

RADIATION PHYSICS NOTE # 115

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Date 6/12/95

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Date 6/14/95

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Accelerator Produced Exclusive Beta Emitters in Fermilab Material

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Introduction.

During the April, 1994 Westinghouse-Hanford audit (ref. 1) of Fermilab's radioactive waste program, the auditors issued a formal finding (Finding A-1) concerning the characterization of accelerator produced radioactive waste. They correctly observed that Fermilab does not quantitatively assess the levels of accelerator produced radionuclides which are exclusively beta emitters such as ^{14}C , ^{59}Ni , ^{63}Ni , and ^{93}Mo in structural materials used at Fermilab. This assessment was made independently of whether measurable quantities of such radionuclides can be reasonably expected to exist in Fermilab structural materials. It is this particular question which this note will seek to address. In order to fully address this question ^{151}Sm will also need to be considered as it is an exclusive beta emitter with a half life of less than 1000 years that is listed in Table 3-1 of the Westinghouse-Hanford document WHC-EP-0063-4. Half lives for these radionuclides and several prominent gamma emitting accelerator produced radionuclides are presented in Table 1. Under most circumstances the presence of exclusive beta emitting radionuclides can be correlated with the presence of gamma emitting radionuclides in a material. For example, the presence of the exclusive beta emitter ^3H in vacuum pump oils removed from beam enclosures at Fermilab can be correlated with the existence of ^7Be in those oils if they are surveyed and/or analyzed within an appropriate time interval after their removal from the beam enclosure (ref. 2).

Analysis.

Past History:

Past investigations at accelerator laboratories have observed only ^{14}C of the long lived exclusive beta emitting radionuclides in their effluents (refs. 3, 4, 5, and 6), particularly cooling waters which typically come in direct contact with structural materials. Table 6.9 of ref. 5 does not list any of these exclusive beta emitters as radionuclides identified in either the soil or water at four different accelerator facilities. Both ^{14}C and ^{11}C are listed in Table 6.7 (ref. 5) as radionuclides which are produced in cooling waters by high energy hadrons (30 GeV protons). However, the activity of ^{14}C produced was more than five orders of magnitude less than the ^7Be and ^{11}C activity and almost four orders of magnitude less than the ^3H activity. Since ^7Be is usually not even detectable in most structural materials at Fermilab, it is reasonable to anticipate that ^{14}C , ^{59}Ni , ^{63}Ni , ^{93}Mo , and ^{151}Sm are well below the detection limits of Fermilab field instruments.

Meteorites contain many of the same materials found in Fermilab structural materials and they are continuously exposed to cosmic ray fields which to some degree mimic the radiation fields present at high energy accelerators. Studies of these meteorites (ref. 7) have shown that the ratio of ^{59}Ni to the total Ni present in these meteorites is on the order of 10^{-11} . This is well below the detection capabilities of standard instrumentation found at most laboratories.

Production Mechanisms:

Typical production mechanisms for ^{14}C involve the irradiation of beryllium nitride or aluminum nitride in the intense thermal and epithermal neutron fields of a nuclear reactor. Such radiation fields simply do not exist at accelerator facilities. Reference 8 points out that ^{14}C is very expensive because of its long half life and the relatively low production cross section for the $^{14}\text{N}(n,p)^{14}\text{C}$ charge exchange reaction.

The primary structural materials used at Fermilab are steel, concrete, and aluminum. While copper, bronze, silver, gold, and other specialty materials are used at Fermilab, they are not typically employed as structural materials. Steels are composed primarily of iron and carbon (0.5% to 2.0%) with varying trace amounts of manganese, chromium, molybdenum, and nickel. If the steel is a hardened variety, it will contain nitrogen in some small percentage as well. Concrete is composed primarily of the hydrated oxides of calcium and silicon (~80%). Hydrated oxides of aluminum, iron, sulfur, magnesium, manganese, potassium, and sodium can also be found in varying amounts. Trace amounts of heavy elements such as bismuth, mercury, and thallium can also be present in the minerals forming the aggregate.

Production of the exclusive beta emitters in Fermilab materials can occur either through direct nuclear reactions such as (n,p) , (n, γ) , $(n,2n)$, (p,n) , or (d,n) , or through spallation reactions of the general form (h, X) , where the h represents any hadron (strongly interacting particle) and the X represents a large piece ($A > 4$) of the initial nucleus. The principle difference between these two reaction types is the time span over which the reaction typically occurs. Direct reactions occur over a time interval on the order of the time it takes the incident particle to transit the nuclear dimensions of the target material. This means that the product particles retain a memory of the incident particle conditions and only a few nucleons in the target nucleus interact with the incident particle. Therefore preferred spatial directions for particle emission are established in the scattering event. During indirect or spallation reactions, the incident particle interacts with many of the target nucleons over a time span long compared to the transit time and in the process loses memory of its initial conditions. Emission of spallation fragments from the target nucleus then tends to be more spatially isotropic. It should be noted here that the distinction between direct and spallation reactions is not always so clear-cut. For example, the (n, γ) and (p,n) reactions at low energies can exhibit features consistent with the formation of an intermediate compound nucleus as one would expect in spallation type reactions.

Table 2 lists some of the more prominent hadronically induced reactions by which both the exclusive beta emitting radionuclides being considered in this note (light colored rows) and some of the commonly observed gamma emitting radionuclides at accelerators (shaded rows) can be produced. Unless otherwise indicated by a reference number in parentheses, the maximum and asymptotic cross sections were taken from ref. 9. Since ref. 9 only lists cross sections out to 20 MeV, the listed asymptotic cross sections are the cross sections at 20 MeV unless otherwise indicated. The only proton induced reaction listed is the $^7\text{Li}(p,n)^7\text{Be}$. It is listed because it is probably the primary production mechanism for ^7Be around the Fermilab antiproton production target. Although this is certainly not an exhaustive list, it is clear that the production cross sections for the gamma emitting radionuclides and the exclusive beta emitters are not significantly different, with the possible exception of the neutron capture cross section on ^{150}Sm . However ^{150}Sm (7.4 % natural abundance) is not generally found even in the speciality materials used at Fermilab.

The (n,p) reaction on ^{14}N is probably the dominant reaction mechanism for the formation of ^{14}C both at reactors and accelerators. However, ^{14}N is not measurably present in most structural

steels, construction grade aluminum, or concrete and only present at 0.1 to 0.3 % in hardened steels (ref. 16). One would therefore be forced to conclude that the production of measurable levels of ^{14}C in structural materials at Fermilab would require extremely high flux neutron fields to compensate for the low number of potential production sites.

Production of the nickel isotopes ^{59}Ni and ^{63}Ni through direct nuclear reactions occurs primarily by neutron capture on ^{58}Ni or ^{62}Ni and the charge exchange reaction (n,p) on ^{63}Cu . As with nitrogen, nickel is a minority constituent of steel and concrete. Structural steels, construction grade aluminum, and concrete typically contain no measurable nickel or copper. In any case, the presence of ^{59}Ni or ^{63}Ni would be tagged by the gamma emitters ^{54}Mn , ^{58}Co or ^{60}Co . The gamma emitters would appear in large quantities before either the ^{59}Ni or ^{63}Ni could be detected because of the large differences in half lives and relatively low abundances of potential target nuclei in typical structural materials. Thus one can conclude that neither of these nickel isotopes can be reasonably expected to be present in Fermilab structural materials at measurable levels due to direct reactions without long (> 5yrs.) continuous exposures to high flux neutron fields and high concentrations of ^{54}Mn , ^{58}Co and ^{60}Co .

Stainless and specialty steels can have nickel concentrations as high as 22% (ref. 16) but are much too expensive to be used as structural materials. Copper and bronze are generally used as plumbing conduits in cooling and vacuum systems. Long exposure times of these materials to accelerator neutron fields can result in the generation of measurable levels of ^{63}Ni . However the presence of ^{63}Ni should be tagged by the concomitantly produced gamma emitters ^{54}Mn , ^{58}Co or ^{60}Co . Reference 17 documents a recent study of ^{63}Ni production in copper and stainless steel conducted at the National Laboratory for High Energy Physics in Tsukuba, Japan. Results from this study verified that the gamma emitters ^{54}Mn , ^{58}Co and ^{60}Co are indeed produced along with ^{63}Ni in copper continuously exposed to accelerator neutron fields for five years. The ^{54}Mn was generated at approximately the same concentrations as the ^{63}Ni and ^{60}Co was generated at approximately 5 times the concentration of ^{63}Ni in the copper. Stainless steel exposed under similar conditions had measurable concentrations of ^{55}Fe , ^{51}Cr , ^{54}Mn , ^{60}Co , and ^{63}Ni . The ^{54}Mn was generated in concentrations approximately 5 times those of the ^{63}Ni and ^{60}Co was generated in concentrations approximately equal to those of the ^{63}Ni . These comparative concentrations can be used to approximately quantify ^{63}Ni in stainless steels and copper from gamma ray spectroscopy if analysis of the irradiated material is conducted within 100 days of its removal from a beam enclosure. Otherwise quantification of ^{63}Ni involves a complex series of chemical separations before it can be counted in a liquid scintillation counter.

The highest concentrations of ^{63}Ni observed in the stainless steel (10% Ni - 10%Cr) were 100 pCi/g. This translates into $8.90 \frac{\text{g}}{\text{cm}^3} \cdot 100 \frac{\text{pCi}}{\text{g}} \cdot 10^6 \frac{\text{cm}^3}{\text{m}^3} \cdot 0.1 = 8.9 \times 10^7 \frac{\text{pCi}}{\text{m}^3} = 8.9 \times 10^{-5} \frac{\text{Ci}}{\text{m}^3}$ of ^{63}Ni . Westinghouse-Hanford document WHC-EP-0063-4 states in section 3.4.2.2 3.b) that only radionuclides which exist in quantities equal to or exceeding .01 of the Table 3-1 category 1 value should be reported. For ^{63}Ni this number is $4.8 \times 10^{-2} \text{ Ci/m}^3$; almost 3 orders of magnitude larger than the highest observed concentration of ^{63}Ni in stainless steel. For copper, the highest observed concentrations of ^{63}Ni were 92 pCi/g. This translates into $8.90 \frac{\text{g}}{\text{cm}^3} \cdot 92 \frac{\text{pCi}}{\text{g}} \cdot 10^6 \frac{\text{cm}^3}{\text{m}^3} \cdot 0.1 = 8.2 \times 10^7 \frac{\text{pCi}}{\text{m}^3} = 8.2 \times 10^{-5} \frac{\text{Ci}}{\text{m}^3}$ of ^{63}Ni . As with stainless steel, this concentration is almost 3 orders of magnitude smaller than the lowest reporting limit. The stainless steel was exposed for 2 years in a 12 GeV beam line at KEK. This is actually a typical residence time for beam line components which need to be periodically replaced. Longer exposure would typically only apply to components which remain in the beam line enclosures

until decommissioning of the accelerator. Since it takes 5 years for ^{63}Ni to buildup to the concentrations relevant to the discussion in the preceding paragraph, it seems reasonable to conclude that ^{63}Ni will appear in negligibly small concentrations in the stainless steel and copper typically shipped to Westinghouse Hanford as radioactive waste for the foreseeable future. Should Fermilab ever undergo decommissioning, it would then become necessary to evaluate the concentrations of ^{63}Ni in stainless steels and copper removed from beam line enclosures.

Molybdenum is generally not found in measurable quantities in structural steels, concrete, or construction grade aluminum, and when it exists as an alloying agent in specialty steels, it is in much lower concentrations than nickel. Again, because of the paucity of production sites in Fermilab materials, it would require unrealistically high flux neutron fields to produce measurable quantities of ^{93}Mo from ^{92}Mo (only a 15% natural abundance) by neutron capture.

Thermal neutron fields at Fermilab have been quantified within the AP0 target vault (ref 18) and directly in the Neutron Therapy Facility neutron beam (ref. 19). The maximum thermal neutron fields extant at Fermilab probably exist in the AP0 target vault during actual running conditions. Measurements of the thermal neutron flux at AP0 using gold foil activation, showed a flux of approximately 2×10^8 neutrons per cm^2 per sec ($\text{n}/\text{cm}^2 \cdot \text{sec}$). Measurements in the neutron beam at NTF using both gold and indium foil activation have yielded a thermal neutron flux of 2.5×10^5 neutrons/ $\text{cm}^2 \cdot \text{sec}$. Typical thermal neutron fluxes at reactors range from 10^{12} to 10^{14} $\text{n}/\text{cm}^2 \cdot \text{sec}$ (ref. 20); four to six orders of magnitude higher than the worst case at Fermilab.

Spallation of large nuclei into ^{14}C , ^{59}Ni , ^{63}Ni , ^{93}Mo , and ^{151}Sm fragments could also result in their production in Fermilab materials. Clearly, the initial nucleus must be significantly larger than the product nuclei and the initiating particle must deposit enough energy in the initial nucleus to allow it to break up for this type of reaction to occur. Unfortunately there are very few experimental data on the production of exclusive beta emitters via spallation (p.97 of ref. 21). In fact, the only experimental activity on spallation reactions initiated by high energy hadrons in recent years has been restricted to studies of monitor reactions for hadron flux.

Fortunately, there is a generally accepted formula for predicting spallation cross sections called Rudstam's formula (p.98 of ref. 21). Due to the scarcity of experimental data, the usefulness of Rudstam's formula for very light ($A < 20$) or very heavy nuclei is still uncertain. According to Rudstam's formula the reaction cross section is independent of the target nucleus proton number Z , and only slightly dependent on its mass number, A . Table 3 lists the cross sections predicted by Rudstam's formula for several representative spallation reactions. The first three columns list potential reactions associated with the production of exclusive beta emitters and the last three columns list some of the reactions associated with the formation of common gamma emitting radionuclides found in radioactive materials at Fermilab. It can be readily seen that the calculated cross sections for proton/neutron induced spallation on both Fe and Cu targets are within 30% of each other, i.e., there are no extreme variations. It should also be clear that for every target group, there is at least one reaction producing a commonly observed gamma emitter with a production cross section exceeding that of the exclusive beta emitters ^{14}C and ^{63}Ni by a factor of 2 or more. Only the production cross section for ^{93}Mo on very large mass targets significantly exceeds the concomitant production cross sections for Fermilab gamma emitters.

Table 4 presents a compilation of measured cross sections for production of ^{24}Na and ^{22}Na during high energy spallation of aluminum and copper. For completeness, the production cross sections for ^7Be on aluminum and ^{11}C on carbon are also included. Agreement between the measurements and predictions of Rudstam's formula are very good for the ^{22}Na production cross sections but rather poor for the ^7Be production cross sections. This suggests that Rudstam's

formula may be very good for predicting cross sections near the median Zs and As for spallation products but loses validity as an accurate representation of the real world for light nuclei. However only a significant amount of new data can provide a definitive basis for evaluating the validity of Rudstam's formula.

Low Specific Activity vs. High Specific Activity:

One can only quantify that which one can measure with some confidence. If the measured levels of radionuclides which decay relatively quickly are measured at levels near the detection limits of the instrumentation used, then it is reasonable to assume that radionuclides which decay very slowly will not be detected by the same instrumentation. To illustrate the effect of a long half life on the detection of a given radionuclide, two examples from Fermilab experience will be used. The first example will be for an aluminum tag (thin 0.5" discs) left in a beam enclosure for an extended period of time (~ 3 months) and the second example will be for a copper tag left in a beam enclosure for a similar time period. In both examples the largest measured concentrations of ^{22}Na and ^{54}Mn respectively over the last three years at Fermilab will be correlated to the estimated corresponding specific activities of ^{14}C and ^{59}Ni respectively through the Rudstam predicted cross sections.

Example 1: Aluminum in a beam enclosure.

- Largest specific activity observed in an aluminum tag was 3100 pCi/g.
- $\sigma_{22} \equiv$ Cross section for ^{22}Na production on $^{\text{nat}}\text{Al}$ by 1 GeV protons/neutrons.
- $\sigma_{14} \equiv$ Cross section for ^{14}C production on $^{\text{nat}}\text{Al}$ by 1 GeV protons/neutrons.
- $\lambda_{22} \equiv$ Decay constant for ^{22}Na .
- $\lambda_{14} \equiv$ Decay constant for ^{14}C .
- $N_{22} \equiv$ Number of ^{22}Na present in aluminum.
- $N_{14} \equiv$ Number of ^{14}C present in aluminum.
- $SA_{22} \equiv$ Specific Activity of ^{22}Na in aluminum.
- $SA_{14} \equiv$ Specific Activity of ^{14}C in aluminum.
- $m =$ Mass of aluminum object.

As a function of time (t), the specific activity is defined as:

$$SA \equiv \frac{A(t)}{m} \quad \text{where} \quad A(t) \equiv \lambda \cdot N(t)$$

Specifically:

$$SA_{22} \equiv \frac{\lambda_{22} \cdot N_{22}(t)}{m} \quad \text{and} \quad SA_{14} \equiv \frac{\lambda_{14} \cdot N_{14}(t)}{m}$$

When the irradiation times are short compared with the half lives of the radioisotopes and the particle flux is approximately constant in time, the ratio of the number of radioisotopes of a given type in a material is approximately proportional to the ratio of their cross sections of formation. At 1 GeV the formation cross sections for ^{22}Na and ^{14}C in aluminum are approximately 12.3 mb and 1.6 mb respectively. Thus:

$$\frac{N_{22}(t)}{N_{14}(t)} \equiv \frac{12.3}{1.6} = 7.67$$

The ratio of the specific activities of ^{22}Na and ^{14}C would then become:

$$\frac{SA_{22}}{SA_{14}} = \frac{\lambda_{22} \cdot N_{22}(t)}{\lambda_{14} \cdot N_{14}(t)} = \frac{\lambda_{22} \cdot (7.67) N_{14}(t)}{\lambda_{14} \cdot N_{14}(t)}$$

By definition, the decay constants are:

$$\lambda_{22} \equiv \frac{\ln 2}{\tau_{22}} \quad \text{and} \quad \lambda_{14} \equiv \frac{\ln 2}{\tau_{14}}$$

Substitution into the ratio of the specific activities then gives:

$$\frac{SA_{22}}{SA_{14}} = (7.67) \cdot \left(\frac{\tau_{14}}{\tau_{22}} \right) = (7.67) \cdot \frac{5730 \text{ yrs}}{2.602 \text{ yrs}} = 1.7 \times 10^4$$

Since the maximum specific activity observed for ^{22}Na in aluminum tags over the last three years at Fermilab is 3100 pCi/g., it is reasonable to expect the maximum specific activity of ^{14}C in that same aluminum material to be approximately 0.2 pCi/g. Since it takes approximately 100 ml of H_2SO_4 to dissolve 5 grams of Al, one can expect the concentration of ^{14}C in any liquid cocktail to be no higher than 0.01 pCi/ml which is more than an order of magnitude lower than the approximate detection limit of 0.5 pCi/ml for routine Liquid Scintillation Counting (LSC).

Example 2: Copper in a beam enclosure.

Using analogous definitions to example 1, one can express the specific activities of ^{54}Mn and ^{59}Ni in the copper as:

$$SA_{54} \equiv \frac{\lambda_{54} \cdot N_{54}(t)}{m} \quad \text{and} \quad SA_{59} \equiv \frac{\lambda_{59} \cdot N_{59}(t)}{m}$$

Under the same assumptions stated in example 1, the number of atoms of ^{54}Mn and ^{59}Ni will be approximately proportional to the production cross section. At 1 GeV, the formation cross sections for ^{54}Mn and ^{59}Ni are approximately 30.9 mb and 20.8 mb respectively.

$$\frac{N_{54}(t)}{N_{59}(t)} \cong \frac{30.9}{20.8} = 1.49$$

Substituting for the number of ^{54}Mn atoms and the appropriate definitions of the decay constants, one obtains the specific activity ratio as:

$$\frac{SA_{54}}{SA_{59}} = (1.49) \cdot \left(\frac{\tau_{59}}{\tau_{54}} \right) = (1.49) \cdot \frac{7.5 \times 10^4 \text{ yrs}}{0.855 \text{ yrs}} = 8.8 \times 10^4$$

The maximum specific activity observed for ^{54}Mn in activated copper tags over the last three years at Fermilab was 150 pCi/g. A concomitant specific activity for ^{59}Ni would be on the order of 0.002 pCi/g. One hundred milliliters (100 ml) of dilute HNO_3 will dissolve approximately 75 grams of Cu. One can thus expect that the concentration of ^{59}Ni in any liquid cocktail to be no higher than 0.0015 pCi/ml which is now more than 2 orders of magnitude below the approximate detection limit of 0.5 pCi/ml for routine LSC

counting.

Thus it is clear that the long half lives of these exclusive beta emitters make them undetectable using the most efficient routine counting methodology available, i.e., LSC systems. Even with the most sensitive measures that technology will currently allow, the predicted levels of ^{14}C and ^{59}Ni are probably not assayable on commercially available LSC systems.

Measurements in the Recent Past at Fermilab:

Samples submitted to the Activation Analysis Laboratory for radionuclide specific analyses which have unusually high concentrations of gamma emitting accelerator produced radionuclides, such as closed loop cooling system water, are routinely subjected to liquid scintillation counting (LSC) analyses as well as gamma ray spectroscopy. To date, no statistically significant evidence of any beta emitters other than ^3H in the region from .001 to 2 MeV has been observed in these relatively high concentration samples during the LSC analyses.

Conclusion:

While these particular isotopes can be typically observed in activated materials from nuclear reactors, the neutron fluences at accelerators are simply not sufficient to overcome the low concentrations of target nuclei from which ^{14}C , ^{59}Ni , ^{63}Ni , ^{93}Mo , and ^{151}Sm can be efficiently produced in most cases. Given our current understanding of the activation process at accelerators, the historical treatment of exclusive beta emitters produced in an accelerator environment, the relatively long half lives of all the exclusive beta emitters, and the absence of any statistically significant evidence for the existence of exclusive beta emitters in samples analyzed for ^3H content, one can only conclude that there is no reasonable expectation that the exclusive beta emitters ^{14}C , ^{59}Ni , ^{63}Ni , ^{93}Mo , and ^{151}Sm exist in measurable quantities in Fermilab waste streams. .

There is, however, the possibility that measurable concentrations of ^{63}Ni can exist in stainless steels or copper which have experienced atypically long resident times in active beam line enclosures. These rare situations can be identified and administratively controlled as in the case of accelerator decommissioning.

TABLE 1
Selected Radionuclide Half Lives

Radionuclide	Decay Mode	Half Life (yrs.)
3H	Very low energy beta (18.6 keV)	12.33
7Be	Electron capture with a 477 keV gamma in 10% of decays.	0.1459
11C	Moderate energy positron (1982 keV)	3.88E-05
14C	Low energy beta (156 keV)	5730
40K	Moderate energy beta (1505 keV)(89.33%) or low energy positron (10.7%) with accompanying 1460.8 keV gamma.	1.28E+09
54Mn	Electron capture with 834.8 keV gamma ray	0.8548
59Ni	Positron decay (1073 keV) with low energy X-rays	7.50E+04
60Co	Moderate energy beta (1491 keV) with two high energy gamma rays	5.271
63Ni	Low energy beta decay (65.9 keV)	100.1
93Mo	Internal conversion/Auger electrons accompanied by low energy X-rays.	3500
151Sm	Beta decay with low energy X-rays.	90

TABLE 2
Direct Reaction Cross Sections

Target	Reaction	Radionuclide	Maximum Cross Section (mb)	Asymptotic Cross Section (mb)
14N	(n,p)	14C	270	30 (10)
7Li	(p,n)	7Be	580 (11)	4 (12)
23Na	(n,2n)	22Na	200 (13)	20 (13)
55Mn	(n,2n)	54Mn	1100 (13)	10 (13)
54Fe	(n,p)	54Mn	500	30 (10)
58Ni	(n,gamma)	59Ni	430	7.5
58Ni	(n,p)	58Co	650 (14)	150 (14)
59Co	(n,gamma)	60Co	18800 (15)	1
59Co	(n,2n)	58Co	900	900
60Ni	(n,2n)	59Ni	400	330
60Ni	(n,p)	60Co	200	30 (10)
62Ni	(n,gamma)	63Ni	14,500 (17)	7.3
63Cu	(n,alpha)	60Co	50	14
63Cu	(n,p)	63Ni	133 (17)	30 (10)
92Mo	(n,gamma)	93Mo	2100	10
150Sm	(n,gamma)	151Sm	1000000 @ thermal	40 @ 300keV
151Eu	(n,p)	151Sm		30 (10)

TABLE 3
Spallation Cross Sections

Reaction	Energy (MeV)	Calculated Cross Section (mb)	Reaction	Energy (MeV)	Calculated Cross Section (mb)
27Al(h,X)14C	100	0.048	27Al(h,X)22Na	100	16.716
	500	1.083		500	14.093
	1000	1.642		1000	12.292
	10000	1.792		10000	9.586
	100000	1.792		100000	9.586
56Fe(h,X)14C	100	3.65E-09	27Al(h,X)7Be	100	7.44E-04
	500	0.012		500	0.293
	1000	0.126		1000	0.721
	10000	0.438		10000	1.055
	100000	0.438		100000	1.055
63Cu(h,X)14C	100	7.35E-11	27Al(h,X)3H	100	8.82E-05
	500	0.004		500	0.179
	1000	0.072		1000	0.58
	10000	0.334		10000	1.003
	100000	0.334		100000	1.003
96Mo(h,X)14C	100	5.95E-19	56Fe(h,X)22Na	100	1.26E-06
	500	2.44E-05		500	0.15
	1000	0.004		1000	0.942
	10000	0.077		10000	2.345
	100000	0.077		100000	2.345
184W(h,X)14C	100	1.21E-40	63Cu(h,X)22Na	100	2.54E-08
	500	2.25E-11		500	0.053
	1000	1.67E-06		1000	0.539
	10000	1.21E-03		10000	1.79
	100000	1.21E-03		100000	1.79
96Mo(h,X)63Ni	100	1.24E-06	96Mo(h,X)22Na	100	2.05E-16
	500	0.097		500	3.18E-04
	1000	0.565		1000	0.031
	10000	1.322		10000	0.41
	100000	1.322		100000	0.41
184W(h,X)63Ni	100	2.52E-28	96Mo(h,X)54Mn	100	5.32E-08
	500	8.95E-08		500	0.166
	1000	2.26E-04		1000	1.798
	10000	0.021		10000	6.14
	100000	0.021		100000	6.14
184W(h,X)93Mo	100	6.72E-20	184W(h,X)22Na	100	4.16E-38
	500	1.09E-04		500	2.93E-10
	1000	0.035		1000	1.25E-05
	10000	0.915		10000	0.006
	100000	0.915		100000	0.006
184W(h,X)151Sm	100	9.28E-08	184W(h,X)60Co	100	1.01E-28
	500	0.007		500	1.23E-07
	1000	0.042		1000	3.82E-04
	10000	0.096		10000	0.04
	100000	0.096		100000	0.04

TABLE 4
Activation Cross Sections from High Energy Protons

Energy (MeV)	X-Section for 11C on C (mb) a)	X-Section for 24Na on Al (mb) a)	X-Section for 22Na on Al (mb) b)	X-Section for 7Be on Al (mb) b)	X-Section for 24Na on Cu (mb) g)	X-Section for 22Na on Cu (mb) g)
50	86.4	6.2				
60	81.1	8.7				
80	70.5	10				
100	61.3	10.2				
150	45	9.4				
200	39	9.3				
300	35.8	10.1				
400	33.6	10.5				
500			14.1 +/- 1.0 a)			
600	30.8	10.8				
1000	28.5	10.94 +/- 0.24 d)				
2000	27.2	9.5	12.3 +/- 0.7		3.40 +/- 0.3	1.90 +/- 0.13
3000	27.1	9.1	11.0 +/- 0.7	7.5 +/- 0.4	2.96 +/- 0.3	1.85 +/- 0.13
6000	27	8.7	14.1 +/- 1.4	6.9 +/- 0.4	3.4 +/- 0.3	2.40 +/- 0.17
10000	26.9	8.6			3.3 +/- 0.3	2.85 +/- 0.20
28000	25.9 +/- 1.2 e)	7.92 +/- 0.18 e)	9.8 +/- 0.3	7.89 +/- 0.29	3.56 +/- 0.07 f)	2.75 +/- 0.19
150000					3.57 +/- 0.03 f)	
300000	24.6 +/- 1.6 e)		9.4 +/- 0.8 c)	8.7 +/- 0.7 c)		
400000					3.64 +/- 0.05 f)	
800000					3.62 +/- 0.04 f)	
a) J.B. Cumming, Monitor Reactions for High Energy Proton Beams, Annual Reviews of Nuclear Science(1963) 261-286.	b) J.B. Cumming, et. al., Phys. Rev. 128, No. 5(1962)2392.	c)S.B. Kaufman, et. al., Phys. Rev. C 19, No. 3(1979)962.	d)J.B. Cumming, Nucl. Inst. & Meth. 180(1981)37-44.	e)S.I. Baker, et. al., Nucl. Inst. & Meth. in Phys. Res. 222(1984)467-473	f) S.I. Baker, et. al., Phys. Rev. C 43, No. 6(1991)2862-2865	g) J. Hudis, et. al., Phys. Rev. 129, No. 1(1962)434-437.

REFERENCES

- (1) Fiscal Year 1994 Waste Management Assessment Report, Westinghouse Hanford Company Assessment Team, Assessment #9453028.
- (2) A. Elwyn, and V. Cupps, Fermi National Accelerator Laboratory Radioactivity Release Criteria for Materials, Equipment, and Waste, Fermilab ES&H Section publication, **Radiation Physics Note #109**, May 1993.
- (3) A. Rindi, S. Charalambus, Airborne Radioactivity Produced at High-Energy Accelerators, **Nuclear Instruments and Methods** **47** (1967) p227.
- (4) A. Rindi, Induced Radioactivity in the Cooling Waters of the 300 GeV SPS, Rep. LABII-RA/NOTE/72-13, CERN, Geneva (1972).
- (5) Ralph H. Thomas and Graham R. Stevenson, Radiological Safety Aspects of the Operation of Proton Accelerators, Technical Reports Series No. **283**, International Atomic Energy Agency, Vienna, 1988.
- (6) H.W. Patterson and R.H. Thomas, Accelerator Health Physics, Academic Press, New York (1973).
- (7) M. Paul, L.K. Fifield, D. Fink, A. Albrecht, G.L. Allan, G. Herzog, and C. Tuniz, Measurements of ⁵⁹Ni in meteorites by accelerator mass spectrometry, **Nucl. Inst. and Meth. in Physics Research B** **83**, (1993)275-283.
- (8) Y. Wang, Handbook of Radioactive Nuclides, The Chemical Rubber Co., Cleveland, Ohio, 1969.
- (9) V. McLane, C.L. Dunford, P.F. Rose, Neutron Cross Sections, Volume 2-Neutron Cross Section Curves, Academic Press, Inc., San Diego, CA. 1988.
- (10) Review of Particle Properties, Physics Letters B **239**, Particle Data Group, North-Holland, III.82 (1990).
- (11) R.R. Borchers, and C.H. Poppe, Physcial Review **129**, 2679 (1963).
- (12) L. Valentin (*in French*), Nuclear Physics **62**, 81 (1965).
- (13) Y. Uwamino, et. al., Nuclear Science and Engineering **111**, 391-403 (1992).
- (14) Neutron Flux Spectra Determination by Multiple Foil Activation-Iterative Method, RSIC Peripheral Shielding Routine Collection, SNL-SAND -II, PSR-345, Radiation Shielding Information Center, Oak Ridge, TN. (1994).
- (15) Handbook on Nuclear Activation Data, Technical Reports Series No. **273**, International Atomic Energy Agency, Vienna, 1987.
- (16) ASM Metals Reference Book, American Society for Metals, Metals Park, Ohio, 1981.

- (17) M. Numajiri, Y. Oki, T. Suzuki, et. al., Estimation of Nickel-63 in Steel and Copper Activated at High-energy Accelerator Facilities, **Appl. Rad. Isot.**, Vol. 45, No. 4, (1994)509-514.
- (18) Unpublished work by W. Freeman and P. Yurista to check calculations for the TeV I Safety Analysis Report.
- (19) V.R. Cupps, A. Elwyn, A. Lennox, T. Kroc, Fermilab Radiation Physics Note, to be published.
- (20) J.R. Lamarsh, Introduction to Nuclear Engineering, 2nd Edition, Addison-Wesley Publishing Company, Reading, MA., 1983.
- (21) M. Barbier, Induced Radioactivity, North -Holland Publishing Company, Amsterdam-London, 1969, (University Microfilms International, Ann Arbor, MI., current source).