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Using Established Regulations to Recycle Contaminated Metals

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DOE restoration projects require acceptable standards for processing volumetrically contaminated metals:

- NRC has no regulations addressing recycling of scrap metal containing residual volumetric radioactivity.
- DOE is currently restricting outside radioactive scrap metal sales; however, previous Fernald and Ohio State clean-ups have released metals with measurable levels of radioactivity into the open market.
- Public sensitivity to the subject of non-governmental disposal of materials with residual radioactivity was heightened with the Below Regulatory Concern (BRC) issue.

There are no clear guidelines for free release of volumetrically contaminated material.

An acceptable *de minimus* standard allows processed materials without a radiation or contamination safety hazard to be recycled or stored in significantly reduced volumes. This standard can be set under existing DOE and NRC standards and IAEA volumetric limits. The nuclear industry could shift the paradigm using existing regulations to free-release metal from existing and proposed thermal treatments; starting with a specific waste stream containing a limited number of radionuclides. The following free release limits are proposed for a nickle matrix: 1) Technetium de minimus limit 0.5Bq/gram (0.5ppb) and 2) Uranium de minimus limit 0.01 Bq/gram (1ppm).

DOE order 5400.5 *Radiation Protection of the Public and the Environment* addresses "release of property having residual radioactive material:"

> "(6) Volume Contamination. No guidance is currently available for release of material contaminated in depth, such as activated material or smelted contaminated metals (e.g., radioactivity per unit volume or per unit mass). **Such materials may be released if criteria and survey techniques are approved by EH-1**[1]." (emphasis added).

Two additional related limits mandated by this regulation are:

- The effective dose equivalent to the public of 100 mrem/year[2] (1mSv/year); and
- The Derived Concentration Guidelines (DCG) for air and water.

The DCGs provide reference values for radiological environmental protection programs. They provide limits for three exposure modes: ingestion of water, inhalation of air, and immersion in a gaseous cloud. These levels are based on a continuous exposure resulting in a dose of 100 mrem/year. The DCG water limits for some radionuclides, including Tc and U, are presented in Table I. The comparable NRC sewer discharge limits are also provided in Table I.

Table I: DOE/NRC Water Release Criteria

Radionuclide	DOE[1] Derived Concentration Guides			NRC[2] Sewer Release		
	UCi/ml	Ppb	Bq/gram	uCi/ml	ppb	Bq/gram
C- 14	7.5E-05	0.016	2.7	3 E-04	6.5 E-02	11.1
K-40	7.0 E-06	1,002	0.25	4 E-05	5,730	1.48
Co-60	1.0 E-05	9 E-06	0.37	3 E-05	2.7 E-05	1.11
Tc-99	1.0 E-04	5.88	3.7	6 E-04	35	20
U-238	6.0 E-07	1818	0.02	3 E-06	9,090	.01

For an order-of-magnitude comparison, the typical concentration of radioactive K-40 in the human body is 0.05 Bq/gram, in an off-the-shelf salt substitute its 60 Bq/gram.

The NRC regulatory guide[3] 1.86 “Termination of Operating Licenses for Nuclear Reactors,” defines acceptable surface contamination levels for free-release of materials (dpm/100cm^2). Note that the surface free-release standards for the DOE are exactly the same[4]. Currently, GTS Duratek, American Ecology (formerly Quadrex), and DOE sites (such as Fernald), use these NRC surface limits to free-release material previously considered surface contaminated. This is allowable only when it is known that the material was surface contaminated and the entire surface is accessible.

Proposed Three-Step “DeMetal-Approach”

Proposed free-release limits are 0.5 Bq/gram (0.5 ppb) for Tc-99, and 0.01 Bq/gram (1 ppm) for U_x in a nickel matrix. These limits are enforced with surface limits, solution volumetric limits, and residual radioactivity limits. This DeMetal-approach is based on homogeneity existing in a molten metal system that processes the contaminated nickel.

Step 1: Surface Limits

The surface contamination guidelines must be converted into volumetric release criteria to verify that Ni contaminated with U and Tc-99 meets free-release criteria. If a metal is thin enough in a chosen volume, then the surface contamination limit will also serve as a volume limit. The determination of the thinness required (dependent on the host material and incident gamma ray energy) is developed below:

The general expression of gamma attenuation through matter has been rearranged:

$$x = \frac{-1}{\mu} \ln \frac{I}{I_o} \tag{1}$$

Where:

- I_o = number of photons without absorption
- I = number of transmitted photons
- μ = linear attenuation coefficient (cm^{-1}) (a function of material and incident gamma ray energy)
- x = thickness of the material (cm).

After the thickness is determined (Eq.1), this is multiplied by the surface contamination limits based on area (100cm^2), producing a discrete volume. This discrete volume provides a measure of activity per mass or volume.

Since the U in the gaseous diffusion cascades is in equilibrium with its daughter products, several gamma rays are emitted. Sample calculations for U in a Ni matrix are presented in Table II:

Table II: Uranium concentration in Ni (NRC surface limits <5000 DPM/100cm^2)

Detection Probability*	Thickness (cm)	Volume (cm^3)**	μCi/gram***	Bq/gram	PPM
0.99	0.03	3	1.46 E-03	6.07	481
0.90	0.32	32	1.39 E-04	0.57	45
0.80	0.68	68	6.57 E-05	0.27	21
0.10	7.0	700	6.37 E-06	0.23	2.1

*For 200 keV gamma
**Based on 100cm^2 detector on both sides
***Uranium specific activity is 3 xE-09 Ci/gram

Consider the calculation of 90% detection probability in Table II. The thickness of 0.32 cm is calculated from Equation 1, then by multiplying that 100 cm^2, a discrete volume of 32 cm^3 is defined. Thus if detectors on both sides of the metal detect 5,000 designations in a minute the U concentration in the discrete volume is 1.39 E-04 Ci/gram (90% probability). Therefore, comparing the 90% detection probability case of 45 ppm in Table II to the proposed U DeMetal limit of 1 ppm demonstrates that the limit can be quantified. Rolling a representative metal sample(s) to a thickness of 0.32 cm allows accurate measurement of U contamination in a Ni matrix.

To measure the Tc component in Ni, the same surface release limit principles apply; however, a different relationship must be used for Beta radiation. The rolled thickness is determined by the following empirical relation[6]:

$$R = 0.412 E_{\max}{}^{(1.265 - 0.0954 E_{\max})} \qquad (2)$$

where R is the range in g/cm^2, and E_{max} is maximum energy in MeV. Due to the limited range of Tc's beta in Ni, the required thickness is order of magnitudes smaller therefore rolling the Ni thin enough for Tc-99 detection is sufficient for U detection.

<u>Step 2: Solution Volumetric Limits</u>

The volumetric activity of the Ni is now compared to the liquid effluent concentrations currently allowed by law. Representative portions of the melt would be dissolved in acid and activity measured by standard scintillation techniques. The first column in Table III provides the current NRC maximum monthly average effluent concentration for Tc and U, column two is a conversion to Bq/g and column three provides the ppb using the specific activity for each radionuclide.

Table III: Effluent Concentrations (in water)[2]

Nuclide	UCi/ml	Bq/gram	ppb
Tc-99	6E-04	20	35
U-238	3E-06	0.1	~3000*

*Assumes 1.5% enrichment to compute specific activity

The limits proposed by the DeMetal approach are a factor of 40 and 2 lower for Tc and U respectively that can currently be put down a domestic sewer.

Step 3: Residual Radioactivity Limits

The final step in the DeMetal demonstrates individual dose equivalent less than 15 mrem/year, (NUREG/CR-6156). This residual radioactivity limit is based on the entire object (self-shielding included), and is measured with a radiation monitor. For the DeMetal approach limits proposed, a person who continuously held this 1 ft^3 block for one year would receive 0.0003 mrem (gammas from the U-decay chain), five orders of magnitude below the proposed limit. The exposure from Tc beta's is less.

The Bottom Line

In summary, the DeMetal-Approach will establish meaningful limits, ensuring protection of the public and the environment from the harmful effects of radiation, while recycling a useful product—using existing standards! Based on current regulations, the following free-release limits are proposed for Tc and U contained in a Ni metal found in a diffusion enrichment process:

- Tc *de minimus* limit 0.5 Bq/gram (0.5 ppb), and
- U *de minimus* limit 0.01 Bq/gram (l ppm).

These limits invoke existing regulations

- DOE and NRC governing surface contamination;
- NRC governing sewage disposal and proposed residual radioactivity limits; and

The consequences of continued inaction must be considered:

- additional spending of public monies to study and/or to continue to contain the problem, and
- environmental impacts of mining new quantities of Ni when such a large untapped reservoir of Ni (two years of U.S. use) lies in wait.

Because resources are not used on radiation problems of real magnitude, failing to act is serious. The release limits proposed are verifiable/measurable, and close to (or well below) background levels of activity in the environment.

VIII. REFERENCES

1. US Department of Energy, Order 5400.5, "Radiation Protection of the Public and the Environment." p. II-11. January 7, 1993.
2. Nuclear Regulatory Commission, 10 CFR Part 20.1, "Water Discharge Limits." 1999.
3. Nuclear Regulatory Commission, Regulatory Guide 1.86, "Termination of Operating Licenses for Nuclear Reactors." June 1974.
4. D.O.E. Order 5400.5, Figure IV-1, IV-3, January 7, 1993.
5. Nuclear Regulatory Commission, Regulatory Guide CR-6156. p. 45. 1996.
6. Shlein, Bernard. "The Health Physics and Radiological Health Handbook", p. 62. 1992.