure 6 gives a very good fit for the two components of a 845.8 KeV radiation (Magnesium and Manganese). similar procedure allows us to identify other lines and the results are presented in the table below.

Table 1. List of peaks with the parents, half-lives and possible processes leading to the isotopes or peaks.

Energy (keV)	Parents	Halflife	Process
511	^{11}C	20min23s	(γ, n) on ^{12}C
	^{15}O	2min2s	(γ, n) on ^{16}O
	^{13}N	10min	(γ, n) on ^{14}N
845.8	^{27}Mg	9min27s	n-capture on $^{26}\text{Mg}/(n, p)$ on ^{27}Al
	^{57}Mn	2h35min	(γ, p) on $^{56}\text{Fe}/n$ -capture on ^{55}Mn
1271	^{29}Al	6min40s	(γ, p) on ^{30}Si or (n, p) on ^{29}Si
1367	^{24}Na	15h	n-capture on $^{23}\text{Na}/(\gamma, p)$ on ^{25}Mg
1434	$^{52\text{m}}\text{Mn}$	21min7s	(p, n) on ^{52}Cr
1460	^{40}K	1,27 billion y	n-capture on ^{39}K
1776	^{29}Al	6min40s	(γ, p) on ^{30}Si or (n, p) on ^{29}Si
1809	^{56}Mn	2h35min	(γ, p) on ^{56}Fe or n-capture on ^{55}Mn
2164	^{38}K	7min56s	(γ, n) on ^{39}K
2422	^{29}Al	6min40s	(γ, p) on ^{30}Si or (n, p) on ^{29}Si
2754	^{24}Na	15h	n-capture on $^{23}\text{Na} / (\gamma, p)$ on ^{25}Mg

There are many other small peaks which are most probably noises from adjacent bigger ones. Some of these lines come from the very same isotope but at different energy or even from a process known in gamma-ray spectroscopy as single or double escape peak[8]. This is the case for the 1653 keV and 1143 keV respectively. We also have a peak at 2675 which comes from a single "break-in" (2164 keV collected simultaneously with a 511 keV gamma ray)

4. Conclusion

We irradiated a kimberlite sample using a 100 MeV electron beam to produce a bremsstrahlung radiation. Note that this energy is higher than the MinPET energy, however this experiment gave us a good idea of the different radioisotopes produced during the activation process of the MinPET. We were able to unambiguously identify the contributors to the dominant gamma lines together with their lifetimes. We could not make a conclusive identification for some of the peaks because they were swallowed in noise. This is also the reason our lifetime spectra are taken after 10 min. We also took into consideration the time taken to get the sample to the detectors and the initial period where the detectors are swamped. The use of the lifetime data can be seen here as a revolution in gamma-ray spectroscopy because without these data, many identifications would not have been possible. The next step in this analysis will be to identify all the remaining gamma lines, and based on the chemical composition of kimberlite use a software like say FISPACT to inventory all the nuclear reactions that led to the production of these radioisotopes.

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