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Challenges and opportunities in high-precision Be-10 measurements at CAMS

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Abstract

We determined the overall efficiency for ^{10}Be of the high-intensity LLNL modified Middleton cesium sputter source in combination with the CAMS FN mass spectrometer. BeO^- ionization efficiency is >3%. Charge exchange efficiency including transmission through the tandem for 7.5MeV Be^{+3} is ~34%, resulting in a total system efficiency of just over 1%. At this efficiency and with very low backgrounds, we estimate our detection limit to be ~1000 ^{10}Be atoms. Cathodes prepared with only ~80 micrograms of ^{9}Be show only an ~33% reduction in ^{9}Be beam current compared to a sample with ~200 micrograms. These same samples, prepared from 07KNSTD1032 standard material, contained 1 x 10^7 and 5 x 10^6 ^{10}Be atoms and exhibited similar ionization and total system efficiency. These results demonstrate the feasibility of pursuing applications that require precise measurement of samples with low ^{10}Be concentrations and/or small sample size.

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Keywords: AMS, Ion source, Efficiency, Beryllium-10

1. Introduction

Beryllium-10 measurements have important applications in the fields of Earth sciences, astrophysics-cosmochemistry, and nuclear chemistry. However, development of new applications often requires and is limited by, small sample size and subsequent efficiencies in sputtering, charge exchange, and transmission from the ion source to the detector. Recent work at CAMS demonstrates that ^{10}Be measurements can consistently be made at <1% precision, from targets producing 30-40 μA ^{9}Be beam currents, and with very low backgrounds (2-5 total ^{10}Be counts in 15 minutes, ~5 x 10^{-17} $^{10}Be/^{9}Be$ ratio). We have explored the sensitivity of low ratio/concentration samples by determining the overall efficiency of the LLNL modified Middleton cesium sputter source in combination with the CAMS FN mass spectrometer. Modifications to and characteristics of the high-intensity CAMS Cs^+ sputter source are reviewed by Southon and Roberts [1] and Fallon et al. [2].

2. Methods

We prepared 5 targets from 07KNSTD1032 beryllium oxide standard material [3] mixed with Nb and packed in stainless steel cathodes. Aliquots of BeO and Nb were transferred into tared quartz crucibles and their masses measured on a balance with an accuracy of approximately +/- 1 μg. Each target contained either ~200 or ~80 ug of Be as BeO (Table 1). The beryllium oxide and Nb were mixed thoroughly, transferred to stainless steel cathodes, and packed for measurement. Assuming that the BeO and Nb were completely homogenous, the mass of the residue in the quartz crucible was subtracted from the original mass of material in order to determine the total BeO in the cathode. After converting the total mass of BeO to Be, we used the published $^{10}Be/^{9}Be$ of the 07KNSTD1032 standard, 972 x 10^{-15} [3], to calculate the initial number of ^{10}Be atoms in the target. ~200 and ~80 μg targets contained ~1.5 x 10^{7} and ~5.3 x 10^{6} atoms, respectively (Table 1).

The targets were analyzed using the high-intensity LLNL modified Middleton cesium sputter source with the LLNL CAMS AMS system based on an HVEC model FN

Van de Graaff accelerator [4]. The ion source was optimized to sputter cones on the target by focusing the cesium beam just forward of the target surface. We collected ^{10}Be data during two different runs (August 2 and 17, 2008) in blocks of 600 seconds until targets were run to "exhaustion" (i.e. the ^{9}Be current fell from the initial 37-13 μA range to ~1 μA so that samples were exhausted to greater than ~93%). Typical ion source settings used for ^{10}Be measurements at CAMS are given in Table 2.

For each sample, we determined the total system efficiency by comparing the calculated ^{10}Be atoms in each target (Table 1) to the number of gated ^{10}Be counts measured in the gas ionization detector. In order to calculate the ionization efficiency from the total system efficiency, we measured the total transmission efficiency at 7.5 MeV by comparing the beam currents of $^{9}Be^{16}O^{-}$ before and $^{9}Be^{3+}$ after the accelerator. This charge exchange efficiency was measured on a diluted 1:90 (BeO:Nb) tuning target in order to limit the amount of sustained beam current transmitted through the accelerator. The total transmission efficiency at 7.5 MeV is ~34% (Table 3), which includes a 10% loss on the gridded lens [5].

3. Results and discussion

Fig. 1 shows the primary current as a function of cumulative measurement time. Targets generally show a pattern of rapidly increasing current to a constant maximum during the first 600-second cycle (e.g. sample 07KNSTD1032 of August 2, 2008) and then slowly and steadily decreasing throughout the experiment. For the August 17, 2008, samples, the ~200 μg and ~80 μg targets were generally exhausted after 9,000 and 4,000 seconds, respectively. On average, targets with ~80 μg of BeO showed only an ~33% decrease in initial primary current compared with ~200 μg targets.

Fig. 2 shows the cumulative ^{10}Be counts measured over time for the experiment duration. For the first hour of the experiments, count rates were 50 cps for the August 2, 2008, sample and 30 cps and 20 cps for the 200 μg and 80 μg samples, respectively, on August 17, 2008. At these count rates, all of these samples would have >1 x 10^4 ^{10}Be counts in 15 minutes, which based on counting statistics should yield a ^{10}Be/^{9}Be measurement with <1% precision.

Table 4 summarizes results for the 6 BeO cathodes (four ~200 μg and two ~80 μg targets) for which efficiencies were measured. Sample 07KNSTD1032-200-5 died after 3,000 seconds, and thus yielded a minimum efficiency value because it was not run to exhaustion. Based on the results for the remaining samples, the total system efficiency is just over 1%. Using our estimated total transmission efficiency of 34%, we calculate an ion source BeO^- ionization efficiency of >3% for targets with ~200 μg and ~80 μg of BeO.

4. Conclusions

Charge exchange efficiency (i.e. total transmission efficiency) for the CAMS FN mass spectrometer is ~34% for 7.5MeV Be^{+3}, resulting in a total system efficiency of just over 1%. BeO^- ionization efficiency for the high-intensity LLNL modified Middleton Cs^+ sputter source is >3%. At this efficiency and with very low backgrounds (~5 x 10^{-17} ^{10}Be/^{9}Be ratio), we estimate our detection limit to be ~1000 ^{10}Be atoms. Cathodes prepared with only ~80 μg of ^{9}Be as BeO (07KNSTD1032 standard material) show only a ~33% reduction in primary current compared to a sample with ~200 μg, and exhibited similar ionization and total system efficiencies. These same targets, prepared with ~80-200 μg of Be, lasted for more than one hour and gave count rates between 50 and 20 cps. With this efficiency, ion source output, count rate, background, and detection limit, the LLNL AMS system can routinely measure samples with <1% precision. The calculated total system efficiency allows us to estimate the total ^{10}Be counts at the detector and the corresponding counting statistics assuming complete consumption (i.e. exhaustion) of the target (Fig. 3). Accordingly, this efficiency demonstrates the potential at CAMS for development of new applications that require small sample sizes (e.g. small amounts of Be carrier addition) and low ^{10}Be concentrations (e.g. young or deeply buried surface exposure samples) with associated precision.

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Figure Captions

Fig. 1. Primary current ($^{9}Be^{3+}$ μA) from the 6 samples versus time (s).

Fig. 2. Cumulative ^{10}Be gated counts versus time from the 6 samples. The total number of counts measured is shown in parentheses in the legend.

Fig. 3. Achievable statistical precision based on counting statistics, assuming complete consumption of a BeO target versus sample size. The counting statistics are calculated using the estimated total ^{10}Be counts at the detector with a 1% total system efficiency. The lines correspond to samples with 9.72×10^{-13} (e.g. 07KNSTD1032), 9.72×10^{-14}, and 9.72×10^{-15} $^{10}Be/^{9}Be$ ratios.

Table 1. Chemical data and calculations for samples

Sample Name	BeO (mg)	Nb (mg)	% transferred	Total BeO in cathode (mg)	Total Be in cathode (μg)	^{10}Be atoms in cathode
KNSTD1032	0.71	5.15	91	0.65	233	1.5E+07
KNSTD1032-200-3	0.64	2.82	90	0.58	209	1.4E+07
KNSTD1032-200-4	0.69	3.09	95	0.66	237	1.5E+07
KNSTD1032-200-5	0.69	2.59	94	0.65	234	1.5E+07
KNSTD1032-50-6	0.24	4.28	97	0.23	84	5.4E+06
KNSTD1032-50-8	0.23	4.28	97	0.22	80	5.2E+06

Table 2. CAMS typical source operating settings

Extraction voltage	40 kV
Cathode voltage	10 kV
Ionizer power	127 W
Cs reservoir temperature	172 degrees C

Table 3. CAMS 7.5MeV total transmission efficiency (including 10% loss on gridded lens)

$^{9}Be^{16}O^{-}$ low energy cup (μA) *	$^{9}Be^{3+}$ high energy cup (μA) *	Total transmission efficiency (%) **
0.084	0.091	36
0.072	0.082	38
0.075	0.084	37
0.85	0.89	35
0.95	0.9	32
1.1	0.95	29

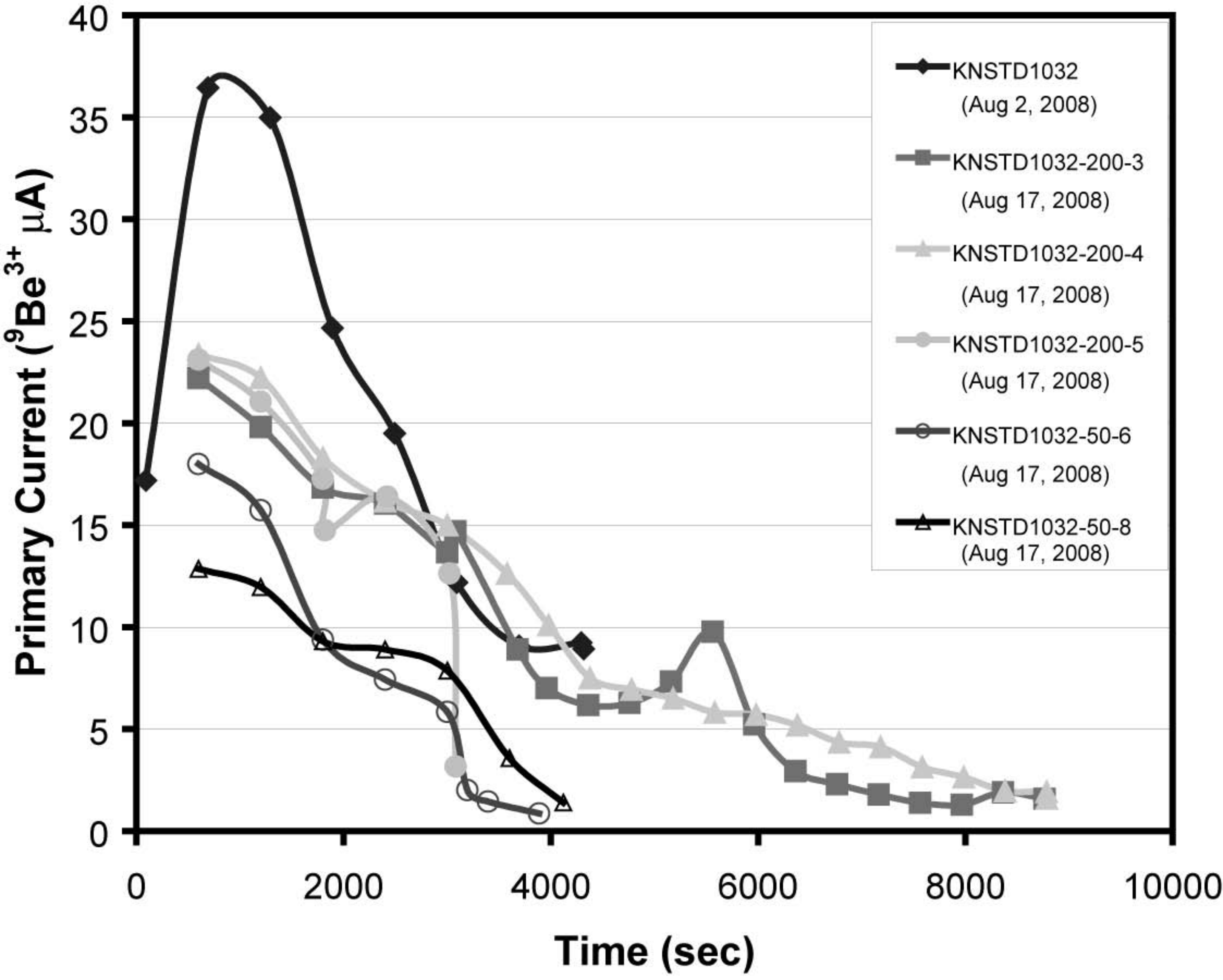
* Currents measured on a dilute 1:90 (BeO:Nb) tuning target

** Calculated conversion = $((^{9}Be^{3+} / 3) / {}^{9}Be^{16}O^{-}) \times 100$

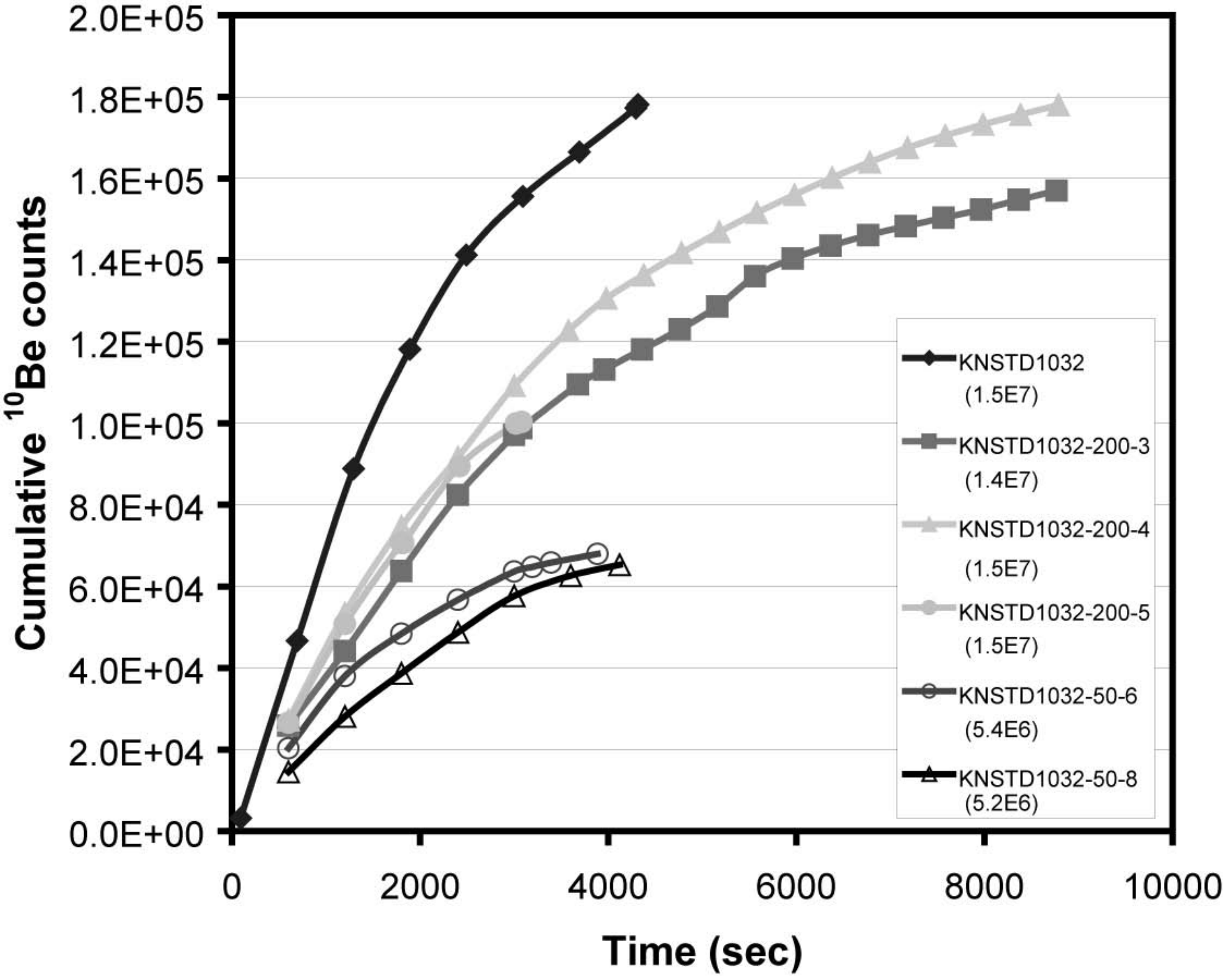
Table 4. CAMS source efficiency

Sample	10Be atoms in target	10Be gated counts	Total system efficiency (%)	Ionization efficiency (%)
KNSTD1032	1.5E+07	1.8E+05	1.2	3.4
KNSTD1032-200-3	1.4E+07	1.6E+05	1.2	3.4
KNSTD1032-200-4	1.5E+07	1.8E+05	1.2	3.4
KNSTD1032-200-5 *	1.5E+07	1.0E+05	0.7	1.9
KNSTD1032-50-6	5.4E+06	6.8E+04	1.2	3.6
KNSTD1032-50-8	5.2E+06	6.5E+04	1.3	3.6

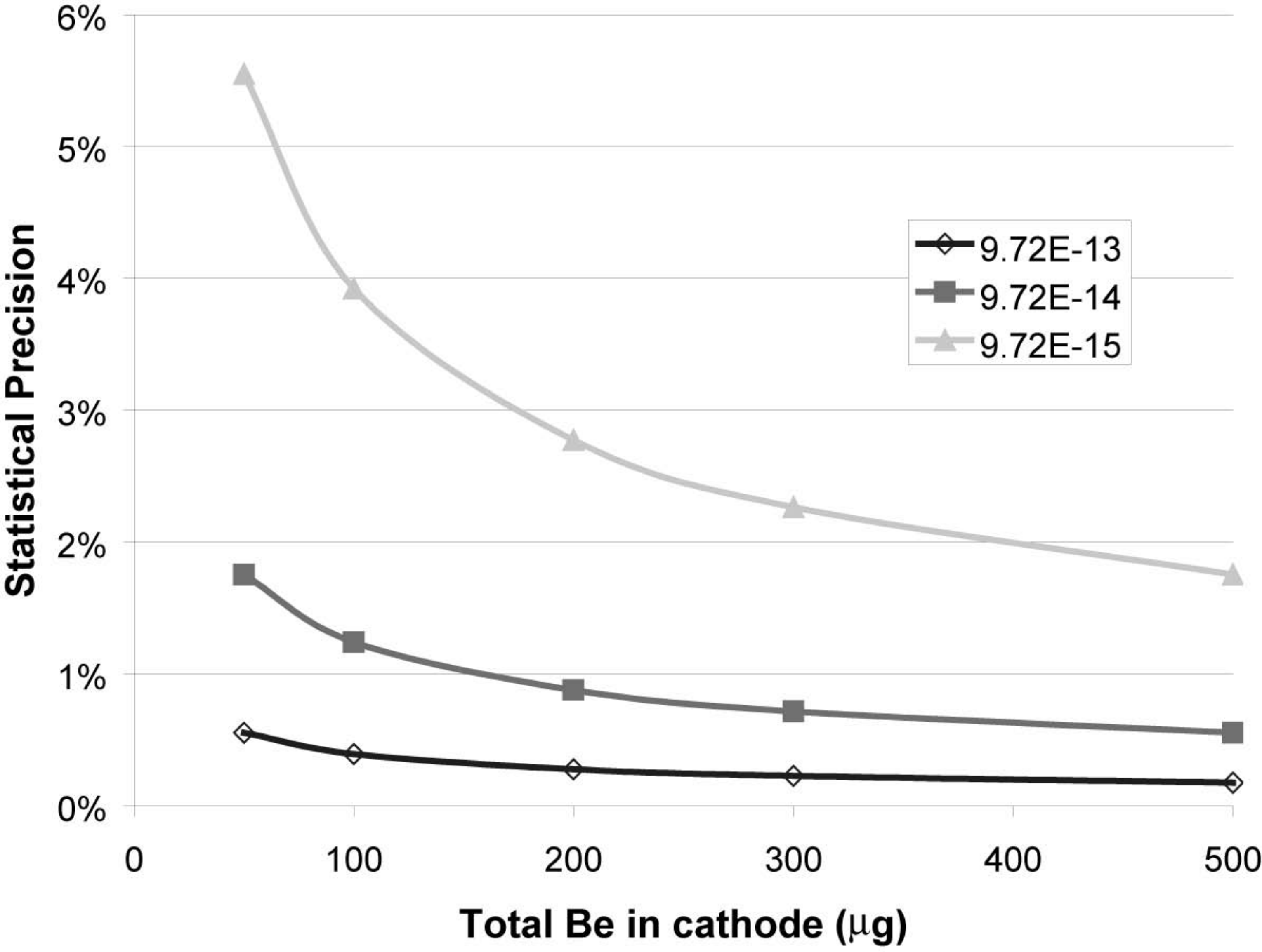
* Target not run to exhaustion, thus minimum efficiencies



Primary Current ($^9Be^{3+}$ μA)
Time (sec)
KNSTD1032 (Aug 2, 2008)
KNSTD1032-200-3 (Aug 17, 2008)
KNSTD1032-200-4 (Aug 17, 2008)
KNSTD1032-200-5 (Aug 17, 2008)
KNSTD1032-50-6 (Aug 17, 2008)
KNSTD1032-50-8 (Aug 17, 2008)
0
5
10
15
20
25
30
35
40
0
2000
4000
6000
8000
10000



Cumulative ^{10}Be counts
Time (sec)
2.0E+05
1.8E+05
1.6E+05
1.4E+05
1.2E+05
1.0E+05
8.0E+04
6.0E+04
4.0E+04
2.0E+04
0.0E+00
0
2000
4000
6000
8000
10000
KNSTD1032
(1.5E7)
KNSTD1032-200-3
(1.4E7)
KNSTD1032-200-4
(1.5E7)
KNSTD1032-200-5
(1.5E7)
KNSTD1032-50-6
(5.4E6)
KNSTD1032-50-8
(5.2E6)



6%
5%
4%
3%
2%
1%
0%
Statistical Precision
0
100
200
300
400
500
Total Be in cathode (μg)
9.72E-13
9.72E-14
9.72E-15