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## **LDRD 140639 Final Report: Investigation of Transmutation Claims**

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## **Abstract**

The Proton-21 Laboratory in the Ukraine has been publishing results on shock-induced transmutation of several elements, including Cobalt 60 into non-radioactive elements. This report documents exploratory characterization of a shock-compressed Aluminum-6061 sample, which is the only available surrogate for the high-purity copper samples in the Proton-21 experiments. The goal was to determine Sandia's ability to detect possible shock-wave-induced transmutation products and to unambiguously validate or invalidate the claims in collaboration with the Proton-21 Laboratory. We have developed a suitable characterization process and tested it on the surrogate sample. Using trace elemental analysis capabilities, we found elevated and localized concentrations of impurity elements like the Ukrainians report. All our results, however, are consistent with the ejection of impurities that were not in solution in our alloy or were deposited from the cathode during irradiation or possibly storage. Based on the detection capabilities demonstrated and additional techniques available, we are positioned to test samples from Proton-21 if funded to do so.



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## NOMENCLATURE

AES	Auger–electron spectroscopy
EDS	energy dispersive spectroscopy
EPMA	electron probe microanalysis
FIB	focused ion beam
g	gram
ICP	inductively coupled plasma
kA	kiloamp
keV	kiloelectron volts
LMS	laser mass spectrometry
micron	( $\mu\text{m}$ ) millionth of a meter
m/z	mass to charge ratio (in mass spectrometry)
MS	mass spectrometry
nm	nanometer
ppb	parts per billion
ppm	parts per million
RBS	Rutherford back scattering of accelerated alpha–particles
SEM	scanning electron microscopy
SIMS	secondary ion mass spectrometry
SNL	Sandia National Laboratories
TOF	time of flight (mass spectrometry)
Z number	atomic number (number of electrons)

### Chemical Element Abbreviations Used:

Al	aluminum	Mn	Manganese
B	boron	Na	sodium
Ba	barium	Ni	nickel
Bi	bismuth	O	oxygen
Ca	calcium	P	phosphorous
Cd	cadmium	Pb	lead
Cl	chlorine	S	sulfur
Cr	chromium	Si	silicon
Cu	copper	Sn	tin
Fe	iron	Ti	titanium
Ga	gallium	Zn	zinc
K	potassium	Zr	zirconium
Mg	magnesium		

## 1. INTRODUCTION

The Proton-21 Laboratory in Ukraine has for many years published theoretical and experimental articles on a shock compression mechanism for the transmutation of elements [1-4]. They focus a moderately intense beam of electrons onto a hemispherical target to produce a converging shock wave in the target material. A significant investment in time, equipment, and analysis is clear. A host of chemical analysis techniques has been applied to a variety of metal targets that have been irradiated with very high energy ( $> 400$  KeV), high current ( $\sim 30$  KA), focused electron beams in short bursts on the order of 50 nanoseconds. It should be noted that few of the experimental articles are in peer reviewed journals. The following lists some of the techniques utilized to demonstrate and validate claims of transmutation:

EPMA — electron probe microanalysis;  
AES — Auger–electron spectroscopy;  
SIMS — secondary ion mass spectrometry;  
LMS — laser mass spectrometry;  
RBS — Rutherford back scattering

The reports suggest many thousands of target irradiations, and the data includes quantitative measurements of changes in the detected elements as well as isotopic measurements that show quite radical changes in the distribution of isotopes such as nickel and iron from a copper target. It is difficult to separate exactly which targets were used for some of the data, and some tables have so many detected elements both before and after irradiation that the purity claim of some targets is questionable.

The facility has also published what is apparently marketing-type literature with a plethora of images and analysis results and filed for a United States patent in 2004 which has not to date been granted[5].

The claims published include using:

1. 99.99% pure copper targets and obtaining tungsten, gold, bismuth, and silicon [4]
2. platinum/bismuth targets and obtaining “unknown” superheavy elements
3. cobalt 60 targets and obtaining reduced quantities of beta particle emission
4. unidentified peaks in Auger analysis suggest the production of long-lived transuranic elements[6]

While the claims may seem to some quite implausible, the possibility of non-nuclear mechanisms for transmutation deserves investigation. The goal of this project was to take a Sandia target previously irradiated with an electron beam of similar energies and determine if we could develop the analysis process to unambiguously validate or invalidate their claims. We would also see if any transmutation could be detected in our sample. The only available Sandia target is an alloy and serves as a ‘dry-run’ for future testing of Ukraine targets or possible future controlled energetic beam tests at Sandia. The

Results and Discussion section of this document presents materials characterization data that show elevated and localized concentrations of impurity elements in the SNL sample, which do not suggest transmutation, however demonstrate the capability to detect such products in follow-on work. Lessons-learned via the Sandia target are also discussed; these lessons should be taken into account to ensure scientifically sound and verifiable measurements of any future suspected transmutation products.

## 1.1 SNL Sample Background

An aluminum alloy (Al 6061) target was irradiated by a pinched electron beam of energy 2 MeV and current 400,000 amps. The deposited energy generated a strong shock wave, material heating, and a crater as a result of the molten material redepositing and solidifying on the anode surface—qualitatively similar to that reported in the Proton-21 experiments. While the irradiation occurred almost 30 years ago, the target was still in hand (see photos below). It is approximately 8 inches across.



**Figure 1. Photo of the SNL aluminum anode with a closeup of the resulting crater area in the center.**

Unfortunately Al 6061 is not remotely chemically pure, and may contain a wide variety of other trace elements as shown in Table 1. On the other hand these impurities present an opportunity to demonstrate the capabilities of the Materials Characterization Department 1822 to analyze the Ukrainian targets. The ability to measure the specified and unknown trace level impurities is critical to the success of measuring low levels of transmutation products.

**Table 1: Chemical composition limits for Al 6061 specified by ASTM B308/B308M [7].**

<b>Element</b>	<b>Composition (%)</b>
Silicon	0.4-0.8
Iron	0.7 maximum
Copper	0.15-0.4
Manganese	0.15 maximum
Magnesium	0.8-1.2
Chromium	0.04-0.35
Zinc	0.25 maximum
Titanium	0.15 maximum
Other (unspecified elements)	0.05 each
Total (of unspecified elements)	0.15 maximum
Aluminum	Remainder

It was determined in the course of analysis that a brass component was part of the Sandia electron beam test. Brass comes in many styles and compositions depending upon its desired function(s) and form. For reference there are many specifications listed in ASTM B249/B259M -06 for copper alloys [8]. Three examples are listed below for different applications: rod/bar, naval castings, and rod for machine screws:

1. ASTM method B453/B453m, the “Standard Specification for Copper-Zinc-Lead Alloy (Leaded-Brass) Rod, Bar, and Shapes”, specifies that leaded brass may contain copper in a range of 61.5 to 63.5%; zinc from 35.3 to 37.1%; and lead from 0.5 to 2.5% [9].
2. Mil spec MIL-B-17511A, for naval castings, specifies brass as 60-65% Cu, 0.5-1.5% Sn, 0.75-1.5% Pb, max 0.75% Iron, max 0.5% Al, and the remainder Zn [10].
3. ASTM method B16/B 16M – 05, the “Standard Specification for Free-Cutting Brass Rod, Bar and Shapes for Use in Screw Machines”, specifies copper from 60.0 to 63.0%; lead from 2.5 to 3.7%; iron at a maximum of 0.35%; and the “remainder” as Zinc [11].

None of the specifications provides or suggests what other impurities are present. Since the grade of brass used in the test is unclear it was impossible to predict what elements--in addition to CU, Zn, and Pb--may or should be present in the redeposited material.

## 2. EXPERIMENTAL DETAILS

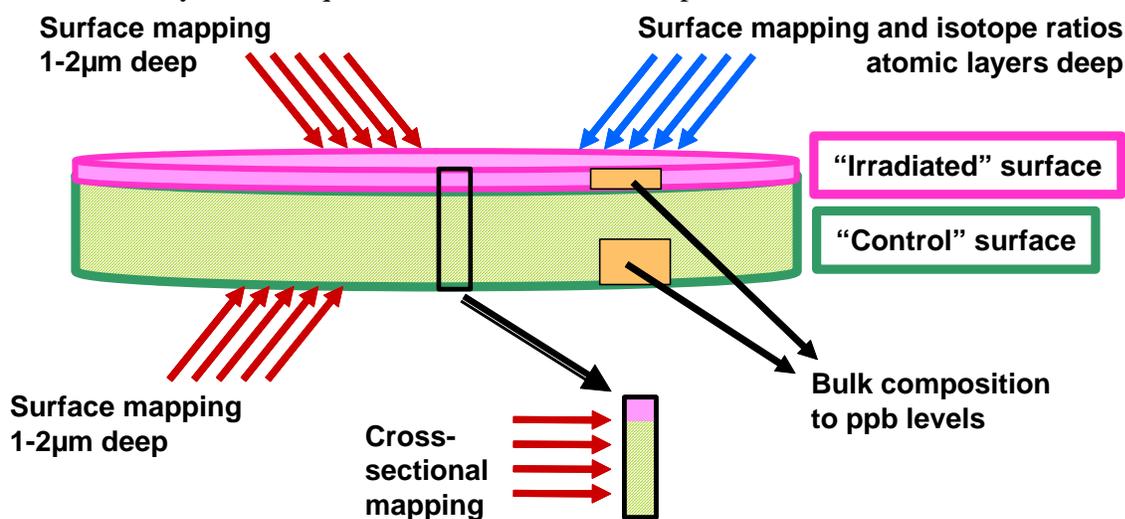
### 2.1 Aluminum target

The SNL disc was sectioned and portioned for subsequent analysis following the initial screening by SEM-EDS. The photo below shows the initial section taken.



**Figure 2: Photo of section cut from SNL Al sample for sub-sampling.**

The following schematic illustrates the general locations of measurements performed on the SNL sample. The backside of the disc serves as the sample control when looking for trace elements. Surface mapping, isotope ratios, bulk composition, and cross-sectional mapping refer to the elemental analysis techniques described later in the report.



**Figure 3: Schematic illustrating general measurement locations.**

### 2.2 Copper samples

Several high purity copper samples were tested by inductively coupled plasma mass spectrometry (ICP-MS) as described in the next section. These included:

1. Wire: Materials Research Corp. (Orangeburg, NY), stock 29/29/199/050 lot 29/25689; 0.050in. diameter wire grade MARZ.
2. Large copper wire with grain structure suggesting moderately high purity
3. Copper powder
4. 99.6% copper

Replicate portions of wire or powder were weighed and dissolved in high purity nitric acid for further analysis. Solution control samples and blanks were also analyzed to ensure data quality.

## 2.3 SEM-EDS

A Scanning Electron Microscope (JEOL 5900LV SEM) using a Noran System Six energy dispersive spectroscopy (EDS) system was used to determine compositional information from a depth of approximately 1-5 micron depending on material and beam energy. An electron beam (15 kV) is directed at the material of interest, which yields a spectrum of x-ray radiation that is element specific. The technique is non-destructive, and produces both a microscopic image of the material and the elemental composition of various spots within that image. The spot size varies depending on the magnification.

## 2.4 ICP-MS

Elemental analysis was performed using a Perkin-Elmer (Waltham, MA) inductively coupled plasma mass spectrometer (ICP-MS, model Elan 6100 DRC). This instrument utilizes a quadrupole mass analyzer for measurement and quantitation. Detection limits are in the parts-per-billion (ppb) range for most elements.

Three pieces of "backside" bulk and "frontside" flaked samples were cut from the SNL target. The frontside sample is referred to as "flaked" because a portion of the molten re-deposited layer could be removed from the surface of the disc.

Samples analyzed were three ~0.1g pieces of the disk and one 0.025g sample of the redeposited material from the surface of the disk. Samples were prepared by digestion in Optima hydrochloric and Optima hydrofluoric acids. Samples were then analyzed semi-quantitatively by ICP-MS. This technique identifies metals that are present and gives an approximate concentration for each identified metal. The accuracy is nominally +/-25% of the value. All values are given in ppm ( $\mu\text{g/g}$ ) of the original material. Due to the digestion procedure a value for silicon could not be determined.

## 2.5 TOF-SIMS

A TOF.SIMS 5 instrument (ION-TOF GmbH, Münster, Germany) performing time-of-flight secondary ion mass spectrometry (TOF-SIMS) used a 25kV Bismuth 1+ primary ion beam to scan a 30 by 30 micron area. Data was collected at 50 primary shots/pixel to generate a 256 by 256 datapoint matrix for each spectrum. This results in an analysis of approximately 3 atomic layers (~1 nm) of the surface, and can resolve oxides a couple nm deep. Averaging of 5.5 minutes per data point was performed.

### 3. RESULTS AND DISCUSSION

#### 3.1 SEM-EDS Elemental Analysis

The advantage of the scanning electron microscopy with energy dispersive spectroscopy (SEM-EDS) technique is that no sample preparation is required and that large areas can be screened in a relatively short time. While not quantitative it provides both an image and a chemical element map of the surface. Image contrast is approximated by elemental Z number, for example the instrument gives a brighter spot for Fe (see Figure 4, right hand side, spot labeled “7”) than it does for Si (same image, spot #2). The corresponding energy spectra for those same spots are shown respectively in Figure 5 and Figure 6. Some elements give emission at overlapping energies, but often there is a second emission line than can be used to provide confirmation.

The roughness and diverse morphology is shown at these magnifications. Higher magnification is possible as well as the ability to cross-section very small features using focused ion beam (FIB) techniques. When quantitative results are desired an Electron Microprobe that utilizes wavelength dispersive detectors is employed as well as material reference standards. SNL has both FIB and wavelength dispersive detection capabilities.

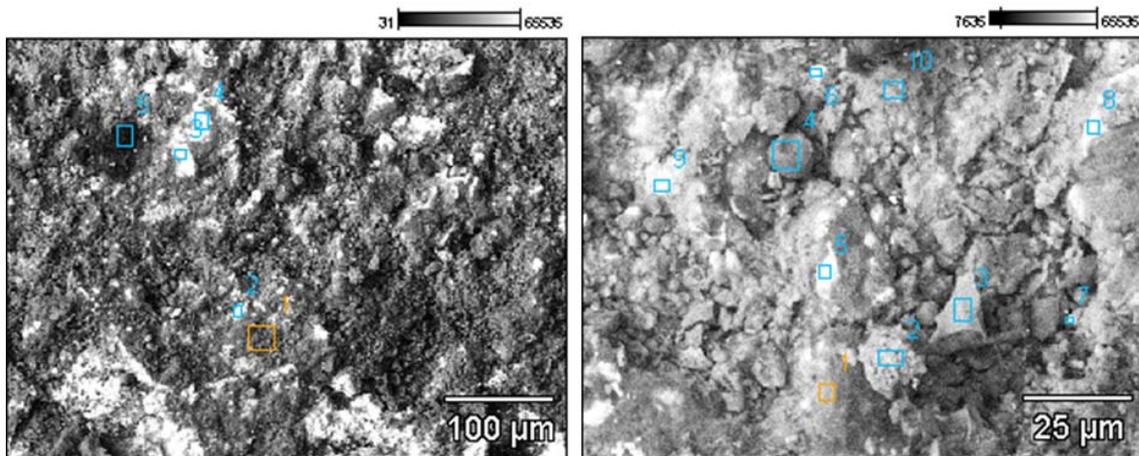


Figure 4. SEM images at 250x and 1000x magnification (views designated 4 and 5) with indication of sub-areas analyzed for elemental composition by EDS.

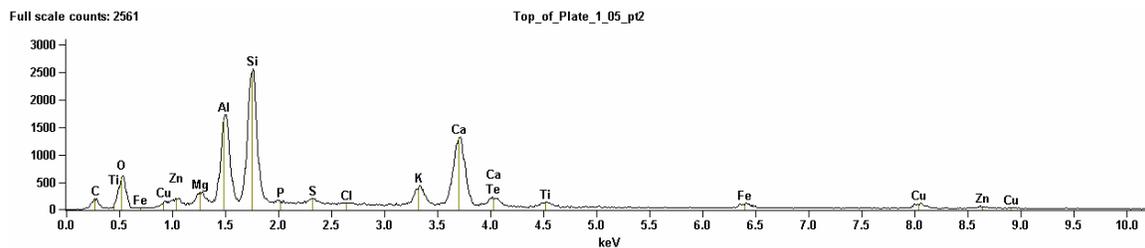
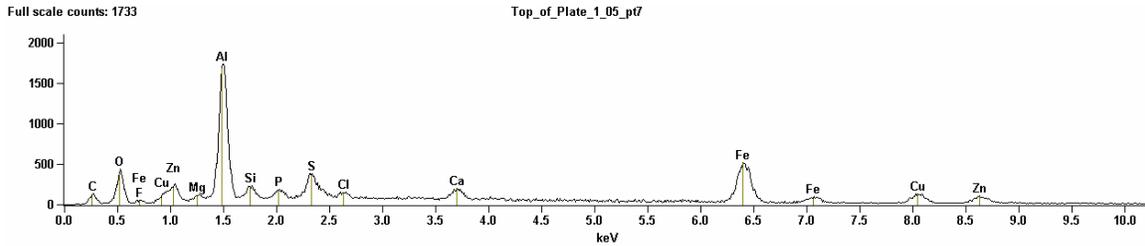


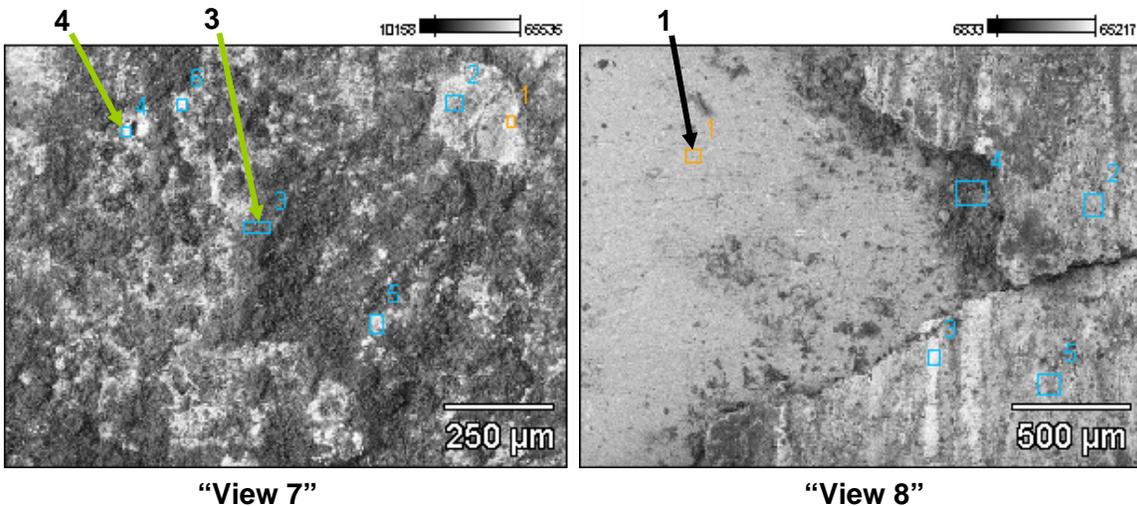
Figure 5: EDS spectrum, counts versus keV, of View 5 point #2, illustrating Si-rich surface.



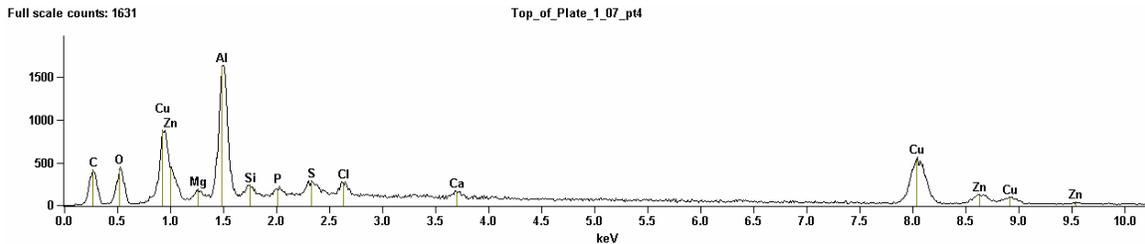
**Figure 6: EDS spectrum, counts versus keV, of View 5 point #7, illustrating Fe-rich surface.**

The image on the left side of Figure 7 (100x magnification, “View 7”) shows the typical surface of the redeposited material on the frontside. The darker areas, for example point #3, are primarily Al. The brighter point #4 (see spectrum in Figure 8) shows evidence of the brass (primarily Cu and Zn) hardware that held parts of the SNL experiment.

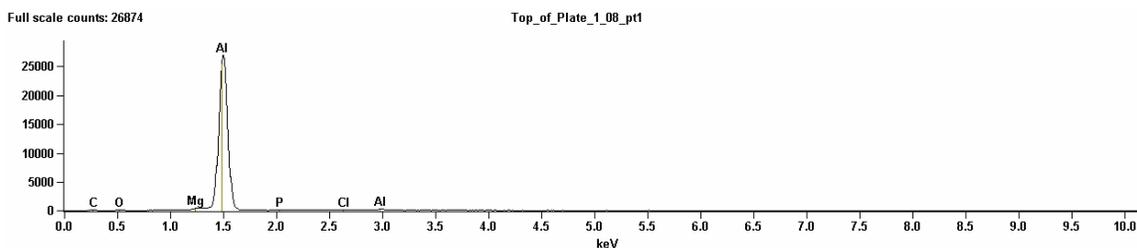
On the right hand side of Figure 7 is a lower magnification view (55x magnification, “View 8”) of a portion of the Al sample where some of the re-deposited material has flaked off over time. This area is at about “11 o’clock” in the macro photo (Figure 1). The EDS spectrum (Figure 9) taken at spot #1 in this image shows almost exclusively aluminum. The EDS spectrum collected in an area scan mode of the backside of the sample is very similar.



**Figure 7. SEM images (views designated 7 and 8) with indication of sub-areas analyzed for elemental composition.**

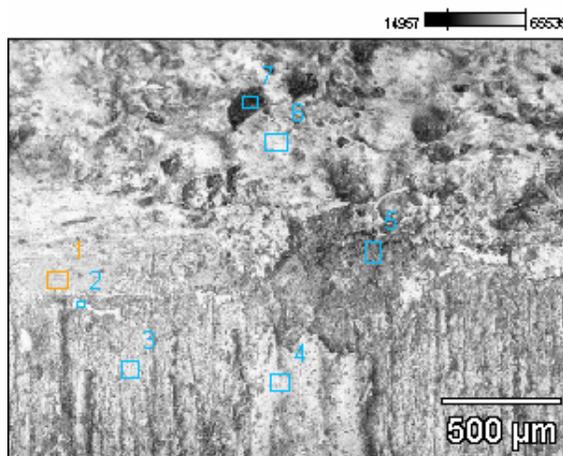


**Figure 8: EDS spectrum, counts versus keV, of View 7 point #4, illustrating prevalent Cu and Zn peaks.**



**Figure 9: EDS spectrum, counts versus keV, of View 8 point #1, illustrating Al surface under re-deposited layer.**

Analysis of several points near the center hole, which clearly has the morphology of a melt (Figure 10), shows primarily Al with much less frequent detection of the Cu and Zn and other elements that were detected farther out from the center.



**Figure 10: SEM image, 55x magnification, (designated View 9) with indication of sub-areas analyzed for elemental composition.**

Given that an unknown composition of brass was used, yet that most common brass grades have Pb, the reader may note the absence of any Pb peaks in these SEM-EDS experiments. In short it is a combination of detection limits, sampling volume, melt and re-deposition characteristics that affect detection of any element. Pb was readily detected, however, in the bulk measurements made using ICP-MS (next section). This points out the benefit of using multiple techniques in the scenario of searching for transmutation products – no one technique is “all knowing” about any sample.

The sample has been unprotected for 30 years. Therefore, surface contamination is the most likely source for the Ca, Si, Cl, P, Mg, and S detected. Obviously, the care of future samples must avoid such contamination. It was deemed unnecessary in the uncontrolled context of this sample to perform additional SEM or FIB/SEM characterization work to elucidate whether these elements were indeed surface contamination or originated in the original experiment.

A summary of the elements other than aluminum detected in this relatively quick sample survey are listed in Table 2. A “Tr.” entry indicates that a small (trace) peak was observed in the EDS spectrum. This does not imply a true quantity as this is a non-quantitative method. This table also provides additional support for the need of multiple techniques, as some of these elements were not detected by other methods. Again, this is due to the combination of limited sampling

(number of spots), sampling volume (of instrument), instrument response factors, and detection limits.

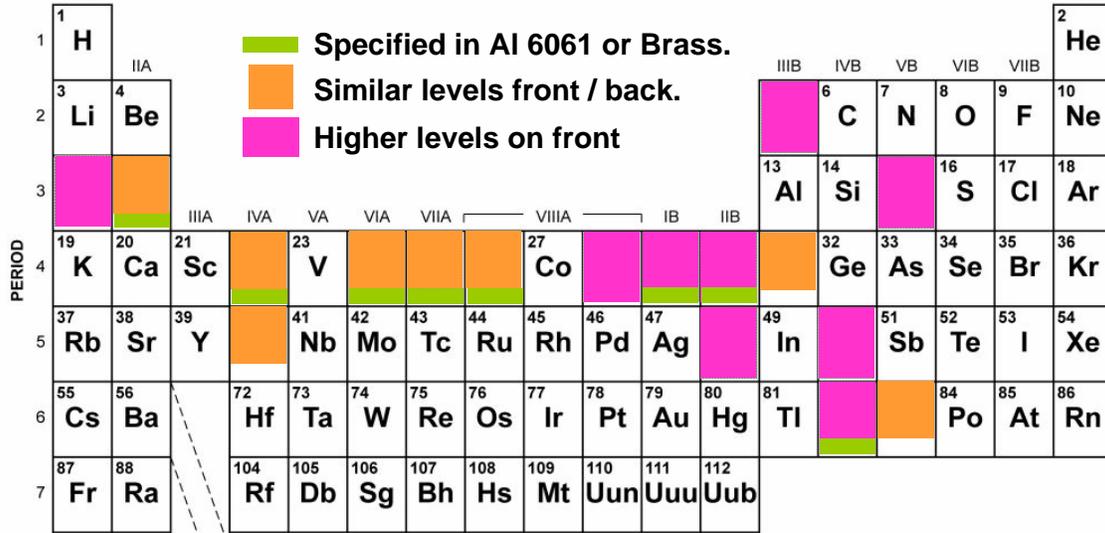
**Table 2. Elements other than Al observed in SEM-EDS experiments (front side of target).**

View / spot(s)	Cu	Zn	O	Fe	Addl. trace
4 / 1, 2	x	x	x		Mg, P, S, C, Ca
4 / 3, 4	x	x	x		C, Si, P, S
4 / 5	x	x	x		Ca, C, Ti/Ba, K, S, Cl
5 / 1, 2	x	x	x	x	Ca, K, Si
5 / 3, 4	x		x		Ca, Cl
5 / 5, 6	x	x	x		Si, S, P, Ca, C
5 / 7, 8	x	x	x	x	C
5 / 9, 10	x	x	x		S, Ca, C
7 / 1, 2	x	x	x		C
7 / 3, 4	x	x	x		C, Mg, Si, P, S, Cl
7 / 5, 6	x	x	x		Si, S, Cl, Ca
8 / 1, 2	Tr.	Tr.	Tr.		
8 / 3, 4	x	Tr.	Tr.		Si
8 / 5	x	Tr.	Tr.		Cl, C
9 / 1, 2	Tr.	Tr.	Tr.	Tr.	C
9 / 3, 4	x	x	x	?	Si, S, Cl, C
9 / 5, 6	Tr.	Tr.	x		C, Cl, S, Si
9 / 7	x	?	Tr.	Tr.	C, Cl, Ca

### 3.2 ICP-MS Elemental Analysis

Inductively coupled plasma mass spectrometry (ICP-MS) results were collected in a “semi-quant” mode, which means numbers were determined using an average instrument response factor rather than element-specific response factors. When more precise levels are needed more laborious (and therefore expensive) quantitative methods are used. It is common to do a “screening” measurement to save the labor of multiple calibration curves on unknown samples. If needed, a single sample preparation can also be used for the quantitative analysis.

A quick summary view of the ICP-MS results is given in Figure 11 in the form of an abbreviated periodic table of the elements. The elements shaded orange (triangle) were detected at similar levels front/back in the Al sample, whereas pink (square) indicates higher levels on the front. Green rectangles indicate those elements that are “allowed” by the specifications for Al 6061 and (common) leaded brass. Note that the specifications leave room for “other” and since the exact brass composition is not known, it is unclear whether the pink (only) shaded elements provide evidence for transmutation. Additional input from metallurgists familiar with brass may be helpful. Additional sampling points and full quantitative testing of these sample points would give tighter confidence levels.



**Figure 11. Summary of elements detected by ICP-MS shown as periodic table graphic.**

Actual detection levels in parts per million (ppm or microgram/gram) for those elements that were detected at similar (Table 3) and higher (Table 4) levels on the front (exposed) compared to the back (unexposed) are given in the following tables. Elements indicated with an asterisk (\*) are those listed in specifications for Aluminum 6061. Each of the three portions sampled of the back are listed individually to provide an indication of the macro level uniformity. Because the sample size is larger for ICP-MS analysis, the heterogeneity observed will be less than with SEM/EDS techniques. The redeposition of copper and zinc from brass components of the original experiment are clearly evident for example in the 27 times higher levels of Cu from front to back.

**Table 3: Metals detected at similar ppm levels front-to-back (\*=specified in Al 6061).**

Element	Back 1	Back 2	Back 3	Front Redeposited Material
* Mg	9300	11100	10100	11800
* Fe	4100	4300	3300	3800
* Cr	650	660	580	630
* Mn	280	270	220	270
Ga	89	110	110	94
* Ti	83	85	75	77
Bi	52	56	50	51
Zr	15	9	9	10

**Table 4: Metals detected at different ppm levels front-to-back (\*=specified in AI 6061).**

<b>Element</b>	<b>Back 1</b>	<b>Back 2</b>	<b>Back 3</b>	<b>Front Redeposited Material</b>
* Cu	2400	2400	2000	65000
* Zn	340	340	270	30000
Pb	190	210	180	4100
P	<5	<5	<5	320
Na	<5	<5	<5	260
Sn	8	8	8	210
Ni	50	54	42	170
B	<5	5	5	27
Cd	<5	<5	<5	14

As discussed in the SEM-EDS results, multiple instruments/techniques are needed when searching for unknown or outlier elements. ICP-MS does not detect Cl well, which was seen in SEM-EDS, and likewise SEM-EDS did not detect Ti nor Bi. These discrepancies are not errors nor flaws, but fall to factors such as sampling volume(s) and detection limits.

Additional work was performed on several high-purity copper materials in order to get an understanding of what common impurities might be found among them. Normal specifications ordinarily have a “miscellaneous” or “other” category for impurities; therefore, results from these direct measurements help define future experiments. Results of replicate measurements are summarized in the table below. Note that the highest specified purity sample (99.999% powder) which is actually an instrument reference sample, has the highest number of elemental impurities. Their concentrations are all very low, but there are many there. Note also that the 99.6 wire and high purity cable have quite a large variance in their total impurity values among the three replicate analyses – this indicates that the samples are not homogeneous.

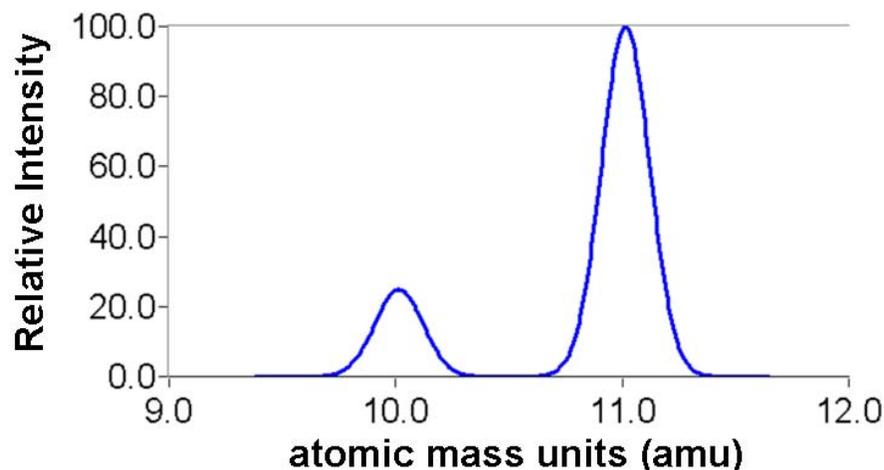
These are somewhat “bulk” samples compared to the very small volumes sampled by SEM and other surface techniques, yet variance (heterogeneity) of the material is apparent. It is likely that this variance could potentially be more drastic at smaller analysis volumes. This highlights the importance of thorough characterization of samples prior to high energy bombardment

**Table 5: Summary of trace elemental analysis of several high purity copper samples (all values in ppm).**

	99.999 Powder			Marz Wire			99.6 Wire			High Purity Cable		
Element	Powder-3	Powder-2	Powder-1	Wire-3	Wire-2	Wire-1	Wire-2	Wire-3	Wire-1	Cable-1	Cable-2	Cable-3
Ag	0.277	0.284	0.261	0.196	0.205	0.196	19.352	20.376	18.411	0.144	0.148	0.167
Al	0.778	0.324	0.425	3.102	0.095	0	0.418			0.076		
As	0.035	0.04	0.029	0.078	0.067	0.072	1.293	1.375	1.203			
Au								0.444				
Ba	0.038	0.036	0.154									
Bi							0.258	0.23	0.227			
Cd	0.121	0.116	0.112									
Co				0.125								
Cr	0.538	0.205	0.23									
Ge	0.039											
In	0.207	0.191	0.188									
La	0.073	0.078	0.068									
Mg	0.603	0.567	0.554									
Mn	0.078	0.086	0.103		0.279		0.05	0.035	0.031			
Mo							0.495	0.42	0.314	1.152	0.658	0.895
Ni	0.42	0.308	0.372		0.046		1.885	4.108	1.852			0.066
Pb	0.089	0.066	0.042	0.973	0.333	0.628	1.995	2.127	1.938	0.13	0.075	0.097
Sb	0.071	0.056	0.056	0.161	0.169	0.167	0.2	0.172	0.173	22.9	3.852	2.233
Sn	0.493	0.435	0.437	0.095	0.101	0.302						
Ti	1.04	1.028	1.088	0.367								
W		0.039										
Zn	2.288	2.313	2.14	1.566		0.866	2.04	1.925	2.018	1.529	0.989	1.069
Zr								0.093				
<b>Total</b>	<b>7.188</b>	<b>6.172</b>	<b>6.259</b>	<b>6.663</b>	<b>1.295</b>	<b>2.231</b>	<b>27.986</b>	<b>31.305</b>	<b>24.149</b>	<b>25.931</b>	<b>5.722</b>	<b>4.527</b>

### 3.3 TOF-SIMS Elemental and Isotopic Analysis

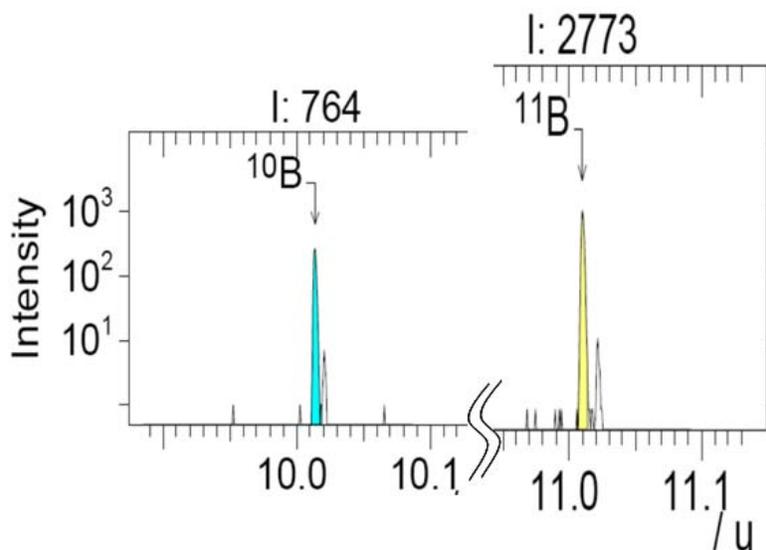
The focus of TOF-SIMS characterization was the elements detected at higher concentrations by ICP-MS on the frontside of the sample. This included Cu, Zn, Pb, P, Na, Sn, Ni, B, and Cd. Several of these elements have unique isotopic envelopes that should provide an indication whether the element is present as a trace contaminant or transfer or whether it may have been formed via transmutation. Elements such as Na and P have only one isotope and therefore are less helpful. Boron for example has two natural isotopes at m/z 10 and 11 in the ratio of 0.248 (19.9/80.1) as shown in a calculated mass spectrum (Figure 12).



**Figure 12: Calculated mass spectrum of natural isotopes of boron.**

TOF-SIMS generates ions from the surface and then measures the mass-to-charge ( $m/z$ ) of the ionized isotopes, but the efficiency at generating only elemental ions is not 100%. This means that there can be interferences from oxides, hydrides, or molecular species which confound accurate measurements. Quantification can be difficult depending on issues like surface morphology and background.

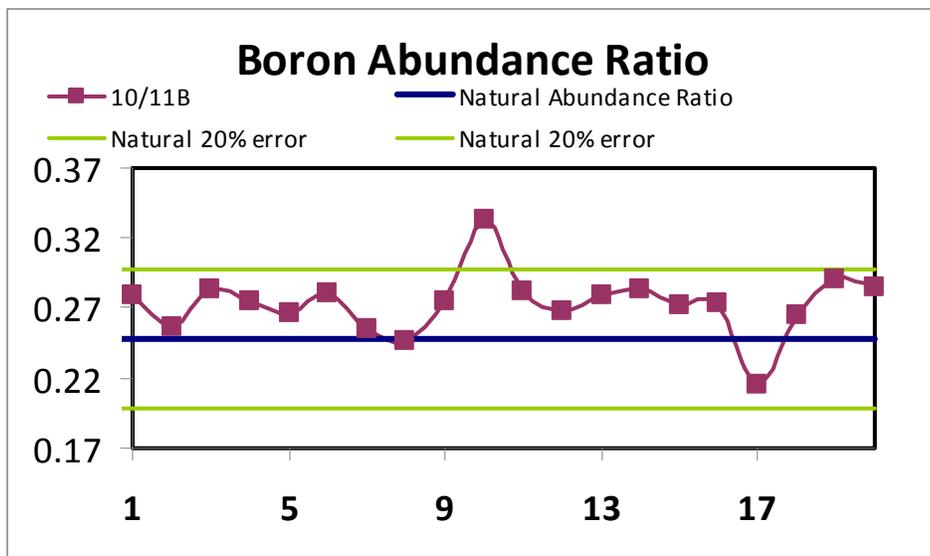
A composite plot of the boron isotope measurements on the front surface of the sample is shown in Figure 13. The peaks are color coded indicating where the instrument calculated peak area. If care is not taken to verify these peak assignments, which can vary spot-to-spot especially on a rough sample, an anomalous isotope ratio can easily be obtained! In large amounts of data this verification can be very time consuming. Boron is a “clean” example as there are few interferences at this low mass value. Using an instrument outfitted with a kinetic energy filter can reduce interferences.



**Figure 13: TOF-SIMS measurement of boron isotopes.**

The isotope ratio of  $^{10}\text{B}$  to  $^{11}\text{B}$  determined for 20 spots on the front surface is plotted in Figure 14 along with lines indicating a 20% confidence interval. The first 5 spots are in the area where the

base aluminum is exposed, serving as a control sample, while the remainder are in areas of redeposited material. From left to right one can observe there are no significant trends between the control and redeposited material. Due to the low number of sampled points the significance of the deviation from the average ratio the high point (#10) and the low point (#17) is unclear.



**Figure 14: Plot of Boron isotope ratio for 20 sample spots, blue line indicating natural ratio and green lines indicating +/- 20% confidence.**

It should be noted that for Boron (and others) there is an instrumental bias resulting in a positive shifts of the B isotope ratio, emphasizing the need for control samples. Within experimental error, the isotope ratios measured for the redeposited area were the same as those measured in the control surface for major Ni, Pb, and Cu isotopes. Also within experimental error, these ratios matched natural ratios. The Ukraine group claims to observe several instances of isotope ratios far from natural abundance. Such deviations are within our experimental error and would be detected.

The primary lessons from the TOF-SIMS isotope analysis include:

1. there is often an instrument bias that must be carefully accounted for
2. energy filtering to reduce hydrides and other interferences should be employed
3. interpretation of isotope ratios should include cross-validation of abundance data

### 3.4 Lessons Learned and Analysis Plan

For the available sample, impurities in a relatively impure Al alloy are not in solution and remain in local deposits after electron beam induced shock, melt, and ejection. The Ukrainian transmutations are also in localized deposits even though their copper is 99.99% pure, which seems unusual.

Of the major lessons learned, proper choice, control, and characterization of starting material(s), including those containing or supporting the target, is crucial. Multiple characterization methods providing complementary sampling volumes, detection limits, mapping and quantification abilities are required for both pre-test certification and characterization of materials and post-test to detect products. Multi-disciplinary expertise in metallurgy, chemistry, and physics would

inform choices in starting materials and ensure full interpretation. Step-wise or serial analysis could reduce costs, ensure cross-validation, and provide guidance for other techniques. Many additional techniques are available at SNL. The following analysis plan is proposed to ensure confident results in future experiments.

With a sample supplied from the Proton-21 group, we propose to:

- Obtain one set of pre-exposed sample material.
- Obtain list (and representative samples) of materials contained within or used as support for targets or deposition screens.
- Provide an independent source of high-purity sample material for a second set of samples.
- Perform in parallel with Proton-21 independent analyses of both sets of material and compare results prior to exposure.
- Handle and expose the samples with care to avoid surface contamination during transfer, storage, and testing.
- Separately and independently analyze samples post-test and compare the results with those analyses performed by Proton-21.

For an SNL-controlled experiment, we propose to:

- Discuss with metallurgists possible target materials, available forms, and potential impurities. A metallurgist's input and knowledge regarding metal solubilities, melt characteristics, and purification techniques are critical.
- Discuss with physicists possible products to expect.
- Discuss with test personnel the available options for chamber and other internal supporting materials.
- Obtain materials in sufficient quantities so that control samples are available for calibrating instrumentation such as TOF-SIMS and SEM microprobe.

## 4. CONCLUSIONS

The Proton-21 Laboratory in the Ukraine has been publishing results on shock-induced transmutation of several elements, including Cobalt 60 into non-radioactive elements. We have developed and exercised a process to detect possible shock-wave-induced transmutation products and to unambiguously validate or invalidate the claims in collaboration with the Proton-21 Laboratory. We found elevated and localized concentrations of impurity elements like the Ukrainian's report in our sample, but all our results are consistent with the ejection of impurities that were not in solution in our alloy, were deposited from the cathode during irradiation, or could be from surface contamination although the surface contamination cannot be validated. We are positioned to test samples from Proton-21 if funded to do so.

If we do proceed with testing the Proton-21 samples, the control of starting sample and hardware is critical. Multiple methods are necessary to provide complimentary sampling volumes, different trade-offs for cost and detection limits, and different mapping and quantification abilities. Multi-disciplinary inputs will be needed in the fields of metallurgy, chemistry, and physics. Step-wise analysis is useful as shown in this work; additional techniques were available but not needed. Initial measurements provide guidance for more sophisticated and expensive measurements.

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