Practical Experience Implementing the Comecer ALCEO Metal Solid Targetry System

CE Erdahl[a,1], BR Bender[a], DW Dick[a]

[a]Department of Radiology, University of Iowa, Iowa City, IA, USA

Introduction

The Comecer ALCEO Metal system is intended to be a comprehensive solid targetry system, capable of all steps necessary to produce copper isotopes ($Cu^{60,}$ Cu^{61} & Cu^{64}) from enriched nickel: plating, transfer to/from cyclotron, irradiation, and dissolution/purification. To develop plating and chemistry methods, we plate natural nickel, and irradiate with deuterons to produce Cu^{61}. This alleviates the need for expensive enriched nickel isotopes, but gives a lower activity yield. We report a few issues with the ALCEO system, and some of our modifications.

Material and Methods

BRIEF DESCRIPTION OF SYSTEM: The ALCEO system uses cylindrical shuttles (28mm ø x 35mm h) comprised of an Al body with a Pt well, onto which the Ni is plated. Shuttles are transferred pneumatically from the hot cell to the irradiation module, on the end of the cyclotron beamline.

The plating and dissolution are both done at the electrochemical cell, located in the hot cell. This cell is connected by capillary tubes to the electrolytic solution reservoir (for plating), or the acid reservoir (for dissolution). These tubes form a recirculation loop, through which the fluid is propelled by an inline micropump throughout plating and dissolution.

The platinum well is 16mm in diameter, while an O-ring is used to plate only the center (6mm in diameter). A constant DC voltage is applied.

PLATING: Prior to plating, we clean the platinum surface with nitric acid.

We dissolve natural nickel nitrate (99.999% pure) into an electrolytic solution comprised of deionized water, ammonium hydroxide and ammonium chloride (pH=9.3). We use 30–100 mg of nat. Ni in a 10mg/mL solution. We have varied the ALCEO electrochemical cell voltage between 2.25 – 3 VDC, and tried to maintain a low pump flow rate between 1 – 2 mL/min.

The electrochemical cell uses a fixed platinum tube as the anode (~3mm above the plating surface). This tube also delivers the electrolytic plating solution to the Pt surface, forming part of the recirculating loop. The Pt surface is in contact with a gold-plated cathode.

Due to issues discussed below, we have built a custom plating rig for the ALCEO shuttles, which does not use the pump/recirculation loop, but leaves the reservoir of electrolytic solution in place, atop the plating surface. A fixed Pt wire (supported by a glass capilary tube) is used for the anode, placed ~2mm above the plating surface. We use the same size O-ring to plate only the center 6mm of the Pt surface, and apply a constant DC voltage.

TRANSFER TO/FROM CYCLOTRON: The pneumatic transfer tube is 50 ft. long between the hot cell and the cyclotron vault, and has a rise of 14 ft. from under the floor to the ceiling of the cyclotron vault.

IRRADIATION: The ALCEO irradiation module holds the plating surface orthogonal to the beam path. The module has a 10-mil-thick (0.010") Al front foil, supported by a hex-grid. Once we realized the thickness, we replaced this with a 1-mil-thick Al foil, followed by a 1-mil-thick Havar foil.

The foil is cooled by a flow of helium, while the shuttle and grid are cooled by a flow of water. The helium and water are cooled in heat exchangers by chilled water. As initially plumbed, the chilled water flowed through the heat exchangers in series, cooling the helium first, then the water. After initial runs, we plumbed the heat exchangers in parallel, teeing the chilled water to the supply of each heat exchanger, and teeing the returns together.

Irradiation is performed with a PETtrace 800 accelerating deuterons to 8.4 MeV on target. The beam current limit is 20 µA for the ALCEO Metal target. A set point of 19 µA is used to avoid the system tripping off.

DISSOLUTION/PURIFICATION: The ALCEO system circulates 5mL of 6M HCl, while heating the shuttle to 100°C for 40 minutes. This solution is loaded onto a column containing 10g of 200-400 mesh chromatographic resin in chloride form.

A separation is performed yielding three solutions: The column is washed with 40mL of 6M HCl to obtain the Recovered Nickel Solution, then 20mL of 4M HCl to obtain the Cobalt Solution, then 10mL of 0.5M HCl to obtain the Copper Product Solution.

Results and Conclusion

PLATING: Using the ALCEO method, the platings obtained had a tendency to mound, (up to 0.75 mm thick for 50mg) giving a lower density of 3–4 g/cm^3. This was attributed to the anode tube being fixed in place over the center of the

[1]Corresponding author, E-mail: colbin-erdahl@uiowa.edu

plating surface. Using the custom rig, almost no mounding was observed, (0.25mm thick for 50mg) giving a density of 7 g/cm^3, closer to nickel's nominal density of 8.9 g/cm^3.

Images 1 and 2 attempt to show the mounding from the ALCEO method, and relative flatness from the custom rig. Both methods give a rough, or "fuzzy" plated surface. Image 2 shows that the custom rig exaggerates this "fuzziness". Using the ALCEO method, a slower pump speed (~1 mL/min) gives a smoother plating surface, but the pump has a tendency to lock up at this lower set point, stalling the plating as often as half of the plating attempts.

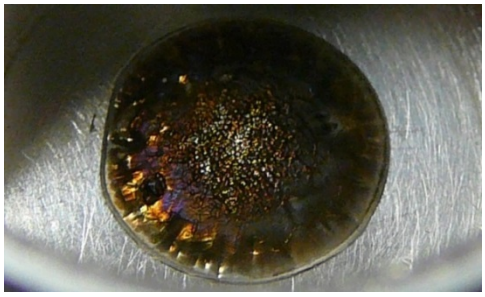

IMAGE 1. Plating with ALCEO method

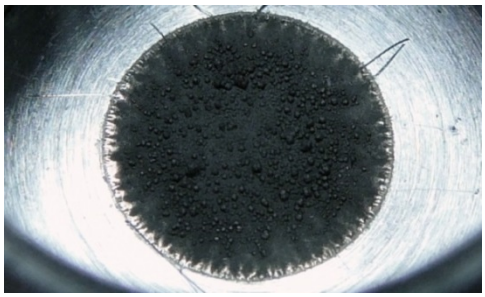

IMAGE 2. Plating with custom rig

Using the ALCEO method, slight increases in the voltage (2.65 V instead of 2.25 V), can form thin stalagmites of nickel, electrically connecting the anode and the plating surface, ending the chance for a useable plating. Using the custom rig, no stalagmites are seen, adjusting the voltage from 2.3V to 3.0V. This leads to the potential for a faster plating.

Both the ALCEO method and the custom rig have obtained plating efficiencies of 95%.

Table 1 summarizes the plating results.

Plating Method	*ALCEO Method*	*Custom Rig*
Plating Time	4-5 days	3-4 days
% Metal Plated	94-98%	94-97%
Plating Shape	Mound	Plateau
Plating Height	650-900 µm	~500 µm
ALCEO often locks up or has stalagmites		

TABLE 1. Comparison of two plating methods

TRANSFER TO/FROM CYCLOTRON: The shuttle typically transfers without issue in under 15 seconds. Once or twice it has remained in the transfer tube, but been retrieved by cycling the compressed air/vacuum a couple of times.

IRRADIATION: During initial testing, the temperature of the return water rose rapidly during irradiation. This was attributed to the chilled water already gaining heat from the helium heat exchanger. Once the parallel chilled water plumbing was implemented, the water temperature rose much more slowly. The initial return water maximum temperature was 30^{o}C. After re-plumbing, this temp plateaus around 33^{o}C, so the maximum was raised to 50^{o}C, with no issues observed.

Initial testing with the 10-mil Al foils gave a very poor activity yield. The 1-mil Al foil ruptured under the 20 psi helium pressure before beam was applied. The 1-mil Havar foil produced 1.57 mCi of Cu^{61} at EOB, giving an activity yield of 0.308 mCi/uAh (results summarized in Table 2). This compares to yields of 1.4 obtained by [1], and 0.29 obtained by [2] for deuterons on natural Ni.

Foil used	*I* (µA)	t_b (h)	A^{EOB} (mCi)	Yield (mCi/uAh)
10-mil Al	15	1	0.019	0.007
1-mil Al	failed under He pressure			
1-mil Havar	19	1.5	1.57	0.308

TABLE 2. Activity yields using different foils

Table 3 shows the results of bombardments with the following specifications:

- Plated with ~100 mg of Nat Ni
- Bombarded with 19µA for 4 hours (Deuterons)
- 3 runs each for ALCEO plating method, and custom rig method were averaged.

After dissolution and purification, several assays were made by dose calibrator. All products, waste, and the dissolved shuttle were assayed several times between 2 and 6 hours EOB to give the total amount of Cu-61 present at EOB.

Plating Method	*Total Cu-61 (corr. To EOB)*	*Saturation Yield*
ALCEO Method	5.56 mCi	0.49 mCi/µA
Custom Rig	8.14 mCi	0.75 mCi/µA

TABLE 3. Comparison of total shuttle results.

[1]Corresponding author, E-mail: colbin-erdahl@uiowa.edu

DISSOLUTION/PURIFICATION: The column supplied for purification did not seal at the top, because of this, the program had to be modified to alternate dripping the dissolution onto the column, and pushing with He. If this alternation is not done, the hot cell can be contaminated by the long-lived cobalt species in the dissolution.
Table 4 shows the dissolution/purification efficiencies for the same 3-run averages described in Table 3.
The post-dissolution/purification product, waste, recovered Ni, and dissolved shuttle were assayed by dose calibrator at:

- ~2 – 6 hours EOB for Cu-61
- > 3 days EOB for Co-56 / Co-58

The amounts of Cu-61 and Cobalt isotopes present in the Cu product were compared to that of the recovered Ni, waste and the dissolved shuttle. These isotopes were confirmed by gamma spectroscopy using a HP-Ge detector for each solution and the shuttle. Very little Ni-65 was present in the Cu product.

Plating Method	*% Cu-61 dissolved*	*% Cu-61 purified*	*diss/pur Eff*	*% Co rmvd*
ALCEO Method	77.1 %	64.8 %	48.4 %	98.8 %
Custom Rig	89.7 %	68.3 %	61.2 %	99.2 %

TABLE 4. Comparison of dissolution/purification results.

The purification is very efficient at removing the starting material, and long-lived Co isotopes from the Copper Product Solution, no matter which plating method is used. However, much of the desired Cu^{61} is removed as well, with only 50-60% remaining in the product.

The results from Tables 3 & 4 show that the custom-rig-plated shuttles outperformed the original ALCEO plating method consistently at every stage of the process. A higher dissolution/purification efficiency, combined with a greater starting activity leads to double the activity in the Cu product, as shown in Table 5.

Plating Method	*Cu-61 in Product (corr. to EOB)*
ALCEO Method	2.48 mCi
Custom Rig	4.96 mCi

TABLE 5. Comparison of Cu product activities.

References

1. R. Strangis, C. Gonzalez-Lepera: *Cyclotrons and Their Applications, Eighteenth International Conference*, pp. 246–247, 2007
2. D. W. McCarthy, L. A. Bass, P. Duffy Cutler, R. E. Shefer, R. E. Klinkowstein, P. Herrero, J. S. Lewis, C. S. Cutler, C. J. Anderson, M. J. Welch: *Nuclear Medicine & Biology*. **26**, pp. 351–358, 1999.

Acknowledgements

Comecer has been forthcoming and prompt in replying to our inquiries about the ALCEO Metal system. Our gratitude to Carlos Gonzalez-Lepera regarding the custom plating rig design.

[1]Corresponding author, E-mail: colbin-erdahl@uiowa.edu