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An Assessment of the Potential Radiation Exposure from Residual Radioactivity in Scrap Metal for Recycling

- 재활용 scrap metal 의 잔류방사능에 의한 방사선 피폭 평가 -

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Abstract

With current waste monitoring technology it is reasonable to assume that much of the material designated as Low Level Waste (LLW), generated within nuclear facilities, is in fact uncontaminated. This may include operational wastes, metal and rubble, office waste and discrete items from decommissioning or decontamination operations. Materials that contain only trivial quantities of radionuclides could realistically be exempted or released from regulatory control for recycle or reuse. A criterion for uncontrolled disposal of low-level radioactive contaminated waste is that the radiation exposure of the public and of each individual caused by this disposal is so low that radiation protection measures need not be taken. The International Atomic Energy Agency(IAEA) suggests an annual effective doses of 10 μSv as a limit for the individual radiation dose. In 1990, new recommendation on radiation protection standards was developed by International Commission on Radiological Protection (ICRP) to take into account new biological information related to the detriment associated with radiation exposure. Adoption of these recommendations necessitated a revision of the Commission's secondary limits contained in Publication 30, Parts 1～4. This study summarized the potential radiation exposure from valuable scrap metal considered to uncontrolled recycle by new ICRP recommendations. Potential exposure pathways to people following were analyzed and relevant models developed. Finally, concentrations leading to an individual dose of 10 μSv/yr were calculated for 14 key radionuclides. These potential radiation exposures are compared with the results of an IAEA study.

1. Introduction

The recycle and reuse of materials and equipment has increased in recent years partly for two reasons: a recognition of the economic opportunities presented and an increased societal awareness of the benefits of conserving raw materials and natural resources. The processes involved in maintaining, refurbishing, and decommissioning nuclear fuel cycle facilities generate materials and equipment that are radioactively contaminated or activated to varying degrees. Materials that contain only trivial quantities of radionuclides could realistically be exempted or released from regulatory control for recycle or reuse.

A criterion for uncontrolled disposal of low-level radioactive contaminated material is that the radiation exposure of the public and of each individual caused by this disposal is so low that radiation protection measures need not be taken. For the purposes of developing radiological control levels for recycling or reuse, the International Atomic Energy Agency (IAEA) recommended the adoption of an individual dose limit to the critically exposed population group on the order of 1 mrem (10 μSv) in a year from each exempt practice[1].

In 1990, new recommendation on radiation protection standards was developed by ICRP to take into account new biological information related to the detriment associated with radiation exposure[2]. Adoption of these recommendations necessitated a revision of the Commission's secondary limits contained in Publication 30, Parts 1～4[3]. Since ICRP Publication 61[4] and 67[5] were issued, ICRP has adopted a revised model for the respiratory tract issued in 1994 as publication 66[6]. Consequently, the inhalation dose coefficients of Publication 56[7] had to be recalculated using the revised model, and age-dependent inhalation dose coefficients for radio-isotopes of the remaining elements of Publications 67 and 69[8] had to be calculated. Recently, committed effective dose coefficients for workers had been calculated with the new respiratory tract model and issued by ICRP in Publication 68[9].

The main objective of this study is to illustrate methodologies for assessing potential radiation exposures, for the recycling and reuse of scrap materials that contain residual radioactive contamination with the new ICRP respiratory tract model. The initial radiological control criteria are the concentrations in or on materials considered for recycling or reuse that meet the individual dose criteria. The analyses include determining relevant radionuclides, potential mechanisms of exposure, and methods to determine possible health-related impacts from residual radioactive contamination in materials considered for recycling or reuse.

The preliminary results described in this paper are based on generic exposure scenarios and pathway analyses using 14 key-radionuclides determined to be potentially present as residual contamination in scrap metals from generic nuclear facilities operations. The scenarios and information developed by the IAEA in Safety Series No. 111-P-1.1 were considered as a primary reference in developing the initial radiation exposure pathway and scenario analysis[10]. Although alternative public dose limits were considered, the initial control criteria in this study are based on a dose of 10 μSv/yr (1 mrem/yr) to a worker in a smelter or an individual who uses consumer products made from recycled materials.

2. Dose Assessment Methods

To determine if radioactively contaminated materials can be released from regulatory controls, it is necessary to first determine the potential future uses for the materials and then the potential radiation doses resulting from those future uses. Generic radiation exposure scenarios were used to conceptually model likely future uses for materials released for recycle or reuse. While these scenarios may not exactly match existing or projected future conditions, they are designed to serve as the basis for conducting a dose analysis for the average member of a critical population group. These scenarios are a combination of radiation exposure pathways that contain specific exposure conditions. This section contains a summary of the basic radiation exposure pathways, scenarios, and methods used to estimate the potential exposure for recycle of scrap metals.

2.1. General Assumption

For the preliminary calculations that follow, it is necessary to assume that 100 t of contaminated steel and aluminum with a normalized unit of initial activity per unit mass are recycled during a year. This assumption allowed a normalized calculation leads to the development of bulk contamination control criteria. A unit concentration of each radionuclide is assumed and control criteria in terms of Bq/g for volume contamination are derived. For the scenario calculations that follow, 14 reference radionuclides were selected. The radionuclides considered and their physical half-lives are listed in Table 1.

Early daughter products in equilibrium with parent radionuclides are assumed in all cases. For smelting, it is probable that the majority of some radionuclides, such as ^{60}Co, remain in the ingot. However, a fraction of material will remain in the slag, and another portion will likely volatilize and be released with fumes and gases. The behavior of a specific radionuclide will depend on the chemistry of the radionuclide in question and the type of smelting process considered. Because the partitioning is not known for most radionuclides during smelting, the dose calculations that follow are based on the conservative assumption that, for each radionuclide, all of the activity retained in each of the three phases of smelting: the metal (steel or aluminum), the slag and gases released out of the stack. The stag is assumed to equal about 10% of initial mass of the steel, or about 10 t in the steel and aluminum analyses. This triple accounting approach will overestimate the true doses; however it will maximize the potential importance of the scenarios and should serve as an adequate basis for the initial development of radiological control criteria for recycling and reuse.

Table 1. Radionuclides Considered in the Recycle or Reuse Analysis.

Group Characteristic	Radionuclides	Half-life(a)
Low Dose Conversion Factors	^{36}Cl	3.0E+5
	^{41}Ca	1.3E+5
	^{55}Fe	2.7E+0
	^{63}Ni	1.0E+2
	^{99}Tc	2.1E+5
Alpha Emitters with Large Inhalation Dose Conversion Factors	^{238}U	4.5E+9
	^{239}Pu	2.4E+4
	^{241}Am	4.3E+2
Photon Emitters with Large External Dose Conversion Factors	^{60}Co	5.3E+0
	^{94}Nb	2.0E+4
	^{152}Eu	1.3E+1
	^{54}Mn	8.5E-1
	^{65}Zn	6.7E-1
	^{137}Cs	3.0E+1
No Photon Emission with Moderated Internal Dose Conversion Factors	^{90}Sr	2.9E+1
	^{241}Pu	1.5E+1

2.2. Radiation Exposure Pathways

Humans may be exposed to radiation in three main ways:

- exposure to external radiation
- inhalation of radioactive gases or small particles
- secondary ingestion of radioactive material.

The following sections describe the specific ways in which these pathways have been used as a part of the assessment methods in this study.

2.2.1. External Radiation Exposure

A self-absorbing, homogeneous, cylindrical volume generally represents the radioactive sources considered in this study or surface contaminated source with the dose point on the central, longitudinal axis of the cylinder. Except for exposure conditions that represent exposure to molten metals contained in a furnace, external absorbers and shields are ignored. This *procedures to maximize the estimated dose equivalents from exposure to external radiation. In* some situations, a half cylinder than by a full cylinder can represent a source better. For these situations, a full cylinder is defined such that the area of its flat surface is twice that actually needed; the equivalent dose is then calculated by using the full cylinder and dividing the resulting dose by two. The computer code MICROSHIELD was used to calculate dose conversion factor for external doses from all operations, for their particular geometry, at varying distances, and for each radionuclides[11].

2.2.2. Inhalation Exposure

Inhalation dose coefficients (committed effective doses per unit intake, *e(50)*) for workers have been calculated with the new respiratory tract model, and issued by ICRP in Publication No. 68. Where revised biokinetic models and data have been given for adults in Publication 56, 67 and 69, they have also been used. Table 2 compares some of these dose coefficients for 1 and 5 μm AMAD aerosols with those calculated for 1 μm AMAD aerosols using the Publication 30 respiratory tract and biokinetic models. From Table 2, it can be seen that most of the inhalation dose coefficients for workers calculated with the new models, at both sizes, are within a factor of three of the corresponding doses on which the Publication 61 ALIs were based. Generally, doses calculated with the new respiratory tract model and AMAD of 5 μm are lower than those calculated using the Publication 30 model and parameters. Although total respiratory tract deposition is higher at 5 μm with the Publication 66 model (82%), than at 1 μm in the Publication 30 model (63%), about 34% of the intake is deposited in ET_1(Extrathoracic airways consists of anterior nose), where no absorption to blood takes place. There are exception (e.g. ^{140}Ba) for which the dose coefficient is up to about 80% higher, because of contributions to bladder wall and/or colon from the excretion pathways now included in the biokinetic models. The concentration of respirable dust in the air will vary depending upon a variety of factors including the physical condition of the material being handled, the quantity of the material present, and the building ventilation. Thus, it is difficult to predict the concentrations that may be present during any recycle step. However, so that a complete analysis may be performed, air concentrations have been assumed based on the information in IAEA Safety Series No. 111-P-1.1 for those recycle steps where the potential for inhalation is most likely. In general, the air concentrations were assumed to vary between about 10^{-3} and 10^{-5} g/m^3.

Table 2. Ratios of Inhalation Dose Coefficient *e(50)* in Publication 68 to that Calculated for a 1μm AMAD Aerosol using Publication 30 Respiratory Tract and Biokinetic Models for a Range of Radionuclides.

Type/Class	F/D		M/W		S/Y	
AMAD	1μm	5μm	1μm	5μm	1μm	5μm
S-35	0.62	0.94	1.96	1.62		
Co-60			1.07	0.79	0.52	0.30
Ni-63	0.51	0.61	0.69	0.48		
Sr-90	0.52	0.65			0.42	0.22
Zr-95	0.60	0.71	1.16	0.92	0.87	0.67
Nb-95			1.08	1.00	1.03	0.81
Mo-99	0.48	0.75			0.74	0.85
Tc-99m	1.09	1.82	2.32	3.54		
Ru-103	0.58	0.80	1.20	1.00	1.10	0.88
I-131	0.50	0.61	0.80	0.52	0.48	0.27
I-132	0.58	0.85				
Cs-137	0.74	1.54				
Ba-140	0.56	0.79				
Ce-144	0.91	1.45	0.69	0.47	0.49	0.29
Po-210			2.96	2.20		
Np-237	0.60	0.71	0.28	0.19		
Pu-239			0.69	0.47	0.23	0.13
Pu-241			0.65	0.45	0.17	0.09

2.2.3. Ingestion Exposure

For this study, ingestion is assumed to occur by one of three separate routes:

- ingestion of removable radioactive materials on surfaces
- ingestion of corroded material from using frying pans or water pipes
- ingestion of food products contaminated by airborne released from a smelter.

Ingestion of removable radioactive contamination found on recycled metals or reused equipment can occur when workers inadvertently transfer contamination from a surface to hands, foodstuffs, cigarettes, or other items that enter the mouth. Since very little information exists on the estimated radiation doses associated with this pathway, the methods outlined by the IAEA for recycle and reuse are used for this study. Adult workers at a smelter assume a quantity of 10 mg of contamination per hour of direct contact exposure for ingestion. Ingestion of contaminated metal corroded from frying pans during cooking or from water pipes is considered as separate ingestion pathways. Since the use of stainless steel and aluminum pans with a much lower corrosion rate is perhaps more consistent with current domestic practice, a lower value of 0.13 mm/yr is used for this study[12].

3. Radiation Exposure Scenarios

The general steps for recycling of steel were divided by the IAEA into the sets of representative exposure scenarios given in Table 3. In the IAEA study, the first seven recycling steps and associated scenarios shown in Table 3 were intended to model metal recycling from delivery of the scrap metal to distribution of the manufactured consumer products. For this analysis, two separate categories of contaminated (or activated) materials and future conditions were considered:

- recycle of steel
- recycle of aluminum

These two categories are further subdivided into various exposure scenarios, describing the activities of a specific individuals or groups of individuals. The ranges of scenarios evaluated is based on previous dose estimates for recycling and reuse to adequately represent those scenarios likely to be of generic importance and relevance to all nuclear facilities. The scenarios presented here yield the highest potential doses for each category of recycled material and radionuclide grouping, as determined from the IAEA study. Details of the scenarios considered relevant assumptions, and the values assigned to the important parameters are discussed in following sections.

3.1. Scenarios for Steel Recycle

Recycled steel may contain both activation products and surface contamination from reactor, coolant or other sources. The three most limiting scenarios identified by the IAEA are for 1) a slag worker at the smelter, 2) consumers who drive an automobile, and 3) consumers who work with a piece of large equipment made of recycled steel. For each scenario, exposure duration and the total number of individuals exposed are estimated based on the analysis procedures of previous study. The IAEA considered scenarios that account for the potential doses resulting from use of consumer products made from contaminated steel or slag. The consumer scenarios considered by the IAEA include slag use in asphalt: sheet steel use in constructing building, appliances, and automobiles: and consumer use of frying pans and large equipment.

Table 3. Data for Individual and Collective Dose Estimates for Metal Recycling Steps and Scenarios.

Recycle Step	Scenarios	External Exposure Category	Internal pathway		Individual exposure duration(h)	Collective exposure duration(h)	No. of exposed individuals	Air concentration (g/m3)
Scrap delivery	1.1 Loader 1	1	Inh	0	4	4	2	0.0005
	1.2 Truck driver	2			4	4	5	0
Scrap processing	2.1 Processor	3	Both	1	12	12	3	0.0001
Smelting	3.1 Worker1	4	Inh	0	80	80	10	0.0001
	3.2 Loader1	5	Both	1	4	4	5	0.001
	3.2 Loader2	5	Both	1	20	20	5	0.001
	3.3 Operator1	6	Both	1	5	5	3	0.001
	3.3 Operator2	7	Both	1	50	50	3	0.001
Industrial product / by product	4.1 Caster1	8	Both	1	2.5	2.5	2	0.001
	4.1 Caster2	8	Both	1	25	25	2	0.001
	4.2 Caster3	9	Both	1	50	50	2	0.001
	4.3 Slag Worker	10	Both	1	25	25	10	0.001
	4.4 Loader2	11			2	2	2	0
	4.5 Truck driver2	12			5	5	5	0
Initial Fabrication	5.1 Worker2	13			40	40	10	0
	5.2 Sheet worker	14	Both	1	1	1	15	0.0001
	5.3 Coil worker	15	Both	1	1	1	1	0.0001
Final Fabrication	6.1 Sheet worker	14			1	1	20	0
	6.2 Coil worker	15			80	80	5	0
Distribution	7.1 Loader2	11			20	20	2	0
	7.2 Truck driver2	12			8	8	5	0
	7.3 Sheet worker	14			20	20	20	0
	7.4 Worker3	16			2000	2000	5	0
Consumer use	8.1 Parking lot	17			2000	2000	1	0
	8.2 Room	18			1500	500	440	0
	8.3 Appliance	19			1000	500	4300	0
	8.4 Automobile	20			2000	300	1200	0
	8.5 Frying pen	21	Ing	1	180	60	3300	0
	8.6 Large equipmer	25			2000	1000	200	0
Stack emmissions	9.1 Downwind individual							

3.2. Scenarios for Aluminum Recycle

Although the long-lived activation of aluminum is negligible, surfaces may become contaminated through contact with reactor coolant or other sources. Thus, the same 14 reference radionuclides considered for steel are also considered for the aluminum recycling scenarios. The surface contamination is assumed to be uniformly spread throughout the melted material with a final concentration of 1 Bq/g, consistent with the analysis method used for steel. The IAEA modified the general recycling steps shown in Table 3 by establishing sets of representative exposure scenarios for aluminum recycling as shown in Table 4. The general steps associated with aluminum recycling re quite similar to those previously described for steel recycling. The IAEA scenarios include conditions that represent loaders, truck drivers, processors, general workers, furnace operators, casters, and sheet workers. Also listed in Table 4 are the generic data used by the IAEA in analyzing aluminum recycling for issues of individual and collective exposure duration, number of people exposed and internal exposure pathways.

Table 4. Data for Individual and Collective Dose Estimates for Aluminum Recycling Scenarios.

Recycle Step	Scenarios	External Exposure Category	Internal pathway		Individual exposure duration(h)	Collective exposure duration(h)	No. of exposed individuals	Air concentration (g/m3)
Scrap delivery	1.1 Loader	1	Inh	0	4	4	2	0.0005
	1.2 Truck driver	2			4	4	5	0
Scrap processing	2.1 Processor	3	Both	1	12	12	3	0.0001
Smelting	3.1 Worker1	4	Inh	0	80	80	10	0.0001
	3.2 Operator	5	Both	1	50	50	3	0.001
Industrial product / by product	4.1 Caster	6	Both	1	25	25	2	0.001
	4.2 Worker2	7	Both	1	48	48	5	0.001
Fabrication	5.1 Sheet worker	8	Both	1	1	1	15	0.0001
	5.2 Worker3	9			80	80	5	0
Consumer use	6.1 Siding	10			1500	1500	500	0
	6.2 Automobile	11			2000	300	3400	0
	6.3 Frying pen	12	Ing	1	180	60	10000	0

4. Results and Discussion

The results of this preliminary study are based on generic exposure scenarios and pathway analyses using 14 radionuclides determined to be potentially present as residual contamination in scrap metals from nuclear facilities operations that may be considered for recycling or reuse. Although alternative public dose limits were considered, the initial control criteria in this study are based on a dose of 10 μSv/yr (1 mrem/yr) to a worker in a smelter or to an individual who uses consumer products made from recycled materials. Table 5 summarizes the comparison of limiting concentrations based on individual committed effective dose for residual radioactivity in (or on) recycled steel. For the radionuclides in this table, doses to slag workers or to users of consumer products provided the most restrictive (i.e., the smallest) derived residual radioactivity concentrations. The detailed results for aluminum are presented in Table 6 for individual dose and the initial control criteria for bulk materials in units of Bq/g. The overall results are summarized as follows:

- For alpha emitters with large inhalation dose conversion factors, the derived potential individual doses are about 60% ~ 70% decreased to the ICRP-30 case because of the impacts of the smaller dose coefficients.
- For photon emitters with large external dose conversion factors, there are no differences between the derived potential individual doses.
- For no-photon emitters with moderate internal dose conversion factors, the derived potential doses are about 40% ~ 60% decreased to the ICRP-30 case because of the impacts of the smaller dose coefficients.
- For other radionuclides, the derived potential doses are about 10% decreased to the ICRP-30 case except Tc-99. In case of Tc-99, it is estimated that potential doses from the ICRP-66 model are about two times larger than those of old model because of the impact of the increased effective dose coefficients.

Table 5. Initial Control Levels Based on a Potential Individual Dose for Recycling and Reuse of Steel Containing Residual Radioactive Contamination.

Characteristics	Radionuclide	Limiting scenario	Limiting Dose(Sv)		Initial Control Level(Bq/g)	
			ICRP-30	ICRP-66	ICRP-30	ICRP-66
Alpha Emitter	U-238	Slag worker	9.78E-06	2.30E-06	1.02E+00	4.34E+00
	Pu-239	Slag worker	3.84E-05	1.47E-05	2.60E-01	6.79E-01
	Am-241	Slag worker	3.85E-05	1.22E-05	2.60E-01	8.20E-01
Photon Emitter	Co-60	Large Equipment	8.80E-05	8.80E-05	1.14E-01	1.14E-01
	Nb-94	Large Equipment	5.70E-05	5.70E-05	1.75E-01	1.75E-01
	Eu-152	Automobile	2.40E-05	2.40E-05	4.17E-01	4.17E-01
	Mn-54	Automobile	2.80E-05	2.80E-05	3.57E-01	3.60E-01
	Zn-65	Large Equipment	1.80E-05	1.80E-05	5.56E-01	5.56E-01
	Cs-137	Automobile	2.20E-05	2.20E-05	4.55E-01	4.55E-01
No-Photon Emitter	Sr-90	Slag worker	2.02E-07	1.15E-07	4.95E+01	8.70E+01
	Pu-241	Slag worker	7.05E-07	2.67E-07	1.42E+01	3.75E+01
Others	Fe-55	Automobile	1.02E-09	1.10E-09	9.80E+03	9.08E+03
	Ni-63	Slag worker	6.02E-10	5.31E-10	1.66E+04	1.88E+04
	Tc-99	Slag worker	1.45E-09	3.12E-09	6.90E+03	3.21E+03

Table 6. Initial Control Levels Based on a Potential Individual Dose for Recycling and Reuse of Aluminum Containing Residual Radioactive Contamination.

Characteristics	Radionuclide	Limiting scenario	Limiting Dose(Sv)		Initial Control Level(Bq/g)	
			ICRP-30	ICRP-66	ICRP-30	ICRP-66
Alpha Emitter	U-238	Operator	1.95E-06	4.60E-07	5.12E+00	2.17E+01
	Pu-239	Operator	7.68E-06	2.95E-06	1.30E+00	3.40E+00
	Am-241	Operator	7.69E-06	2.44E-06	1.30E+00	4.10E+00
Photon Emitter	Co-60	Automobile	3.00E-05	3.00E-05	3.33E-01	3.33E-01
	Nb-94	Automobile	1.94E-05	1.94E-05	5.15E-01	5.15E-01
	Eu-152	Automobile	8.40E-06	8.40E-06	1.19E+00	1.19E+00
	Mn-54	Automobile	9.80E-06	9.80E-06	1.02E+00	1.02E+00
	Zn-65	Automobile	6.20E-06	6.20E-06	1.61E+00	1.61E+00
	Cs-137	Automobile	7.40E-06	7.40E-06	1.35E+00	1.35E+00
No-Photon Emitter	Sr-90	Frying Pen	7.20E-08	5.04E-08	1.39E+02	1.98E+02
	Pu-241	Operator	1.41E-07	5.34E-08	7.09E+01	1.87E+02
Others	Fe-55	Automobile	6.80E-09	6.80E-09	1.47E+03	1.47E+03
	Ni-63	Frying Pen	2.52E-10	2.70E-10	3.97E+04	3.70E+04
	Tc-99	Frying Pen	6.12E-10	1.40E-09	1.63E+04	7.12E+03

5. Conclusion

As with any generic analysis, there are uncertainties in the results that are associated with the generic parameter values and assumptions chosen for use in the analysis. As results of this study and the study performed by the IAEA, the following areas were identified where additional data are needed:

- Partitioning of radionuclides during smelting
- Detailed descriptions of smelters expects to use for the various recycled metals
- Acceptable levels of the lower limit of detection of radionuclides in bulk material
- Information on the maximum permissible levels of contaminants in metals

In establishing radiological control levels for unrestricted release, decisions should be based on limiting conditions for doses to individuals, dose to public, and collective dose. In this study, the first two conditions have been addressed, but because of uncertainty about the appropriate generic population distribution and meteorology to use, collective doses have not been assessed. As additional data are collected, this assessment will need to be revised.

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