

Experiments on a Possible γ -Ray Emission Caused by a Chemical Process

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Abstract — We report here results from Geiger-Mueller and NaI(Tl) γ -ray counting of a mixture of several chemicals before and after burning compared with a control sample of KNO_3 . The experimental results did not show non-trivial increase in γ -emissions after burning of the mixture.

Introduction

Enhancement in the low-energy end of the γ -ray spectrum incited by non-nuclear processes is increasingly being reported. Probably one of the first to report such non-trivial low energy γ -ray emissions was Matsumoto (1990). Unfortunately, in addition to the experimental results he has presented an obviously untenable theoretical explanation of three-body collision of deuterons and protons. He has also misinterpreted the $^{208}_{81}\text{Ti}$ (2.615 MeV) peak, interpreting it incorrectly as a $^{208}_{83}\text{Bi}$ (2.615 MeV) peak. Other groups also report low-energy (Karabut *et al.*, 1992; Scott *et al.*, 1990; Takahashi, 1990) and high-energy (Prelas *et al.*, 1990) γ -ray emissions induced in an unexpected way. Recently Bockris and collaborators reported unexpected effects during burning of a mixture of chemicals. Bockris and colleagues' reports caused a certain controversy (cf. Bishop, 1993) and some further scientific inquiry into the problem is warranted. In this paper we present some experimental results with regard to the claimed increased γ -ray emission as a results of a chemical process. No experiments were carried out concerning other claimed effects (Bishop, 1993; Sundarasan & Bockris, 1994) in this system.

Experiments

The experiment was carried out by mixing of 300 g C, 900 g KNO_3 , 80 g S, 120 g SiO_2 , 100 g FeSO_4 , 30 g Cd, 100 g Hg_2Cl_2 , 50 g PbO , 5 g Ag and 30 g CaO . The resulting mixture was burned in a hood. We performed five burns, four of which were successfully completed (burn #2 contained approximately one-half the amount of KNO_3 , resulting in very slow burning and was discarded). The burning was vigorous and lasted from about 30 s (burn #1) to about 3 min (burn #3). After burning was complete and the sample was cooled down, random samples were taken from the bulk and were ground in a mortar with a

pestle. The initial experiments were carried out by transferring the sample into a crucible. To ensure that the configuration (area, thickness, and geometry) of sample viewed by the detector was the same for all samples, the same size crucible was used, and care was taken to level each sample in the same way. The crucible was then inserted under the active window (–1 cm diameter) of a Radalert Geiger-Mueller counter. The Radalert was connected to a laptop IBM PC, where a special program was written to monitor the counts every minute throughout many hours. In the initial experiments the Geiger-Mueller counter was surrounded by a 0.2 cm lead shield. Later, in addition to the Geiger-Mueller counter a NaI(Tl) scintillation detector (3 cm x 3 cm) was set up. The sample, prepared according to the above procedure, was transferred into a watch-glass, levelled using a piece of glass and inserted into the gap below a NaI(Tl) scintillation detector. The configuration (area and thickness geometry) of the sample in this case was also the same for all samples used; the watch-glass was the same throughout. The detector and the sample were surrounded by 5 cm thick lead bricks. The detector was connected to a Model 35 Canberra Multichannel Spectrum Analyzer. In all experiments measures were taken to maintain the same counting geometries. The obtained spectra were dumped through the serial port of an IBM PC, where they were observed and analyzed using MathCad 5.0. Besides the natural calibration of the NaI(Tl) detector system by the prominent $^{40}_{19}\text{K}$ line appearing at 1.460 MeV in the background and the 0.511 MeV e^+e^- annihilation γ -ray, we also calibrated the system using $^{60}_{27}\text{Co}$ (1.33 MeV and 1.17 MeV) and $^{22}_{11}\text{Na}$ (0.511 and 1.274 MeV) sources. Although a Ge(Li) detector would have given better resolution, we believe that the NaI(Tl) detector available in our laboratory should be considered capable of giving reliable results.

Experimental Results

The typical Geiger-Mueller count rate per minute of the sample before burning was 19.07 ± 0.86 over a 24-h period. This was clearly lower than that after burning which was 24.64 ± 0.43 . The control sample, KNO_3 , exhibited 22.85 ± 0.32 counts per minute. It is to be noted that these determinations were carried out on random days during February and March, 1994, thus influences of cosmic events on the results were expected to be excluded. The increase in counting rate after burning provoked our interest and we continued our studies with a NaI(Tl) detector.

In Figure 1 we show the background taken for 12 hours starting on March 13, 1994 at 11:32 am (curve 1). In the background trace the prominent $^{40}_{19}\text{K}$ peak at 1.460 MeV, the e^+e^- annihilation peak at 0.511 MeV, the $^{214}_{83}\text{Bi}$ at 1.12 MeV, the smaller peaks of $^{214}_{83}\text{Bi}$ at 1.764 MeV, $^{214}_{83}\text{Bi}$ at 2.117 MeV, and $^{208}_{81}\text{Tl}$ at 2.615 MeV are observed. In Figure 1 the γ -ray spectrum from the sample before burning (curve 2), after burning (curve 3), and the control KNO_3 sample (curve 4) are also shown.

Figure 1 shows that the background counts at all energies are lower than the

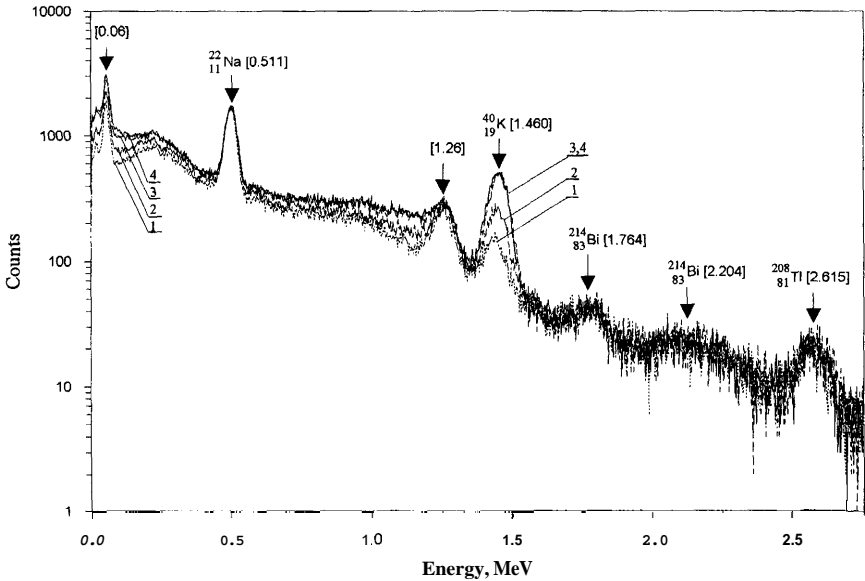


Fig. 1. The γ -ray spectrum measured with a 3 cm x 3 cm NaI(Tl) detector. Curve 1 — background, curve 2 — mixture before burning, curve 3 — mixture after burning, curve 4 — pure KNO_3 .

counts of the studied samples. Even the mixture before burning (curve 2) shows higher counts vs. background, especially at energies corresponding to the ^{40}K peak and the low-energy γ -ray peak (0.06 MeV). This fact, however, is easily explainable by the higher concentration of ^{40}K in the mixture. As expected from the results of the preliminary Geiger-Mueller experiments, the curve corresponding to the sample after burning (curve 3) lies at still higher counts at almost all energies compared to background and the mixture before burning. Practically no change is observed for the e^+e^- γ -ray annihilation peak at 0.511 MeV and for energies higher than that of ^{40}K peak. The higher counts for the mixture after burning were confirmed during several independent trials. We believe that the obtained data (of which Figure 1 is only one example) contains a statistically significant number of counts for this difference to be considered real.

Discussion

In order to explain the above increase in the γ -ray emission after burning of the sample, we carried out some additional experiments. For clarification, standard isotope data references were used to determine if any significant

decay of the observed γ -ray peaks would occur (Walker *et al.*, 1989). The NaI(Tl) detector was mounted vertically inside the lead-brick housing and a weighed amount of the burned sample #5 (several hours after the burn) was placed directly on top of the detector. The sample was monitored during the course of one month. No decay was observed during the period of study which indicates that no radioactive isotopes of short lifetime were created during the burn of the sample.

Of all the elements initially present in our mixture K is the most important since it has a naturally occurring radioactive isotope ($^{40}_{19}\text{K}$) with a long half-life. Other elements that have naturally occurring radioactive isotopes are $^{199}_{80}\text{Hg}$ (16.9%), $^{204}_{82}\text{Pb}$ (1.4%), $^{207}_{82}\text{Pb}$ (22.1%), $^{107}_{47}\text{Ag}$ (51.8%), $^{109}_{47}\text{Ag}$ (48.1%), $^{111}_{48}\text{Cd}$ (12.8%, half-life 48.5 min) (Walker *et al.*, 1989). These elements have isomeric transitions (states) which usually decay through γ -emissions but $^{107}_{47}\text{Ag}$ and $^{109}_{47}\text{Ag}$ decay by β^- . The half-life of $^{199}_{80}\text{Hg}$ is 42.6 min, $^{207}_{82}\text{Pb}$ is 0.8 s, $^{204}_{82}\text{Pb}$ is 1.12 hours, $^{107}_{47}\text{Ag}$ is 44.2 s, $^{109}_{47}\text{Ag}$ is 39.8 s (Walker *et al.*, 1989). Isotopes with even numbers of neutrons may decay through β^- to $^{17}_8\text{O}$ and neutrons (delayed neutrons) are emitted with a half-life of 4.17 s. Thus, the short half-lives of the naturally occurring isotopes of elements other than $^{40}_{19}\text{K}$ cannot account for the observed effect. Therefore, we focused our attention on the concentration of K in the samples. As is already observed from Figure 1 an enhancement of the intensities of all peaks is seen after burning of the mixture. This, however, is a trivial effect due to concentrating the amount of available K (resp. $^{40}_{19}\text{K}$). Increase of K concentration was independently confirmed by atomic absorption analysis using Perkin-Elmer 3100 Atomic Absorption Spectrometer (AAS). One gram of the mixture before and after burning was dissolved in 50 ml HNO_3 , and the solution was diluted to 1 l. Of these, 5 ml were diluted to 50 ml, and the absorbances of the diluted solutions were determined using AAS with a K lamp at $\lambda=766.5$ nm. For our purposes it was sufficient to just compare the AAS absorbance signals rather than to determine the exact concentrations of the two studied samples. Since, as mentioned, it was 1 g of each sample that was used for the AAS determinations, a difference in density of the two studied samples might have resulted in differences in K concentrations due to differences in volumes of samples. The increase in K concentration was not due to differences in sample density, because the density of the burned sample was at least of the same order of magnitude as that of the mixture before burning. We have noticed, however, that during weighing of the burned sample constant weight was not able to be reached, because of a certain hygroscopicity of the burned sample. Obviously, the mentioned difficulty due to hygroscopicity even further enhances the conclusion that the increased γ -ray emission signal is due to increased K concentration in the burned sample.

In another experiment designed to explain the above effect, the γ -ray spectrum of pure KNO_3 was taken under the same conditions as used for previous experiments. Figure 1 (curve 4) shows the γ -rays from this experiment. Comparison of curve 3 and 4 shows that there is practically no difference between

these curves. It was also determined that the density of the ash was slightly lower than the density of the KNO₃, by weighing equal volumes of these substances on a five-place electronic balance. It was determined by AAS that the concentration of K was practically the same both in the ash and in the pure KNO₃.

Conclusions

The above experimental results provide no basis to conclude that any unexpected effects, such as appearance of radioactive isotopes not present initially, are occurring due to a chemical process (burning of the sample).

Although we did not observe non-trivial effects during the present studies there are several conclusions which we were able to draw. First, we now understand that the results are very reproducible. The methods we apply are sensitive enough for the purpose and we would have been able to observe effects should there have been such. Secondly, we do not exclude the possibility that we have overlooked some hidden effects since we were looking for the broad picture of the events. Also, there may be some intricacies, unknown to us, for preparation of the samples which may cause positive results to appear. We also note that such claimed effects should be resolved only experimentally and no theoretical arguments should supercede or prevent the carrying out of experiments. Theoretical arguments should in general be used as mere guidelines and should be suspended once effects contrary to theoretical expectations are observed. This is not unusual in science. In this respect even experiments such as the above, the negative outcome of which can be predicted based on contemporary knowledge, are worth carrying out because the boundaries of those parts of contemporary knowledge that constitute absolute truths are not always clear cut.

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Response to Noninski *et al.*: Observation of β Radiation Decay in Low Energy Nuclear Reaction

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A decay of β^- radiation from burning a chemical mixture has been observed experimentally. The chemical burn was performed on April 14, 1992. The chemical composition of the mixture was as follows:

C	100 g	(Johnson Matthey, 300 mesh, 99.5%)
KNO ₃	300 g	(Baker, 99.2%)
S	30 g	(Spectrum)
SiO ₂	30 g	(EM Science, 60-200 mesh)
FeS	40 g	(Chempure)
Hg ₂ Cl ₂	10 g	(Fisher, 99.98%)
PbO	10 g	(Johnson Matthey, 99.99%)

The chemicals were weighed separately, and then mixed together. The mixture was ignited in a hood. The burn lasts about 60 sec. The whole product was ground in a mortar with a pestle after cooling down. Samples from both the original mixture and the product were taken and examined.

The β emission of the samples was detected by a β detector (Model 44-1, Ludlum Measurements, Inc.), and recorded by a multichannel pulse height analyzer (Model 800, The Nucleus, Inc.) and a data system (Model ZVM-123A, Zenith). The β detector was covered by a black box to shield the room light.

The β counts of the product was higher than that of raw material, about 1.4 times greater. The β counts of the raw material were stable. On the other hand, β radiation counting decay of the product was observed. The β count of the product (background counts subtracted) as a function of time is shown in figure 1. An exponential β decay was observed, and a half-life time of 17.7 hours was calculated.

Several experiments were performed. Not every experiment gave the same results, i.e., conditions for the reproduction of the experiment are not yet well defined.

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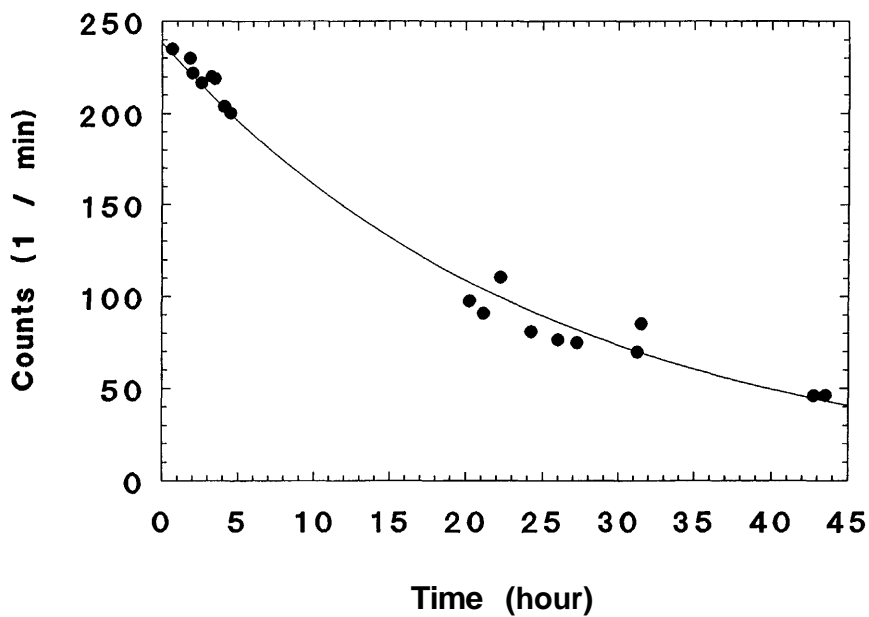


Fig. 1. β counts as a function of time.