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Holmium and Samarium Detectability in Post-Blast Residue

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Abstract. The creation of an identification taggant that survives detonation and can easily be recovered would prove invaluable in situations such as a terrorist attack where the explosives have been detonated and need to be traced by law enforcement. This paper shows that traces of two elements, samarium (Sm) and holmium (Ho), can be identified from explosive post-blast residue. This is the first step to creating a viable taggant system that survives detonation. The two elements were introduced to the explosive before detonation, and the post-blast residue was analyzed by neutron activation analysis (NAA). These data indicate the presence of these elements when they are added to explosives. Importantly, these experiments also show that these elements are not present when they are not added to the explosives as well. Combining NAA with the doped explosive could potentially lead to the development of an identification taggant that is suitable for widespread use in commercial explosives.

INTRODUCTION

Tracking explosives to minimize their potential misuse started, in the United States, in 1917 [1]. The concern at the time of implementing the Explosives Act of 1917 was the use of legally obtained explosives by saboteurs and what would be called terrorism today [1]. To address these concerns, improved methods of tracking them have been developed and investigated throughout the last 100 years [2,3]. Additionally, in this span of time, domestic explosive use has increased nearly 10 times. In 1917, the manufacturers in the United States produced just over 264,000 metric tons of explosives for use within the country [1]. By 2014, that number increased to 3.1 million metric tons of explosives consumed domestically [4]. This increased use of explosives also leads to increased access to explosives. Unfortunately, explosives represent a significant potential for misuse in, for example, terrorist actions. Preventing the misuse of explosives continues to be a major focus of regulatory agencies in the United States. One way to minimize the potential for misuse of explosives is to make it possible to track explosives by some means that cannot be separated from the explosive.

One approach that improves tracking of explosives is the introduction of "identification taggants" into the explosive itself. An identification taggant is anything added to an explosive that encodes information about the explosive, such as manufacturer, type of explosive, or batch. The creation of an identification taggant that survives detonation and can be recovered would prove useful in situations where the explosives have been detonated and need to be traced by law enforcement [2,3]. Detection taggants, on the other hand, are materials added to the explosive that makes the explosive easier to detect: by scent, sight, or other detection systems such as mass spectroscopy [5]. Detection taggants play a significant role in explosive safety and security. However, the NAA technique is intended for identification taggants and therefore this paper will focus on identification taggants only, throughout the remainder.

Taggants that allow identification of explosives up to the batch level of production can also be used in other than identification taggants. This taggant method can help manufacturers by allowing them to identify products more

specifically. This assists quality control operations by providing more detailed information by allowing manufacturers to compare batches in the case of performance issues. It also aids in reducing the cost of product recalls or defects by providing exact batch numbers that are affected by the problem that required the recall or caused the defect.

Previous taggant research has shown two areas to be potentially problematic: recovering the taggant from the post-blast environment, and surviving the detonation process [2]. A “survivable” taggant is one that is not significantly altered by the detonation process, and can therefore be recovered at the site of the detonation. Further, a “survivable” taggant must be able to have the information that is encoded in it read after detonation to be of use.

Initial efforts in creating identification taggants, inserting a material or chemical into an explosive to enable identification at a later time, date back to at least the early 1970s [6-9]. Research into the field has been mostly reactionary in response to major terrorist activity involving explosives [3]. In the event of a terrorist attack using explosives, taggants would make the job of the Bureau of Alcohol, Tobacco, Firearms, and Explosives (BATFE) easier [2,3]. One type of identification taggant, a plastic particle made from several colored layers that encode information based on the sequence, has been used in explosives commercially produced for the use in Switzerland since 1980 and is credited by officials there for aiding investigations into bombings [6]. The key to using this taggant is recovering the small plastic particles, which does not always happen in the post-blast environment [6].

The price of incorporating some type of taggant technology into commercial explosives is a key reason for industry opposition [2,10]. Research into the same particulate taggant that is now used in Switzerland was performed in the late 1970s, and the report *Taggants in Explosives* by the Office of Technology Assessment reproduces those results [2,3]. The conclusion was that the costs of implementing a taggant program in the United States were much higher than in Switzerland due to both the size of the explosives industries and the types of explosives used [2]. For taggants to gain commercial acceptability, a taggant scheme needs to be developed that reduces the cost of tagging the quantity of explosives used in the United States. An alternative taggant scheme based on the detectability of rare earths is currently under consideration.

In the research presented in this paper, samarium (Sm) and holmium (Ho) were added to a commercial binary explosive as taggants. Elements were added to the explosive since the high pressure and temperatures of detonation will not destroy elements, but will destroy almost all chemical compounds. Additionally, the elements chosen allow for the use of very low concentrations when combined with Neutron Activation Analysis (NAA), which makes removing the taggant before detonation difficult. As a first step towards developing a comprehensive taggant scheme for use with explosives, the objective of this research will investigate the partitioning of these taggant elements into the post-blast residue (1) and determine if they can be recovered and identified using NAA (2).

NEUTRON ACTIVATION ANALYSIS

Neutron Activation Analysis is a powerful technique that can identify both the elements present in a sample and the quantities of those elements at very low concentrations. NAA identifies the elements present and their masses, without providing information about their chemical states. As such, this technique is useful for an identification taggant since mass is unaffected by the detonation process. NAA is sensitive [11] and in optimal conditions can identify some elements at the 1 to 4 parts per billion (ppb) level [12]. While this level of precision is not demonstrated in these experiments, they represent the lower bounds for useful concentrations with this method of analysis.

Neutron Activation Analysis starts with collecting a sample and exposing it to a neutron flux. The elements within the sample interact with the neutron flux, and some of these elements will capture a neutron. Often, when a neutron is captured by a stable, naturally occurring, element, the nucleus becomes a radioactive isotope. These newly radioactive isotopes then decay according to their characteristic half-life. Additionally, the decay process produces various kinds of radiation. NAA works by measuring the gamma radiation produced by the decaying isotopes. In this experiment, a high purity germanium (HPGe) detector was used to count the gamma rays and discriminate them by energies. Figure 1, below, shows a schematic diagram of the process.

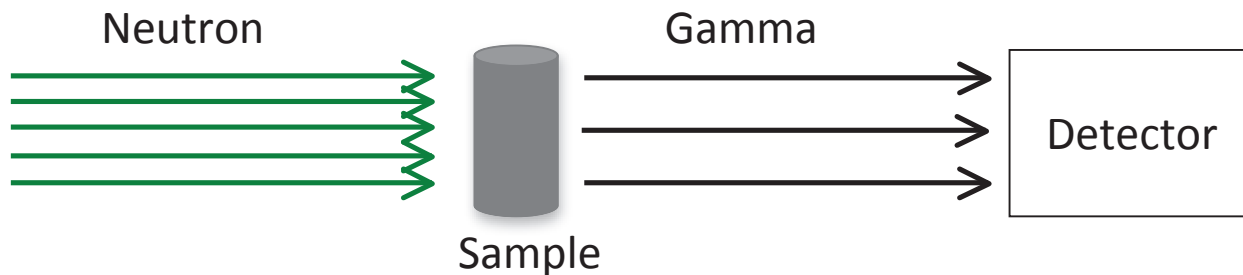


FIGURE 1. Neutron Activation Analysis schematic.

When performing NAA, each radioactive isotope releases characteristic gamma rays that can be used to uniquely identify the isotope. Additionally, the number of gammas that the detector counts is linearly related to number of atoms of the isotope that is emitting the gammas allowing quantitative determination [11,12].

The two main advantages of NAA are that it is extremely sensitive to the two elements being studied and that it detects elements by their mass and not their chemistry. The high sensitivity allows for low concentrations of elements to be used. Low concentrations are useful for a taggant, since it makes them harder to defeat: low concentrations of an element are harder to separate from the explosive than higher concentrations. The sensitivity of NAA to the mass of these two taggant elements and not their chemistry also enables simple sample preparation. NAA can be used on solids, liquids, gases, and dusts; as long as the elements are present [11].

METHODS

A series of experiments have been conducted to determine the detection of samarium and holmium in post-blast residue. The taggant elements were added to a commercial binary explosive of ammonium nitrate and nitromethane (referred to as a binary explosive throughout the rest of this paper). Table 1, summarizes the tests in terms of the taggant element used, the as-prepared concentration of the element, and the mass of explosive used.

TABLE 1. Nuclear barcode experimental setup summary

Test	Taggant Element	Witness Plate Geometry	Taggant Concentration (ppm)	Mass of Explosive (g)
1	Holmium	Cylinder	4.9	167
2	Holmium	Plate	4.9	167
3	Samarium	Cylinder	19.7	169
4	Samarium	Plate	20.1	166
5	Control	Cylinder	0.0	169

All tests used a witness ‘plate’ from which the post-blast residue was collected. The charge was suspended in the approximate center of the plate. Tests 2 and 4 used a square witness plate approximately one-foot square, whereas tests 1, 3, and 5 used a cylinder approximately one foot in length, with the charge being located approximately six inches from the witness plate. The cylindrical witness plate was used to capture almost all the post-blast residue, except for the small amount that goes directly upward and out of the tube. The square witness plates were used to capture the post-blast residue that goes straight down. The two witness plate techniques provide insight into the required post-blast residue collection conditions.

The post-blast residue from each test was sampled by repeatedly dabbing a cotton ball around the inside of the cylinder (for tests 1, 3, and 5), and the surface of the plate that was exposed to the detonation (for tests 2, and 4). The cylinders were sampled at the midpoint of where the charge was suspended, as well as just above and below the edges of the charge. The plates were sampled in the center, and a 6-inch by 6-inch square surrounding the center of the plate. For each test, two additional samples were taken. The first was a cotton ball set aside from the start as a control. The second sample was a sample of the witness plate or cylinder taken before detonation. These pre-detonation samples were taken in the same way as the post-blast samples.

RESULTS

The samples were irradiated using the Missouri University of Science and Technology's Research Reactor (MSTR). The MSTR is a 200 kW, MTR fuel type, swimming pool reactor. The samples were irradiated using a pneumatic tube system to insert and remove the samples from the core. Each sample was irradiated for 10 minutes at 200 kW of power. The samples were counted for one hour using a high purity germanium (HPGe) detector, and the results plotted using Canberra's Prospect software, which was also used to determine the net counts of the various peaks. It must be noted that the time between irradiating and counting the samples is not the same for each test. This means the number of net counts across different tests is not comparable, since the number of radioactive atoms will decrease by half for each half-life that passes. This time difference means that each of the samples had a different amount of decay, so for figure 2, the counts are scaled as a percent of their maximum values within the energy range of 30 keV to 120 keV. However, the number of net counts of holmium and samarium are comparable within the same test. The results of performing NAA on each of the samples collected are shown in figure 2.

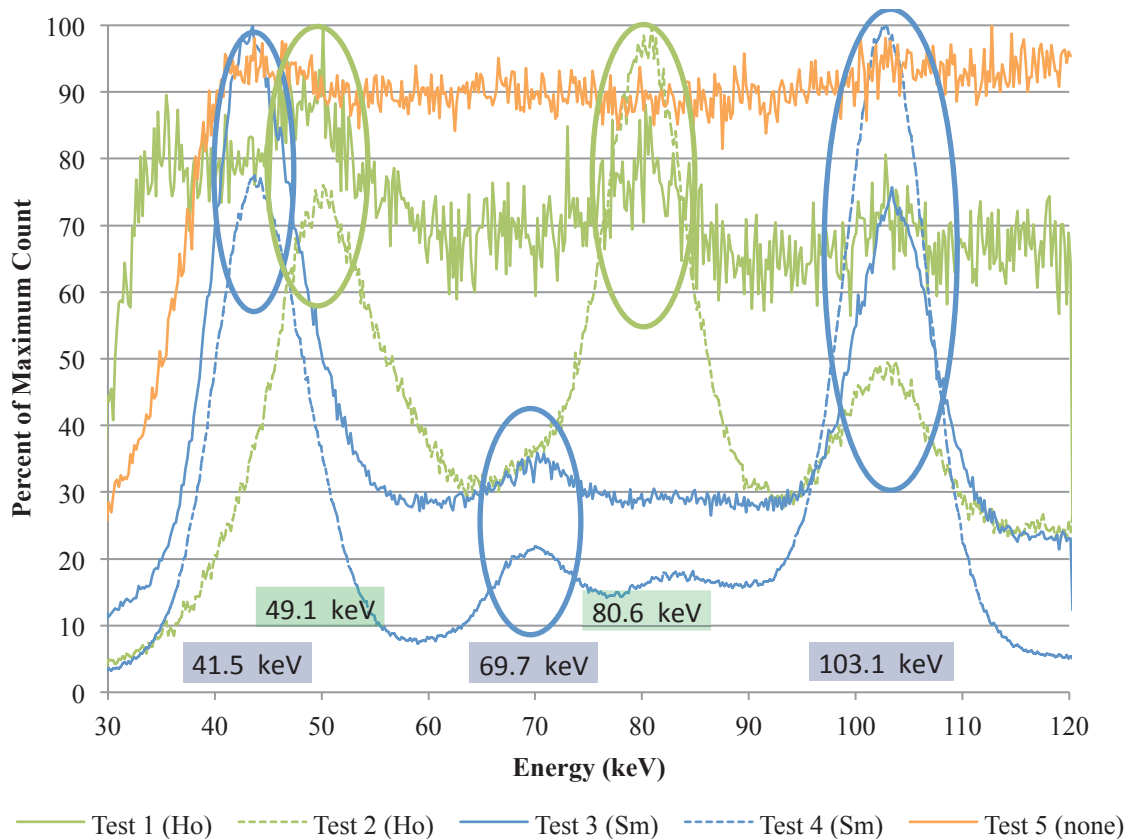


FIGURE 2. Scaled net counts showing the locations of NAA peaks for the elements Samarium and Holmium.

The key features in figure 2 are the peaks in the holmium tagged samples at 49.1 and 80.6 keV, and the peaks in the samarium tagged samples at approximately 41.5, 69.7, and 103.1 keV. The 80.6 keV peak is the strongest peak for holmium in the detector used, and thus indicates the presence of holmium in the sample [12]. The 69.7 keV peak is the strongest indicator peak for samarium, since the peaks at 41.5 keV and 103.1 keV are not unique to samarium and appear in other rare-earth elements when irradiated [13].

Test 1 appears different from tests 2 through 4 because it was measured substantially later than the other tests due to some technical difficulties with the detector at the time of measurement. This gave the sample a much longer time to decay than in tests 2 through 4. This, in turn, reduced the magnitude of the peaks, since the number of radioactive nuclei that create them is significantly lower. Thus, the spectrum becomes more dominated by noise, and resembles the control sample more than test 2.

In tests 1 and 2 there is a smaller peak at 103.1 keV that could potentially indicate the presence of samarium. This is most likely the result of the difficulty in separating rare earth elements, and is not the result of samarium contamination during detonation. The first possibility is that there is some samarium contained in the holmium that was used as a taggant. If there was a significant amount of samarium, then the other peaks at 69.7 and 41.5 keV should also be visible, and should have the same overall shape as the results from tests 3 and 4 where the 41 keV peak and the 103.1 keV peak are roughly equal in magnitude and the 69.7 keV peak is much lower. Looking at the results from test 2 first, there is no peak at 41.5 keV, and there also does not appear to be one at 69.7 keV. The second, more likely possibility is that the peak at 103.1 keV in test 2 is from another element. Test 1 is harder to analyze due to being a noisier, less active sample. In test 1, there is less of an indication of a 103.1 keV peak as well, though this is likely due to the higher noise. There is no strong indication then, that peak at 103.1 keV is from samarium and is not from another element. The results from the collected post-blast residue from tests 1 through 5 are shown below in Table 2.

TABLE 2. Net counts of taggant elements

Test	Taggant Element Added	Holmium Net Counts	Holmium Present	Samarium Net Counts	Samarium Present
1	Holmium	2,236 ± 668	Yes	290 ± 1,306	No
2	Holmium	98,026 ± 1,744	Yes	-	No
3	Samarium	63 ± 1,490	No	10,872 ± 1,299	Yes
4	Samarium	3,372 ± 1,762	Yes	66,381 ± 2,589	Yes
5	Control	766 ± 1,802	No	849 ± 988	No

These results show that samarium and holmium show up in the tests when they were added. Additionally, they also show that they do not occur in the tests where they were not added. Notice that the magnitude of the uncertainty in the net counts for the element that was not added is greater than the magnitude of the net counts measured for all tests except number 4. The results from test are complicated by the apparent peak at 82.5 keV, above the holmium 80.6 keV peak, which affects the net count. Under the test conditions, no cross contamination has occurred, and any naturally occurring quantities of the taggant elements used are low. Additionally, the quantity of post-blast residue collected from both collection geometries, plate or cylinder, appear to be sufficient to analyze through NAA.

CONCLUSION

This series of experiments shows that samarium and holmium survived the detonation process and were identifiable in the post-blast residue from a commercially manufactured binary explosive. This is the first step in making a survivable taggant for explosives.

This research had two objectives: determine if holmium and samarium will partition into the post-blast residue (1), and determine if holmium and samarium can be identified in the post-blast residue by NAA (2). The presence of holmium found in the post-blast residues from tests 1 and 2, and the presence of samarium found in tests 3 and 4 show that both objectives have been met. This is the first step towards making both are viable taggants that can be used with explosives.

FUTURE WORK

Being able to find samarium and holmium in the post-blast residue means they are deposited as a result of the detonation. This is a first step for creating a taggant system that uses these elements. Determining other elements to use with this technique will require selecting elements that are rare in nature and are detectable at low concentrations, in the parts per billion range. Future testing will be required to evaluate if candidate elements will also be identifiable in the post-blast residue. If they prove to be so, then determining how to use them to encode the identifying information would be the next step towards developing an identification taggant based on this technique.

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