

Conf-920808--5

FLWSHEET FOR ⁶³Ni PRODUCTION

CONF-920808--5

D. F. Williams, J. B. Knauer, G. D. O'Kelley,
J. T. Wiggins, and C. E. Porter

DE92 019282

Chemical Technology Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831

Manuscript for Paper 96a to be presented at the
"Extraction Flowsheets for Radioactive Materials" Symposium
during the 1992 AIChE Summer National Meeting
Minneapolis, Minnesota
August 12, 1992

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

RECEIVED
AUG 13 1992

Managed by Martin Marietta Energy Systems, Inc., under contract DE-AC05-84OR21400 for, the U.S. Department of Energy.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

402

FLWSHEET FOR ^{63}Ni PRODUCTION

D. F. Williams, J. B. Knauer, G. D. O'Kelley,
J. T. Wiggins, and C. E. Porter

Chemical Technology Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831

The production of large quantities of high specific activity ^{63}Ni (>10 Ci/g) requires both a highly enriched ^{62}Ni target and a long irradiation period at high neutron flux. Trace impurities in the nickel and associated target materials are also activated and account for a significant fraction of the discharged activity and essentially all of the gamma activity. While most of these undesirable activation products (mainly transition metals) can be easily removed as chloride complexes during anion exchange, chromium, present as ^{51}Cr , and manganese, present as ^{54}Mn , are exceptions and require solvent extraction of the in-cell product to achieve the desired purity. In addition to summarizing the current development and production experience, optimized flowsheets are discussed.

Introduction

The purpose of this paper is to summarize the development and processing experience associated with a special isotope project for the recovery and purification of significant quantities of high specific activity ^{63}Ni from HFIR¹-irradiated targets. Nickel-63 ($t_{1/2} \sim 100$ years) is one of the few isotopes which decays solely by β^- emission, making it especially well suited as a source for use in electron-capture chromatography detectors (Ayala, 1980) and other applications with an exclusive need for low-energy (<67 keV) electrons. Since most of the isotopes of nickel are either stable ($A = 58, 60, 61, 62,$ and 64), short-lived ($A = 65, t_{1/2} = 2.5$ h), or very long-lived ($A = 59, t_{1/2} = 76,000$ years), ^{63}Ni remains as the only useful nickel isotope of intermediate half-life. While the relatively large (n,γ) cross-section of ^{62}Ni ($\sigma_{\text{thermal}} = 15$ barns) provides for a practical production path, its low natural abundance (3.6%) requires targets which are isotopically enriched.

Some additional remarks about the nature of this work and its relation to the Symposium topic, "Extraction Flowsheets for Radioactive Materials," should be included here. This paper stands somewhat apart from the others in this session, since (a) solvent extraction was (and is) only used as a last resort when the potential for separations by ion-exchange was exhausted, and (b) this work focuses on the separation of transition metals, rather than actinide separations (which dominates the balance of this session). However, from a broader perspective this paper addresses the relevant issues of (a) the degree to which solvent extraction separations are unavoidable and are the "method of choice," and (b) the importance of separations on streams dominated by transition metal activity such as activated targets, activated structural materials, and decladding solutions. The flowsheet developed here is also readily applied to the recovery of other transition metal elements² or the complete removal of transition metals from a waste stream.

¹High Flux Isotope Reactor located at Oak Ridge National Laboratory.

²Co, Fe, and Zn were all readily fractionated during the anion exchange step described later.

Flowsheet Development

Separations Strategy

The radiochemical purification of nickel has been achieved by exploiting either (a) the highly selective precipitation of nickel with dimethylglyoxime (Singer and Kurbatov, 1954; Kirby (1961), or (b) the complete absence of nickel sorption from HCl solutions on anion exchange resin—an exception to the typical strong sorption behavior of the transition metals (Kraus and Moore, 1953; Kraus and Nelson, 1959). As will be seen in later sections, the choice of separations chemistry is largely dependent upon the particular impurities which must be removed. At the very minimum a nickel/cobalt separation must be effected, since ^{58}Co and ^{60}Co are inevitably produced in conjunction with the irradiation of nickel.³

Besides palladium, only nickel forms a chelate compound with dimethylglyoxime (DMG) which is removed by precipitation in ammoniacal solution. This separation process produces a voluminous, amorphous nickel precipitate. While this manner of purification has been refined (Singer and Kurbatov, 1954) and carried out on large quantities of nickel (Kirby, 1961), it requires extensive washing and handling of solids and was judged as a difficult and risky hot-cell operation (especially given the cross-contamination hazards present in the cell bank).

While not as specific a separation as DMG precipitation, anion-exchange purification of Ni(II) in 9 M HCl has many desirable features. A separations scheme based upon anion exchange offers the opportunity to purify nickel by a series of simple fluid operations which are easily adapted to hot-cell operations and can provide for isolation from the cell contamination. The well-known adsorption chart of Kraus and Nelson (1959), reproduced in Figure 1, shows that with the exception of Ni(II), all of the transition metals form anionic chloride complexes and are sorbed to varying degrees upon Dowex-1 resin [although very slight for Mn(II), Cr(III), and Ti(III)]. Ni(II) "pours" through the anion

³The primary source of radioactive cobalt is usually the activation of the trace amount of cobalt that is typically associated with even the purest nickel. (n,p) reactions of the nickel isotope provide an additional, but usually less significant, pathway.

exchange column, exhibiting essentially no affinity for adsorption. In this manner most of the common transition metal contaminants ($^{58,60}\text{Co}$, ^{59}Fe , ^{65}Zn) can be removed in a relatively small resin bed, since the major metal present, nickel, does not sorb at all.

The balance of the flowsheet addresses both (a) the early processing steps, targeted at limiting the amount of contamination and undesirable activation products dissolved with the nickel, and (b) the final processing necessary to complement anion exchange purification (i.e., removal of Cr, Mn, and non-transition metal species).

Flowsheet Basis

The basis for flowsheet development should include both a complete physical, chemical, and radiochemical description of the starting (i.e., "feed") materials and a corresponding set of consistent product specifications. With these end-points defined, a flowsheet can be developed which results in an acceptable product. The nature and detail of feed-material description needed also depends on how stringent the product specifications are. The specifications given in Table 1 for the two ^{63}Ni products processed in the first half of 1992 were fairly demanding.

The physical form of the starting material was essentially the same for both batches (denoted A and B): ~ 10 g of nickel pellets (pressed from metal powder) encapsulated in a standard aluminum HFIR target (Bigelow et al., 1981). However, the metallic impurities present (and associated activation) in each batch were quite different (Table 2). Based upon the activation levels displayed in Table 2, it is evident that anion exchange alone is not sufficient to effect the necessary purification. In particular, the ^{46}Sc activation constitutes a considerable gamma source which must be removed before hands-on (i.e., glove box) work can begin. While the ^{51}Cr activity represents much less of an intrinsic radiation hazard,⁴ it is obviously the controlling isotope with respect to the radionuclidic

⁴The gamma yield for ^{46}Sc is 20 times that of ^{51}Cr , and the average gamma energy for the decay of ^{46}Sc is roughly 3 times that of ^{51}Cr .

purity specification. Since neither ^{46}Sc nor ^{51}Cr are removed by anion exchange, additional steps are needed to remove these isotopes.

Flowsheet Overview

The ^{63}Ni flowsheet can be separated into four major sequential operations: (1) target handling, (2) target dissolution (Al, Ni) (3) nickel purification, and (4) product assay and analysis. Target handling covers the transfer of the target(s) from HFIR to the REDC cell bank (Building 7920), target handling within the cell bank, and sectioning of the target into active and waste segments. After the target is sectioned into practical lengths, a set of leaches is performed to remove any surface contamination that may have been introduced during handling within the cell bank. The aluminum is removed from the cleaned segments by a standard caustic dissolution (Martens et al., 1964), leaving just the nickel pellets and a small dissolution heel as solids. The powdery heel is removed from the nickel by a series of rinses/leaches with dilute caustic, water, and dilute HCl. Finally, the cleaned nickel pellets are dissolved in nitric acid. The simple equipment used for the dissolutions and all subsequent in-cell purifications is outlined in Figure 2.

Before purification of nickel can proceed, a nitrate-to-chloride conversion step must be carried out. Nitrate is removed by first distilling off as much HNO_3 as possible and then destroying the remainder by boiling in the presence of excess HCl. After adjustment to a high-acid concentration (9 M HCl), most of the transition metal activation products are removed by a simple "pour-through" anion exchange column. The anion exchange product is adjusted to a low acid concentration by boil-down and dilution for ^{46}Sc removal by cation exchange. Because of the high affinity of scandium (as compared to nickel) for adsorption onto cation resin, it is not necessary to use a column large enough to load the entire nickel inventory. Operation with nickel breakthrough (i.e., with a small "pour-through" column) provides excellent separation from scandium and many actinides (if present).

At this point the penetrating radiation associated with the product is low enough to permit the final purification, adjustment, and analysis of the nickel solution to be done in a shielded glove box. Chromium-51 removal is accomplished by extraction of a 1 M HCl product solution with 0.1 M TOPO (trioctylphosphine oxide) in cyclohexane. The final product analysis consists of (a) obtaining accurate weights and volumes on the product fraction(s), (b) determining the total nickel concentration by absorption spectrophotometry, and (c) determining the ^{63}Ni content by both scintillation counting and mass analysis (i.e., isotopic analysis by mass spectrometry).

Description of Purification Operations

Anion Exchange

In practice, the anion-exchange (AIX) purification is quite straightforward and consists of the typical sequential steps of conditioning, loading, "washing" (i.e., displacement), and stripping. The loading-acid concentration of 9 M HCl was chosen to maximize the distribution coefficient for cobalt, which was presumed to be the limiting isotope from a radiochemical separations standpoint. Since cobalt is not strongly adsorbed even in 9 M HCl ($D_v \sim 70$), a rather oversized column⁵ [Dowex 1-X8 (100-200 mesh), 2 cm ID x 20 cm long] was used to insure that the cobalt band would not bleed into the initial product fraction.

The first step in the AIX separation is to condition for high-acid operation by pumping about 5 bed volumes (~150 mL of 9 M HCl) through the column to displace the resident dilute acid (0.5 M HCl) used for resin storage. The feed solution (~300 mL of 40 g/L Ni, 9 M HCl) is slowly pumped (2.5 mL/min) to the column to insure the formation of tight adsorption bands. During actual hot-cell operations, the spread of these bands is monitored with a portable gamma probe. The

⁵Based upon the 10-mg cobalt "hold-back" carrier addition to the AIX feed solution and the impurities present in the aluminum and nickel feeds, the potential loading of complexed ions is less than 10 meq (<10 mL of resin) and is probably much less due to previous processing steps. No other carriers were used.

adsorbed bands are not visible until the dark-green nickel solution is displaced from the column by the 9 M HCl washing acid (~ 150 mL of 9 M HCl at 2.5 mL/min). The remaining adsorbed bands, primarily Co and Fe, can still be difficult to distinguish because of the yellow background of the anion resin. The blue chloride complex of cobalt actually appears "nickel-green" on the yellow resin, and the yellow iron complex is almost invisible. During the washing step, the mobile green nickel band separates from the adsorbed "green" cobalt band. A product cut is normally made during washing when no green color is detected in the column effluent. The next fraction usually contains the balance of the washing acid and a portion of the first stripping acid (~ 150 mL of 3 M HCl at 2.5 mL/min). A third collection fraction is started when the "green" cobalt band approaches the bottom of the column during the first column strip. Upon breaking through the bottom of the column, the cobalt band displays its true blue color but eventually reverts to the pink hexaquo complex when diluted with sufficient 3 M HCl. After the cobalt collection appears complete, a fourth collection fraction is started for the dilute acid strip (~ 150 mL of 0.5 M HCl at 2.5 mL/min) of the column. This final strip is continued until all of the iron (and any ⁶⁵Zn) is removed, placing the column in readiness for either reuse or storage.

Two separate "cold" tests with natural nickel were performed to assess the performance of the ion-exchange column prior to hot-cell operations. The first test, a ⁶⁰Co tracer test, was designed to provide an accurate estimate of the cobalt decontamination factor⁶ (DF), while the second test, truly a cold test (i.e., no tracer), had the broader focus of providing an estimate of the likely separation achieved from the host of contaminants present (and the ultimate chemical purity of the product). Both experiments used a nickel feed solution derived from simulated target rods which had been processed (approximately) according to the previously described operations (i.e., Al dissolution, leaches, etc.). The stand-in nickel pellets were probably no purer than the actual enriched pellets,

⁶For the purpose of this paper, the decontamination factor for species *i* is defined as: $DF_i = (\text{feed inventory})_i / (\text{product inventory})_i$.

and the aluminum alloys were of the identical lot actually used in the actual target fabrication and accurately reflect the chemical impurities likely to be carried over into the AIX processing.

The tracer experiment yielded a cobalt DF of about 2×10^5 , which is certainly more than adequate for the predicted cobalt inventory in the AIX feed. The analytical (ICP/MS) results from the second cold test provide both an estimate of the likely chemical impurities in the AIX feed and the associated AIX decontamination factors. These results are shown in Table 3 and are in accord with the predictions of Kraus and Nelson (1959): all of the transition metals *except Ni, Cr, and Mn* are effectively removed by AIX, while the alkali metals, pre-transition metals, and rare earths/actinides pass through the column.

While the results from hot-cell operation were generally carried out as previously described and performed as predicted by the cold tests, some additional description of the operations is required. The fact that the AIX feed for both targets (and especially Target A) contained far more ^{51}Cr and ^{46}Sc than originally anticipated posed a significant radiation exposure problem for the final glove-box finishing steps. In-cell removal of ^{46}Sc by cation exchange is described in the next section. Some additional steps were added to the AIX operations to effect the oxidation of Cr(III) to Cr(VI) and thereby remove chromium as the chromate anion.

For the processing of Target A, the first cycle of anion exchange was carried out precisely as described earlier and resulted in a product dominated by ^{46}Sc and ^{51}Cr activity as shown in Figure 4. In order to effect some degree of chromium removal, the product solution was treated at low-acid concentration to oxidize the chromium to chromate and fed (at low acid) once more to the anion exchange column. A modification of the recipe suggested by Burgus (Pijck, 1964) was used to oxidize the chromium after the product solution had been adjusted to less than 0.2 M HCl by boiling down and redilution. A reduction of the ^{51}Cr activity in the product by a factor of about five is inferred from the subsequent cation exchange results, since no separate analysis was performed on this second cycle product. For the second target, the chromium oxidation step was attempted before the

adjustment to high acid which precedes anion exchange. Apparently (Table 5) the subsequent adjustment to 9 M HCl reduced the chromium back to Cr(III) and prevented removal as chromate. This preliminary oxidation step did not interfere with the retention of other elements during AIX. It simply was not effective in helping to remove chromium. More effective ion-exchange strategies for in-cell removal of chromium are presented in Section IV.

Cation Exchange (CIX)

The primary purpose of the cation exchange step is to remove the ^{46}Sc activity from the product solution. A small "pour-through" column (i.e., the nickel is not quantitatively loaded) is adequate to retain the trivalent scandium, while the displaced and eluted nickel is collected as product. An added benefit of this final hot-cell separation is that the cation column also removes any trivalent actinide contamination from the product solution just prior its removal from the cell bank. Considerable care is exercised in handling the product solution so as not to contaminate it before removal from the cell bank.

As in the previous operations, the same basic set of hot-cell equipment is used (Figure 2) except for the cation exchange column, which is essentially a copy of the anion exchange column but loaded with 60 mL of Dowex 50W-X8 (200-400 mesh) resin. The sequence of steps during CIX is also essentially the same as during AIX, even though nickel loads on the column in this instance. Since only the retention of Sc(III) is required, the divalent, monovalent, and Cr(III) ions are simply displaced from the column during loading, washing (100 mL of 0.1 M HCl), and elution (300 mL of 2.0 M HCl) of the nickel. Thus, only the rare earths (including scandium) and actinides remain to be removed during the final strip (300 mL of 6 M HCl). As before tight adsorption bands are maintained by operating at a fairly low flowrate (2.5 mL/min), and the initial and final product cuts are readily made based upon the color of the effluent from the column. No ^{46}Sc was detected in either CIX product (A or B) and ^{51}Cr was the dominant activity, as expected.

Chromium Removal by TOPO Extraction

Most techniques for the selective removal of chromium (Pijck, 1964) from aqueous solutions require the oxidation of the more stable Cr(III) to the strongly oxidizing Cr(VI), since in this higher oxidation state chromium⁷ displays a more unique chemistry and is more readily separated from other metals. The details of the oxidation recipe used in this work is based upon the recommendations of Burgus [8]. The oxidation must be carried out at a low-acid concentration [(HCl) <0.1 M] so that chromium, rather than Cl⁻, is attacked by the oxidant (NaBrO₃), and so that the chromate formed is not reduced back to Cr(III) by additional oxidation of the chloride. After oxidation the acidity is raised to the optimal value (at 1 M HCl, D_v = 100) for removal of chromium by extraction with TOPO.

It should be emphasized that the feed preparation step is usually more critical to the success of this separation than the details of the extraction procedure. The excellent potential for chromium removal by TOPO extraction is only be achieved if (a) the oxidation is carried out in very dilute HCl, (b) the oxidation is carried to completion (boil the oxidation mixture for at least 15 min) with no reduction back to Cr(III), and (c) the extractant is presaturated with 1 M HCl and a small amount of NaBrO₃ oxidant.

The extraction of chromium was carried out in a 1-L separatory funnel with provisions for agitation by an air-driven stirring rod. Equal 250-mL phase volumes of aqueous nickel and TOPO extractant (0.1 M TOPO in cyclohexane) were used. Agitation was conducted for at least 15 min to insure effective contacting. The first extraction was immediately followed by a second using fresh extractant. For the first target, two series of oxidation/extraction steps (two contactings per extraction) were required to remove the large inventory of ⁵¹Cr. The separation factor for each complete extraction cycle was only about ten (Table 4), which was somewhat disappointing since a literature value (White and Ross, 1961) of about 100 is quoted for the distribution coefficient of

⁷Present as CrO₄²⁻ chromate, HCrO₄⁻, Cr₂O₇²⁻ dichromate, or H₂CrO₄ depending on the pH.

chromium. For the second target, special care was taken to assure complete oxidation of the chromium, and the extra precaution of pre-treating the solvent with a small amount (~1 mmol) of NaBrO₃ oxidant⁸ was taken. In just one contacting a separation factor of 100 was realized, and an overall factor of 154 for the first cycle was achieved after the follow-up contacting (Table 5). A second oxidation/extraction cycle was not necessary. The recovery of nickel in the final product was about 95% for both targets, with most of the losses occurring during the pellet leaches. The other "losses" were due to sampling, nickel burnout (i.e., transmutation), and uncertainties in the feed assay.

Improved Flowsheet Possibilities

While the purification flowsheet described in this report was satisfactory, it seems somewhat cumbersome given the nature and number of steps involved. The use of a large [i.e., large enough to load all 200 meq (12 g) of the nickel] cation exchange column immediately after the nickel dissolution offers the opportunity to simplify and improve operations by:

1. Providing for a simpler and faster NO₃⁻ to Cl⁻ conversion (eliminates at least two boil-down steps).
2. Improving product purity by removing monovalent impurities (no step at present accomplishes this).
3. Automatically providing for Sc(III) removal (eliminating the need for CIX after AIX).
4. Providing an opportunity to remove chromium as:
 - a. Cr(VI) (i.e., as an anion requires a preliminary oxidation step).
 - b. Cr(III) (i.e., as a cation requires careful elution, more tenuous).

The use of a large column for NO₃⁻ to Cl⁻ conversion was avoided because of the much larger volumes associated with a 500-mL "loading" column as compared with the 60-mL "pour-through" columns used for anion-exchange purification and scandium removal. However, if the possibility exists

⁸Presumably to counteract the effect of any reducing sites present in the solvent.

to combine the NO_3^- to Cl^- conversion, the scandium removal, and the chromium removal steps all in one operation (and achieve higher chemical purity as a bonus), then the penalty associated with a large CIX column should be reconsidered in light of these potential benefits. Items 1, 2, and 3 above are typical cation exchange features and require little comment, except to note that it should be possible to elute nickel from the CIX column with 9 M HCl and eliminate the need for AIX feed preparation steps (i.e., the CIX product can be tailored to be the AIX feed).

The subject of chromium removal requires the most attention. As mentioned earlier, the separation of trivalent chromium from nickel is particularly difficult, as is evident from the curves of Kraus and Nelson (Figure 1). While the removal of Cr(III) in nitric acid by cation exchange is possible, it is difficult and likely to prove impractical (Massart, 1971). Ion-exchange separation of chromium as Cr(VI) is a much easier task and has been the focus of considerable efforts in the metal plating industry (Morrison, 1956). Cation exchange has been used to remove cationic contaminants from chromate plating solutions (purification), while anion exchange has been proposed to concentrate and retain the chromates from dilute rinse solutions (recovery). From the standpoint of our flowsheet, a chromate-specific anion exchange step is preferable, since this would not require a large capacity column. Provisional anion-exchange removal of chromium was actually attempted during processing (p. 8) and found to be only moderately successful. The fundamental problem with this approach is that chromic acid attacks and is reduced by strong-base anion resin. The remedy adopted by the plating industry, operating under basic conditions so as to suppress the formation of chromic acid, is not a viable option since the nickel would surely precipitate.

The prospects for chromium removal during cation exchange are better. Cation resin is much more resistant to oxidation by chromic acid, and nitric acid is a more desirable (i.e., less reducing) medium for chromate removal than HCl. After washing the chromium and monovalents away from the nickel using intermediate acid concentrations, elution with 9 M HCl should produce an acceptable feed for anion exchange. Even at this high acid concentration, Sc(III) is still strongly sorbed on the

resin. Implicit in this proposed operation is a preliminary chromium oxidation step, analogous to the procedure referenced earlier, except with HCl replaced by HNO₃. So the "improved" flowsheet would have the CIX and AIX steps reversed as shown below:

- Nickel Dissolution (as before)
- CIX Feed Preparation
 - boil down to dryness
 - acid adjust to 0.1 M HNO₃
 - chromium oxidation step
- Cation Exchange
 - load all Ni in feed at 0.1 M HNO₃
 - wash Ni with:
 - a. 0.1 M HNO₃ (chromate removal)
 - b. 0.5 M HNO₃ (or HCl, monovalent removal)
 - elute Ni with 9 M HCl
- Anion Exchange (as before).

The issue of manganese removal has been, up to this point, neglected because it did not constitute a significant contaminating activity. However there are surely cases (highly activated iron streams) where this is not the case. In the present context the easiest way to remove manganese is to run (or re-run) the anion exchange column at an optimally high-acid concentration ($D_v \sim 2$ at 12 M HCl). A long, narrow "separation" column may be required in this case.

Summary

A high degree of ⁶³Ni purification was demonstrated by using a series of simple (fluid) operations that are particularly well suited for use in the hot-cell environment. The bulk of the activated transition metal impurities were removed during anion exchange, while the troublesome ⁴⁶Sc activity was completely retained on a cation exchange column, leaving only ⁵¹Cr to be removed by

TOPO extraction. The potential exists to simplify the flowsheet and eliminate the extraction step by a judicious integration of the anion and cation exchange steps.

Literature Cited

- Ayala, J. A., *J. of Chromatography*, **195**, 1 (1980).
- Bigelow, J. E., B. L. Corbett, L. J. King, S. C. McGuire, and T. M. Sims, "Production of Transplutonium Elements in the High Flux Isotope Reactor," *Transplutonium Elements - Production and Recovery*, J. D. Navratil and W. W. Schulz, eds., American Chemical Society, p. 6 (1981).
- Kirby, L. J., *Radiochemistry of Nickel*, Nuclear Science Series Monograph, Subcommittee on Radiochemistry of the National Academy of Sciences - National Research Council, Report No. NAS-NS-3051 (1961).
- Kraus, K. A., and G. E. Moore, *J. of the Amer. Chem. Soc.*, **75**, 1460 (1953).
- Kraus, K. A., and F. Nelson, "Anion Exchange Studies of Metal Complexes," *The Structure of Electrolytic Solutions*, W. J. Hamer, ed., John Wiley & Sons, Inc., New York, p. 340 (1959).
- Martens, R. I., W. L. Pie, and A. E. Wible, "Preparation of Aluminum Clad Fuels for Solvent Extraction," *Nuclear Engineering Part XI*, F. J. Van Antwerpen, ed., p. 44 (1964).
- Massart, D. L., *Cation-Exchange Techniques in Radiochemistry*, Nuclear Science Series Monograph, Subcommittee on Radiochemistry of the National Academy of Sciences - National Research Council, Report No. NAS-NS-3113 (1971).
- Morrison, W. S., "Chromium Recovery from Plating Solutions (Chapter 13)", *Ion Exchange Technology*, F. C. Nachod and J. Schubert, eds, Academic Press, New York, p. 321 (1956).
- Pijck, J., *Radiochemistry of Chromium*, Nuclear Science Series Monograph No.30, pt. 7, Subcommittee on Radiochemistry of the National Academy of Sciences - National Research Council, Report No. NAS-NS-3007 (rev. 1964), p. 49 (1964).
- Singer, S. E., and M. H. Kurbatov, *J. of the Amer. Chem. Soc.*, **76**, 4739 (1954).

White, J. C., and W. J. Ross, *Separations by Solvent Extraction with Tri-n-octylphosphine Oxide*, Nuclear Science Series Monograph, Subcommittee on Radiochemistry of the National Academy of Sciences - National Research Council, Report No. NAS-NS-3102 (1961).

Table 1. ⁶³Ni Product Specifications

⁶³Ni Specific Activity	>10 Ci/g
(Alpha Activity/⁶³Ni Activity)	<3 × 10⁻¹⁰
Radionuclide Purity	<0.001%
(Non-Nickel Activity/⁶³Ni Activity)	
Chemical Purity	>99.9% Ni
Solution Concentration	500-2000 mCi/mL in 0.1 <u>M</u> HCl

**Table 2. Important Impurities and Associated Activation Products in the Nickel Pellets
(activation levels derived from analysis of dissolved nickel: 4 weeks after 8-cycle
irradiation of Target A; 11 weeks after 4-cycle irradiation of Target B)**

Elemental Impurities	Activated Isotopes	Target A		Target B	
		"Reported" Impurity Level in Feed Ni (wt %)	Measured Activation (mCi)	"Reported" Impurity Level in Feed Ni (wt %)	Measured Activation (mCi)
Ca	⁴⁶ Sc	0.033	25	0.005	0.5
Cr	⁵¹ Cr	0.032	675	0.004	5
Fe	⁵⁴ Mn	—	0.2	<0.003	0.07
	⁵⁹ Fe	—	57		15
Co	⁵⁸ Co	—	89	<0.005	18
	⁶⁰ Co	—	125		24
Cu		—		0.008	
Zn	⁶⁵ Zn	—	7	0.005	17
Cd	^{110m} Ag	—	—	<0.003	0.1
Sn	¹²⁴ Sb	—	—	<0.001	2

Table 3. Analytical (ICP/MS) Results for Second AIX Cold Test

Element ^a	Amount in AIX Feed (mg)	Amount in AIX Product (mg)	DF (decontamination factor)
Al	5.10	5.20	1.0
B	0.48	0.52	0.9
Ba	0.06	0.02	2.5
Ca	0.45	0.30	1.5
Co	19.50	<0.02	>975.0
Cr	0.14	0.06	2.1
Cu	3.60	<0.03	>128.6
Fe	360.00	0.02	18000.0
Mg	0.13	0.11	1.2
Mn	0.93	0.80	1.2
Na	1.23	0.56	2.2
Ni	13500.00	12800.00	1.1
Si	1.98	0.80	2.5
Sn	0.33	<0.20	>1.7
Total non-nickel mass	393.9	8.1	48.4

^aReported at levels below the detection limits in both the feed and product were: Ag, As, Be, Cd, Li, Mo, P, Pb, Sb, Se, Sr, Ti, V, Zn, Zr.

Table 4. Analytical Results for the Purification of Nickel (Target A)
 (the source of the Cs activity is unknown; however, the isotopic distribution suggests an activation mechanism rather than fission product contamination)

Isotope	Activity in AIX Feed (mCi)	AIX DF	CIX DF	TOPO Extraction DF		Activity in the Final Product (mCi)
				1st cycle	2nd cycle	
⁴⁶ Sc	25	~1	Complete	—	—	—
⁵¹ Ci	675	~1, 4.7	~1	11	5.5	2.0
⁵⁴ Mn	0.2	~1	~1	~1	~1	0.15
⁵⁸ Co	89	3×10^4	~1	~1	~1	0.003
⁵⁹ Fe	57	2000	~1	Complete	—	—
⁶⁰ Co	125	3×10^4	~1	~1	~1	0.004
⁶⁵ Zn	7	Complete	—	—	—	—
¹³⁴ Cs	0.2	~1	~1	~1	~1	0.2
¹³⁶ Cs	0.1	~1	~1	~1	~1	0.06
¹³⁷ Cs	0.005	~1	~1	~1	~1	0.004

Table 5. Analytical Results for the Purification of Nickel (Target B)
(Note: Co levels are elevated in final product due to minor cross contamination after AIX)

Isotope	Activity AIX feed (mCi)	AIX DF	CIX DF	TOPO Extraction DF	Activity in the Final Product (mCi)
⁴⁶ Sc	0.5	~1	Complete	—	—
⁵¹ Cr	5	1.2	1.3	154	0.02
⁵⁴ Mn	0.07	~1	~1	~1	0.06
⁵⁸ Co	18	1.6×10^4	~1	~1	0.03
⁵⁹ Fe	15	2000	~1	Complete	—
⁶⁰ Co	24	1.6×10^4	~1	~1	0.046
⁶⁵ Zn	17	Complete	—	—	—
^{110m} Ag	0.1	~1	2	3	0.02
¹²⁴ Sb	1.8	60	~1	~1	0.03

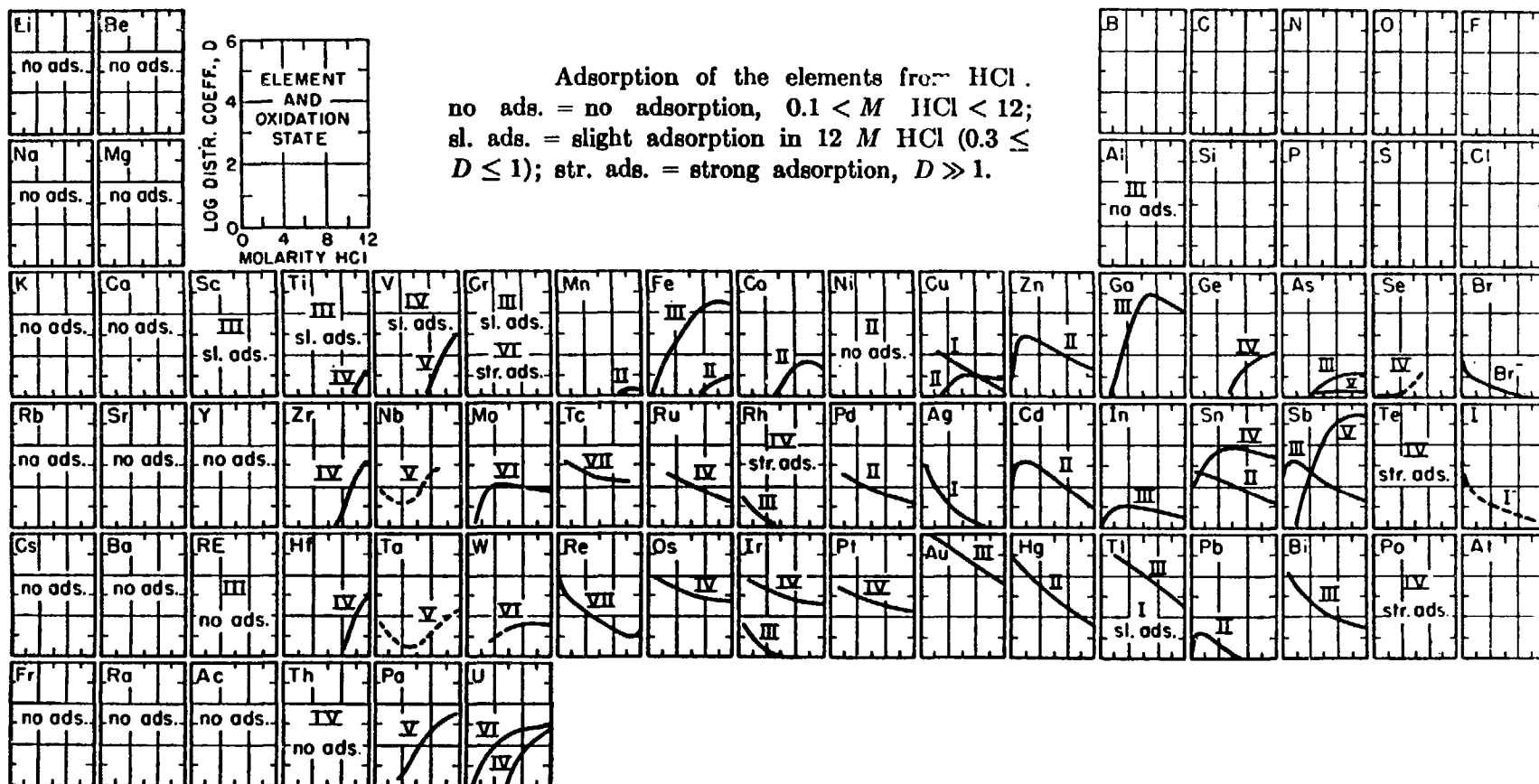


Figure 1. Anion Exchange Behavior of the Elements in HCl Solutions on Dowex-1 Resin.
 [reproduced from Kraus and Nelson (1959)]

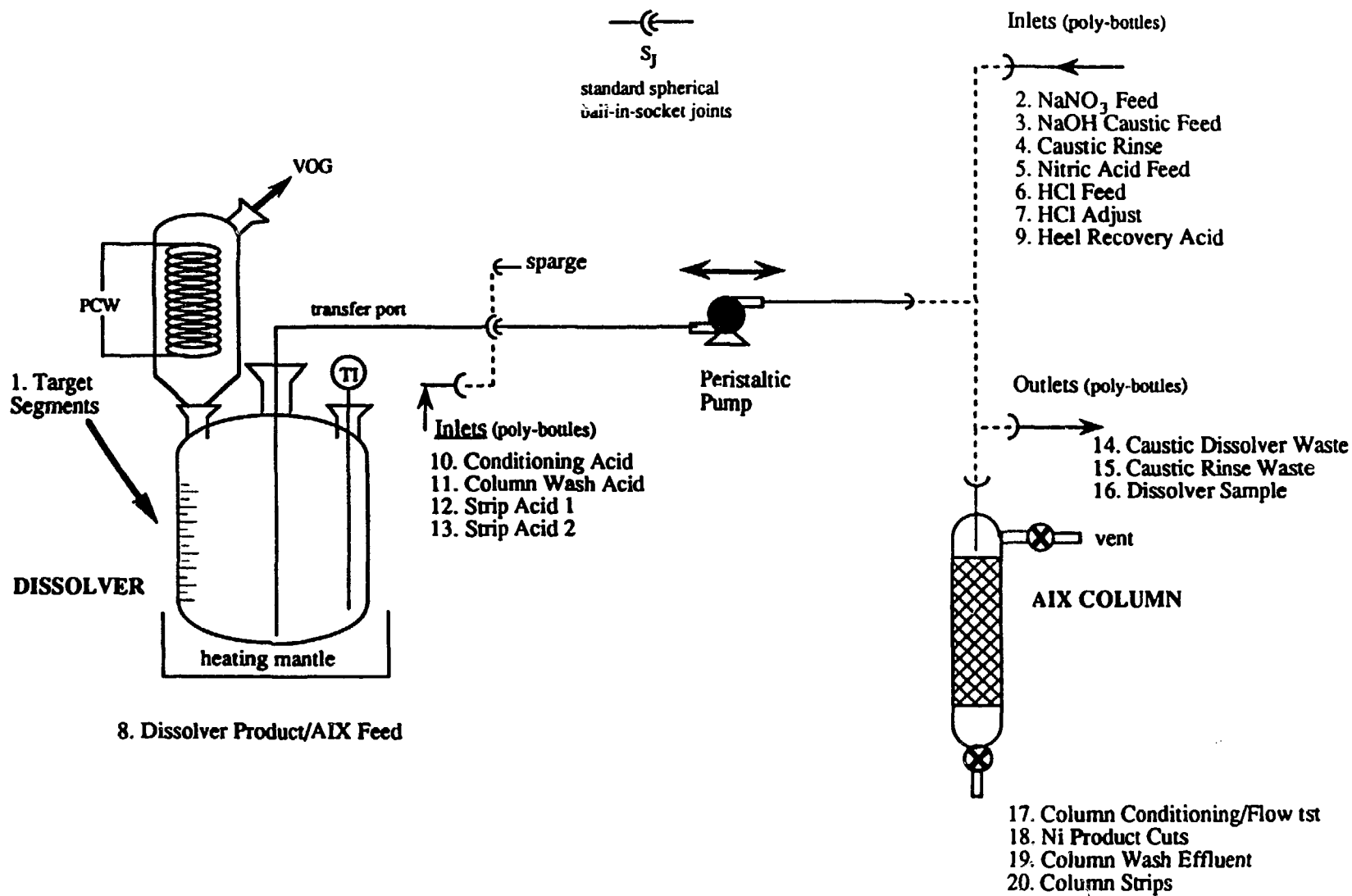


Figure 2. Equipment Diagram for In-Cell Dissolution and Purification Operations.