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Abstract

Disarmament agreements and discussions between the United States and the Russian Federation for reducing the number of stockpiled nuclear weapons require verification of the origin of materials as having come from disassembled weapons. This has resulted in the identification of measurable "attributes" that characterize such materials. It has been proposed that the 871 keV gamma ray of ^{17}O can be observed as an indicator of the unexpected presence of plutonium oxide, as opposed to plutonium metal, in such materials. We have shown that the observation of the 871 keV gamma ray is not a specific indicator of the presence of the oxide, but rather indicates the presence of nitrogen. Some additional measure of the important oxide attribute will need to be found for disarmament purposes.

Introduction

Disarmament agreements such as START II, and the proposed START III, and other discussions are leading to the disassembly of a large number of nuclear weapons. Weapons in both the United States and the Russian Federation will be disassembled, and the components or reprocessed material will be placed in sealed canisters. The canisters will be measured to verify that they contain weapons origin material before being placed into storage. Eventually the material may be reprocessed into reactor fuel or otherwise disposed of, such as through glassification. The Mayak Fissile Material Storage Facility project, funded by the United States, is constructing a facility in Russia to receive plutonium removed from weapons. The international agreements for disassembly and storage of material require the verification of the origin of the material as having actually come from disassembled weapons. This has resulted in the identification of measurable "attributes" that characterize weapons material, such as the presence of plutonium of a certain threshold mass and isotopic composition. Technology is being developed to measure these attributes reliably using non-destructive, non-invasive techniques. Such measurements include observation of neutrons and gamma spectra, and the identification of specific characteristics of such data.

One characteristic of weapons origin plutonium is that it is in metallic form rather than in oxide form. Normally, since a lot of processing is required, plutonium would only be in metallic form if it had been intended for weapons use. This observable characteristic of weapons origin material is termed the "oxide" attribute, and measurement methods have been proposed to distinguish between the metallic and oxide forms of plutonium. The purpose of this paper is to

document recent results that clarify the interpretation of gamma ray measurements on plutonium oxide, and the impact of that interpretation upon measuring the “oxide” attribute.

The Oxide Attribute

In material containing plutonium, alpha particles are released in the decay chain of plutonium, and neutrons are present from spontaneous and induced fission as well as from (α,n) reactions.

The presence of plutonium oxide might be demonstrated through neutron spectroscopy since the neutron energy distribution is effected by the material undergoing an (α,n) reactions. Since neutron spectra of sufficiently high-resolution cannot easily be obtained, there are severe limitations to this method, such as the presence of other impurities in the material masking the presence of the oxide.

Alternately, gamma rays might be observable from plutonium oxide through neutron or alpha induced excitation or reactions on the oxygen. Excited states of ^{16}O are unreachable with inelastic scattering of neutrons or alphas with the available energy, but excited states of ^{17}O (0.038% abundance) and ^{18}O (0.20% abundance) could potentially be reached.

Previous work by Martin [1] indicated that plutonium oxide might be observed by gamma ray emission from the reactions $^{17}O(\alpha,n)^{20}Ne^*$ (1634 keV) or $^{18}O(\alpha,n)^{21}Ne^*$ (1396 keV, 2438 keV, and 2794 keV) where the material is "self-interrogated" by the alphas released in the plutonium decay chain. However, recent measurements at Pacific Northwest National Laboratory (PNNL) as part of the work reported here, have shown that these gamma ray peaks are of very low intensity and are thus unlikely to be useful in a spectral measurement as an indicator of plutonium oxide.

The 871 keV Gamma Ray As An "Oxide" Attribute

The 871 keV gamma ray from ^{17}O has been observed by Martin [1] in high-resolution gamma ray spectra from many, but not all, plutonium oxide samples. Martin attributed the 871 keV line to the $^{14}N(\alpha,p)^{17}O^*$ reaction. Hsu [2], utilizing an americium alpha source, concluded that the 871 keV line observed in plutonium oxide samples was produced via the reaction $^{17}O(\alpha,\alpha')^{17}O^*$ involving the small amount of ^{17}O in the natural oxygen in the oxide. It has thus been suggested that the 871 keV gamma ray of ^{17}O could be produced in plutonium oxide by alphas from plutonium decay via this $^{17}O(\alpha,\alpha')^{17}O^*$ reaction. This 871 keV gamma ray has been proposed as an indicator of the “oxide” attribute in the disarmament community. There has recently been considerable discussion in the community about the “oxide” attribute and the reliability of the 871 keV line as a measure of the presence of oxide.

Nitrogen is a likely low-level contaminant of plutonium oxide since plutonium nitrate is a typical intermediary in the production of plutonium oxide. It is possible that the samples used by Hsu contained nitrogen contamination from the chemical processing (such as nitric acid), which led to the observed 871 keV gamma ray by the $^{14}N(\alpha,p)^{17}O^*$ reaction. We have observed that

removal of such nitrogen contamination is difficult and requires special chemical processing. The work reported here was initiated to determine the relative importance of americium and plutonium alpha decay in the production of the 871 keV gamma ray. Both these sources of alpha particles are present in weapons origin material, since americium is in the decay chain of plutonium. Plutonium mostly produces a 5.15 MeV alpha particle, while americium mostly produces a 5.48 MeV alpha particle. The experiments reported here have led to a determination that the $^{14}N(\alpha,p)^{17}O^*$ reaction is the actual origin of the observed 871 keV gamma ray line, consistent with Martin, but inconsistent with Hsu.

Possible Origins of the Observed 871 keV Gamma Ray

If the 871 keV gamma ray is observed from plutonium oxide, what is its origin? Several reactions are possible.

The presence of an 871 keV gamma ray might be explained by neutron-capture by ^{16}O in a plutonium oxide sample. But, the values of the oxygen thermal neutron-capture cross sections and resonance capture integrals are far too small to effectively compete with either plutonium neutron capture or induced fission. In addition, the other possible gamma ray peaks from neutron-capture on ^{16}O at 1088 keV, 2184 keV and 3271 keV are not found in typical spectra from PuO_2. The conclusion is that the 871 keV peak is not due to neutron capture.

It might be possible to produce the 871 keV peak by inelastic neutron scattering, $^{17}O(n,n')^{17}O^*$. However, there are far fewer neutrons than alpha particles produced in the material and this combined with small cross-sections and the low abundance of ^{17}O makes this mode an unlikely source of the observed 871 keV gamma ray.

It might also be possible to produce the 871 keV peak by inelastic alpha scattering, $^{17}O(\alpha,\alpha')^{17}O^*$ as suggested by Hsu. For a direct reaction, the 5.15 MeV ^{239}Pu alpha particle energy exceeds the 4.6 MeV Coulomb barrier for oxygen by less than 0.6 MeV [3]. Thus, a ^{239}Pu alpha is energetically unable to excite the 871 keV state of ^{17}O. However, the ^{241}Am alpha at 5.48 MeV exceeds the 4.6 MeV Coulomb barrier by about 0.9 MeV and could possibly excite the 871 keV state. If the ^{239}Pu alpha excited this state, the outbound very low energy alpha particle would have to tunnel through the Coulomb barrier before escaping. The tunneling process generally significantly lowers the reaction probability making this explanation unlikely. Coulomb excitation by inelastic alpha scattering, which does not require barrier penetration, is still a possible mechanism for exciting this state [4].

It is also possible that the 871 keV gamma ray of ^{17}O observed in plutonium oxide could arise from the reaction $^{14}N(\alpha,p)^{17}O^*$ involving nitrogen contamination in and around the material. While there is far less nitrogen in plutonium oxide than oxygen, the nitrogen content may approach or even exceed the concentration of ^{17}O. The $^{14}N(\alpha,p)^{17}O$ reaction has a Q value of -1.2 MeV. Both the initial alpha particle and the exiting proton have sufficient energy to surmount the 4.1 MeV and 2.7 MeV Coulomb barriers for the nitrogen and oxygen nuclei, respectively. Any alpha particle with energy greater than roughly 4.8 MeV would be able to penetrate the Coulomb barrier and initiate the reaction, yielding a proton with an energy of at least 2.7 MeV. Heaton's recent paper [5] on alpha reactions with light nuclei discusses alpha

reactions with nitrogen to the higher energy states of ^{17}O, but data is not presented for the low energy spectral region of interest here.

Experiments

Preliminary Indications

To test the origin of the 871 keV gamma, initial measurements were made at PNNL in low background counting caves [6] using high-resolution, high-purity germanium (HPGe) spectrometers. The samples counted consisted of ^{241}Am or ^{239}Pu dissolved in aqueous solutions of zero to 16 molar nitric acid. The isotopic distribution of this plutonium sample was that expected for plutonium containing about 2% ^{240}Pu (typical weapons grade plutonium is about 6% ^{240}Pu). The nitrogen concentration in the sample was directly proportional to the molarity of the nitric acid solution. The oxygen concentration, however, was only very weakly dependent on the acid concentration since the vast majority of the oxygen was found in the aqueous solvent (water is about 55 molar in oxygen). Since both the source strength and nitrogen molarity were varied, the product of these quantities was used as the expected production rate of gamma rays, assuming that the nitrogen was the source of the 871 keV gamma.

The results of these initial measurements are seen in Figure 1, which indicates a nominally linear relationship between 871 keV gamma yield and the product of nitric acid molarity and source strength (either americium or plutonium). The horizontal axis of Figure 1 is the known product of molarity times alpha decay rate in disintegrations per second. The vertical axis of Figure 1 is the observed rate of 871 keV gamma rays per second corrected for geometry and detector efficiency to give an absolute emission rate. These initial results strongly indicated that the 871 keV gamma ray might arise from the $^{14}N(\alpha,p)^{17}O^*$ reaction rather than from the previously postulated $^{17}O(\alpha,\alpha')^{17}O^*$ reaction on the small amount of ^{17}O in the oxide.

Plutonium Oxide Measurements

These initial experiments spurred the need for precision measurements to determine if nitrogen contamination was responsible for Hsu's observed gamma ray from PuO_2. In all cases, a high-resolution gamma-ray spectrometer was used in conjunction with a sufficient thickness of lead attenuator to reduce dead time to acceptable levels. Counting times ranged from minutes to hours, in order to acquire spectra with adequate statistics for measuring and comparing the 871 keV gamma flux under varying conditions. In one case, a sophisticated low-background counting cave was used to lower background and permit measurement of the flux of 871 keV gamma rays from a weak 11 μCi ^{239}Pu source.

The goal of these experiments was to determine whether the nitrogen present in a plutonium oxide sample from entrained air or as a contaminant was the source for the 871 keV gamma ray observed from PuO_2. A source of roughly 5.2 grams of dry plutonium oxide powder was used. The sample's gamma spectrum was measured after a variety of physical and chemical manipulations. The sample was weapons-grade plutonium oxide, with 0.42% ^{241}Am grown in

from the decay of ^{241}Pu. Typical new weapons grade plutonium is composed of ^{238}Pu (0.01%), ^{239}Pu (93.3%), ^{240}Pu (6.0%), ^{241}Pu (0.6%), and ^{242}Pu (0.16%).

The atmosphere surrounding this powder was controlled and variously selected to be ordinary air, pure argon, and pure nitrogen. After the initial characterization of the PuO_2, with its indications of nitrogen impurities, the PuO_2 powder sample was successively treated to remove those nitrogen impurities:

1. The PuO_2 was washed five times with 0.1 molar oxalic acid to remove the soluble fluoride and nitrate impurities, dried at 110ºC, and calcined[1] at 1350ºC to convert any remnant of oxalate or carbonate into oxide and to decompose any residual plutonium nitrate or nitride. The washed oxide was then counted.
2. The oxalic acid washed PuO_2 was i) fused in a sodium hydroxide and sodium peroxide mixture; ii) the fusion was dissolved in hydrochloric acid; iii) the plutonium oxidation state was adjusted to Pu^{3+} with ascorbic acid; iv) the oxalate was precipitated, and the oxalate precipitate calcined at 700ºC. This treatment changed the physical character of the powder (density, granularity, appearance, etc.) as a result of these manipulations. We believe these changes arose because of residual sodium chloride in the oxalate precipitate. These plutonium solids were then counted for gamma ray activity.

Discussion

Although the region of the plutonium spectrum near 871 keV contains many peaks, the 871 keV line can be sufficiently strong to determine its presence with high statistical confidence in less than an hour using a typical HPGe spectrometer. Figures 2 and 3 show spectra of the plutonium oxide powder sample under various conditions taken during the course of this work. Because of small variations in measurement geometry of the various samples, the spectra were all normalized to the 769 keV peak originating from ^{239}Pu, which would be unaffected by the chemical processing.

Figure 2 shows the data with the strongest nearby peaks from ^{239}Pu and ^{241}Am to illustrate the normalization process. Note the recoil-broadened ^{72}Ge peak that starts at 834 keV. This feature is due to fast neutrons from the sample that undergo inelastic scattering on ^{72}Ge within the detector crystal itself. These fast neutrons arise largely from (α,n) reactions on a ^{19}F contaminant in the sample. Figure 3 shows the same data over a narrower energy window.

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[1] N.B. — "Calcine" is the chemical term for heating a refractory material to a high temperature to drive off volatile components or to convert a compound to a simpler chemical form. For the plutonium sample used in this study, the purpose was to convert any nitrate, oxalate, or carbonate to the oxide [7]. The process here does not involve any reactions with calcium, but rather, the term "calcination" derives historically from the production of lime (calcium oxide) by roasting calcite (i.e., limestone, chalk, or marble as principal sources of calcium carbonate), which decomposes the carbonate, driving off the resulting carbon dioxide.

The “pre-wash” spectra shown are the oxide as originally received under a nitrogen or air atmosphere. Spectra were taken (not shown) following the simple chemical washing with an oxalic acid solution and are similar to the pre-washed spectra. The two “purified” oxide spectra are the highly purified powder under nitrogen and argon atmospheres. The spectra clearly show that the 871 keV peak is removed through purification and immersion in an argon atmosphere. The chemical cleaning removed contaminants such as ^{19}F and the ^{228}Th chain leading to ^{208}Tl. The resulting gamma rays from these sources are thus removed from the spectra. The removal of fluorine also reduces significantly the number of neutrons, and thus the ^{206}Pb line from neutron excitation of the shield is removed. The chemistry of ^{241}Am is similar to plutonium in the process used, and it is thus not removed.

There are a number of peaks in the general region of the 871 keV peak, as seen in Figure 3. These peaks arise not only from plutonium and americium, but also from ^{228}Th daughters, inelastic neutron scattering gamma rays, and other (α,n) reactions [8]. Note that both the 873 keV and 874 keV peaks from ^{241}Am and ^{240}Pu are close enough to the 871 keV peak that resolution of the two separately is very difficult. This fact complicates statistical identification of the 871 keV peak when it is weak.

We conclude that the observed 871 keV flux arises from nitrogen impurities and from nitrogen gas surrounding the powder, and not from the oxide. Clearly the ratio of the 871 keV flux to the 769 keV flux (from ^{239}Pu) would not change (as was observed) were oxygen the dominant source of the 871 keV gamma ray emission. Instead, we find that the amplitude of the 871 keV peak varies with the atmosphere at each of the three stages in our purification process. There is a decrease in the 871 keV line intensity between the nitrogen-atmosphere spectra before and after the washing and calcining steps. The powder’s void fraction changed after each step in cleaning, as did the amount of powder that adhered to the sides of the container. However, the 871 keV line returned again to nearly its original intensity after the purification steps when in a nitrogen atmosphere, but disappeared under an argon atmosphere. This result implies that the 871 keV line arises primarily from alpha-particle interactions with the nitrogen gas rather than nitrogen-containing chemical impurities in the original powder, though both contributions could be originally present.

The clearest evidence for the prevalence of gas-phase interactions is shown in Figure 4. This figure shows the entire spectra of the purified oxide powder with argon and nitrogen atmospheres with only a fine dusting of the powder on the walls of the counting vial. The spectra are identical except for the 871 keV line. The remaining nitrogen impurity concentration for these spectra should be both greatly reduced relative to the original concentration and completely insignificant in the context of 871 keV gamma-ray generation. Based upon these results, we attribute the 871 keV gamma ray flux to alpha-particle interactions with gas-phase nitrogen.

These results are not surprising upon careful consideration. Our preliminary experimental measurements [6] found that 1.1 mCi of ^{239}Pu in an 8 molar solution of nitric acid emitted the 871 keV gamma ray at a rate of roughly 2.1 per second. The same amount of plutonium produces the 769 keV gamma ray at a rate of 4.6 per second as a result of plutonium decay. We saw that the 871 keV flux from the nitric acid solution arises from nitrogen interactions since the flux scaled linearly with the molarity of the nitric acid solution, and no

measurable 871 keV flux was present when americium was dissolved in a hydrochloric acid solution.

It is reasonable to expect that the ratio of the 871 keV flux to the 769 keV flux should scale linearly with both the nitrogen concentration and the range of the alpha-particle. The alpha-particle range for the aqueous solution was roughly 40 microns. The alpha-particle range in plutonium oxide depends on the powder density. Although the density of highly packed plutonium oxide is roughly 11 g/cm^3, the density for our powder was in the range of 3-4 g/cm^3, giving an alpha-particle range in our oxide powder also close to 40 microns. The ratio of the 871 keV flux to the 769 keV flux for both aqueous acid solutions and fine, low-density oxide powders should scale approximately with the nitrogen concentration. The nitrogen concentration in the 8M nitric acid solution was 4.8 x 10^{21} /cm^3. The presence of air within the powder resulted in a nitrogen concentration of roughly 3 x 10^{19} /cm^3 (computed for air at normal pressure and temperature). One would therefore expect the gamma line ratio to be roughly 160 times lower in the oxide powder than in the acid solution (ratio of 4.8 x 10^{21}/ 3 x 10^{19}). In fact, the ratio was observed to be roughly 18 times lower, leaving a discrepancy of a factor of roughly 9.

We believe this additional factor of 9 arises from alpha-particle interactions in the gaseous nitrogen *external*, rather than internal, to the powder. We compute that there was only about 0.8 cm^3 of nitrogen gas within the powder, whereas there was roughly 10 cm^3 of gas in the space above the powder. Since the range of alpha particles in air is roughly 3.5 cm, a nitrogen reaction would be possible during roughly the first centimeter of a 5 MeV alpha particle's track through the air. While exact calculations are not possible given the unknown distribution of oxide on the sides of the container, it seems that such interactions with the air above the powder could account for the bulk of the 871 keV gamma ray generation.

Two quantitative results of importance can be gathered from these experiments. First, we can state that the nitrogen impurity concentration necessary to lead to significant observable 871 keV gamma ray peak generation is roughly 3 x 10^{19} /cm^3, which corresponds to 200 ppm. Amounts significantly less than this would not be able to generate a measurable 871 keV gamma ray flux.

Second, we can estimate the maximum contribution to the 871 keV flux that could be accounted for by the $^{17}O(\alpha,\alpha')^{17}O^*$ reaction in the oxide. Measurement of a hydrochloric acid solution with 86 μCi of dissolved americium placed an upper bound of 0.004 gamma rays per second on the 871 keV emission rate. Assuming that americium alpha particles produce 871 keV gamma rays at the same rate as plutonium alpha particles, we can place an upper bound on the generation rate from the ^{17}O in our powder sample. Based on our data, the 871 keV flux from ^{17}O in the plutonium oxide can be no more than a few percent of the value observed for the purified oxide powder in a nitrogen atmosphere. Even at this maximum possible level, the 871 keV gamma ray flux would be difficult to measure in a reasonable time, especially in light of the nearby ^{241}Am and ^{240}Pu peaks in the gamma spectrum. These peaks will be even more pronounced for plutonium containing more ^{240}Pu than the 6% of our oxide sample. We believe that this conclusion can be extrapolated to bulk samples of plutonium oxide.

A measurable 871 keV gamma-ray flux should be observable only when the oxide either contains significant nitrogen as an impurity, or when air is present within or around an oxide powder. Based on these considerations, the 871 keV gamma ray line should not be observed from oxide when the purified oxide is surrounded by a gas that contains little nitrogen, when the oxide is compacted and sealed with little extra space for air, or when the oxide is immersed in or dissolved in a liquid such as oil or water that does not contain nitrogen.

The 871 keV peak is not observed from a plutonium metal sample. Since measures are taken to prevent oxidation of plutonium components (e.g. plating or an inert gas atmosphere) alpha-particles from the metal surface would not reach any nitrogen around a metal sample, and thus no 871 keV gamma rays would be expected.

Conclusion

The experimental data resulting from our high resolution spectra of plutonium oxide powder and aqueous solutions of plutonium and americium allow us to conclude that the 871 keV gamma ray observed for many oxide samples arises from the reaction $^{14}N(\alpha,p)^{17}O^*$. There are three different ways that nitrogen comes to be present in plutonium oxide. First, nitrogen may be present as an impurity. Our samples showed impurities up to approximately the 1000-ppm level, as inferred by our observation of the 871 keV gamma ray in measurements of samples of weapons grade plutonium oxide. Such nitrogen would lead to an 871 keV gamma ray flux that is easily measurable under ordinary conditions. Second, immersion of a powder in air causes significant amounts of nitrogen to permeate the pore spaces between the individual oxide particles. Third, a measurable 871 keV gamma ray flux could arise merely from the interaction of alpha particles from plutonium decay with air surrounding an oxide sample. Thus, a measurable 871 keV gamma ray flux should be observable only when the oxide either contains significant nitrogen as an impurity, or when air is present within or around an oxide powder. We have concluded that the observed nitrogen is dominated by the gas within and around the powder sample. This is the only hypothesis that can quantitatively explain the full spectrum of experimental observations:

- The linear scaling of the 871 keV flux with nitric acid molarity.
- The similar 871 keV gamma ray production rates for americium and plutonium sources.
- The lack of an observable 871 keV flux for americium in a nitrogen-free solution.
- The lack of an observable 871 keV flux for a variety of plutonium oxide samples [1].
- The lack of an observable 871 keV flux for purified plutonium oxide powder in a nitrogen-free atmosphere.
- The linear scaling of the 871 keV flux with nitrogen concentration and alpha-particle range for both aqueous and powder samples.

The presence of an 871 keV gamma-ray flux from a plutonium sample would probably indicate that a sample is not metallic, but the converse assertion is not true. That is, the lack of a measurable 871 keV gamma-ray flux does not necessarily indicate that the sample is metallic. Plutonium in the oxide form can easily be “disguised” either via compaction, purification, or atmospheric control. A more appropriate name for this attribute would be “presence of nitrogen,” which accurately conveys the strengths and weaknesses of this measurement.

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Footnotes

1) N.B. — "Calcine" is the chemical term for heating a refractory material to a high temperature to drive off volatile components or to convert a compound to a simpler chemical form. For the plutonium sample used in this study, the purpose was to convert any nitrate, oxalate, or carbonate to the oxide [7]. The process here does not involve any reactions with calcium, but rather, the term "calcination" derives historically from the production of lime (calcium oxide) by roasting calcite (i.e., limestone, chalk, or marble as principal sources of calcium carbonate), which decomposes the carbonate, driving off the resulting carbon dioxide.

Figure Captions

Figure 1. The 871 keV gamma ray emission rate (measured and corrected for geometry) as a function of the product of the nitric acid molarity and the total alpha emission rate. The line is drawn to indicate the approximate linear relationship of the data.

Figure 2. Comparison of normalized spectra for pre-washed and purified plutonium oxide samples. Note the indication of the 769-keV ^{239}Pu peak used for normalization. The background in the pre-wash data is significantly higher than the purified data due to contaminants and neutron generation.

Figure 3. Comparison of normalized spectra for the same raw and purified plutonium oxide sample. Note that the spectra for the purified sample have been raised by 10 counts/1000s to aid in comparison. Comparing the two purified spectra, the only significant difference is the missing 871 keV peak in the argon atmosphere. The thallium and its parents were removed in the purification, eliminating the ^{208}Tl line at 860 keV. Also removed in purification was the fluorine which gives a gamma from ^{19}F(α,n)^{22}Na* at 891 keV. This reaction is also the source of the neutrons that give rise to the ^{206}Pb peak at 881 keV through inelastic scattering.

Figure 4. Normalized spectra for dusting of purified plutonium oxide powder under argon and nitrogen. Note that the intensity of the 871 keV line is the only significant difference between the two spectra.

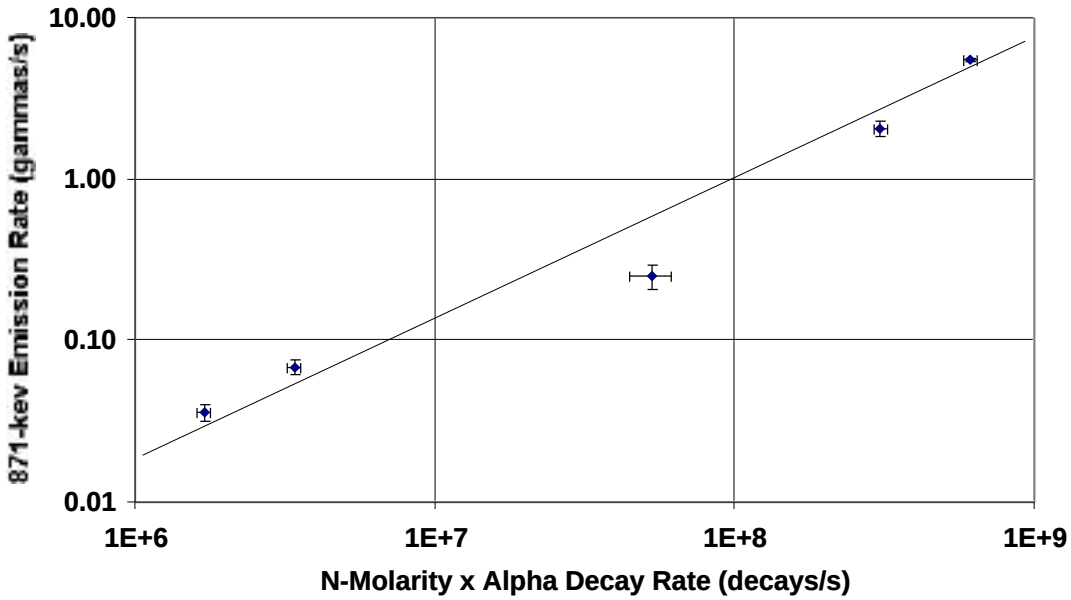
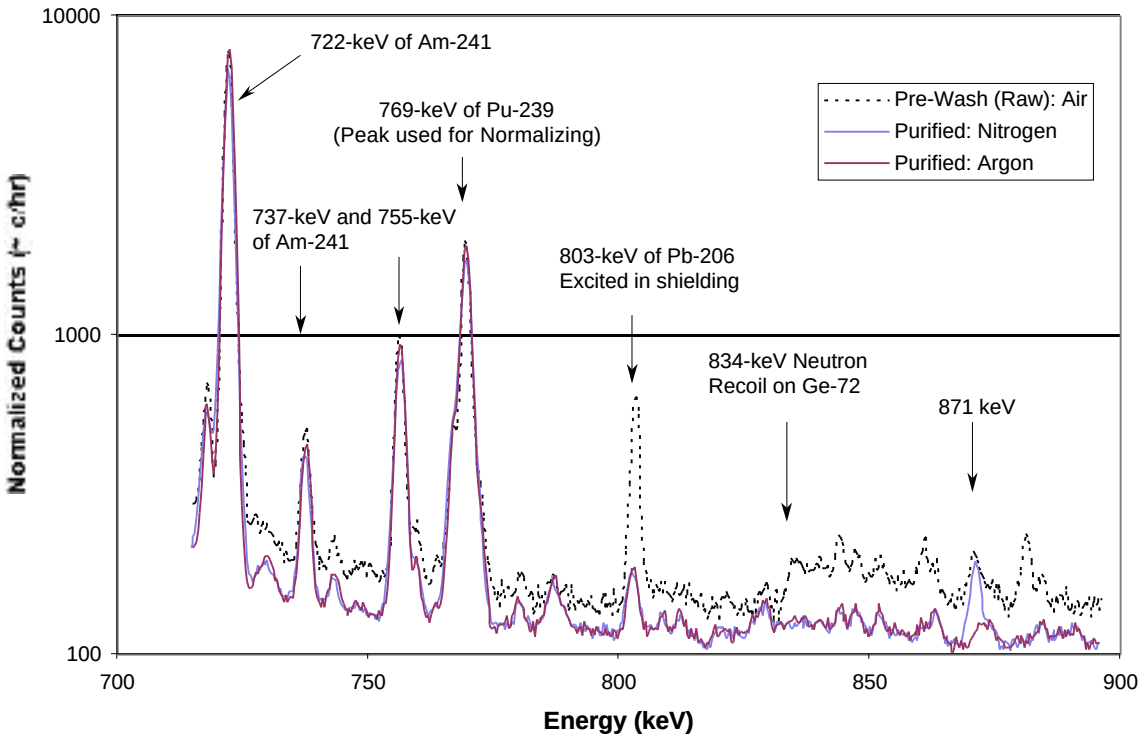


Figure 1. The 871 keV gamma ray emission rate as a function of the product of the nitric acid molarity and the total alpha emission rate. Statistical error bars are shown for the data points. The line is drawn to indicate the approximate linear relationship of the data.



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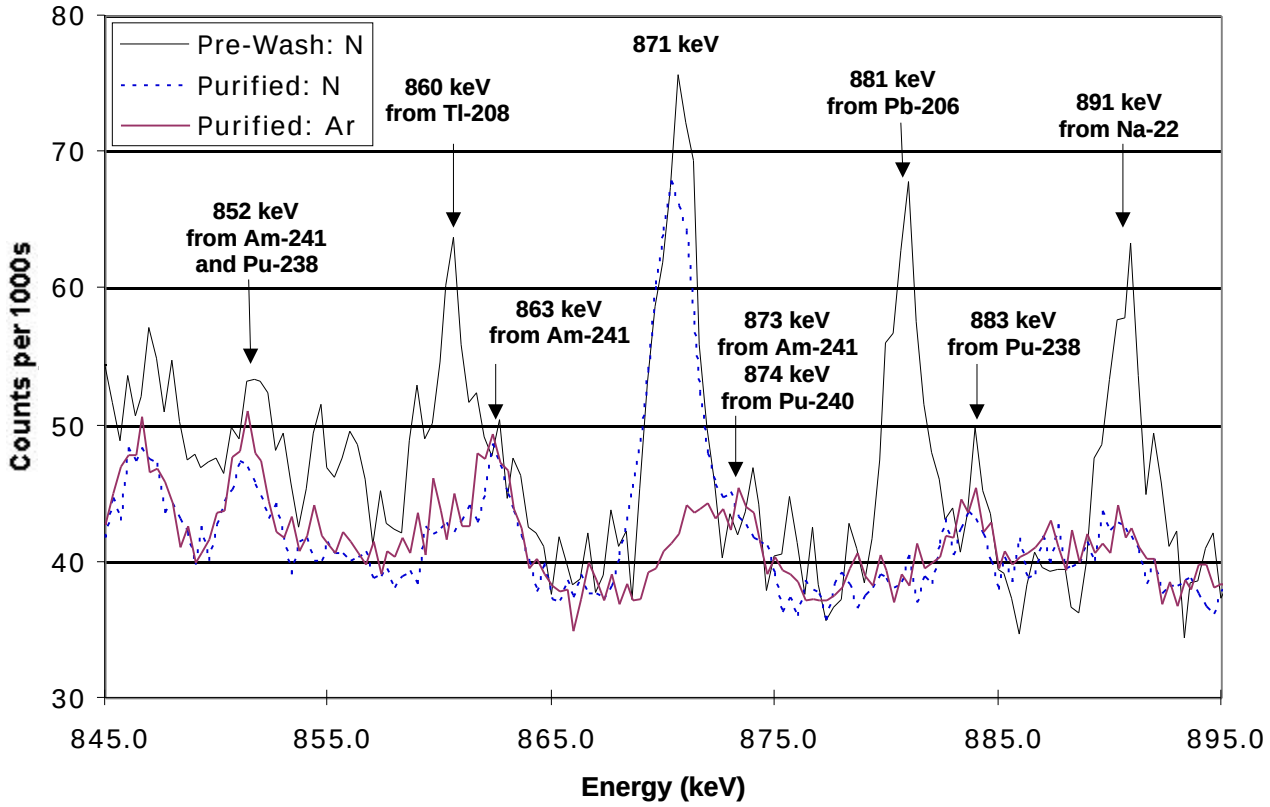

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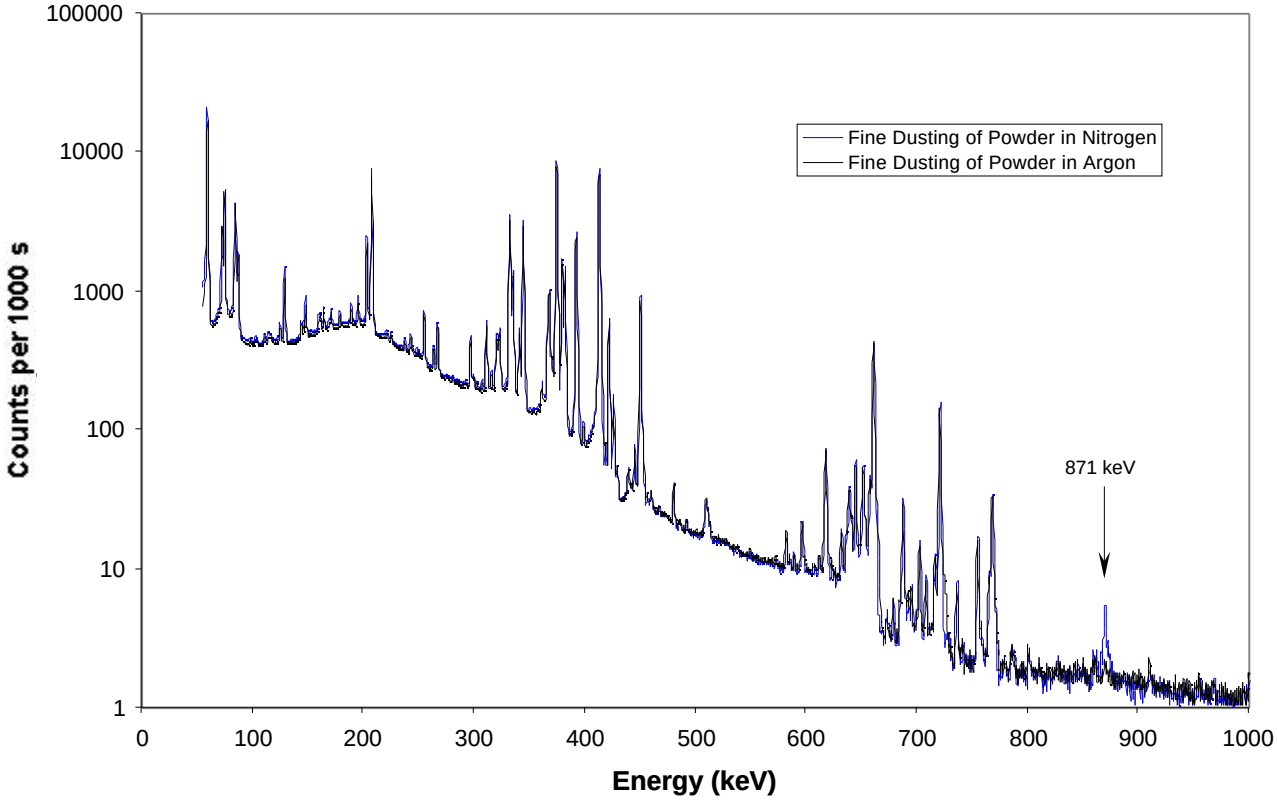

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