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PARAMETRIC STUDIES FOR ^{233}U GAMMA SPECTROMETRY

Candice C. Scheffing and Alan Krichinsky

ABSTRACT

Quantification of special nuclear material is a necessary aspect to assuring material accountability and is often accomplished using non-destructive gamma spectrometry. For ^{233}U, gamma rays are affected by matrix and packaging attenuation and by a strong Compton continuum from decay products of ^{232}U (inherently found in ^{233}U) that obscure ^{233}U gamma photopeaks. This project, based on current work at the national repository for separated ^{233}U located at Oak Ridge National Laboratory (ORNL), explores the effects of various parameters on the quantification of ^{233}U– including material form and geometry. Using an attenuation correction methodology for calculating the mass of ^{233}U from NDA analysis, a bias of almost 75% less than the actual ^{233}U mass was identified. The source of the bias needs to be understood at a more fundamental level for further use of this quantification method. Therefore, controlled experiments using well characterized packages of ^{233}U were conducted at the repository and are presented in this paper.

INTRODUCTION

Non-destructive assay (NDA) has been a useful tool for many years to quantify nuclear material by measuring the interaction of emissions such as neutrons and gamma rays from radioactive decay with matter [1]. Gamma ray ionization is detected using a gamma ray detector that measures the amount of gamma radiation emitted as well as the energy of release. NDA can be very useful in applications such as waste characterization, nuclear Material Control and Accountability (MC&A), and criticality safety. Gamma spectrometry can help determine the type and mass of a nuclide present without destructively altering or damaging the material of interest or its packaging.

Inherent in gamma ray interactions is attenuation, which must be carefully considered when performing NDA measurements. The fundamental law of gamma ray attenuation states that the transmitted gamma ray intensity is a function of gamma ray energy, absorber composition, and absorber thickness [2]. The absorber can be any number of things from packaging and shielding to the material itself (self-attenuation). Uranium and plutonium have high self-attenuation factors due to their high density, which promotes a large number of gamma ray interactions within the sample before a photon is detected. In order to calculate and characterize the amount of attenuation occurring, information about the sample's matrix (packaging and the material) including size and shape needs to be known. Different isotopes and forms of uranium and plutonium (and their impurities) cause varying degrees of self-attenuation, which may be calculated readily for pure or well characterized materials and packaging configurations [1]. Unfortunately, factors such as impurity content, material distribution and packaging configuration often are not well known and complicate determining their detailed influence on attenuation. The attenuation of ^{233}U is important due to the need for accurate quantification of this special nuclear material to ensure accountability and criticality safety.

The man-made isotope ^{233}U is produced from neutron capture in ^{232}Th in a nuclear reactor. A small amount of ^{232}U 'contaminant' is always present with ^{233}U depending on the

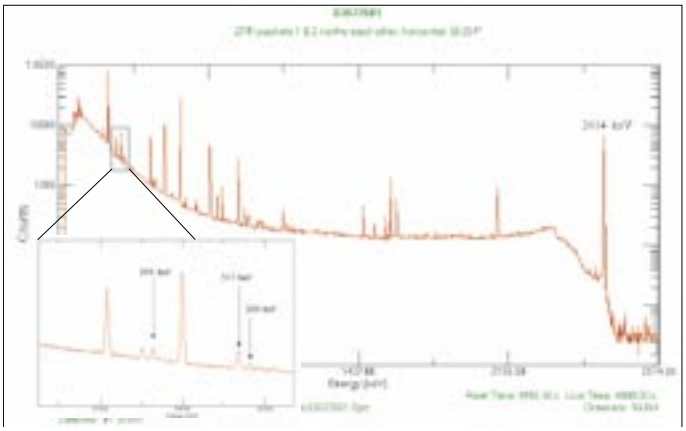


Figure 1: Sample gamma scan showing Compton continuum from 2614 keV photopeak. Inset shows size and location of ^{233}U photopeaks.

reactor conditions under which it is produced. The attenuation and response of ^{233}U photons is somewhat more difficult to characterize due to the presence of its associated isotope, ^{232}U. These two isotopes along with their progeny, present a range of gamma emissions leading to a high source-related background which makes it difficult to separate the peaks that can be used for quantification of ^{233}U directly [3]. A typical problem with quantifying ^{233}U relates to the 2.6-MeV photopeak emission from ^{208}Tl, a decay product of ^{232}U. This photopeak is easy to resolve but establishes a Compton continuum, or range of energies corresponding to Compton-scattered electrons, that often overwhelms the lower energy ^{233}U peaks (Figure 1) [1]. A previous method [4] for NDA of waste drums used this 2.6-MeV photopeak from ^{208}Tl and an external gamma source

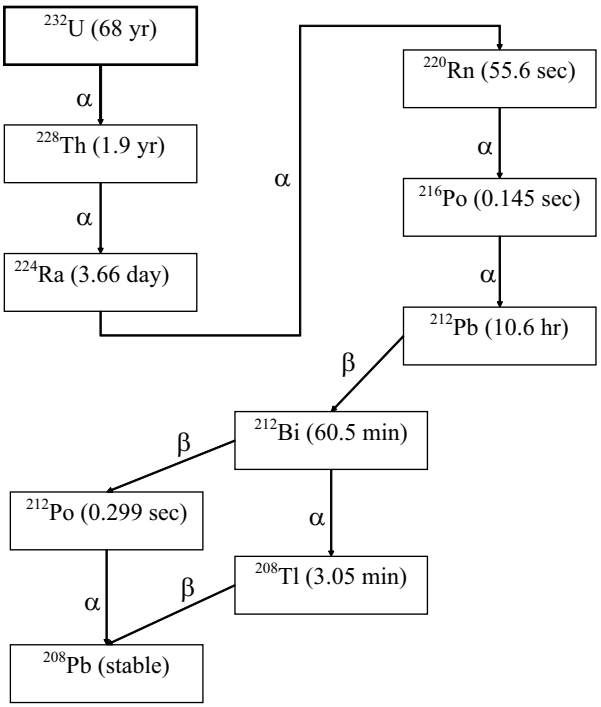


Figure 2: ^{232}U Decay chain showing that ^{232}U half-life is much, much greater than decay product half-lives, which is the basis for secular equilibrium assumption.

to determine a correction for attenuation. This method was acceptable for quantities of ^{233}U that were less than 200 grams. Regulations for MC&A measurements of quantities greater than 200 grams, however, must be measured directly (with ^{233}U emissions rather than ^{208}Tl emissions), requiring a new method for accurately quantifying the nation's inventory of ^{233}U.

In search for a reliable method of attenuation correction using a high-resolution detector for MC&A measurements, ORNL researchers [3] created a technique that utilizes the broad energy range of product emissions from ^{232}U. The activities of ^{232}U and its decay products were assumed to be in equilibrium, for this case defined as secular equilibrium. This occurs because the half-life of ^{232}U is much greater than its decay products and after about 5.4 half-lives of the longest lived product has passed (~10 years in this case), the chain activity reaches equilibrium [5]. Figure 2 shows the decay chain and half-lives for ^{232}U.

Isotope	Energy (keV)
^{233}U	291.34
	317.13
	320.54
^{212}Pb	238.63
	300.09
^{224}Ra	240.99
^{208}Tl	277.36
	510.77
	583.19
	763.13
	860.56
	2614.53

Table 1. Selected photopeak energies for gamma spectrometry measurements of ^{233}U and ^{232}U progeny [8].

The experiments conducted for this report pick up with analyzing the results of the technique created by ORNL researchers [3] and goes further with parametric studies using three small Zero Power Reactor (ZPR) packets of ^{233}U (Figure 3). The ZPR packets were fabricated by ORNL along with more than 1700 others for Argonne National Laboratory more than 20 years ago. Each packet contains approximately 27 g of ^{233}U in the form of U_3O_8 with less than ten ppm of ^{232}U. The packet itself forms a 3"x 2" x ¼" nickel-plated stainless steel cladding [6].

The objective of the present experiment was to analyze gamma spectrometric data of ^{233}U material and accurately quantify the amount present using an attenuation function of ^{232}U progeny for ^{233}U (without extensive and detailed knowledge about the materials matrix). The parametric experiments were used as a means to study the conditions of the material and determine factors significantly affecting the ^{232}U and ^{233}U attenuation. In the long term, it will be useful to find a consistent method of quantification for the special nuclear material ^{233}U.

MATERIALS AND METHODS

Activity Measurements

Two different sets of results are included in this paper, those for cans of ^{233}U and those for the ZPR packet experiments. The cans of ^{233}U were scanned in what will be referred to as the operating area. These scans and their quantification results led to the decision to perform more controlled experiments in a laboratory using the ZPR packets.

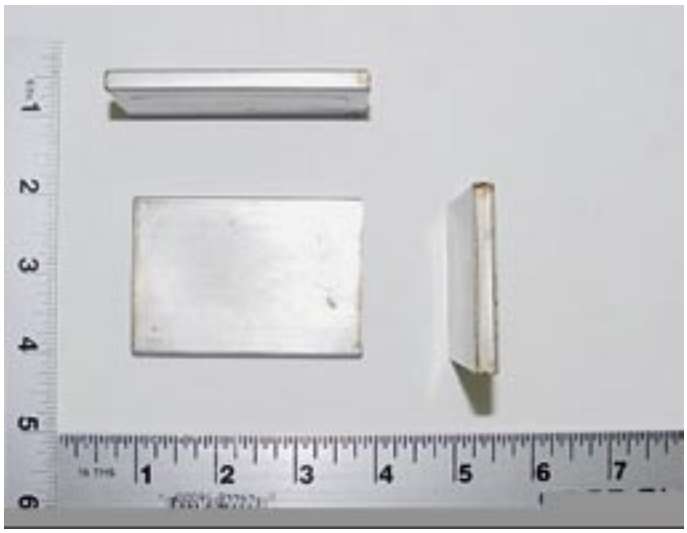


Figure 3: Photo of sample ZPR packets.

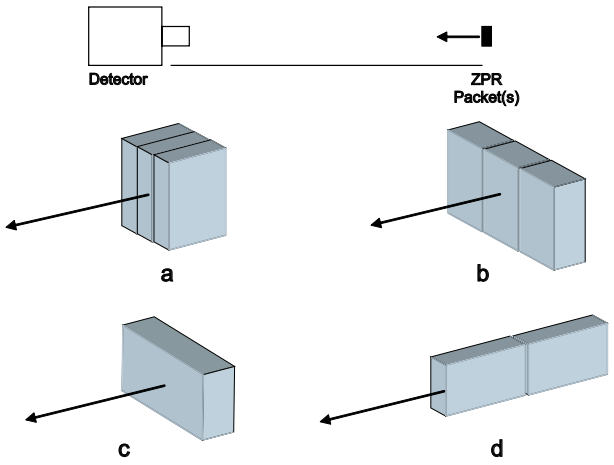


Figure 4: ZPR experimental setup, views a-d are all examples of how ZPR packets were arranged to face the detector. (a) three packets stacked vertically with the narrow face toward the detector; (b) three packets vertical, end to end with broad face toward the detector; (c) one packet horizontal with broad face toward the detector; (d) two packets horizontal, end to end with the narrow face toward the detector (more depth).

Gamma ray measurements were completed using an n-type High-Purity Germanium detector [HPGe; EG&G Ortec Model CFG-GG (GEM-15180-P), 50.0-mm-diameter x 57.7-mm-thick crystal dimensions] coupled with EG&G ORTEC counting electronics and a computer that collects and analyzes the data using GammaVision software [7]. Calibration for the detector was performed using a point source calibration standard to correspond photon peak energies and abundance efficiencies [8]. For all measurements, the activity for nuclides at corresponding energies of interest listed in Table 1 were calculated using the GammaVision software and standardized for the background, branching ratio, and source-to-detector distance vs. source-to-calibration standard distance. The branching ratio is the probability of various de-excitation transitions (Figure 2, i.e. α or β decay from the same isotope); the detector-to-source distance accounts for the inverse square rule where a photon source diminishes as $1/r^2$, with r the distance from the detector to the source [1].

For typical site operations (measurements made on stored cans of ^{233}U), cans were taken from storage and measured inside the operating area at distances ranging from 75 inches to 206 inches. The material was always inside its storage can and, depending on the specific scan location, sometimes had a ¼-inch-thick steel radiation confinement barrier located between the material and the detector crystal. This operation area presented a high radiation background due to its close proximity to the stored ^{233}U and due to radioactive contamination internal to equipment in the area.

For the parametric studies, ZPR packets were scanned in a separate laboratory which presented a lower background radiation than the normal operating area. The lower background laboratory provided less photon noise while collecting spectra and presented a less hazardous environment in which to conduct the experiments. A majority of the ZPR packet scans were performed with no shielding, at a distance of either 192 inches or 254 inches from the detector face. Each packet was scanned with its broad-face or narrow-face projected toward the detector, and then packets were placed next to each other in different combinations of two and three packets together (Figure 4). The packets were either stacked one behind another or placed next to each other to increase depth or projected area of material, respectively. Measurements were also made with different attenuators in front of the packets: lead (2" thick) and steel (1" and $^3/_8$" thick) were chosen because of their relatively high attenuation coefficients and their availability. Data recorded with each gamma scan included measurement time, distance, packet placement and shielding.

Attenuation Correction

The activity values as reported by GammaVision for the energies of each ^{232}U decay product were analyzed using the

EQ. 1 $$A = \frac{A_0\, r^2_{measured}}{B\, r^2_{calibration}}$$

methodology as described by [3]. The activities for ^{232}U and its decay products were corrected for branching and distance using,

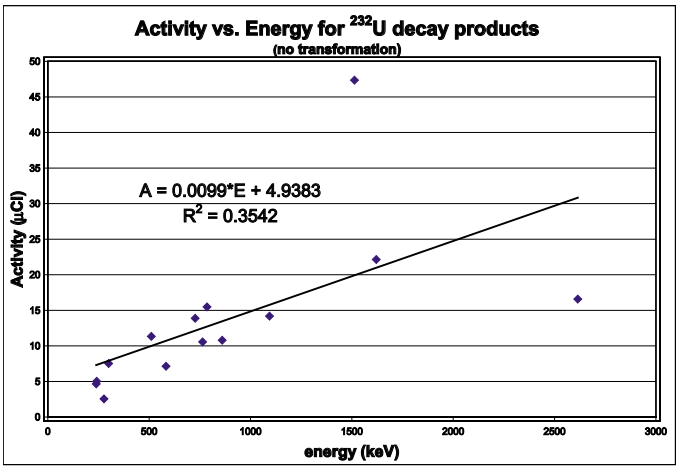
where A_0 is the original activity reported by GammaVision, B is the branching ratio (0.3593 for ^{208}Tl and 1 for all others), and r is the distance from the source to the detector. Using these corrected activities, a plot of the natural logarithm of activity versus the inverse of the energy was made and a linear fit to the data was calculated, $\ln(A) = m/E + b$ (Figures 5a and 5b). The intercept of this plot, b represents the natural logarithm of the true ^{232}U activity corresponding to the mass of ^{232}U present. The slope, m was then used as a proportionality constant for an attenuation correction of ^{233}U.

The activities of each ^{233}U photopeak as reported by GammaVision were corrected for distance and then the proportionality constant (slope = m) was applied to the corresponding data points (activities and energies). A corrected (non-attenuated) ^{233}U activity was calculated by solving for the intercept:

EQ. 2 $$A_{true} = \exp\left(\ln(A) - \frac{m}{E}\right)$$

An overall average of the various photopeak's (three peaks for ^{233}U, Table 1) true activities was taken to be the calculated activity for the experiment. Using the specific activity of ^{233}U (0.0096 Ci/g) and this corrected activity, the mass of ^{233}U was calculated, *mass ^{233}U (g) = A_{true}(Ci) / SA (Ci/g)*. The calculated mass for ^{233}U was then compared to the book value of the material, and the percent differences, $-(x_{calc.} - x_{book})/x_{book}$ were plotted for each can and/or packet. Again, this methodology was developed by previous ORNL researchers [3] and the experiments in this paper used the same methodology to determine its usefulness.

Due to the varying quality of the photopeaks at different energies, an objective criterion to the attenuation function was made by eliminating questionable peaks (insufficient peak area, skewed shape, or bad resolution from other photopeaks). This usually led only to the elimination of the 510.77 keV peak from ^{208}Tl due to its proximity to the annihilation photons of energy 511 keV [1]. For data collected on cans of ^{233}U, physical conditions that may have contributed significantly to a poor scan were also identified promoting their removal from the results (typically, uncertain calibration distances).



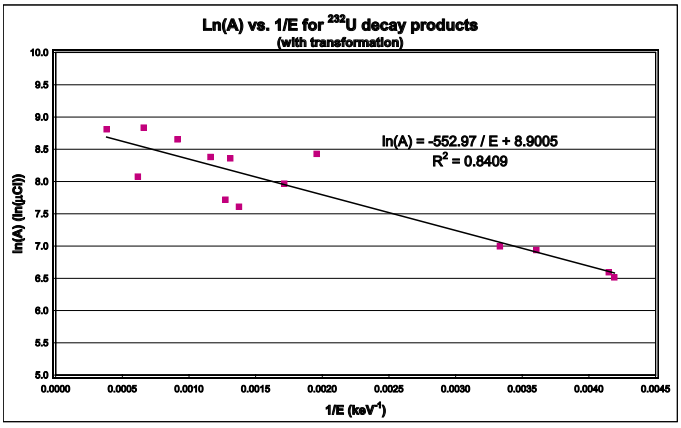


Figure 5a and 5b: sample plot of data for attenuation equation and EQ. 2.

RESULTS

The cans of ^{233}U that were gamma scanned in the operating area (i.e. scans performed before ZPR experiments) all included a background correction determined the day of the scan due to the elevated and occasionally variable background in the area. Early scans used a calibration with the point source located only 10 cm (about 4 inches) from the detector face; this resulted in a larger scaling error than later calibrations which were performed at 15-16 inches. The attenuation correction for cans of ^{233}U indicated a bias of approximately 72 % (± 25 %)

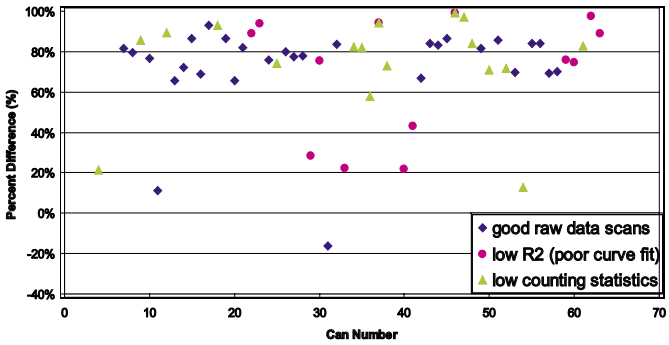


Figure 6: Percent Difference in mass for Cans of U-233

mass difference below the actual book value recorded for each can (Figure 6). Only one scan gave a result higher than the recorded mass value.

As further analysis, the linear fits for each scan were reviewed using the coefficient of determination, or R^2 value which is included in MSExcel™ data analysis software. This value indicates the extent of a linear relationship between two data sets and ranges from 0 to 1, with 1 indicating a good relation (or linear fit to the data) and 0 indicating no linear relation at all [9]. There were 12 cans with an R^2 value below 0.64, but this did not seem to affect the overall percent difference. It should be noted that results from the vast majority of scans clustered around a percent difference in mass bias of 81% (± 10 %) with only a few points showing less bias.

The ZPR packets produced an average error in mass of approximately 49 % (± 19 %) below the book value as shown in Figure 7. There was a difference in R^2 values of the ln*(A)* vs. *1/E* plots between scans of the broad-face and narrow-face of the packet (Figure 8). The narrow-face scans (more material depth) resulted in high R^2 values (>0.8 in 92% of the scans) while the broad-face scans (less material depth) predominately resulted in notably lower R^2 values (<0.8 in 77% of the scans). A closer look at these plots showed that there were three photopeaks that were consistently and pronouncedly below the linear function estimation: 727.19 keV, 785.37 keV, and 1620.50 keV all of ^{212}Bi, requiring further investigation. The majority of the ZPR scans showed a percent difference in mass bias greater

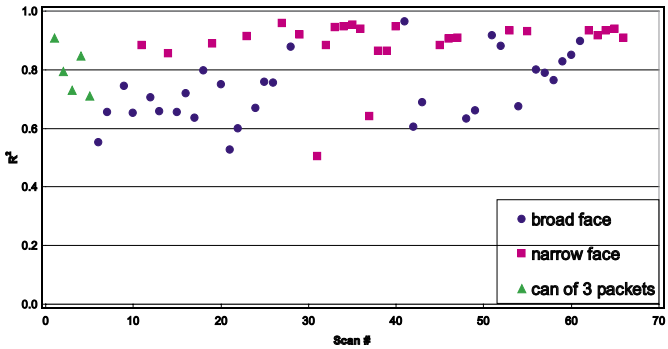


Figure 7: R^2 trends for ZPR Packet Analysis (Raw Data)

than 30%, but there was not a single number around which the bias seemed to cluster.

There was a relationship of the coefficient of determination, R^2 between un-attenuated and steel-attenuated packets. The R^2 value for broad-faced measurements increased for those with steel placed between the packet and the detector. The R^2 value for broad-face scans increased as the thickness of steel increased, which can be seen in Figure 8 (increased R^2 corresponds to an increased thickness of steel). No useful results such as those from the steel were obtained when lead was used as an attenuator.

DISCUSSION / CONCLUSION

The percentage difference between the calculated ^{233}U masses and the book values is too large for the secular equilibrium methodology (proposed by earlier researchers [3] and described in this paper) to be employed in its current state of development, as can be seen in Figure 6. A Department of Energy manual requires uncertainties of <10% for inventories (the actual required uncertainty may be as low as <3% depending upon the quantity and form of material being measured) [10]. A large bias can be tolerated if it is consistent and can be factored into the correction as a systematic function, e.g. the bias is discovered to be a systematic error. However an uncertainty of 25% is not acceptable from a nuclear materials and accountability standpoint. There are a number of factors that can cause this large difference including the geometry of the material,

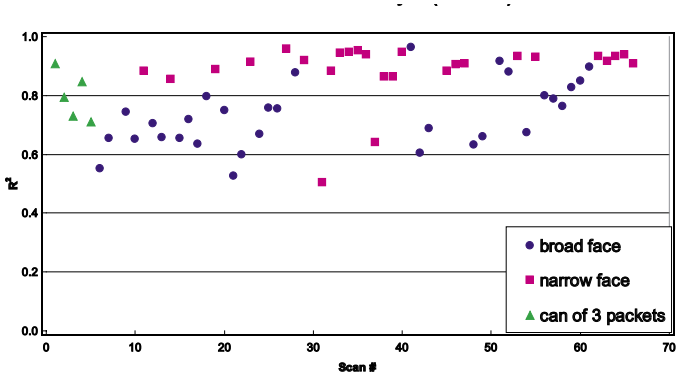


Figure 8: R^2 trend for ZPR Packet Analysis (Raw Data)

count time, photopeak statistics, and the fit of the function for the ln(A) vs. 1/E plot.

The counting time and background have a direct effect on the abundance (net peak area) of certain energy peaks. The GammaVision software calculates and includes an uncertainty for each identified peak which is inversely proportional to the net peak area for activity calculations. Even when scans with high uncertainties (as reported by GammaVision) were discarded, the bias was still in the 60% to 80% range. For scans with high uncertainty peaks, the inclusion or omission of these peaks did not have a noticeable effect on the percent difference or the R^2 values, making it difficult to show that these scans could or should be discarded.

An important factor contributing to the attenuation function is the fit of the ln(A) vs. 1/E plot. The attenuation correction is based on a fit of the ^{232}U progeny activity to their corresponding photopeak energy, and it does not seem reasonable to use a poor fitting function (i.e., low R^2) as a correction. There does not, however, seem to be a correlation between R^2 values and percent difference in mass from the book value. All the ZPR packet scans (with both low and high R^2 values) still had approximately 57% difference from the actual ^{233}U mass. The difference in coefficients of determination values between broad-face and narrow-face ZPR packet scans and between added attenuation material (steel) shows that there is a correlation between the quality of fit (R^2) and attenuation. It is not obvious, however, why this correlation exists.

The three ^{212}Bi peaks that were consistently and pronouncedly below the function estimation did not become important factors that warranted further experimental investigation because of a later discovered error in the GammaVision library. After fixing the library for the error, the ^{212}Bi photopeaks were no longer consistently below the function estimation. Since these experiments and the provided results were completed, the GammaVision libraries have been scrutinized and compared to referenced materials [8] to ensure accurate analysis. Even with these errors corrected, there is still a considerable bias warranting more experiments to discover the major factors affecting the coefficient of correlation values and attenuation correction methodology.

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