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PROOF-OF-CONCEPT DEVELOPMENT OF PXAMS (PROJECTILE X RAY ACCELERATOR MASS SPECTROMETRY)*

White Paper prepared as part of the final report for the Laboratory Directed Research and Development Program project "Proof-of-Concept Development for Accelerator Mass Spectrometry Inverse PIXE" (tracking code 95-ERP-126).

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EXECUTIVE SUMMARY

Prior to the current work, accelerator mass spectrometry (AMS) was limited to a set of ~8-10 cosmogenic isotopes. This limitation is caused primarily by the inability to discriminate against stable atomic isobars. An analysis scheme that combines the isotopic sensitivity of AMS with similar isobar selectivity would open a large new class of isotope applications. This project was undertaken to explore the use of characteristic x rays as a method for the detection and identification of ions, and to allow the post-spectrometer rejection of isobaric interferences for isotopes previously inaccessible to AMS.

During the second half of FY94 (with Advanced Concepts funding from the Office of Non-Proliferation and National Security), we examined the feasibility of this technique, which we are referring to as PXAMS (Projectile X ray AMS), to the detection of several isotopes at Lawrence Livermore National Laboratory (LLNL). In our first exploratory work, we measured the x ray yield vs. energy for ^{80}Se ions stopped in a thick Y target. These results, shown in Fig. 1, demonstrated that useful detection efficiencies could be obtained for Se ions at energies accessible with our accelerator, and that the count rate from target x rays is small compared to the Se K α rate. We followed these measurements with a survey of x ray yields for $Z = 14-46$.

With support from this LDRD project during FY95, we successfully developed the PXAMS concept and applied it for the detection of ^{63}Ni , which we will apply in the determination of the fast neutron fluence from the Hiroshima bomb. This first application to ^{63}Ni is in collaboration with Tore Straume

and Alfredo Marchetti of the Health and Ecological Assessment Division at LLNL. In a demonstration experiment, ^{63}Ni was measured in Cu wires (2-20 g) which had been exposed to neutrons from a ^{252}Cf source. We successfully measured ^{63}Ni at levels necessary for the measurement of Cu samples exposed near the Hiroshima hypocenter. To our knowledge, this work represents the first attempt to apply AMS to the detection of this long-lived isotope, and was the first application of PXAMS to any isotope at LLNL. We have also begun to extend the methods developed for ^{63}Ni to the detection of ^{59}Ni as a biomedical tracer in living systems.

Our success in the detection of ^{63}Ni in Hiroshima wire samples — a pathological technical challenge — will have several major impacts on future development of isotopes for AMS. As the first application of PXAMS at CAMS, this work has demonstrated the effectiveness and simplicity of the technique, has illuminated both the advantages and the drawbacks, and has revealed many of the parameters which determine the sensitivity. The work also demonstrates the applicability of analytical chemistry techniques to AMS, and the value of considering novel chemical methods and sampling handling. Our efforts to reduce instrumental Cu background in the ion source will be directly applicable to other isotopes. Primarily however, this work should encourage the AMS community to explore what at first glance appears to be a set of near intractable problems.

PXAMS holds the promise of enabling the application of AMS to long-lived isotopes throughout the periodic table, pointing to a wide range of potential applications. In just the range $Z=22-50$, there are 15 long-lived isotopes which were previously unmeasurable at LLNL. PXAMS can be an effective detection technique for these isotopes and, as a general statement, PXAMS can be expected to provide 100-1000 times better sensitivity for these isotopes than would be possible with any other mass spectrometric technique (for a given sample and a given preparation chemistry). Further feasibility studies for these isotopes will now concentrate on potential applications, funding opportunities, required sensitivities, options for chemical sample purification and preparation, and availability of material for production of standards and tracers.

In the near future, there are plans at LLNL to extend AMS to several new isotopes with applications in biomedical tracing (^{53}Mn , ^{60}Fe , ^{79}Se , ^{126}Sn , ^{194}Hg , $^{202,205}\text{Pb}$), effluent migration (^{90}Sr , ^{99}Tc , ^{93}Zr , ^{205}Pb), and non-proliferation (^{90}Sr , ^{99}Tc). The success of the current work has significantly increased the feasibility of detecting these isotopes by AMS. More importantly, it has spawned a new interest in developing new isotopes at LLNL.

1. INTRODUCTION

Stable atomic isobars are the dominant background limiting the sensitivity of AMS for most isotopes. For isotopes of atomic number $Z < 20$, the post-spectrometer rejection of these backgrounds relies on the energy loss of ions in matter. The rate of energy loss dE/dx is a function of Z . It is therefore possible to distinguish isobars by sampling the energy deposited at different depths in the material. The standard AMS detector is the multiple anode gas counter, in which the ionization of a low pressure (50-200 Torr) gas is used as a measure of energy deposition. The ΔE -E telescope using silicon surface barrier detectors is similar. Another alternative is the gas-filled magnet, in which a magnetic field combines with energy loss and charge changing interactions to physically separate the isobars.

Isobar rejection techniques that rely on energy loss suffer from two intrinsic features of the energy loss process — charge changing interactions and the statistical nature of multiple collisions — which cause the width of the energy distribution to increase as the ions slow down. At lower Z this spreading, known as energy straggling, is small compared to the difference in energy loss between isobars. With increasing atomic number, the straggling increases relative to the energy loss difference, so that isobar separation becomes progressively less effective, and at some point unworkable. For the energies accessible with larger tandem accelerators, this point is somewhere in the range $Z \approx 25$ –30.

We are exploring an alternative method for isobar rejection which does not rely on energy loss. In this method, a variant of PIXE (particle induced x ray emission), projectile ions are identified by the characteristic x rays they emit when slowing down in matter. These x rays are of atomic origin, and can therefore be used to distinguish the ions by atomic number. Artigas *et al.*^{1,2} examined this approach for the AMS detection of ^{36}Cl , ^{59}Ni , and ^{94}Nb at 15-21 MeV, and Wagner *et al.*³ explored its application in the detection of ^{59}Ni and ^{60}Fe at 55 MeV. In both works, the purpose was to find a method for detecting the radioisotopes given the constraints of the available accelerators.

During the second half of FY94,⁴ we examined the applicability of this technique, which we are referring to as PXAMS (Projectile X ray AMS), to the detection of several isotopes at Lawrence Livermore National Laboratory (LLNL). In our first exploratory work, we measured the x ray yield vs. energy for ^{80}Se ions stopped in a thick Y target. These results, shown in Fig. 1, demonstrated that useful detection efficiencies could be obtained for Se ions at energies accessible with our accelerator, and that the count rate from target x rays is small compared to the Se $K\alpha$ rate. We followed these measurements with a survey of x ray yields for $Z = 14$ –46.

During the course of this LDRD project in FY95, we successfully developed PXAMS for the detection of ^{63}Ni , which we will apply in the determination of the fast neutron fluence from the Hiroshima bomb, in collaboration with Tore Straume and Alfredo Marchetti of the Health and Ecological Assessment Division at LLNL. To our knowledge, this work represents the first attempt to apply AMS to the detection of this long-lived isotope, and was the first application of PXAMS to any isotope at LLNL. We have also begun to extend the methods developed for ^{63}Ni to the detection of ^{59}Ni .

In this paper we present the results of our x ray yield measurements, describe the experimental arrangement currently used for PXAMS at LLNL, and discuss our work on the detection of ^{63}Ni , ^{59}Ni , and other isotopes.

2. MEASUREMENT OF THE X RAY YIELDS

X ray yields were measured using the 10 MV FN tandem accelerator at the Center for Accelerator Mass Spectrometry (CAMS) at LLNL.⁵ Picoamp currents of selected ions were stopped in thick targets, and the resulting x rays were detected with a lithium-drifted silicon detector ($\text{Si}(\text{Li})$) normally used for PIXE analyses. The number of incident ions was determined by collecting the charge deposited on the targets, which were mostly elemental metal foils of 10 mg/cm^2 thickness or greater. Absolute x ray detection efficiencies were measured using thin ($\sim 50 \text{ } \mu\text{g/cm}^2$) PIXE standards. In general, the uncertainties in the measured $K\alpha$ yields were insignificant compared to the $\sim 10\%$ uncertainty in the charge integration. For the $K\beta$ yields, the uncertainties were somewhat larger because of increased statistical uncertainty and increased uncertainty in the peak extraction.

Results of the yield measurements vs. ion energy for Ni and Se ions are shown in Figs. 1 and 2. The Ni results exhibit a trend similar to the Se results, but with significantly higher yields for the same energy per unit mass. Also seen in Fig. 2 is the good agreement of the present Ni data to the x ray yields of Ref. 2.

Projectile and target yields for Se ions on various targets are displayed in Fig. 3. The optimum target is either Y or Zr, and the shape of the yield curves is essentially the same for different energies (compare the 80 and 100 MeV results). The projectile and target $K\alpha$ yield curves illustrate the cross-section resonances which results from the formation of molecular orbitals between the projectile and target atoms when there is an energy overlap between the respective K shells. This atomic physics phenomenon has been studied in detail by Meyerhof *et al.*,⁶ and the present data follow the trends in that reference. In addition, we studied the second resonance region (near $Z=80$), in which there is an overlap between the projectile K shell and the target L shell. The maximum yield in the second resonance region is about 75% of the Y yield, however this is a more difficult region for PXAMS applications because of the intensity and number of target $L\alpha$ lines.

In Table 1, we present additional measured yields which are representative of the efficiencies that will be accessible with our accelerator. Yields as high as 1 x ray per incident ion are accessible for light ions such as Si. The yields are significantly reduced for heavier ions, primarily because of the reduced energy per unit mass. However, even for ^{107}Pd ions, where the best yield is less than 1 x ray per 300 incident ions, we expect that PXAMS will allow more sensitive detection than is possible with other techniques.

We have also examined the use of projectile $L\alpha$ lines, which have significantly higher yields, however with standard x ray detectors these lines are not resolved from the target $L\alpha$ lines. Spectrum

fitting is an option, however this would likely require significant intervention and analysis time per sample on the part of the experimenter. We have considered the possibility of detecting $L\alpha$ lines using a wavelength dispersive x ray detector, which could provide increased isobar rejection, but the small acceptance of these detectors is expected to be a significant drawback.⁷

3. PXAMS AT LLNL

For further PXAMS development, we have purchased a high resolution high-purity germanium (HPGe) detector[†] and which is installed on the AMS beamline at CAMS. A schematic of our setup is shown in Figs. 4 and 5. The target and absorbers are placed immediately in front of the Be window of the detector. This design was chosen to maximize the solid angle intercepted by the detector. While the solid angle in this geometry could in principle approach 2π , the solid angle at present is $\sim 0.08 \times 4\pi$ because of the ~ 10 mm distance between the crystal face and the outer edge of the cryostat, plus the combined thicknesses of the foils.

The 0° detector position leads to significant shifting and broadening of the x ray lines due to Doppler shifts. A somewhat smaller effect (which acts at all angles) is the shifting of the atomic levels caused by the high average charge state of the ions as they slow down in the target. Because of the Doppler broadening, a careful choice of target thickness and accepted angular range is important to find a balance between maximizing the x ray yield and minimizing the width of the peaks to improve for isobar rejection.

Examples of the observed x-ray spectra for a ^{63}Ni standard ($^{63}\text{Ni}/\text{Ni} = 1 \times 10^{-6}$) and blank ($^{63}\text{Ni}/\text{Ni} = 0$) are shown in Fig. 6. A fraction of the Cu counts extend through the Ni region as a low energy tail, so that subtraction of background Cu counts from the Ni sum is necessary for ultimate sensitivity. Ni and Cu counts are extracted using the indicated summing regions. The amount of isobar rejection, defined as the ratio of Cu counts in the Cu region to Cu counts in the Ni region (ie., the "peak-to-tail" ratio), is ~ 300 in this case. For measurements with a signal-to-background of >0.1 , $^{63}\text{Cu}/^{63}\text{Ni}$ levels can be as high as ~ 3000 (larger ratios are possible with sufficient statistics). It was found that the Cu peak-to-tail ratio is stable to $<0.5\%$ over periods of hours, and for count rates up to ~ 3 kHz. It is therefore conceivable that ^{63}Ni could be detected at the 3σ level for samples in which the $^{63}\text{Cu}/^{63}\text{Ni}$ ratio was as large as ~ 10000 .

4. PXAMS APPLIED TO THE DETECTION OF ^{63}Ni AND ^{59}Ni

The long-lived isotopes of nickel have current and potential use in a number of applications including cosmic ray studies,^{8,9} biomedical tracing,¹⁰ characterization of low-level radioactive wastes,^{1,2} and

[†] An Igelet-X detector, 100mm^2 active area and 145 eV FWHM resolution at 5.9 keV, purchased from EG&G Ortec, Oak Ridge, TN.

neutron dosimetry. As our first application of PXAMS, we are developing methods for the routine detection of these isotopes by AMS.

One intended application of ^{63}Ni is in Hiroshima dosimetry, in support of work by Tore Straume and Alfredo Marchetti in the Health and Ecological Assessment Division of LLNL. Currently there is a discrepancy in the neutron fluence emitted by the Hiroshima bomb which may be as large as a factor of ten.^{11,12} Resolution of this discrepancy has important implications in studies of the long-term health effects of radiation exposure which follow the health histories of Hiroshima and Nagasaki survivors.¹³ The reaction $^{63}\text{Cu}(n,p)^{63}\text{Ni}$ has been identified as one of a small number of reactions which might be used for the direct determination of the Hiroshima fast neutron fluence.¹⁴ The current (1994) level of ^{63}Ni in copper samples exposed near the Hiroshima hypocenter has been estimated to be ~ 1.4 fg ^{63}Ni per g Cu.¹⁵

The detection of ^{63}Ni in copper samples from Hiroshima is in many ways a pathological, worst-case challenge for AMS. The sample consists of only femtograms of analyte in grams of the isobaric interference, the isobar easily produces high negative ion currents in a Cs sputter source, the isobar is relatively ubiquitous in nature, and a cursory look at the chemistry of Cu and Ni would indicate that their separation at ultratrace levels is difficult if not impossible.

AMS measurement of ^{63}Ni ($t_{1/2} = 100$ y) requires the chemical removal of ^{63}Cu (natural isotopic abundance 68%), which is a stable isobar of ^{63}Ni . PXAMS will allow a post-spectrometer rejection of ^{63}Cu by a factor of ~ 3000 , so that, for the measurement of a 10 g copper sample (~ 14 fg ^{63}Ni), the Cu must be reduced to < 50 pg. Strict demands are therefore placed on the chemical separation technique, which must provide a reduction of Cu by $\sim 10^{12}$, with quantitative retention of the ^{63}Ni .

For sample preparation, two chemical steps were used: electrodeposition of bulk Cu, and reaction of Ni with carbon monoxide. The first step relies on the large difference in electropotential between Cu and Ni to preconcentrate the trace Ni from gram-sized copper samples.¹⁶ Samples are dissolved in an electrolytic cell, and the Cu is simultaneously electrodeposited on a platinum electrode, leaving Ni in solution. Cu concentrations are reduced from grams to sub-microgram levels in this step.

Following the electrochemical separation, the Cu concentration is further lowered using the reaction of Ni with carbon monoxide to form the gas nickel tetracarbonyl ($\text{Ni}(\text{CO})_4$). This reaction, suggested by an external collaborator (Prof. P. Jones, Chemistry Department, University of the Pacific), is highly selective for Ni, and has been exploited by other researchers for the detection of sub-nanogram quantities of Ni by graphite furnace-atomic absorption spectrometry.^{17,18,19,20} We have adapted this technique for the preparation of AMS samples (see Fig. 7). Samples containing 1 mg Ni (used as a chemical carrier for the ^{63}Ni) in weak nitric acid solution are adjusted to pH $\approx$ 10 using concentrated ammonia. A mixture of carbon monoxide and helium is bubbled through the solution. A solution of sodium borohydride is added, reducing Ni^{II} ions to Ni^0 , which then react with the CO to form $\text{Ni}(\text{CO})_4$. The $\text{Ni}(\text{CO})_4$ is carried in the gas flow to a cold trap immersed in liquid nitrogen. Following completion

of the reaction, the trap is allowed to warm to room temperature and the $\text{Ni}(\text{CO})_4$ is transferred in a He flow to the AMS sample holder, where it is thermally decomposed to Ni metal at $\sim 160^\circ\text{C}$ (Fig. 8).

The $\text{Ni}(\text{CO})_4$ reaction has proved to be an exceedingly effective method for the removal of Cu. For samples containing 1 mg Ni carrier, the Cu concentration is consistently reduced to $< 2 \times 10^{-8}$ (Cu/Ni), or < 20 pg Cu. This Cu level appears to be the background limit of the AMS ion source. These levels are regularly achievable even for samples which have been spiked with 1 mg Cu, demonstrating a reduction in Cu by $> 10^8$. Processing times are ~ 15 -30 min per sample. Decomposition of the $\text{Ni}(\text{CO})_4$ directly in the sample holders obviated the need for any additional handling of the sample following purification. Any such handling would have caused additional Cu contamination, and would likely have required clean-room conditions.

For AMS measurements, the terminal of the accelerator was set to +9.0 MV, and the high energy spectrometer was tuned to select the 99 MeV $^{63}\text{Ni}^{10+}$ ions. Using a 2.0 mg/cm^2 Zr foil and a 10 mm foil to detector distance, the total ion detection efficiency for this arrangement is $\sim 3.4 \times 10^{-3}$ K α counts per incident $^{63}\text{Ni}^{10+}$ ion.

To fully demonstrate our ability to measure ^{63}Ni induced in Cu samples, we prepared a set of demonstration samples for measurement by AMS. This experiment was performed in collaboration with T. Straume and A.A. Marchetti. Cu wires (2-20 g) were exposed to neutrons from a ^{252}Cf source at LLNL. Fast neutron fluences were ~ 2 and 20 times the estimated fluence near the hypocenter of the Hiroshima bomb. The results of this experiment are listed in Table II, and are presented graphically in Fig. 9. We successfully measured ^{63}Ni at levels necessary for the measurement of Cu samples exposed near the Hiroshima hypocenter. For the demonstration samples, the Cu content was chemically reduced by a factor of 10^{12} with quantitative retention of ^{63}Ni . Detection sensitivity (3σ) was ~ 24 fg ^{63}Ni in 1 mg Ni carrier ($^{63}\text{Ni}/\text{Ni} \approx 2 \times 10^{-11}$). The linearity of the results for wires of different sizes and different neutron fluences illustrates the accuracy of the technique. Significant improvements in sensitivity are expected with planned incremental changes in the methods. Because of precious, historical nature of the archived Hiroshima samples, measurements of the actual samples will be delayed until these improvements have been made.

We have begun to modify our methods for the detection of ^{59}Ni ($t_{1/2} = 1.0 \times 10^5$ y) which we plan to use as a biomedical tracer in living systems. Initial results indicate that only minor changes will be required. In a first experiment using a series of ^{59}Ni standards, we demonstrated a detection sensitivity (3σ) of < 10 fg ^{59}Ni in 1 mg Ni carrier ($^{59}\text{Ni}/\text{Ni} \approx 1 \times 10^{-11}$). This corresponds to a ^{59}Ni activity of ~ 20 μBq (~ 0.6 fCi), well below the limits of decay counting techniques.

5. CONCLUSIONS

Prior to the present work, AMS at LLNL was limited to isotopes of the elements with atomic number

$Z \leq 20$ (Ca), allowing a set of ~ 8 long-lived isotopes to be accessed.[‡] Even with the largest accelerators suitable for AMS, there are <15 available isotopes. PXAMS holds the promise of enabling the application of AMS to long-lived isotopes throughout the periodic table, pointing to a wide range of potential applications. In just the range $Z=22-50$, there are 15 long-lived isotopes which were previously unmeasurable at LLNL. We now know that PXAMS can be an effective detection technique for these isotopes. As a general statement, PXAMS can be expected to provide 100-1000 times better sensitivity for these isotopes than would be possible with any other mass spectrometric technique (for a given sample and a given preparation chemistry). Further feasibility studies for these isotopes will now concentrate on potential applications, funding opportunities, required sensitivities, options for chemical sample purification and preparation, and availability of material for production of standards and tracers.

Our success in the detection of ^{63}Ni in Hiroshima wire samples — a pathological technical challenge — will have several major impacts on future development of isotopes for AMS. As the first application of PXAMS at CAMS, this work has demonstrated the effectiveness and simplicity of the technique, has illuminated both the advantages and the drawbacks, and has revealed many of the parameters which determine the sensitivity. The work also demonstrates the applicability of analytical chemistry techniques to AMS, and the value of considering novel chemical methods and sampling handling. Our efforts to reduce instrumental Cu background in the ion source will be directly applicable to other isotopes. Primarily however, this work will encourage the AMS community to explore what at first glance appears to be nearly intractable problems.

In the near future, our plans at LLNL are to extend AMS to several new isotopes with applications in biomedical tracing (^{53}Mn , ^{60}Fe , ^{79}Se , ^{126}Sn , ^{194}Hg , $^{202,205}\text{Pb}$), effluent migration (^{90}Sr , ^{99}Tc , ^{93}Zr , ^{205}Pb), and non-proliferation²¹ (^{90}Sr , ^{99}Tc). The success of the current work has significantly increased the feasibility of detecting these isotopes by AMS. More importantly, it has spawned a new interest in developing new isotopes at LLNL.

[‡] This count includes ^{129}I , $Z=53$, which is an exception because there is no stable isobar with mass 129.

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TABLE I. Representative measured xray yields for various ions and targets.

Isotope	Half-Life (y)	Competing Isobar	Measured Ion	Target	Energy (MeV)	K α x rays per incident ion
³¹ Si	100	³¹ S (Z+2)	³¹ Si 7+	CaCO ₃	56	1.0
			7+	Ti	56	0.3
⁶⁰ Fe	1.5×10 ⁶	⁶⁰ Ni (Z+2)	⁶⁰ Fe 11+	Cu	102	0.8
⁵⁹ Ni	1×10 ⁵	⁵⁹ Co (Z-1)	⁵⁸ Ni 11+	Zn	102	0.5
				Ge	102	0.4
⁷⁸ Se	~10 ⁵ -10 ⁴	⁷⁸ Br (Z+1)	⁸⁰ Se 10+	Y	100	0.05
			12+	Y	120	0.08
⁹³ Mo	4×10 ³	⁹³ Nb (Z-1)	⁹² Mo 11+	Rh	106	0.008
¹⁰⁷ Pd	7×10 ⁴	¹⁰⁷ Ag (Z+1)	¹⁰⁶ Pd 11+	Ag	106	0.003

TABLE II. Results of AMS measurement of ^{63}Ni in irradiated copper wires.

Irradiation Time (d)	Expected $^{63}\text{Ni}/\text{Cu}$ (fg/g)	Dissolved Wire Mass (g)	Total ^a Nickel (mg)	Measurement Time (min)	Measured ^{63}Ni (fg)	Statistical ^b Uncertainty (fg)	Systematic ^c Uncertainty (fg)	Expected ^{63}Ni (fg)
0	2.7	2.92	1.11	24	-9	± 14	± 14	0
1.00	27	19.81	1.17	22	59	± 9	± 8	54
10.0	27	2.24	1.11	30	65	± 14	± 10	60
10.0	27	4.33	1.11	24	133	± 10	± 7	116
10.0	27	8.73	1.13	17	243	± 13	± 7	233

^a Includes 1.1 mg Ni added as carrier and trace nickel from the wire sample ($3.4 \pm 0.5 \mu\text{g Ni} / \text{g Cu}$).

^b Uncertainty resulting from counting statistics.

^c Uncertainty resulting from uncertainty in the calibration.

FIGURE CAPTIONS

FIG. 1. X ray yields vs. incident ion energy for ^{60}Se ions on a thick Y target. Shown are the Se K α (squares) and K β (diamonds) yields, and the Y K α yields (crosses). For the K α yields, error bars are smaller than the symbols. The curves are guides to the eye.

FIG. 2. X ray yields vs. incident ion energy for ^{58}Ni ions on a thick Zn target. Shown are the Ni (squares) and Zn (crosses) K α yields of the present work, and the Ni (inverted triangles) K α yields of Ref. 2. For the present data, error bars are smaller than the symbols. The curves are guides to the eye.

FIG. 3. X ray yields vs. target atomic number for 80 and 100 MeV ^{60}Se ions. Shown are the Se K α yields at 80 MeV (squares) and 100 MeV (open triangles). Also shown for 80 MeV are the Se K β yields (diamonds), and the Y K α (crosses) and La (plusses) yields. For the K α and La yields, error bars are smaller than the symbols. The curves are guides to the eye.

FIG. 4. Schematic of the AMS spectrometer at LLNL. For PXAMS measurements, the ions are detected and identified via characteristic x rays (see also Fig. 5).

FIG. 5. Detection and identification of ions via characteristic projectile x-rays (PXAMS). Following analysis in the AMS spectrometer (Fig. 4), the ions are incident on a Zr foil, inducing characteristic x-rays which are detected with a HPGe detector. Additional absorbers (Be and mylar) are included to stop the ions and attenuate low-energy x-rays.

FIG. 6 ^{63}Ni PXAMS x-ray spectra for a ^{63}Ni standard ($^{63}\text{Ni}/\text{Ni} = 1 \times 10^{-4}$) and a blank. 99 MeV $^{10}\text{B}^+$ ions were incident on a $2.0 \text{ mg}/\text{cm}^2$ Zr foil. For these samples, the copper concentration was $^{63}\text{Cu}/\text{Ni} = 1 \times 10^{-7}$.

FIG. 7. Apparatus for generation of $\text{Ni}(\text{CO})_4$. Sample solution containing Ni is placed in the reactor. A mixture of He and CO is bubbled through the reactor. BH_4^- is added, reducing Ni^{2+} to Ni^0 , which reacts with CO producing $\text{Ni}(\text{CO})_4$. The $\text{Ni}(\text{CO})_4$ condenses on silanized quartz wool in the LN_2 trap. After completion of the reaction, the trap is warmed and the $\text{Ni}(\text{CO})_4$ is transported with He to the AMS sample holder for thermal decomposition.

FIG. 8 Thermal decomposition of $\text{Ni}(\text{CO})_4$ in target holder. $\text{Ni}(\text{CO})_4$ is carried in helium to the sample holder, where it is thermally decomposed to Ni.

FIG. 9 Measured vs. expected ^{60}Ni contents in the demonstration experiment. The results demonstrate the accuracy and sensitivity of the methods. Wires were irradiated with fast neutrons from a ^{252}Cf source. Fast neutron fluences corresponded to ~ 1.4 and ~ 14 times the estimated fast neutron fluence near the hypocenter of the Hiroshima blast. The estimated fluence is from Ref. 15, which is based on the DS86 evaluation (Ref. 13). Crosses: ^{60}Ni standards and blanks. Open square: unirradiated wire. Filled square: 20 g wire irradiated with ~ 1.4 times the estimated Hiroshima fluence. Open triangles: wires of various masses irradiated with ~ 14 times the estimated Hiroshima fluence.

FIG. 1

FIG. 2

FIG. 3

FIG. 4

FIG. 5

FIG. 6

FIG. 7

FIG. 8

FIG. 9

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 <950> ABSTRACT

Prior to the current work, accelerator mass spectrometry (AMS) was limited to a set of {approximately}8--10 isotopes. This limitation is caused primarily by the inability to discriminate against stable atomic isobars. An analysis scheme that combines the isotopic sensitivity of AMS with similar isobar selectivity would open a large new class of isotope applications. This project was undertaken to explore the use of characteristic x rays as a method for the detection and identification of ions, and to allow the post-spectrometer rejection of isobaric interferences for isotopes previously inaccessible to AMS. During the second half of FY94 (with Advanced Concepts funding from the Office of Non-Proliferation and National Security), we examined the feasibility of this technique, which we are referring to as PXAMS (Projectile X ray AMS), to the detection of several isotopes at Lawrence Livermore National Laboratory (LLNL). In our first exploratory work, we measured the x ray yield vs energy for {sup 80}Se ions stopped in a thick Y target. These results, demonstrated that useful detection efficiencies could be obtained for Se ions at energies accessible with our accelerator, and that the count rate from target x rays is small compared to the Se K{alpha} rate. We followed these measurements with a survey of x ray yields for Z = 14-46.