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Emission factors are modified for heated solids.

Modification in Applying Appendix D of 40 CFR Part 61 to Heated Solid Radionuclide Materials With High Melting and Boiling Points

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Abstract: Appendix D of Title 40 Part 61 of the US Code of Federal Regulations (CFR) provides a procedure that US Department of Energy (US DOE) facility owners and operators can use to estimate radionuclide emissions to the atmosphere for dose calculations instead of measuring emissions for minor sources under the 40 CFR Part 61, Subpart H, National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities, regulation. The procedure assumes that any radioactive material heated above 100 °C is completely vaporized and emitted to the atmosphere. In 1991, the US DOE Oak Ridge Reservation (ORR) requested approval to use different release fractions (RFs) for uranium because of its high melting and boiling points. In response to the request, the US Environmental Protection Agency (US EPA) Region IV approved the use of modified RFs for elemental uranium provided no reaction had taken place to alter its chemical form. In 2015, the ORR requested approval to use different RFs for tungsten, again because of its high melting and boiling

points. EPA Region IV approved the use of modified RFs for heated radioactive tungsten metal. In accordance with the two precedents set for heating uranium and radioactive tungsten metals, in 2016, the ORR requested approval to use modified RFs in a similar fashion for other radioactive solid metals and compounds with melting and boiling points above 500 °C that might be heated above 100 °C in future research projects and experiments. EPA Region IV again granted approval to use modified RFs for the list of compounds. This note discusses the proposed modified RFs and their development. *Health Phys.* 124(5):431–437; 2023

Key words: operational topics; emissions; atmospheric; radioactive materials; regulations

INTRODUCTION

APPENDIX D of Title 40 Part 61 of the Code of Federal Regulations (CFR), Methods for Estimating Radionuclide Emissions (US EPA 1989a) provides a US Environmental Protection Agency (US EPA) procedure that facility owners and operators can use to

estimate, rather than measure, atmospheric radionuclide emissions for dose calculations from minor sources under the National Emission Standards for Emissions of Radionuclides Other Than Radon From Department of Energy Facilities regulation (40 CFR Part 61, Subpart H; US EPA 2002). The regulation limits the annual effective dose equivalent that any member of the public can receive from atmospheric emissions of radionuclides from US Department of Energy (US DOE) facilities to 0.1 mSv (10 mrem).⁴

A minor source is a release point (i.e., emission point) that has the potential to release radionuclides into the air resulting in a dose to the public of less than 1% of the standard, or less than 0.001 mSv y⁻¹ (0.1 mrem y⁻¹). In evaluating potential radionuclide emissions, 40 CFR 61.93(f) requires that “the estimated radionuclide release rates shall be based on the discharge of the effluent stream that would result if all pollution control equipment did not exist, but the facilities operations were otherwise normal.”

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The authors declare no conflicts of interest.

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⁴The US regulatory values/limits are generally stated in non-SI units; therefore, they are included as parenthetical values for tracking to the regulatory criteria.

Table 1. Typical decontamination factors (DFs) for various abatement controls (US EPA 1989a).

Abatement Control Device	Types of Radionuclides Controlled (i.e., Form)	DF to Emissions
HEPA filters	Particulates	0.01
Fabric filters	Particulates	0.1
Sintered metal	Particulates	1
Activated carbon filters	Iodine gas	0.1
Venturi scrubbers	Particulates	0.05
Packed bed scrubbers	Gases	0.1
Electrostatic precipitators	Particulates	0.05
Xenon traps	Xenon	0.1

The emissions estimate without the pollution control equipment is often referred to as the *unabated emission result*.

In the Appendix D procedure, radionuclide emissions to the atmosphere are estimated by first determining the amount of radioactive material that was used, handled, or in inventory (not emitted) for a calendar year in the facilities. Radioactive materials that are sealed and remain unopened during the assessment period do not need to be included. The following release fractions (RFs) based on the physical state of the radionuclide are then applied by multiplying the appropriate factor by the amount of material used:

- 1 for gases;
- 10^{-3} for liquids or particulate solids; and
- 10^{-6} for solids.

Finally, a decontamination factor (DF) can also be applied for control devices installed between the place of use and the point of release by multiplying the DF by the factored emissions as described. The DF is referred to in the Appendix D regulation as an *adjustment factor*, but here, *DF* is used as a more descriptive term. Thus, typical DFs (i.e., adjustment factors) for control devices also are identified in Appendix D, and typical DFs are provided in Table 1. Emissions estimates that include DFs are often referred to as the *abated emission result*.

Appendix D states, "If any nuclide is heated to a temperature of 100 degrees Celsius or more, boils at a temperature of 100 degrees Celsius

or less, or is intentionally dispersed into the environment, it must be considered to be a gas" (US EPA 1989a). It is similarly stated and applicable by EPA for certain facilities licensed by the Nuclear Regulatory Commission and non-DOE federal facilities (US EPA 1989b). Gases have a DF of 1. The temperature-dependent definition of a gas becomes problematic when heating radioactive materials with melting and boiling points significantly higher than 100 °C.

METHODS

On August 1, 1991, the US DOE Oak Ridge Reservation (ORR) requested approval to use different RFs for the heating of uranium because the melting point is 1,132 °C and the boiling point is 3,818 °C.⁵ On December 2, 1991, EPA Region IV granted the ORR approval to use modified RFs for the heating of elemental uranium, provided that no reaction altering its chemical form takes place.⁶ The resulting modified RFs are as follows:

- 1 for uranium heated above 3,000 °C;
- 10^{-3} for uranium heated above 1,100 °C but below 3,000 °C; and
- 10^{-6} for uranium at ambient temperature or heated below 1,100 °C.

⁵US Department of Energy. 1991. *ORR Compliance Plan Alternative Monitoring Methods*. Letter from R. R. Nelson to W. A. Smith, August 1, 1991, Oak Ridge National Laboratory Site Office, Oak Ridge, TN.

⁶US Environmental Protection Agency. 1991. *Request to Modify 40 CFR Part 61, Appendix D, RFs for Uranium at the Department Of Energy (DOE) Oak Ridge Reservation (ORR) and Paducah Gaseous Diffusion Plant (PGDP)*. 4APT-AEB. Letter from W. A. Smith to R. R. Nelson, December 2, 1991, EPA Region IV, Atlanta, GA.

The DFs for control devices (e.g., Table 1) presented in Appendix D will still be allowed.

In practical terms, the estimated emissions to the atmosphere of radioactive material is multiplied by both an RF and a DF as given in eqn (1). If there is more than one abatement control device in series, then multiple DFs are applied (Barnett 2011).

Therefore, in applying the approved US EPA modification, it is necessary to know the form of the radioactive material and any applicable abatement controls. When abatement control devices are not applicable to gas emissions, a special DF case exists in which the DF is set equal to 1:

$$A_{Estimated\ Release} = A_{Inventory} \times RF \times \prod (DF)_i (Bq), \quad (1)$$

where:

$A_{Estimated\ Release}$ = estimated release of a given isotope (Bq);

$A_{Inventory}$ = activity of given isotope used in the facility (Bq);

RF = release fraction for the material; and

DF_i = decontamination factor (s) for each device used in series.

New project and research experiments at US DOE's Oak Ridge National Laboratory (ORNL), located on the ORR, presented a dilemma in that additional radioactive materials with high melting and boiling points had to be considered gases when heated above 100 °C in accordance with the Appendix D procedure. ORNL researchers contended that this was problematic because many materials have boiling points greater than 100 °C that when heated above 100 °C are not gases. As mentioned previously, the following definition of a gas in the procedure is as follows: *If any nuclide is heated to a temperature of 100 degrees Celsius or more, boils at a temperature of 100 degrees Celsius or less, or is intentionally dispersed into the environment, it must be considered a gas.*

The researchers believed that a conjunction (either *and* or *or*) is

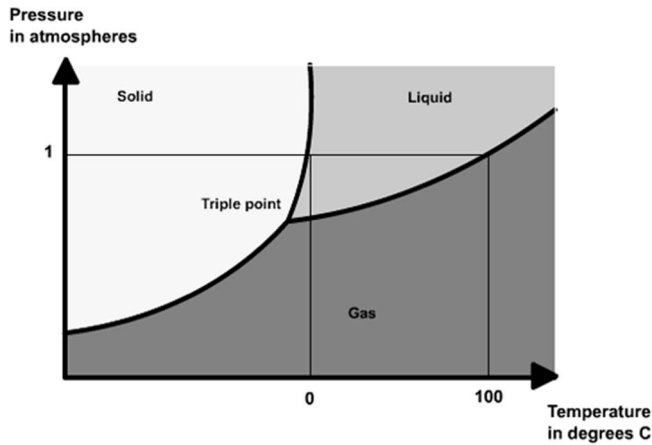


FIG. 1. Phase diagram for water (Kurtus 2021; used by permission).

missing after the first comma. If the missing conjunction is *and*, then the sentence assumes the following form: *If any nuclide is heated to a temperature of 100 degrees Celsius or more, [and] boils at a temperature of 100 degrees Celsius or less, or is intentionally dispersed into the environment, it must be considered a gas.*

At first glance, the sentence with the conjunction *and* after the first comma may solve the problem and could be acceptable; however, it still is not technically valid because of the complex and non-linear relationship between temperature and vapor pressure of a substance near, at, or above the substance melting point. If the missing conjunction is *or*, then the sentence assumes the following form: *If any nuclide is heated to a temperature of 100 degrees Celsius or more, [or] boils at a temperature of 100 degrees Celsius or less, or is intentionally dispersed into the environment, it must be considered a gas.*

However, considering the three criteria separately, the missing conjunction cannot be *or*. Discussions regarding the intent of the requirements in the Appendix D procedure motivated environmental compliance staff at ORNL to ensure future RF values were based on actual physical properties of materials being used. Upon further reading of Appendix D (US EPA 1989a) and EPA guidance (US EPA 1989b), the researchers determined that the RFs in Appendix D are likely based on the physical state of the radionuclide.

The physical state of a material depends on its temperature and surrounding pressure. A phase diagram is a graphical representation of the different physical states of a substance at varying temperatures and pressures. A constant pressure line can indicate the temperature at which the material changes from a solid to a liquid, which is its melting point, and the temperature at which it changes from a liquid to a gas, which is its boiling point. Conversely, a constant pressure line on the phase diagram can show the temperature at which a gas condenses to a liquid, which is its condensation point, and the point at which a liquid freezes into a solid, which is its freezing point. The freezing point is the same temperature as the melting point, and the boiling point is the same temperature as the condensation point.

The phase diagram for water (Fig. 1) shows that at a constant pressure of 101.3 kPa (1 atm), water changes from a solid (ice) to a liquid at 0 °C and changes from a liquid to a gas (water vapor) at 100 °C. Therefore, 0 °C is the melting point for solid water as the material is heated at a constant pressure of 101.3 kPa (1 atm) or the freezing point for liquid water as the material is cooled at a constant pressure of 101.3 kPa (1 atm). The boiling point of water at 101.3 kPa (1 atm) is therefore 100 °C, which is the same temperature as the condensation point for water vapor. Increasing the pressure of a

material will increase the required temperature to change the state of that material because the pressure—whether it is air pressure, water pressure, or pressure exerted by some surrounding material—will inhibit or prevent molecules of the material from breaking free from the material’s molecular attraction (Kurtus 2021). Therefore, water boils at a lower temperature at higher elevations because the surrounding pressure or atmospheric pressure is lower, and it boils at higher temperatures at elevations below sea level because the surrounding pressure is higher. Table 2 illustrates this phenomenon for water.

Sublimation is a phase change that occurs when a material transitions directly from a solid to a gas, completely skipping the liquid phase. The opposite of this phase change is deposition, which occurs when a gas changes phase to a solid without passing through the liquid phase. This occurs at pressures and temperatures below the triple point. The triple point of a substance is the temperature and pressure at which the three phases (gas, liquid, and solid) of that substance coexist together in equilibrium. An example of sublimation is the transition of dry ice (solid carbon dioxide) from a solid to a gas. The phase diagram of dry ice presented in Fig. 2 shows that dry ice sublimates at temperature and pressures below its triple point, which occurs at 526.9 kPa (5.2 atm) and – 56.6 °C (Rumble 2017).

Table 2. Boiling Point of Water Versus Altitude (The Engineering Toolbox 2021).

Altitude — Compared to Sea Level	Boiling Point
(m)	(°C)
0	100
152	99.5
457	98.4
762	97.3
1,067	96.3
1,372	95.2
1,676	94.2
1,981	93.1
2,286	92.1
2,591	91.1
2,896	90.1
3,048	89.6

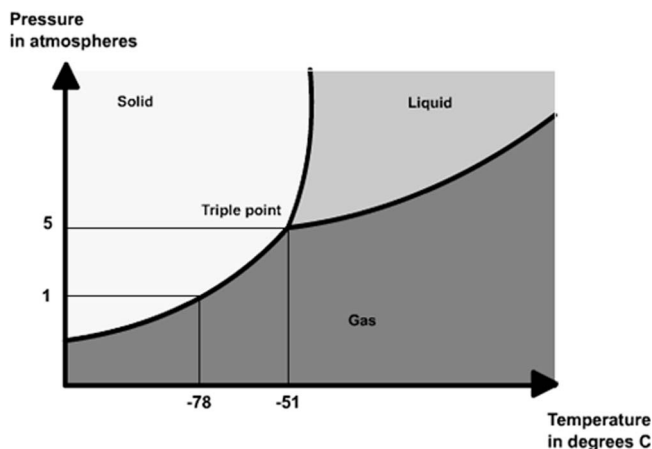


FIG. 2. Phase diagram for dry ice (Kurtus 2021; used by permission).

Studying the states of matter and changes that occur under different conditions demonstrates that the melting point is a very important property for solid materials. Volume changes are very small when a solid is heated or when the surrounding pressure changes, thus making the melting point a test method often used to identify and characterize solid materials. The melting point is also used as a criterion for purity. A sharp melting point (with a range less than about 1 °C) is often taken as evidence that the sample is fairly pure, whereas a wide melting point range is evidence that the sample is not pure. Impurities can even lower the melting point of a substance, which is a condition called melting point depression (Loudon 1988).

If the physical state of a material is needed to determine appropriate RFs, the Appendix D procedure for estimating radionuclide emissions likely falls short when solid radioactive materials with melting and boiling points significantly above 100 °C are heated. Clearly, if RFs are to be based on the physical state of a material, setting an arbitrary temperature such as 100 °C is not sufficient to account for the heating of many solid materials with high melting and boiling points that are not in the gas phase at that temperature. This conclusion was the basis for the original ORR request to use modified RFs for heating solid uranium metal and was the justification for additional requests to use

modified RFs for heating other solid radioactive materials that have high melting and boiling points.

RESULTS

On June 3, 2015, the ORR requested approval to use different RFs for heating radioactive tungsten, again because of its high (significantly above 100 °C) melting point (3,410 °C) and boiling point (5,660 °C).⁷ EPA Region IV granted approval to the ORR on June 25, 2015, to use modified RFs for heating radioactive tungsten metal, again provided that no reaction altering its chemical form occurs.⁸ This is the same condition of approval as for the heating of uranium metal that was granted in 1991. The modified RFs for heating radioactive tungsten metal are as follows:

- 1 for elemental tungsten heated to temperatures above 5,000 °C;
- 10^{-3} for elemental tungsten heated to temperatures above 3,400 °C but below 5,000 °C; and
- 10^{-6} for elemental tungsten heated to temperatures above ambient air temperature but below 3,400 °C,

⁷US Department of Energy. 2015. *Request for Approval for a Modification in Applying Appendix D of 40 Code of Federal Regulations (CFR) Part 61 to Heated Radioactive Tungsten Metal and Approval for Surrogate for Niobium (NB)-91 M*. Letter from J. O. Moore to G. Worley, June 2, 2015, Oak Ridge National Laboratory Site Office, Oak Ridge, TN.

⁸US Environmental Protection Agency. 2015. *Untitled*. Letter from B. H. Banister to J. Moore, June 25, 2015, EPA Region IV, Atlanta, GA.

provided the tungsten is in the solid phase.

DFs for effluent abatement controls as presented in Appendix D (see Table 1) are allowed. In accordance with the two precedents set for heating uranium and radioactive tungsten metals, on February 3, 2016, US DOE and ORNL requested approval to use modified RFs in similar fashion for other radioactive solid metals and compounds with melting and boiling points above 500 °C that might be heated above 100 °C, at ambient pressure, in future research projects and experiments.⁹

A list of about 95 radioactive solid metals and compounds was submitted to US EPA for consideration with the request for approval (Table 3). Most of these compounds are materials associated with nuclear fuel rods, spent fuel, depleted uranium, and uranium enrichment processing (e.g., compounds of actinium, barium, neptunium, plutonium, radium, thorium, tungsten, uranium, zirconium).

On February 23, 2016, EPA Region IV granted approval to ORR for ORNL to use modified RFs for the list of compounds that was submitted.¹⁰ The modified RFs, generally based on boiling and melting points rather than specifically stated temperatures or pressures, are as follows:

- 1 for radioactive solid metals and compounds heated to temperatures above or equal to the boiling point of the solid
- 10^{-3} for radioactive solid metals and compounds heated to temperatures above or equal to 90% of the

⁹US Department of Energy. 2016. *Request for Approval for a Modification in Applying Appendix D of 40 Code of Federal Regulations (CFR) Part 61 to Heated Radioactive Solid Materials With High Melting and Boiling Points*. Letter from J. O. Moore to G. Worley, February 3, 2016, Oak Ridge National Laboratory Site Office, Oak Ridge, TN.

¹⁰US Environmental Protection Agency. 2016. *Untitled*. Letter from B. H. Banister to J. Moore, February 23, 2016, EPA Region IV, Atlanta, GA.

Table 3. Listing of potential radioactive solid materials that ORNL may heat above 100°C in future research experiments (Rumble 2017; Speight 2017).

Radioactive Compound	Melting Point (°C)	Boiling Point (°C)
Tungsten Metal	3,410	5,660
Uranium Metal	1,132	3,818
COMPOUNDS OF ACTINIUM		
Actinium Metal	1,050	3,200
Actinium Bromide	800 (sublimes)	NA
Actinium Iodide	700 – 800 (sublimes)	NA
Actinium Trichloride	960 (sublimes)	NA
COMPOUNDS OF BARIUM		
Barium Metal	725.0	1,640
Barium <i>ortho</i> Arsenate	1,605	Not known
Barium Bromide	847	Not known
Barium Bromide Dihydrate	880	Not known
Barium Carbonate (α)	1,740	Decomposes
Barium Carbonate (β)	982 (transition point to α)	NA
Barium Carbonate (γ)	811 (transition point to β)	NA
Barium <i>perchlorate</i>	505	Not known
Barium Chloride	963	1,560
Barium Fluoride	1,355	2,137
Barium Hydride	675 (decomposes)	1,400 (estimated)
Barium Hexaboride	2,270	Not Known
Barium Iodide	711	2,027
Barium Iodide Hydrate	539; 740 (decomposes)	NA
Barium Molybdate	1,480	Not known
Barium Niobate	1,455	Not known
Barium Nitrate	592	Decomposes
Barium Nitride	Not Known	1,000 (decomposes)
Barium Oxide	1,973	3088
Barium Selenide	1,780	Not known
Barium <i>metasilicate</i>	1,604	Not known
Barium Sulphate	1,580	1,149 (transition point)
Barium Monosulphide	1,200	Not known
Barium Trisulphide	554	Not known
Barium Titanate	1,625	Not known
Barium <i>pyrovanadate</i>	863	Not known
Barium Zirconate	2,500	Not known
COMPOUNDS OF NEPTUNIUM		
Neptunium Metal	644	>3,900
Neptunium Tribromide	800 (sublimes)	NA
Neptunium Tetrachloride	538	Not known
Neptunium Trichloride	800 (sublimes)	NA
Neptunium Oxide	2,547	Not known
Neptunium Octatrioxide	500 (decomposes)	NA
COMPOUNDS OF PLUTONIUM		
Plutonium Metal	641	3,232
Diplutonium Trioxide	2,085 (in Helium)	Not known
Plutonium Dihyride	-727	Not known
Plutonium Dioxide	2,390	2,800 (decomposes)
Plutonium Oxide	1,900	Not known
Plutonium Sulphide	1,727	Not known
Plutonium Tetrafluoride	1,037 (decomposes)	NA
Plutonium Tribromide	681	>1,300 (decomposes)
Plutonium Trichloride	760	1,767
Plutonium Trifluoride	1,425	2,000 (decomposes)
Plutonium Triiodide	777	Not known
COMPOUNDS OF RADIUM		
Radium Metal	700	1,737
Radium Bromide	728	900 (sublimes)
Radium Carbonate	>1,100	Not known
Radium Chloride	1,000	Not known
COMPOUNDS OF THORIUM		
Thorium Metal	1,750	4,788
Thorium Hexaboride	2,195	Not known
Thorium Bromide	610 (sublimes)	NA
Thorium Carbide	2,655	5,000
Thorium Chloride	770	928 (decomposes)
Thorium Fluoride	1,110	1,680
Thorium Iodide	570	837
Thorium Nitrate	500 (decomposes)	NA

Continued next page

Table 3. (Continued)

Radioactive Compound	Melting Point (°C)	Boiling Point (°C)
Thorium Oxide	3,320	4,400
Thorium Sulphide	1,925	Not known
COMPOUNDS OF TUNGSTEN		
Ditungsten Carbide	2,860	6,000
Tungsten Carbide	2,870	6,000
Tungsten Diboride	2,900	Not known
Tungsten Dioxide	1,550	1,724 (decomposes)
Tungsten Pentoxide	800 (sublimes)	1,530 (decomposes)
Tungsten Trioxide	1,472	1,837
Tungsten Silicide	>900	Not known
Tungsten Sulfide	1,250 (decomposes)	NA
COMPOUNDS OF URANIUM		
Uranium Diboride	2,365	Not known
Uranium Dicarbide	2,350	4,370
Uranium Dioxide	2,827	Not known
Uranium Disulfide	>1,100	Not known
Uranium Mononitride	2,630	Decomposes
Uranium Monosulphide	>2,000	Not known
Uranium Tetrabromide	519	777
Uranium Tetrachloride	590	790
Uranium Tetrafluoride	1,036	1,414
Uranium Tetraiodide	506	757
Uranium Tribromide	730	Not known
Uranium Trichloride	837	1,657
Uranium Trifluoride	>1,000 (decomposes)	NA
Uranium Trioxide	1,300 (decomposes)	NA
Uranyl Acetate Chloride	577	Not known
Triuranium Octaoxide	1,300 (decomposes to UO ₂)	NA
COMPOUNDS OF ZIRCONIUM		
Zirconium Metal	1,852	3,577
Zirconium Carbide	3,532	5,100
Zirconium Chloride	727	1,292
Zirconium Diboride	3,245	4,193 (decomposes)
Zirconium Fluoride	932	912 (sublimes)
Zirconium Nitride	2,980	Not known
Zirconium Oxide	2,678	4,300
Zirconium Silicate	1,540 (decomposes)	NA
Zirconium Sulphide	~1,550	Not known

- melting point and below the boiling point of the solid
- 10^{-6} for radioactive solid materials heated to temperatures above ambient air temperature but below 90% of the melting point of the solid

Table 4 summarizes the RFs discussed in this paper. McCarter (2022) provides a summary of the ORR letters and US EPA approvals. Other US DOE sites would need to use a similar approach within their US EPA region to obtain their own site-specific

approval to use these factors or similar ones.

A conservative factor of 90% of the melting point was chosen to mitigate the effect of the melting point depression phenomenon in which impurities in the material

Table 4. US EPA and ORNL modified release fractions (RFs) for estimating radionuclide releases.

Material Form	40 CFR 61, Appendix D RFs ^a	EPA Region IV Approved ORNL Modified RFs ^b
Gas	1 — Applied to radioactive gases and any radionuclide that is heated to a temperature of 100 °C or more, boils at a temperature of 100 °C or less, or is intentionally dispersed into the environment, it must be considered to be a gas.	1 — Applied to approved radioactive gases and radioactive solid metals and compounds heated to temperatures above or equal to the boiling point of the solid.
Liquids/Particulates	10^{-3} — Applied to liquids and particulate solids.	10^{-3} — Applied to approved radioactive solid metals and compounds heated to temperatures greater than or equal to 90% of the melting point and less than the boiling point of the solid.
Solids	10^{-6} — Applied to solids.	10^{-6} — Applied to approved radioactive solid materials heated to temperatures above ambient air temperature but below 90% of the melting point of the solid.

^a EPA 1989a^b McCarter 2022

can lower the melting point. As with previous approvals, DFs for effluent controls are allowed. By way of example, using 1×10^{11} Bq of ^{227}Ac metal heated to 950°C (i.e., 90.5% of the $1,050^\circ\text{C}$ melting point of the metal) with two stages of HEPA filtration in the system, eqn (1) becomes

$$A_{(\text{Ac-227}) \text{ Estimated Release}} = 1 \times 10^{11} \times 1 \times 10^{-3} \times \Pi(0.01 \times 0.01) \text{ (Bq)}, \quad (2)$$

$$A_{(\text{Ac-227}) \text{ Estimated Release}} = 1 \times 10^4 \text{ (Bq)}. \quad (3)$$

CONCLUSION

The science behind the determination of RFs using the procedure in 40 CFR Part 61, Appendix D, Methods for Estimating Radionuclide Emissions, is likely based on the physical state of a material and is sufficient for establishing RFs for materials that are not heated. However, the actual physical properties of materials being used must be considered when the solids being heated have melting and boiling points significantly above 100°C . The modified RFs approved for the ORR and ORNL as described in this paper are based on the phase changes of the materials resulting from the individual melting and boiling points of each material. Also included is a conservative factor for the melting point to mitigate the effect of the melting point depression phenomenon. The procedure in Appendix D would be more accurate if

it incorporated the melting and boiling points of materials as the basis for determining the physical states of the material as opposed to using arbitrary temperatures such as 100°C .

Acknowledgments—This manuscript has been authored by UT-Battelle, LLC, under contract DE-AC05-00OR22725 with the US DOE, and the Pacific Northwest National Laboratory, which is operated for the US Department of Energy by Battelle under Contract DE-AC05-76RL01830. The US government retains and the publisher, by accepting the article for publication, acknowledges that the US government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this manuscript or allow others to do so, for US government purposes. US DOE will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (<http://energy.gov/downloads/doe-public-access-plan>).

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