Do pyrotechnics contain radium?

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Received 23 April 2009
Accepted for publication 31 July 2009
Published 24 August 2009
Online at stacks.iop.org/ERL/4/034006

Abstract

Many pyrotechnic devices contain barium nitrate which is used as an oxidizer and colouring agent primarily for green-coloured fireworks. Similarly, strontium nitrate is used for red-coloured pyrotechnic effects. Due to their chemical similarities to radium, barium and strontium ores can accumulate radium, causing a remarkable activity in these minerals. Radium in such contaminated raw materials can be processed together with the barium or strontium, unless extensive purification of the ores was undertaken. For example, the utilization of ‘radiobarite’ for the production of pyrotechnic ingredients can therefore cause atmospheric pollution with radium aerosols when the firework is displayed, resulting in negative health effects upon inhalation of these aerosols. In this study, we investigated the occurrence of gamma-photon-emitting radionuclides in several pyrotechnic devices. The highest specific activities were due to K-40 (up to 20 Bq g\(^{-1}\), average value 14 Bq g\(^{-1}\)). Radium-226 activities were in the range of 16–260 mBq g\(^{-1}\) (average value 81 mBq g\(^{-1}\)). Since no uranium was found in any of the samples, indeed, a slight enrichment of Ra-226 in coloured pyrotechnics can be observed. Radioactive impurities stemming from the Th-232 decay chain were found in many samples as well. In the course of novel developments aiming at the ‘greening’ of pyrotechnics, the potential radioactive hazard should be considered as well.

Keywords: fireworks, inhalation, natural radioactivity, \(^{238}\)U decay chain, \(^{226}\)Ra, \(^{228}\)Ra

1. Introduction

Fireworks are probably the application of chemistry with the best resonance with the general public. Nonetheless, fireworks are increasingly raising environmental concerns. Although the problem of pollution caused by fireworks (and other civil and military pyrotechnic applications) had been identified many years ago [1, 2], the number of environmental studies focusing on this problem has dramatically increased quite recently, e.g. [3–11]. Also the search for environmentally benign pyrotechnic formulations exhibits a rapidly expanding scientific field and has not hit its peak yet [5].

Pyrotechnics are thermodynamically metastable mixtures which consist of at least two basic constituents: the reductant/fuel (e.g. magnesium, aluminium, magnalium alloy, sulfur, charcoal, red phosphorus, etc) and the oxidizer (alkali metal or alkaline earth metal nitrates, perchlorates, chromates, metal oxides, etc). Several additives may find application in pyrotechnics in order to obtain a certain intended effect (e.g. colouring agents, propellants, smoke or sound generators, etc). Colours in pyrotechnics are obtained by the addition of compounds of elements with the desired flame colour. For red light, strontium nitrate is used; barium nitrate for green light; sodium oxalate or cryolithe (Na\(_3\)AlF\(_6\)) for yellow; and any copper/chlorine system (compounds or mixtures) for blue (see table 1 for some typical compositions of pyrotechnics). During combustion, very short-lived and unstable compounds, such as the monochlorides of alkaline earth metals (SrCl\(_2\), BaCl\(_2\)) are formed, which emit light in the desired spectra [5, 12, 13]. The formation of the monochlorides thus depends on the presence of a chlorine source. If no chlorine donor is added to a pyrotechnic formulation, barium nitrate causes combustion under the emission of almost white light. This is the reason why barium nitrate is not only used as an oxidizer in green or greenish flares (with a chlorine donor, which is typically PVC powder) but also for white and yellow flares (without a chlorine donor), as shown in table 1. In the presence of chlorine, barium nitrate acts as a combined pyrotechnic oxidizer and colouring agent.
From an environmental and toxicological point of view, the formation of barium-rich aerosols following the display of a firework is a problem. The inhalation of barium-rich aerosols has adverse affects on the lungs and heart and causes muscle cramps [14, 15]. In cases of fireworks and pyrotechnics, barium compounds are set free in the form of mostly water-soluble and thus bioavailable compounds: BaO, BaCl₂ and undecomposed Ba(NO₃)₂. The raw material of barium compounds is generally barium sulfate (barite). In varnish, rubber, etc. This mineral is also used as a constituent of petroleum and ores have remarkable 226Ra activities. On geological timescales, however, 226Ra has a relatively short half-life. If radiobarite minerals, therefore, are older than 10–20 ka and isolated from any further radium supply, they slowly lose their radioactive properties. The radiobarite-rich sludges and scalings at oil-field-production sites investigated by Zielinski et al. [17], have 226Ra activities in the range between 3 and 130 Bq g⁻¹, with one sample as active as 4.9 kBq g⁻¹. In their study, the 226Ra activities have been found to be always lower than the 228Ra values. Radiobarite ores in the Ohře Rift (Bohemian Massif) have activities between 0.02 and 7.80 Bq g⁻¹ [18]. The scales and tailings in Polish hard coal mining sites were reported to contain radiobarites with activities in the range of 40–100 Bq g⁻¹ for 226Ra and 27–62 Bq g⁻¹ for 228Ra (barium-rich Romotk scale), and 5.3–6.4 Bq g⁻¹ for 226Ra and 6.4–8.5 Bq g⁻¹ for 228Ra (barium-poor Bojszowy tailings), respectively [19]. The ambient γ-dose rates are strongly elevated with more than 1 μSv h⁻¹ at both sites.

Previous studies [20, 21] have investigated the trace element content of pyrotechnics and their poisoning potential. From an economic point of view, it is clear that raw materials for the production of fireworks are usually not purified beyond the grade which is necessary for the intended effects. This explains why the fireworks investigated in those studies contained significant traces of heavy metals which do not have a pyrotechnic function. The utilization of radium-rich barium and strontium ores would, therefore, involve the risk that radium might be processed together with barium and strontium into the final product. The display of such radium-containing pyrotechnics would set the radioactive material free in the form of easily inhalable aerosols. The incorporation of α-emitting radionuclides (such as 226Ra) is a major health threat in human radiation protection. The ingestion or inhalation of α-emitters should thus be avoided under all circumstances. In order to examine this potential hazard, we applied radioanalytical methods to investigate the radioactivity of pyrotechnics purchasable in Austria.

### 2. Materials and methods

Fourteen samples of pyrotechnic devices (sky rockets, shell-type rockets, volcanoes) have been investigated with γ-spectrometry in this study (see table 2). The samples were weighed and filled into cylindrical polyethylene (PE) containers (comparable filling level). In principle, for the quantification of the 226Ra activity, two methods are possible: the 186 keV γ-photon emitted by the nuclide itself can be measured. Alternatively, the γ-photons of its decay products 214Pb and 214Bi can be measured, as they are in equilibrium with 226Ra after three or four weeks (due to the short half-lives of 214Pb and 214Bi as well as the intermediate 226Ra-daughter.

**Table 1.** Some typical barium nitrate-or strontium nitrate-containing pyrotechnic compositions (data taken from [5, 12, 27]). Values in wt%.

| Ingredient         | Mk 117 green navy flare | Mk 118 yellow navy flare | Turquoise formulation | Chartreuse formulation | White formulation | Mk 124 red navy flare | Red highway flare |
|--------------------|-------------------------|--------------------------|-----------------------|------------------------|------------------|-----------------------|-------------------|
| Barium nitrate     | 22.5                    | 20.0                     | 75                    | 75                     | 55               | —                     | —                 |
| Strontium nitrate  | —                       | —                        | —                     | —                      | —                | —                     | —                 |
| Magnesium          | 21.0                    | 30.3                     | —                     | —                      | —                | 24.4                  | 74                |
| Potassium perchlorate | 32.5                  | 21.0                     | —                     | —                      | —                | 20.5                  | 6                 |
| Sodium nitrate     | —                       | —                        | —                     | —                      | —                | —                     | —                 |
| Potassium nitrate  | —                       | —                        | —                     | —                      | —                | —                     | —                 |
| PVC                | 12.0                    | —                        | 5                     | 10                     | —                | —                     | —                 |
| Sodium oxalate     | —                       | 19.8                     | —                     | —                      | —                | —                     | —                 |
| Copper powder      | 7.0                     | —                        | —                     | —                      | —                | —                     | —                 |
| Asphaltum          | —                       | 3.9                      | —                     | —                      | —                | 9.0                   | —                 |
| Sulfur             | —                       | —                        | 10                    | 10                     | 20               | —                     | 10                |
| Cuprous chloride   | —                       | —                        | 10                    | —                      | —                | —                     | —                 |
| Binder             | 5.0                     | 5.0                      | —                     | —                      | —                | —                     | 10                |

G Steinhauser and A Musilek

Environ. Res. Lett. 4 (2009) 034006
nuclides $^{222}\text{Rn}$ and $^{218}\text{Po}$, respectively), see scheme 1. Since radon is known to diffuse through many materials (sample vials), causing a loss of activity, the latter method appears to be the less reliable for our analytical purposes.

When the 186 keV $\gamma$ peak of $^{226}\text{Ra}$ is used, the possible interference of $^{235}\text{U}$, which also emits $\gamma$ photons in this energy region, has to be considered. However, since we can assume that uranium in environmental samples must be present in its natural isotopic ratio, a $\gamma$ spectrum showing a $^{235}\text{U}$ peak should also show the $\gamma$ peaks of the short-lived $^{238}\text{U}$ granddaughter $^{234}\text{Pa}$ (with several $\gamma$ photons at 1001, 743, 786 keV, etc), as shown in scheme 1. Since we did not detect any $^{234}\text{Pa}$ in our samples (detection limit approx. 20 mBq g$^{-1}$), the uranium content in pyrotechnics can be regarded as negligible. Consequently, any radium in the sample cannot be due to a contamination with uranium minerals being in equilibrium with the daughter $^{226}\text{Ra}$. Rather, it must be a significant enrichment of radium itself in one of the raw materials.

Gamma-spectrometry was performed on the novel low-level counting facility of the Atominstitut, consisting of a 226 cm$^3$ HPGe detector (Canberra™, detector model GC5020; 2.0 keV resolution at the 1332 keV $^{60}\text{Co}$ peak; 52.8% relative efficiency), connected to a PC-based multi-channel analyser with preloaded filter. The measurement position of the sample was fixed at a distance of approximately 11 cm on top of the detector. The new detector system is characterized by only approximately one-tenth of the background of the other $\gamma$ detectors of the radiochemistry group in the same institute. This is due to the improved shielding of the detector by the ORTEC™ HBLBS1 shielding (solid-cast virgin lead with steel casing, total weight 1134 kg). For calibration of the detector’s efficiency for $^{226}\text{Ra}$, 50 $\mu$l of QCY48 (Amersham® Ltd) solution in hydrochloric acidic solution (comparable bulk density) was used. The measurement times of the pyrotechnics were at least 1 week, or longer, until no significant improvement of the counting error of the most interesting peaks could be yielded by a—reasonably—longer measurement time. The standard solution was measured for 328 000 s. A background spectrum was recorded (1 816 000 s) and considered for the evaluation of the $\gamma$ spectra of the pyrotechnics. For quantification, the $\gamma$ photons with characteristic energies were used as listed in table 3. All nuclear data in this paper are taken from the National Nuclear Data Center [22].

### Table 2. Samples investigated in this study.

| Sample code | Sample name       | Pyrotechnics type | Potential radium carrier |
|-------------|-------------------|-------------------|--------------------------|
| R1          | Weco green glamour| Sky rocket        | Barium                   |
| R2          | Weco green glamour| Sky rocket        | Barium                   |
| R3          | Weco green flower | Sky rocket        | Barium                   |
| R4          | Weco red glamour  | Sky rocket        | Strontium                |
| R5          | Weco red glamour  | Sky rocket        | Strontium                |
| R6          | Weco red flower   | Sky rocket        | Strontium                |
| R7          | Weco pink flower  | Sky rocket        | Strontium                |
| R8          | Weco yellow flower| Sky rocket        | Barium                   |
| R9          | Weco white glamour| Sky rocket        | Barium                   |
| R10         | Wolm Pyrostar Kugelblitz | Shell-type rocket | Barium and/or strontium |
| R11         | Wolm Pyrostar Kugelblitz | Shell-type rocket | Barium and/or strontium |
| R12         | Wolm Pyrostar Kugelblitz | Shell-type rocket | Barium and/or strontium |
| R13         | Weco Riesen Flimmer-Vulkan | Volcano         | Barium and/or strontium |
| R14         | Weco Fegefeuer    | Volcano           | Barium and/or strontium |

3. Results and discussion

The results of the $\gamma$ spectrometry are shown in table 4.

The main activity in pyrotechnics is due to $^{40}\text{K}$ (up to 20 Bq g$^{-1}$, mean value 14 Bq g$^{-1}$). The presence of $^{40}\text{K}$...
except for 40K (Bq g\(^{-1}\)) luminescent pyrotechnics, where we actually expected much lower activities due to the less obvious chemical similarities of strontium (red colourant) and radium. However, it seems that both alkaline earth ore deposits, strontium and barium, respectively, similarly accumulate radium from the geological environment.

Comparing the 214\(^{\text{Pb}}\) activities to the respective 214\(^{\text{Bi}}\) activities shows very good agreement. This observation comes as expected, because the short-lived nuclide 214\(^{\text{Bi}}\) must be in radioactive equilibrium with its mother 214\(^{\text{Pb}}\). Since the principal \(\gamma\) photons of both nuclides have their energy in different regions of the spectrum (352 keV for 214\(^{\text{Pb}}\) and 609 keV for 214\(^{\text{Bi}}\)), this observation leads to the conclusion that our simplified approach of 'comparable bulk densities' of samples and QCY48 standard solution is a valid approach for these measurements. If the \(\gamma\)-photon self-absorption properties of sample and standard were significantly different, this would have led to a deviation in the evaluation of \(\gamma\) photons with different energies (and thus different detector efficiencies). Some occasionally higher deviations are due to higher counting errors.

The activities of 214\(^{\text{Pb}}\) and 214\(^{\text{Bi}}\) were in the same range (at least in the same order of magnitude) as the 226Ra activities. Remarkably, the 226Ra activities were a little higher than the 226Ra activities. In such environmental samples, 228Ac is in secular radioactive equilibrium with its longer-lived mother nuclide 226Ra (a poor \(\gamma\) emitter). The 228Ac activities thus correspond directly to the 226Ra activities.

Table 3. Nuclear data of the radionuclides measured by \(\gamma\) spectrometry.

| Decay chain member | 232Th | 238U | 238U | 238U | 232Th |
|--------------------|-------|------|------|------|-------|
| Half-life          | 1.00  | 7.51 | 7.51 | 7.51 | 1.00  |
| Principal \(\gamma\)-photon energy (keV) | 2288.4 | 2288.4 | 2288.4 | 2288.4 | 2288.4 |
| \(\gamma\)-photon yield (%) | 10.66 | 10.66 | 10.66 | 10.66 | 10.66 |
| 214Pb activities | 120 mBq g\(^{-1}\) | 120 mBq g\(^{-1}\) | 120 mBq g\(^{-1}\) | 120 mBq g\(^{-1}\) | 120 mBq g\(^{-1}\) |
| 214Bi activities | 120 mBq g\(^{-1}\) | 120 mBq g\(^{-1}\) | 120 mBq g\(^{-1}\) | 120 mBq g\(^{-1}\) | 120 mBq g\(^{-1}\) |

Table 4. Results of the \(\gamma\)-spectrometric measurement of commercially available pyrotechnics. Specific activities are given in mBq g\(^{-1}\), except for 40K (Bq g\(^{-1}\)). Errors are due to counting statistics and the efficiency curve error and are given in % relative. 'n.d.' stands for 'not determined'.

| Sample (colour) | \(40\text{K}\) (Bq g\(^{-1}\)) | Error (mBq g\(^{-1}\)) | Error (mBq g\(^{-1}\)) | Error (mBq g\(^{-1}\)) | Error (mBq g\(^{-1}\)) | Error (mBq g\(^{-1}\)) | Error (mBq g\(^{-1}\)) |
|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| R1 (green)      | 11.6            | 1.6             | 35              | 9.2              | 77              | 2.1             | 79              | 2.1             | 110             | 7.0             | 29              | 21              |
| R2 (green)      | 11.8            | 1.7             | 32              | 4.3              | 48              | 3.4             | 54              | 8.5             | 87              | 43              | 32              | 9.2              |
| R3 (green)      | 17.5            | 1.7             | 33              | 12               | 48              | 5.1             | 41              | 5.4             | 16              | 30              | 37              | 14              |
| R4 (red)        | 12.4            | 1.7             | n.d.            | 46               | 3.8             | 48              | 3.7             | 97              | 12              | 51              | 70              |                  |
| R5 (red)        | 11.8            | 1.8             | 48              | 3.5              | 48              | 11              | 41              | 13              | 92              | 63              | 43              | 8.1              |
| R6 (red)        | 17.5            | 1.8             | n.d.            | 36               | 17              | 40              | 5.2             | <50             | 38              | 13              |                  |                  |
| R7 (pink)       | 15.8            | 1.7             | 48              | 18               | 39              | 5.5             | 45              | 5.1             | 120             | 47              | 41              | 40              |
| R8 (yellow)     | 12.9            | 1.8             | 45              | 15               | 54              | 12              | 55              | 13              | 120             | 21              | 37              | 38              |
| R9 (white)      | 11.3            | 1.8             | 32              | 4.6              | 49              | 3.6             | 50              | 3.5             | 73              | 14              | 20              | 16              |
| R10 (multi-coloured) | 15.9         | 1.7             | 11              | 34               | 28              | 20              | 22              | 18              | <40             | 12              | 74              |                  |
| R11 (multi-coloured) | 14.0        | 1.7             | 150             | 2.1              | 110             | 2.5             | 120             | 2.5              | 250             | 6.7             | 120             | 4.0              |
| R12 (multi-coloured) | 11.4        | 1.7             | 18              | 13               | 57              | 4.4             | 66              | 12              | 120             | 15              | <25             |                  |
| R13 (multi-coloured) | 19.3        | 1.7             | 25              | 23               | 26              | 4.8             | 21              | 5.4             | 42              | 69              | 18              | 16              |
| R14 (multi-coloured) | 9.23        | 1.7             | 3.9             | 46               | 11              | 5.4             | 9               | 23              | <20             | <10             |                  |                  |

\(^{a}\) In such environmental samples, 228Ac is in secular radioactive equilibrium with its longer-lived mother nuclide 226Ra (a poor \(\gamma\) emitter). The 228Ac activities thus correspond directly to the 226Ra activities.

in the mixtures can easily be explained by the application of oxidizing potassium salts (nitrate, perchlorate) in pyrotechnics. Black powder, for example, consists of some 75% of potassium nitrate; the increased 40K activity, therefore, did not come as a surprise.

Much more noteworthy is the presence of 226Ra (and/or its daughter nuclides 214Pb and 214Bi, respectively) in almost all pyrotechnics investigated. According to the hypothesis of this study, 226Ra in the samples must be the result of the utilization of slightly active radiobarite ores for the production of the pyrotechnic raw material barium nitrate. As stated in the introduction, the use of barium salts in pyrotechnics is not restricted to green luminescent formulations, as it may be applied as an oxidizer with 'neutral colour', if no chlorine donor is added to the mixture. Accordingly, this explains one of the highest specific radium activities (120 mBq g\(^{-1}\)) in sample R8—a rocket with a yellow effect.

The highest specific 226Ra activity was 260 mBq g\(^{-1}\) (sample R11), whereas only in three samples was the detection limit not exceeded. The average value of all 14 samples was 81 mBq g\(^{-1}\). Pyrotechnics thus exhibit a specific radium activity that is approximately one order of magnitude lower than the lowest active radiobarite sample investigated by Zielinski et al [17]. The specific radium activities of our samples, however, correspond to those found in the radiobarites of the Ohře Rift (Bohemian Massif). Some pyrotechnic samples exceeded the lowest active samples from the Ohře Rift (0.02 to 7.80 Bq g\(^{-1}\) radium) [18].

We could also find similar levels of 226Ra activity in red luminescent pyrotechnics, where we actually expected much lower activities due to the less obvious chemical similarities of strontium (red colourant) and radium. However, it seems that both alkaline earth ore deposits, strontium and barium, respectively, similarly accumulate radium from the geological environment.
presence of $^{228}$Ra (and its short-lived daughter nuclide $^{228}$Ac) in pyrotechnics, therefore, can hardly be the result of a natural, selective incorporation of $^{228}$Ra into the barium or strontium ore. Radium-228 ($T_{1/2} = 5.75$ a), without a supply from the mother nuclide $^{232}$Th, would decay completely after a few decades.

The specific activities of $^{228}$Ac and $^{212}$Pb correlate to a high degree, although, as with the $^{238}$U chain, a gaseous decay product lies in between, namely $^{220}$Rn. In this specific case, however, we believe that the much shorter half-life of the noble gas $^{220}$Rn ($T_{1/2} = 55.6$ s) does not allow a significant diffusion through the PE container. Consequently, $^{220}$Rn and all its decay products are trapped in the container and measured without any losses.

According to the Swiss Administration [24], 1700 tons of pyrotechnics are annually consumed in Switzerland. We can safely assume that these numbers compare to Austria as well. Assuming an average radium content in pyrotechnics of 81 mBq $^{226}$Ra g$^{-1}$ and 14 Bq $^{40}$K g$^{-1}$, these numbers will correspond to an annual emission of 138 MBq $^{226}$Ra and 23.8 GBq $^{40}$K year$^{-1}$ from pyrotechnics only (in countries like Switzerland or Austria).

4. Conclusions

Pyrotechnics do contain radium—not extreme activities but certainly enough to be detected in our new low-level $\gamma$ spectrometer. The $^{226}$Ra activities are in the range of 16–260 mBq g$^{-1}$ and must be due to the selective incorporation of radium into the barium and strontium ore body, as no traces of the mother nuclide $^{238}$U and its early daughters) could be found.

In contrast to this, $^{232}$Th and its decay chain members (like $^{228}$Ra) probably is present in the form of thorium-containing minerals, which are an impurity in the raw materials of pyrotechnics. Due to its much shorter half-life, $^{228}$Ra is unlikely to accumulate significantly in the ore body of an alkaline earth metal deposit. This hypothesis is further supported by the very weak or non-existing correlation of the activity concentrations of the two radium isotopes.

Although the radium activities were relatively low, we believe that this potential hazard should be considered as well in the development of environmentally friendly pyrotechnics [5]. The incidental utilization of highly active radiobarite ores as pyrotechnic raw materials would probably exhibit a greater health hazard than the toxicity of the poisonous pyrotechnic constituents (heavy metals, perchlorate) [5]. In particular, when applied as indoor pyrotechnics, the health aspects of pyrotechnic devices should be taken seriously with respect to air pollution by toxic and radioactive substances. This finding justifies the efforts that are currently being undertaken in the search for barium-free pyrotechnics [25]. One possible method to remove radium from the radiobarite ore, however, could be to set it under reducing conditions which causes the complete mobilization of radium from the barite [26].

Acknowledgments

GS thanks the Austrian Science Fund (FWF) for financial support (Erwin Schrödinger-Stipendium, project no. J2645-N17). We also thank the Vienna University of Technology for supporting this work.

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