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In-Situ Silver Acetylide Silver Nitrate Explosive Deposition Measurements Using X-Ray Fluorescence

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Abstract

The Light Initiated High Explosive facility utilized a spray deposited coating of silver acetylide-silver nitrate explosive to impart a mechanical shock into targets of interest. A diagnostic was required to measure the explosive deposition in-situ. An X-ray fluorescence spectrometer was deployed at the facility. A measurement methodology was developed to measure the explosive quantity with sufficient accuracy. Through the use of a tin reference material under the silver based explosive, a field calibration relationship has been developed with a standard deviation of 3.2%. The effect of the inserted tin material into the experiment configuration has been explored.

ACKNOWLEDGMENTS

The author would like to acknowledge the technical expertise of Phil Miller of Innov-X Systems and Dave Walters of Summit Scientific. Their support and technical assistance helped to overcome the initial accuracy issues. Their involvement contributed to the program's ultimate success.

Daniel Dow and Michael Willis were instrumental in the formulation and testing using the silver acetylide silver nitrate explosive.

CONTENTS

1	Introduction.....	9
2	X-Ray Fluorescence Spectrometer	11
3	X-Ray Fluorescence Measurement Development	15
3.1	Absolute Measurement	15
3.2	Compton Reference Measurement.....	15
3.3	Tin Referenced Measurement.....	18
3.4	Instrument Characterization.....	22
3.5	Measurement Effect on Explosive Impulse Conditions.....	24
4	Conclusions.....	25
5	References.....	27
6	Distribution	28

FIGURES

Figure 2.1	Fox-I X-ray fluorescence spectrometer.....	12
Figure 2.2	X-ray fluorescence spectrometer mounted on a robotic arm for in-situ explosive measurements.....	12
Figure 2.3	XRF spectrometer mount adjacent to the explosive spray gun on the robotic arm, explosives coupons are also shown.....	13
Figure 3.1	Spectral data (energy bins) for SASN explosive on an aluminum substrate.....	16
Figure 3.2	SASN XRF data relative to measured areal density.....	17
Figure 3.3	Error of areal density calculated from XRF data relative to measured areal density..	17
Figure 3.4	Explosive impulse degradation caused by tin	19
Figure 3.5	XRF spectral measurements of silver, silver/tin, and silver/aluminum/tin	19
Figure 3.6	SASN explosive over aluminum covered tin sample	20
Figure 3.7	Field calibration curve fit of silver to tin ratio compared with silver only curve fit ...	20
Figure 3.8	Improvement of silver to tin ratio based measurement relative to silver only	21
Figure 3.9	XRF calibration sample to be coated with SASN explosive.....	21
Figure 3.10	Compilation of XRF calibration data and resultant curve fit	22
Figure 3.11	XRF measurement sensitivity to stand-off distance.....	23
Figure 3.12	XRF measurement spot size	23
Figure 3.13	Effect of measurement time.....	23
Figure 3.14	Hydrocode model of the effect of the tin reference material on mechanical shock results	24

NOMENCLATURE

CTH	Chart to the three halves, hydrocode program
DOE	Department of Energy
LIHE	Light Initiated High Explosive
SASN	Silver acetylide-silver nitrate
SNL	Sandia National Laboratories
keV	kilo-electron volts
XRF	X-ray fluorescence

Variables

ρ_{AD}	Areal density (mg/cm^2)
R	Ratio of silver to tin

1 INTRODUCTION

The Light Initiated High Explosive (LIHE) facility generates an impulsive load by detonating a spray-applied explosive coating. The magnitude of the impulsive loading is directly related to the quantity of explosive deposition. A deposition diagnostic is critical to the LIHE testing process. The simplest and most common explosive diagnostic are removable explosive samples or coupons. These coupons are magnetically attached on and/or near the impulsively loaded surface. These coupons are spray coated with the explosive material. The deposited explosive quantity is determined by weighing the coupon accounting for the tare weight of the uncoated coupon. From the measured explosive weight, knowing the surface area of the deposition, the areal density of explosive coating can be determined. The areal density is mass per area given in units of g/cm^2 . Explosive coupons are a proven and effective technique to determine explosive deposition.

However, there are test conditions and configuration requirements that prohibit the use of coupons. Coupons create a local impulse and pressure perturbation that may be unacceptable to the test customer. The presence of the coupon measurement technique actually modifies the measured phenomenon: local impulse and pressure. Additionally, removing and replacing coupons on delicate surfaces such as thin flyer plates may actually damage the surface or damage the explosive coating. For such test requirements, a non-contact explosive deposition diagnostic is required. After an industry search of available technologies in year 2007, an X-ray fluorescence (XRF) spectrometer was selected to measure the explosive material deposition in-situ.

The explosive coating is a silver based explosive with the silver content approximately 79% based on mass. The range of deposition is less than $15 \text{ g}/\text{cm}^2$ to approximately $40 \text{ g}/\text{cm}^2$ total mass or 12 to $32 \text{ g}/\text{cm}^2$ silver mass. The balance of the explosive is carbon, oxygen, and nitrogen. Prior to spraying the explosive, a thin Viton rubber film is applied with an areal density of 0.4 to $1 \text{ g}/\text{cm}^2$. The most common substrate sprayed with these coatings is aluminum foil ranging in thickness from 0.006 to 0.012 inch.

2 X-RAY FLUORESCENCE SPECTROMETER

On an atomistic scale, all atoms can be represented as a “cloud” of electrons surrounding a nucleus of protons and neutrons. These electrons occupy specific energy levels or orbits around the nucleus that is specific to each element in the periodic table. A stable, neutral atom exists with a balanced distribution of negatively charged electrons in orbit around the positively charged nucleus. The potential energy of each electron is dependent upon its orbital position. Only specific, discrete energy levels or orbit may be occupied by an electron for any specific element.

When an atom is exposed to X-rays of sufficient energy, a finite probability exists that an electron will absorb energy from the incident X-ray, exciting the electron sufficiently to break its electrical charge bond with the nucleus causing the electron to escape from the atom. The escaped electron leaves a vacancy in the orbital configuration of the atom. To fill the vacancy, an electron in a higher orbital configuration, with a higher potential energy, will move into the lower orbital vacancy, with a lower potential energy. To satisfy conservation of energy, when the electron drops into an orbit with a lower potential energy, it releases a photon of energy. This release of photonic energy is a photoelectric effect and is called fluorescence. The potential energy difference between electron orbits of a specific element is exact and constant. Therefore, the photonic energy released during electron orbit changes is also exact and constant for any particular element. The X-ray photons emitted during this process are called characteristic X-rays.

An element may have two to more than 10 orbital transitions possible within an atom depending upon the atomic number defining the total number of electron in the element. Therefore each element will emit a unique set of characteristic X-rays as a response to incident X-ray energy. By measuring the fluorescence X-ray spectrum, the elemental composition can be determined. When a material is subjected to a known intensity, the rate of fluoresced characteristic X-rays will be proportional to the concentration of the elements present in the material. Therefore the composition of the material can be quantitatively determined from the fluoresced X-rays.

In addition to the photoelectric effect described above, a second X-ray interaction can occur and is noteworthy. The incident X-ray is affected by the collision with the target electron. This collision may result in the total energy transfer from the incident X-ray to the electron that may be ejected from the atom. However, the collision may also result in the partial transfer of energy. The incident X-rays may be scattered by the interaction with the target material. The incident X-rays may change direction and lose energy to the electron. The resultant X-ray will have a different energy (wavelength/frequency). This X-ray interaction is called Compton scattering. Compared with the discrete and precise spectrum of fluoresced X-rays, Compton scattering produces a broad distribution of X-ray energies.

The X-ray fluorescence spectrometer operates by generating source X-rays that bombard the target material and then measures the fluoresced X-ray emitted from the target material. The instrument described in this report is the Fox-I instrument manufactured by Innov-X Systems. The instrument is shown in Figure 2.1 and Figure 2.2. The X-ray excitation source is a 35keV tungsten X-ray tube. This instrument uses a silicon PIN detector.

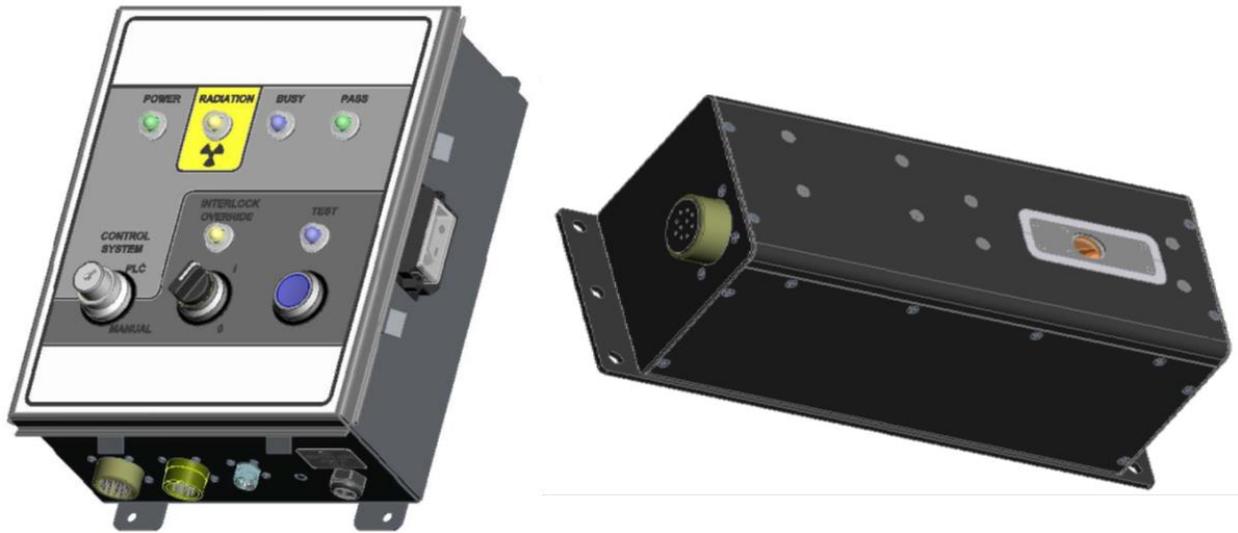


Figure 2.1 Fox-I X-ray fluorescence spectrometer

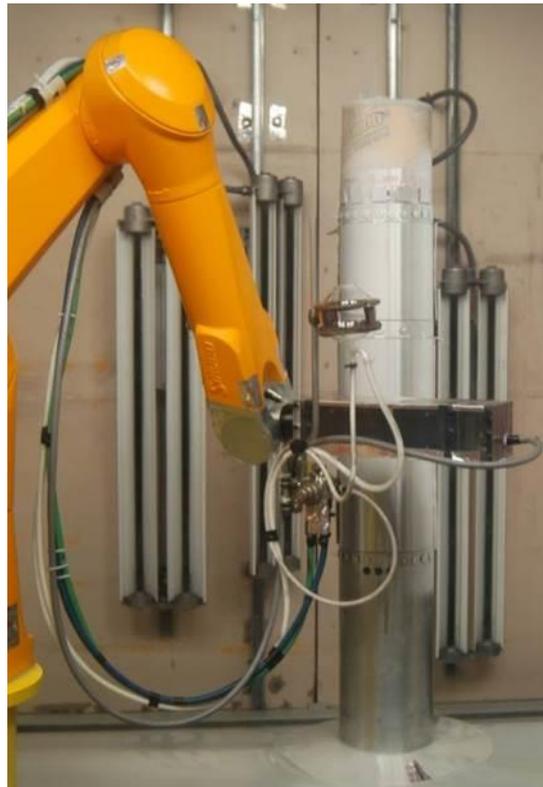


Figure 2.2 X-ray fluorescence spectrometer mounted on a robotic arm for in-situ explosive measurements



Figure 2.3 XRF spectrometer mount adjacent to the explosive spray gun on the robotic arm, explosives coupons are also shown.

3 X-RAY FLUORESCENCE MEASUREMENT DEVELOPMENT

3.1 Absolute Measurement

The X-ray fluorescence measurement technique evolved to achieve the required measurement accuracy. The initial data collected was based strictly on the absolute counts, or count rate, of silver fluoresced X-rays. This absolute measurement was inherently inaccurate. Any variability in source X-rays would correspond to variability in fluoresced X-rays. The interaction of source X-rays producing fluoresced X-rays in the direction of the detector was probabilistic in nature. And finally, any inaccuracy in spacing between the target and the detector would be evident in the accuracy of the X-rays measurement.

Another potential source of error was the penetration ability of the fluoresced X-rays. Silver will fluoresce 25.516 keV (Ka) X-rays. This energy of X-rays has an attenuation length of 60 μm into silver with a nominal density of 10.49 g/cm^3 . This corresponds to an equivalent silver areal density of 63 mg/cm^2 . As discussed previously, a silver areal density of 32 mg/cm^2 corresponds to a SASN areal density of 40 mg/cm^2 , a reasonable upper measurement limit for this application. Therefore, the silver thickness is not self-limiting for this measurement technique.

3.2 Compton Reference Measurement

In an attempt to improve the accuracy of the XRF measurements, a comparative reference was needed. By comparing the fluoresced measurements to a reference, sources of inaccuracy are counteracted. For example, variability in source X-rays that are manifest in fluoresced X-rays and the reference measurement offset. For the application described here, there were no other fluoresced X-rays in the spectral response range of the instrument. The silver based explosive was typically deposited on an aluminum substrate. However any aluminum fluoresced X-rays were beyond the range of the Fox-I instrument. Figure 3.1 shows the spectral measurement of SASN explosive on an aluminum substrate. The silver content of the explosive is evident in the two "K" shell orbits characteristic to silver. There are no other characteristic peaks measured by the XRF spectrometer. A very low level, broad spectral distribution is evident in the data. Therefore the measured silver spectrum was normalized to the total spectral measurement that included the Compton scatter.

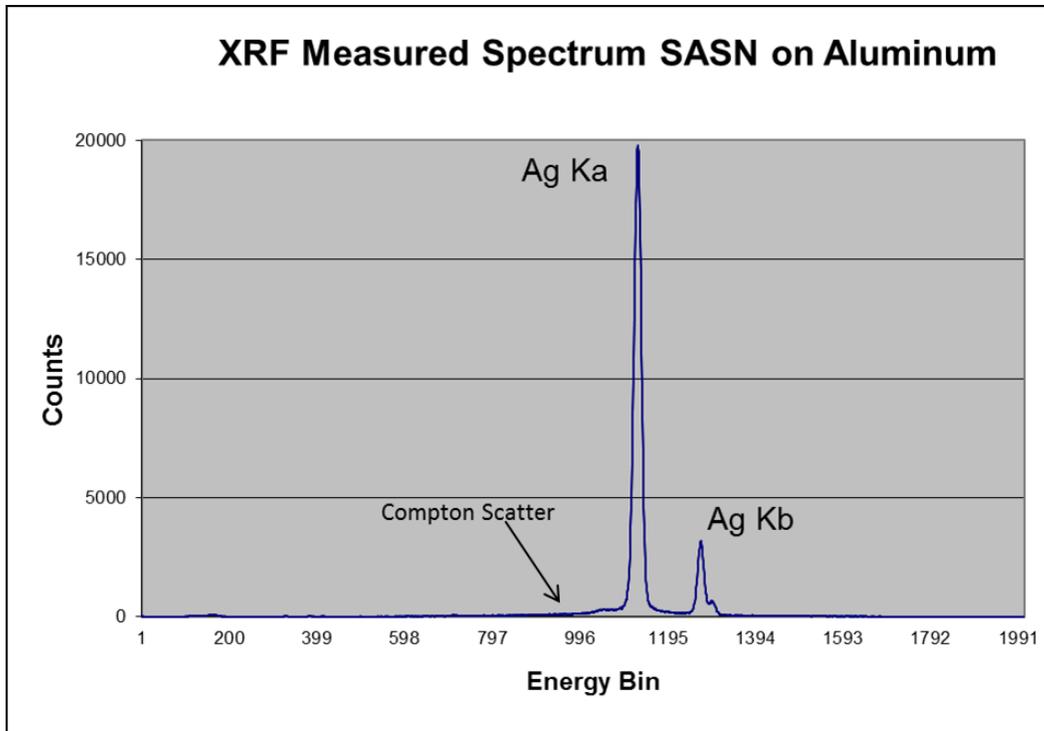


Figure 3.1 Spectral data (energy bins) for SASN explosive on an aluminum substrate

Using the reference methodology, a field calibration was initiated. Large 38mm diameter aluminum coupons were coated with SASN explosive. The coating was measured using the XRF instrument. Then the explosive areal density was determined from the explosive weight and the coupon area. From the explosive areal density and XRF data, a field calibration relationship was developed. The SASN XRF data relative to the measured areal density is shown in Figure 3.2. From this data set, a calibration relationship was developed shown by the curve fit. Using the developed field calibration relationship, areal density was calculated from the measured XRF data. The calculated areal density was compared with the measured areal density revealing significant inaccuracy, greater than 15%. The accuracy of the field calibration relationship is shown graphically in Figure 3.3.

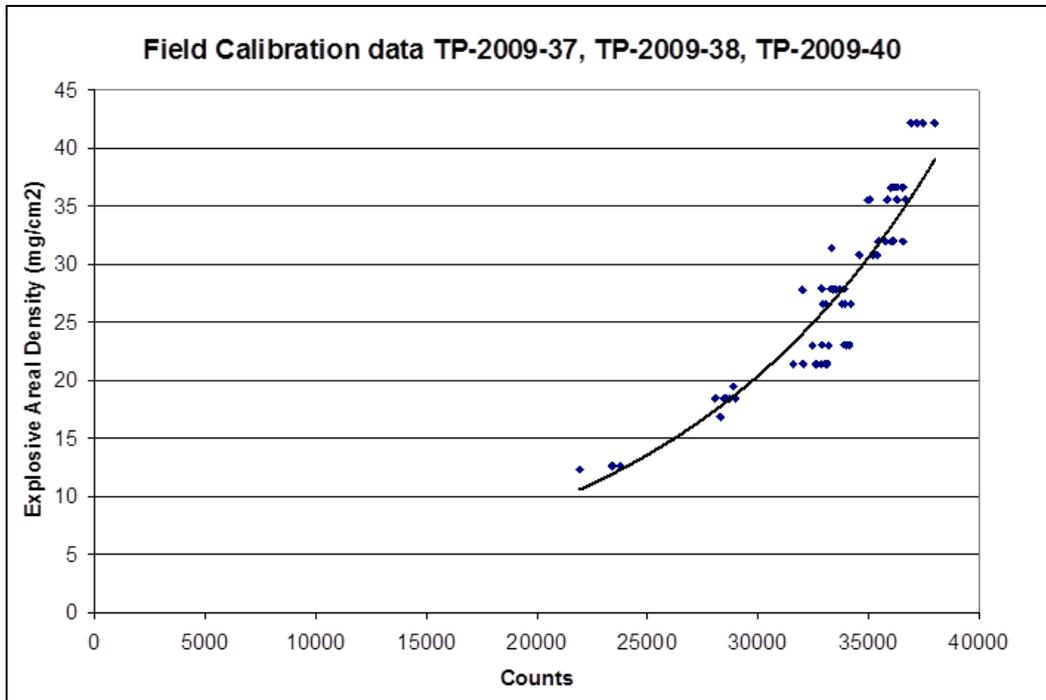


Figure 3.2 SASN XRF data relative to measured areal density.

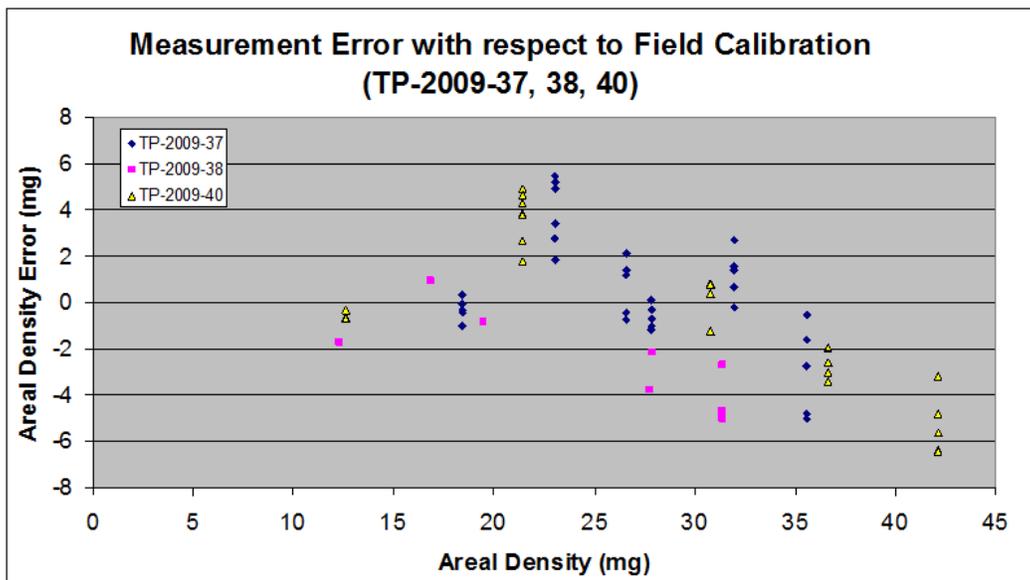


Figure 3.3 Error of areal density calculated from XRF data relative to measured areal density

3.3 Tin Referenced Measurement

The Compton scatter reference is a low level measurement greatly affected by the noise floor of the instrument. In order to improve the accuracy of the XRF measurement, an improved reference was needed. However, no reference was available in this application. Therefore a reference material was introduced into the measurement system. The reference material needed to meet several criteria. The reference material needed to yield characteristic X-rays that were in the measurement range of the XRF spectrometer. The fluoresced X-rays were required to have sufficient energies to penetrate through the silver based explosive. Compatibility of the reference material with the explosive material was required. The material needed to be pliable and conformal since the substrate is not always flat. The effect of the reference material on the explosive application must be minimized. Availability and cost of the material was also a consideration. Several elements were considered: tin, cadmium, antimony, barium, tellurium, iodine, platinum and gold. Elements were discarded from selection based on their X-ray energies and penetration depth through silver, such as gold films. Gold emits characteristic X-rays at 81 keV (K shell) which are beyond the measurement range of the Fox-IO instrument. Gold also emits characteristic X-rays at approximately 13 keV which are unable to penetrate through the silver explosive. Tin emits characteristic X-rays at approximately 29 keV (k shell). The tin X-rays are higher energy than silver and therefore more penetrating. Tin was the most promising candidate in terms of X-ray energy, pliability, availability and cost. However, tin was incompatible with the SASN explosive. Experiments demonstrated the degradation of SASN explosive in contact with tin. Figure 3.4 shows the explosive impulse degraded by approximately 19% due to interaction with tin. To overcome this limitation, a thin aluminum chemical barrier was applied over the tin. Variability in tin thickness could affect the XRF measurement unless the tin was “infinitely” thick. Based on X-ray energies, 60 μm tin was calculated as “infinitely” thick. A very inexpensive supply of 51 μm Tin was identified and selected as a suitable target reference material. In the final configuration, the Tin reference material was covered with 18 μm thick aluminum foil chemical barrier. Although aluminum characteristic X-rays are not measured by the XRF spectrometer, the aluminum foil will attenuate the characteristic X-rays emitted by the tin foil. Figure 3.5 shows the spectral measurements of 51 μm tin foil as well as a lay-up of 51 μm tin covered with a 12 μm silver foil. This figure also shows the attenuation effect of a thick (254 μm) aluminum layer between the tin and silver. Subsequent calibration efforts must include the aluminum barrier.

The initial measurements used silver, tin, and aluminum foils. Varying thicknesses of silver foil were placed on a 51 μm tin foil and 254 μm aluminum foil. Figure 3.6 shows the silver and tin measurements. As expected, the silver measurement increases with increased silver thickness. The tin fluoresced X-rays are attenuated as the silver quantity increases. By using the ratio of silver to tin, the overall measurement sensitivity is increased. A curve fit was obtained based on the silver to tin methodology. For comparison, a curve fit was also obtained for the unreferenced silver only data. Figure 3.7 shows the accuracy of the two methodologies with significant improvement using the silver to tin ratio.

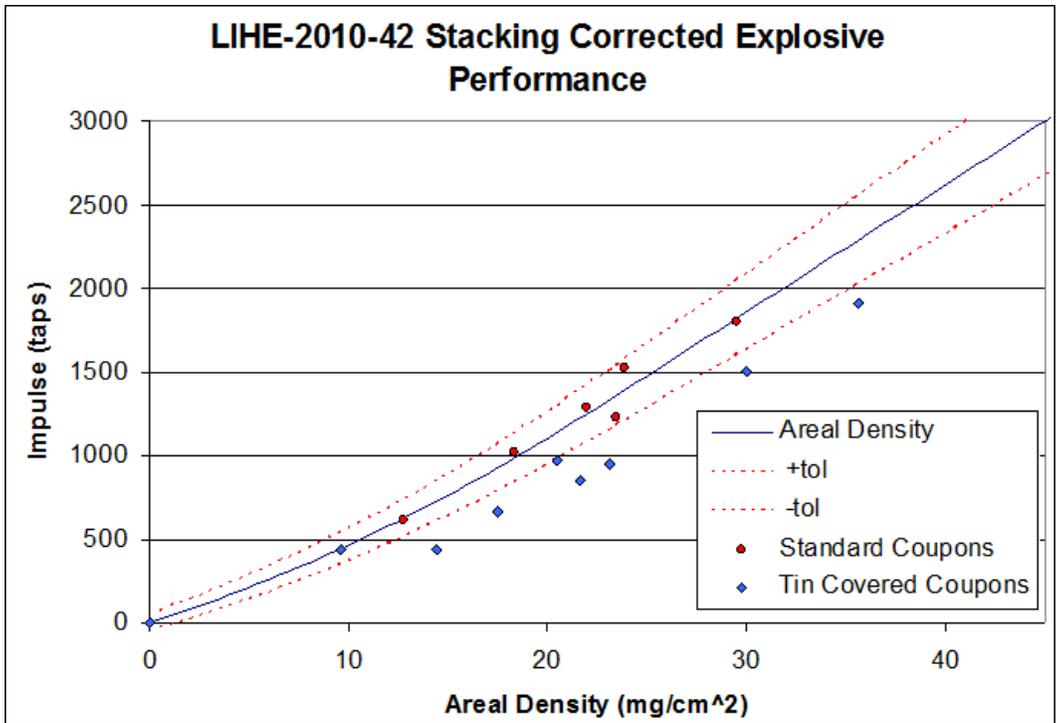


Figure 3.4 Explosive impulse degradation caused by tin

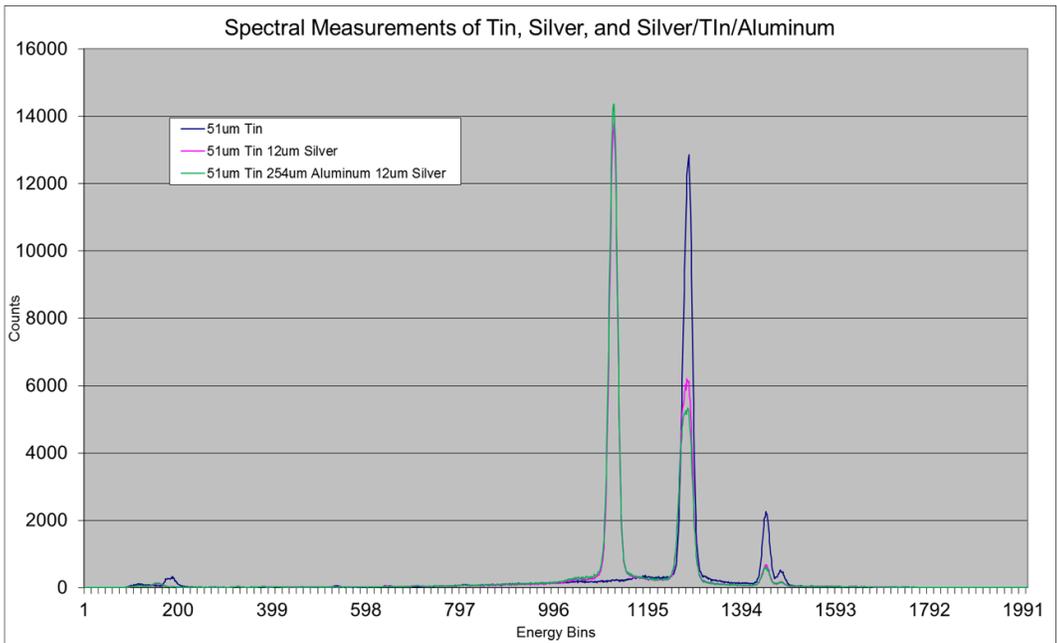


Figure 3.5 XRF spectral measurements of silver, silver/tin, and silver/aluminum/tin

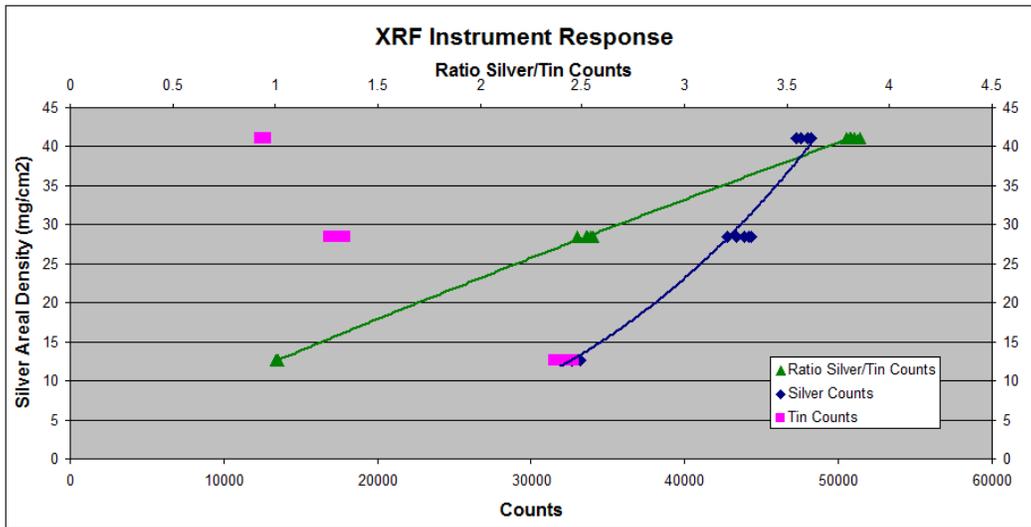


Figure 3.6 SASN explosive over aluminum covered tin sample

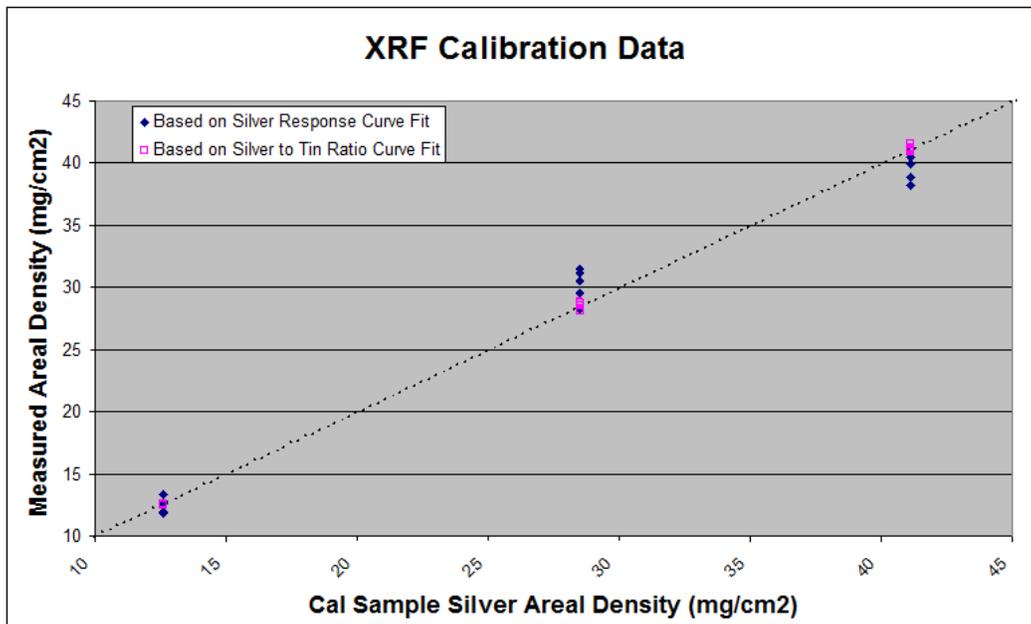


Figure 3.7 Field calibration curve fit of silver to tin ratio compared with silver only curve fit

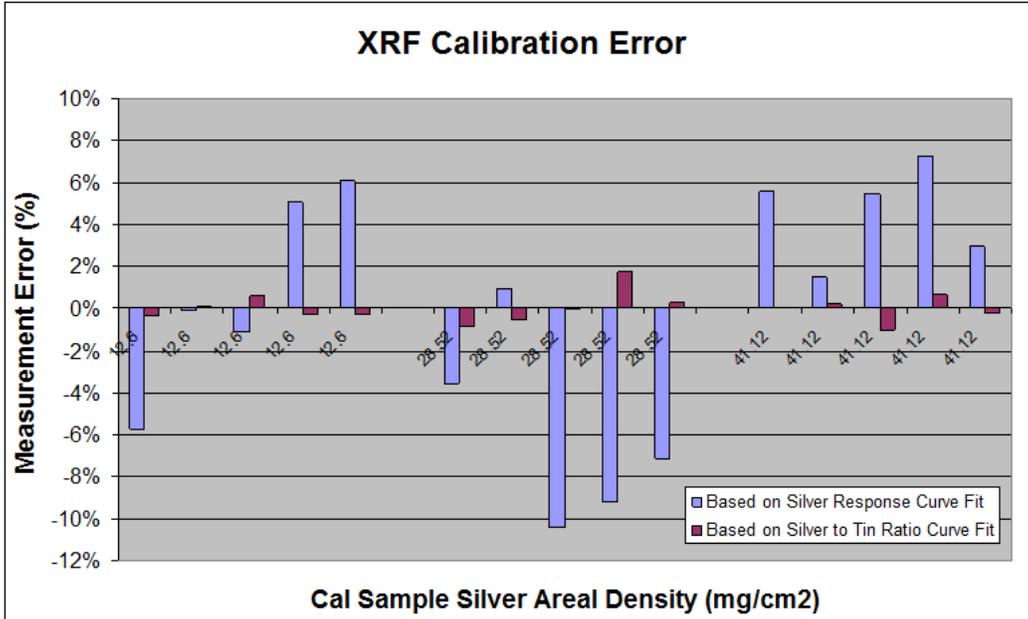


Figure 3.8 Improvement of silver to tin ratio based measurement relative to silver only

A field calibration effort was completed for SASN explosive measurements using a tin reference. The calibration samples that were coated with SASN are shown in Figure 3.9. The coated samples were measured with the Fox-I spectrometer. Then the explosive areal density was determined from the measured SASN weight and the known area. The calibration data is shown in Figure 3.10. The calibration relation is shown below, where ρ_{AD} is SASN areal density in mg/cm^2 and R is the measure silver to tin ratio. The overall standard deviation was 3.2%.

$$\rho_{AD} = 18.162 \cdot R^{0.8027}$$

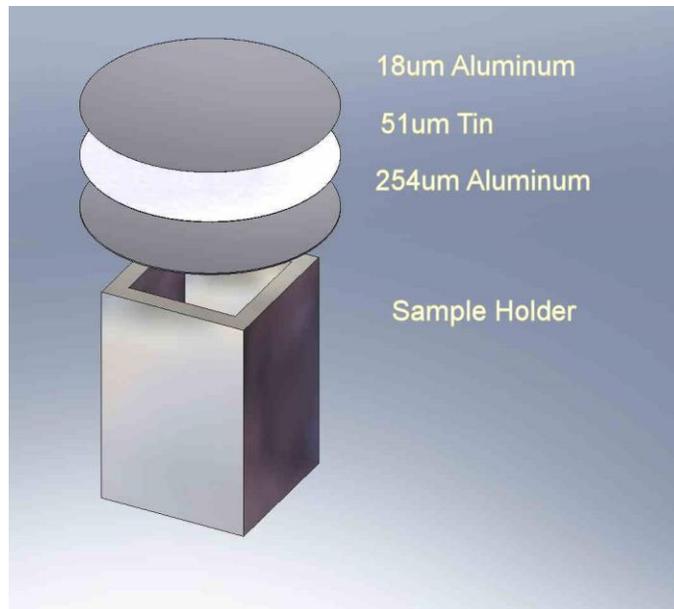


Figure 3.9 XRF calibration sample to be coated with SASN explosive

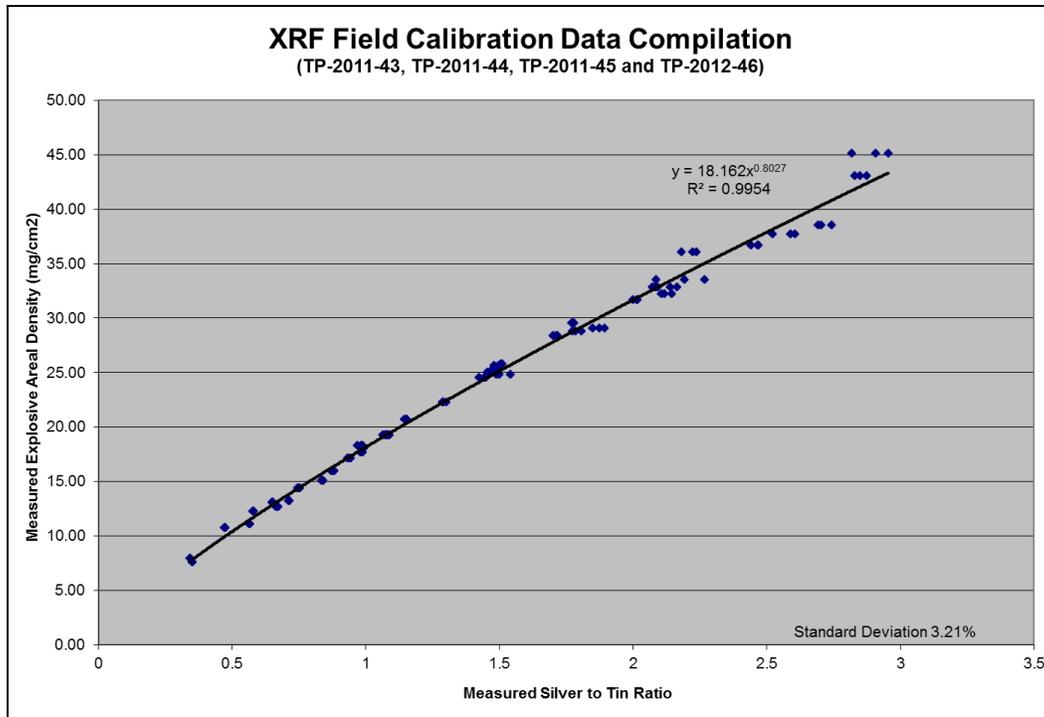


Figure 3.10 Compilation of XRF calibration data and resultant curve fit

3.4 Instrument Characterization

A number of instrument operating parameters and configurations were investigated to determine their affect the data: measurement instrument to target spacing, spot size, and measurement time. The instrument is mounted inside a protective enclosure on the end of a robotic arm. The nominal distance between the instrument measurement window (inside the enclosure) and the target surface is 9.5mm. From this nominal distance, measurements were made a varying offset distances. These measurements used SASN explosive over aluminum covered tin foil. The very limited data set is shown in Figure 3.11. The data indicate stand-off positional accuracy of approximately 1 mm is required. The XRF instrument measures over a finite spot size that is defined by an internal aperture and the stand-off distance. The spot size was determined by using a constant silver foil over varying diameter tin foils. The data shown in Figure 3.12 indicate a spot size of 19 mm. In order to accommodate inaccuracy in target preparation, a tin foil spot size of 25 mm will be used. Finally, the instrument measures the fluoresced X-rays over a prescribed time. When selecting the measurement time, measurement efficiency must be balanced with measurement accuracy. Figure 3.13 shows the effect of measurement time on measurement accuracy. For this particular SASN measurement, the standard deviation of the 60 second measurement was 0.7% compared to 1.5% for the 20 second measurement.

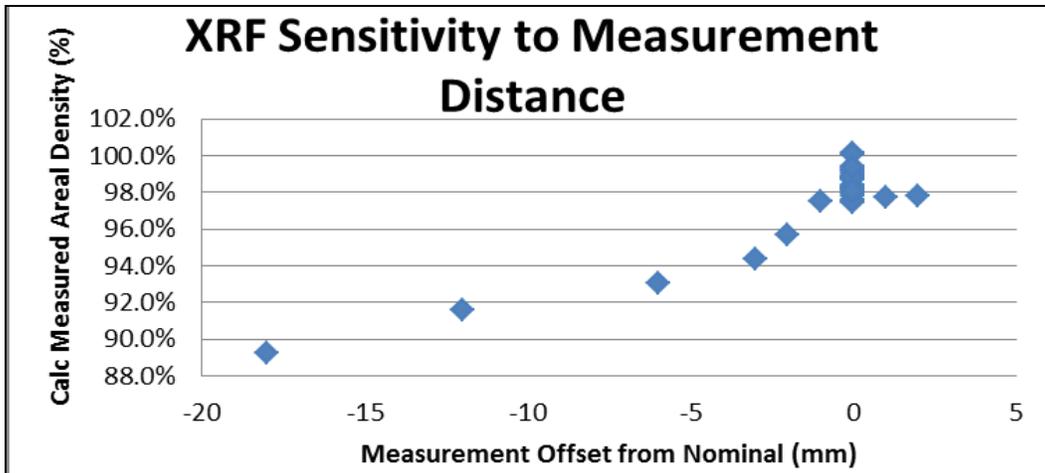


Figure 3.11 XRF measurement sensitivity to stand-off distance

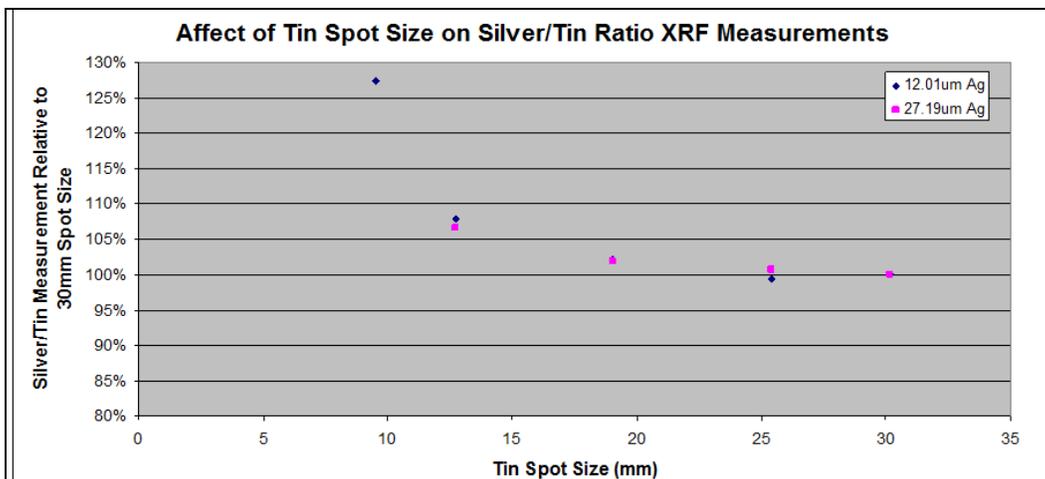


Figure 3.12 XRF measurement spot size

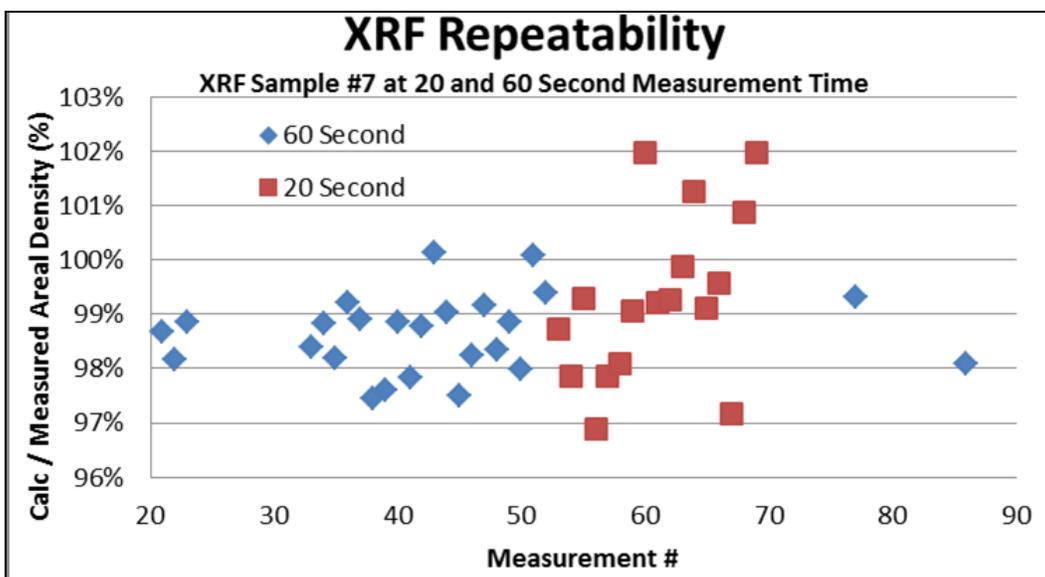


Figure 3.13 Effect of measurement time

3.5 Measurement Effect on Explosive Impulse Conditions

An application using SASN explosive is the impartation of a mechanical shock load into a target of interest. The SASN explosive may be used to accelerate a flyer plate to the required impact conditions. The insertion of the tin reference material affects and modifies the shock loading application. In a flyer plate application, the addition of the tin decreases the achieved flyer velocity due to the increased, localized mass of the flyer. The impact results are thus modified because the mass of the flyer is increased and the velocity of the flyer is decreased. A one-dimensional CTH hydrocode analysis was performed to examine the effect of the tin reference material on the resultant impact conditions. A 254 μm aluminum flyer was accelerated using a nominal SASN explosive model to achieve impulse and pressure conditions in an impacted tape would carbon phenolic (TWCP) target. The results are shown in Figure 3.14. The hydrocode model was modified by the addition of 50.8 μm of tin on the aluminum flyer as also shown in Figure 3.14. The tin layer increases the delivered impulse by 3%, but decreases the peak pressure by 13% and results in a 1 μs impact delay time.

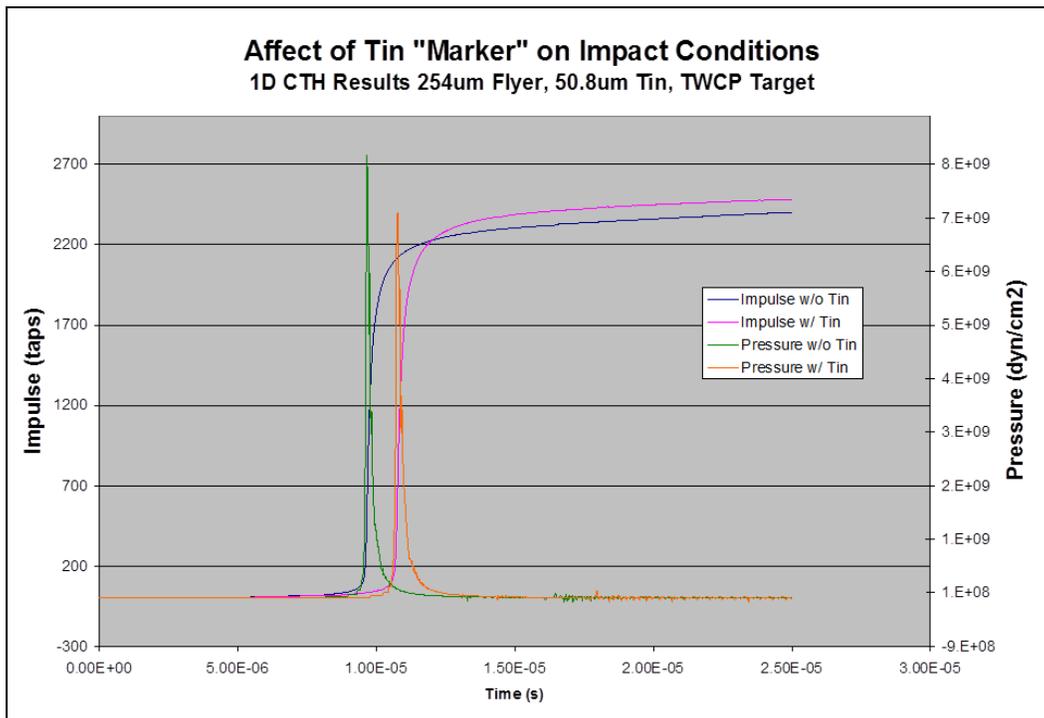


Figure 3.14 Hydrocode model of the effect of the tin reference material on mechanical shock results

4 CONCLUSIONS

The X-ray fluorescence spectrometer instrument and measurement technique have been successfully developed and demonstrated to measure the deposition of silver acetylide-silver nitrate explosive in-situ. In order to achieve the required accuracy a reference material has been incorporated into the measurement configuration. Tin has been selected as an appropriate reference material for this application. The incompatibility of the tin with the SASN explosive has necessitated the use of an aluminum foil chemical barrier. In the final configuration, SASN explosive is deposited on an 18 μm aluminum foil barrier that covers the tin reference layer. As the SASN layer increases, the XRF measures increasing silver content while the tin X-rays are increasingly attenuated by the silver. By using the silver to tin ratio, the sensitivity is greatly improved. A field calibration relationship has been developed with a standard deviation of 3.2%. The insertion of the tin affects the mechanical shock imparted in this application and has been modelled with CTH hydrocode to examine the effect.

5 REFERENCES

1. "Technology Guide, X-Ray Fluorescence Metrology Tools," Matrix Technologies, Revision 08, 2004.
2. "Factory On-Line System: Fox-I Series, User Guide," Innov-X Systems, Revision A.1, 2007.

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