Microdosimetry and Spectral Emissions of Metal Tritide Particles

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Abstract

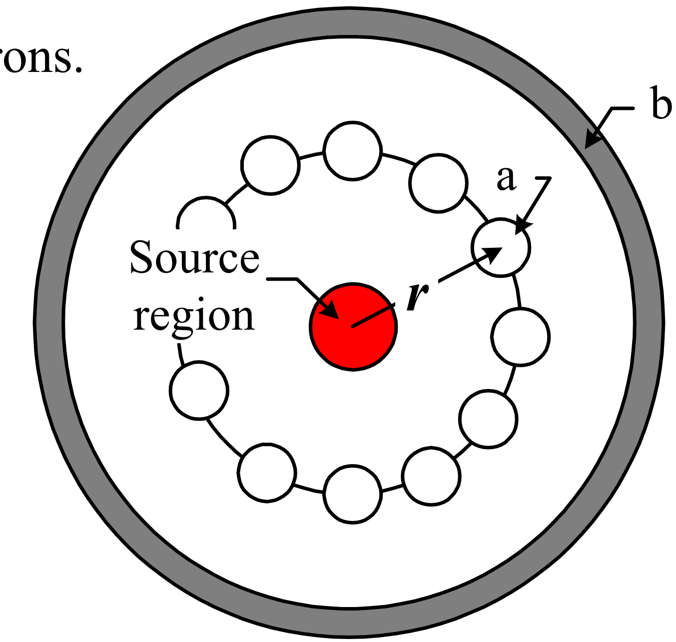
Inference of intakes and doses from inhalation of metal tritide particles has come under scrutiny as a result of decommissioning and decontamination of U.S. Department of Energy facilities. Since self-absorption is very significant for larger particles, interpretation of counting results of metal tritide particles by liquid scintillation requires information about emission spectra. Similarly, inference of dose requires knowledge of charged particle and photon spectra.

Using the PENELOPE Monte Carlo radiation transport computer code, we calculated various dosimetric, microdosimetric and spectral emissions from tritides of Sc, Ti, Zr, Er, and Hf. For metal tritide particles of various sizes, we present emission fractions, distributions of microdosimetric quantities, and the emitted spectra of electrons and bremsstrahlung x-rays. We also present results characterizing the effects of uncertainties associated with the composition and density of the tritides. Emissions from metal tritide particles are weakly penetrating and the emission spectrum tends to “harden” as the particle size increases. Microdosimetric considerations suggest that the radiation emitted by metal tritides can be classified as a low Linear Energy Transfer (LET) radiation source.

Methodology

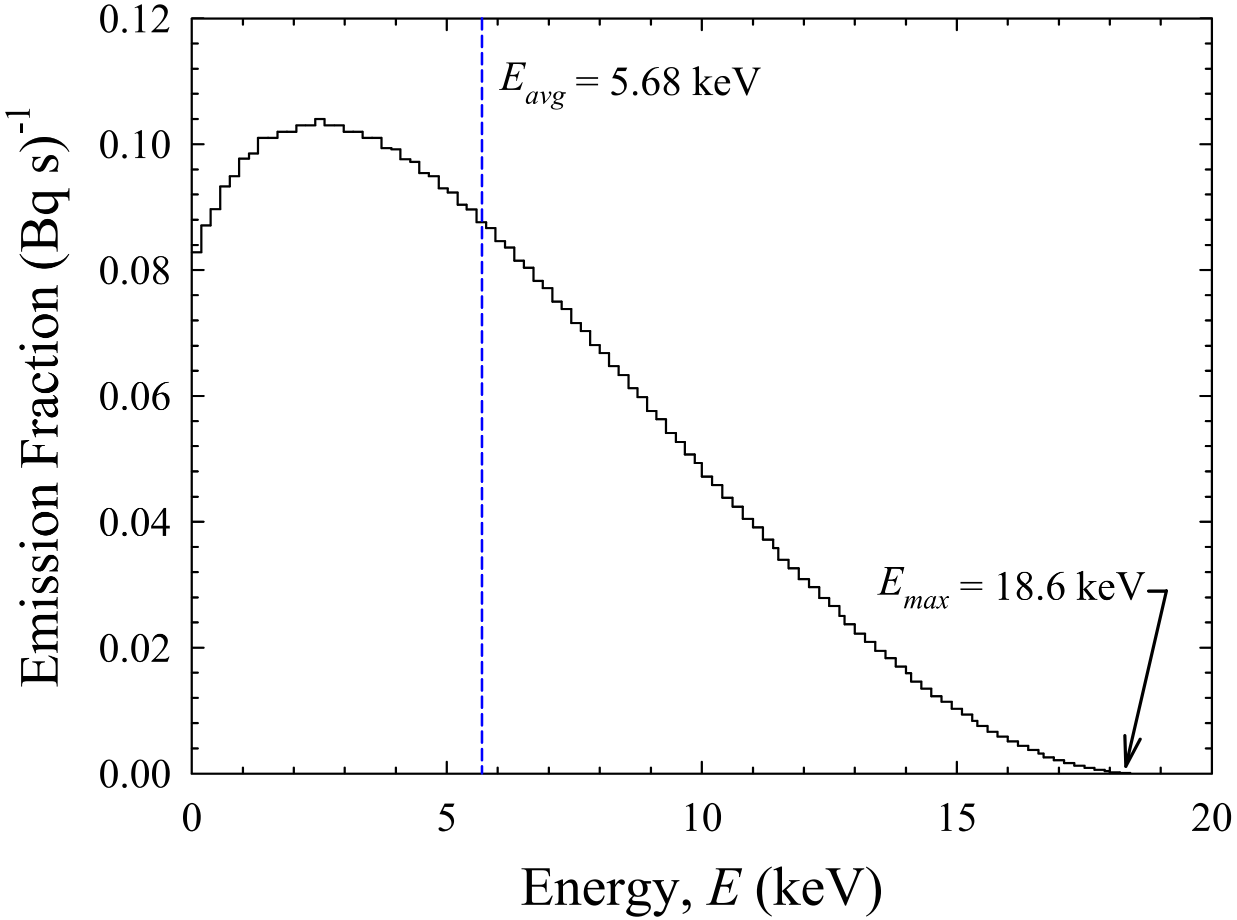
- Electrons and photons transported through the metal tritide and surrounding soft tissue using the PENELOPE (Salvat *et al.* 1999) Monte Carlo radiation transport code.
  - 100 eV cutoff energy for electrons and positrons.
  - 1 keV cutoff energy for photons.

- Absorbed dose and fluence calculations compare favorably to the EGS4, ETRAN, and the ITS3 code systems (Sempau *et al.* 1997).
- Microdosimetric quantities comparable to those computed using the PITS analog Monte Carlo code system (Stewart and Wilson 2000, unpublished).
  - Site sizes greater than about 200 to 500 nm.



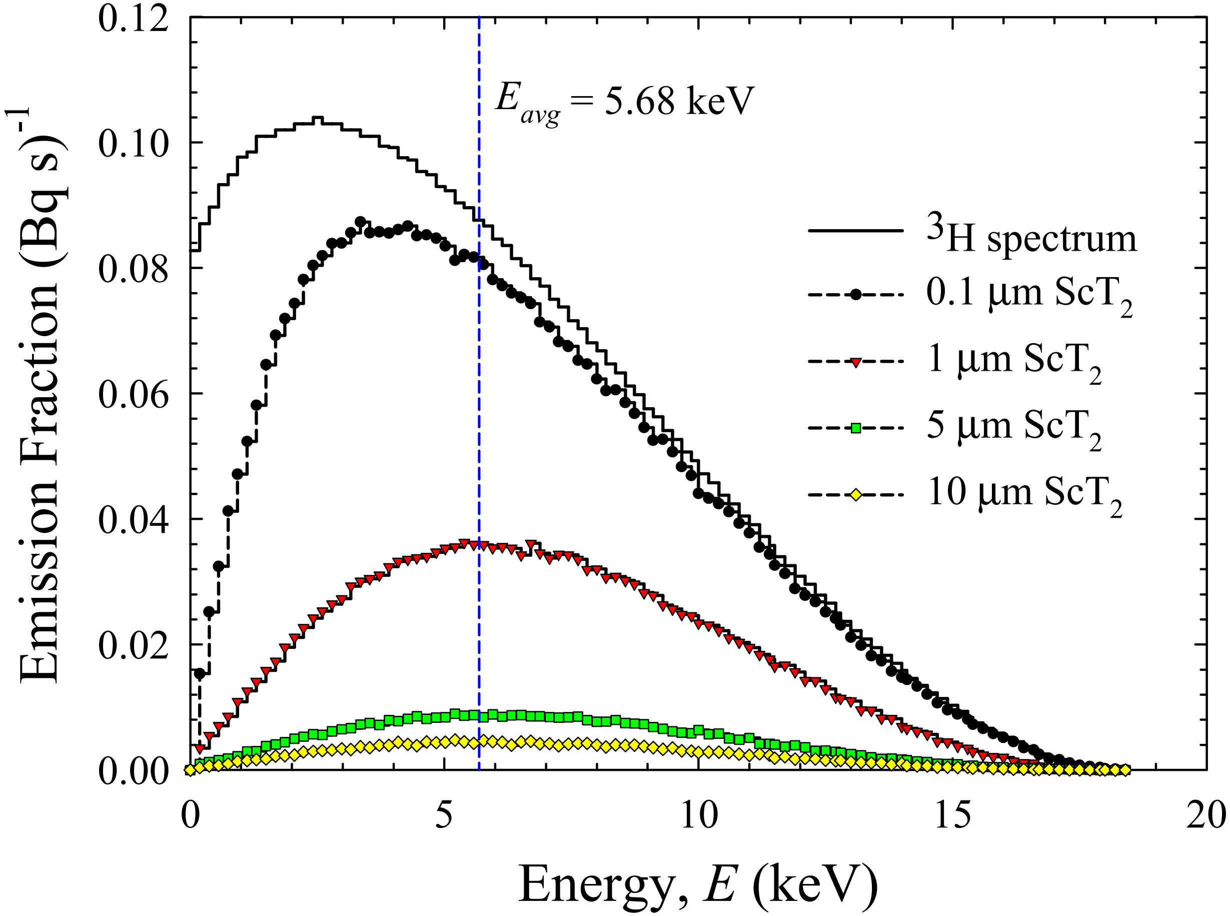
(a) spherical sites used to tabulated microdosimetric quantities (b) annular region used to tally the absorbed dose.

Tritium Spectrum



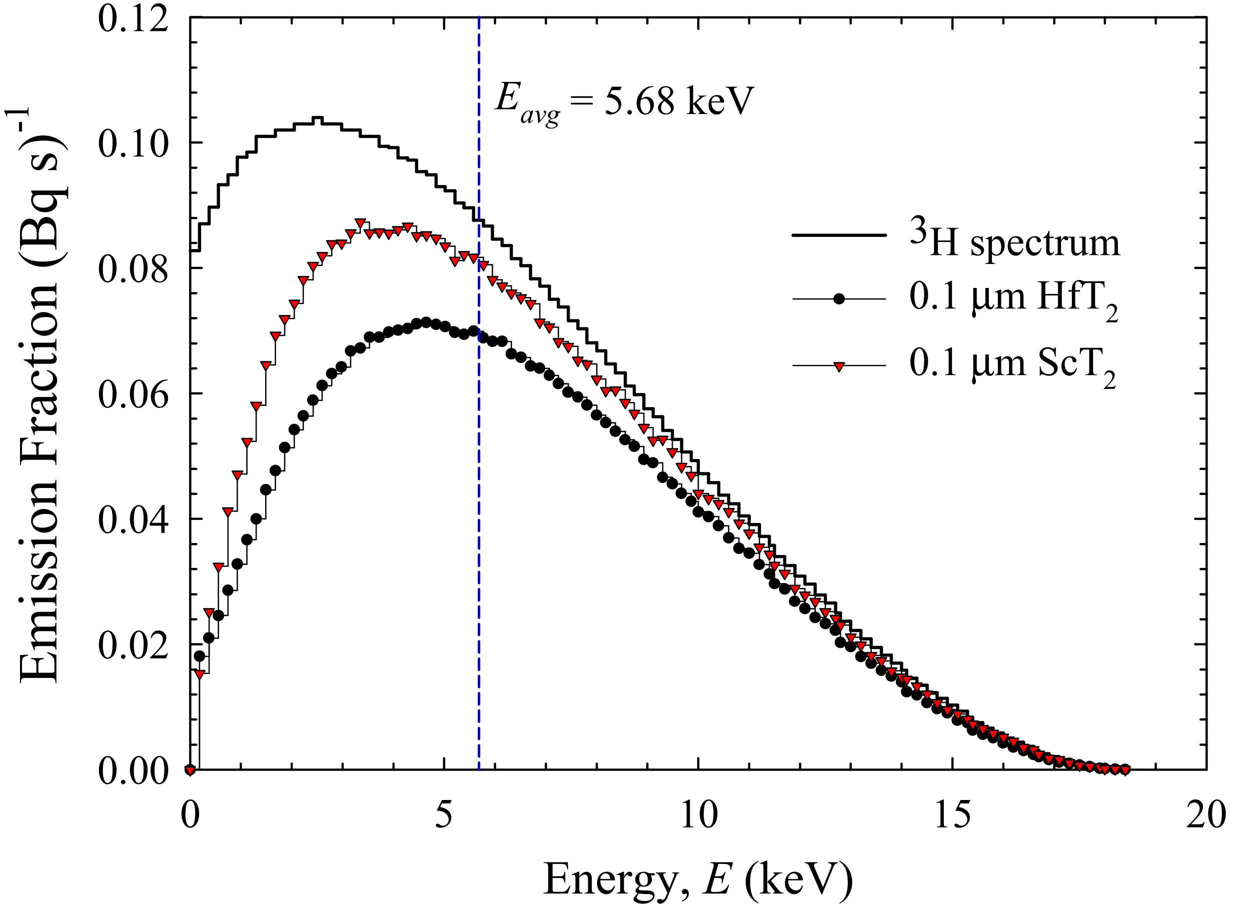
Beta-particle emission frequency for ^3H (Eckerman 1995). For the Monte Carlo simulations, primary electron energies are sampled from a cumulative distribution function with 100 equal-energy bins.

Electron Emission Spectra (Sc)



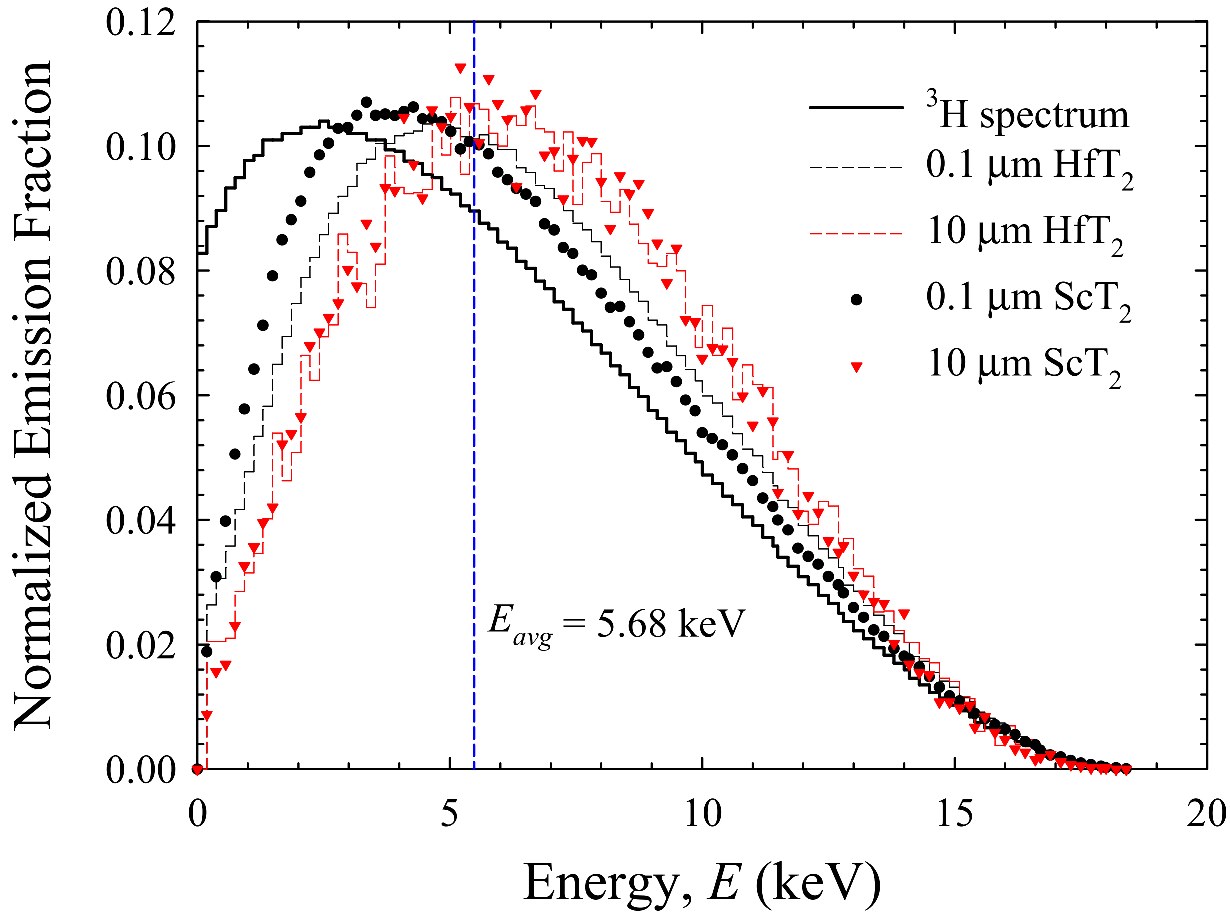
- The larger the particle, the smaller the fraction of radiation that escapes

Electron Emission Spectra (Sc and Hf)



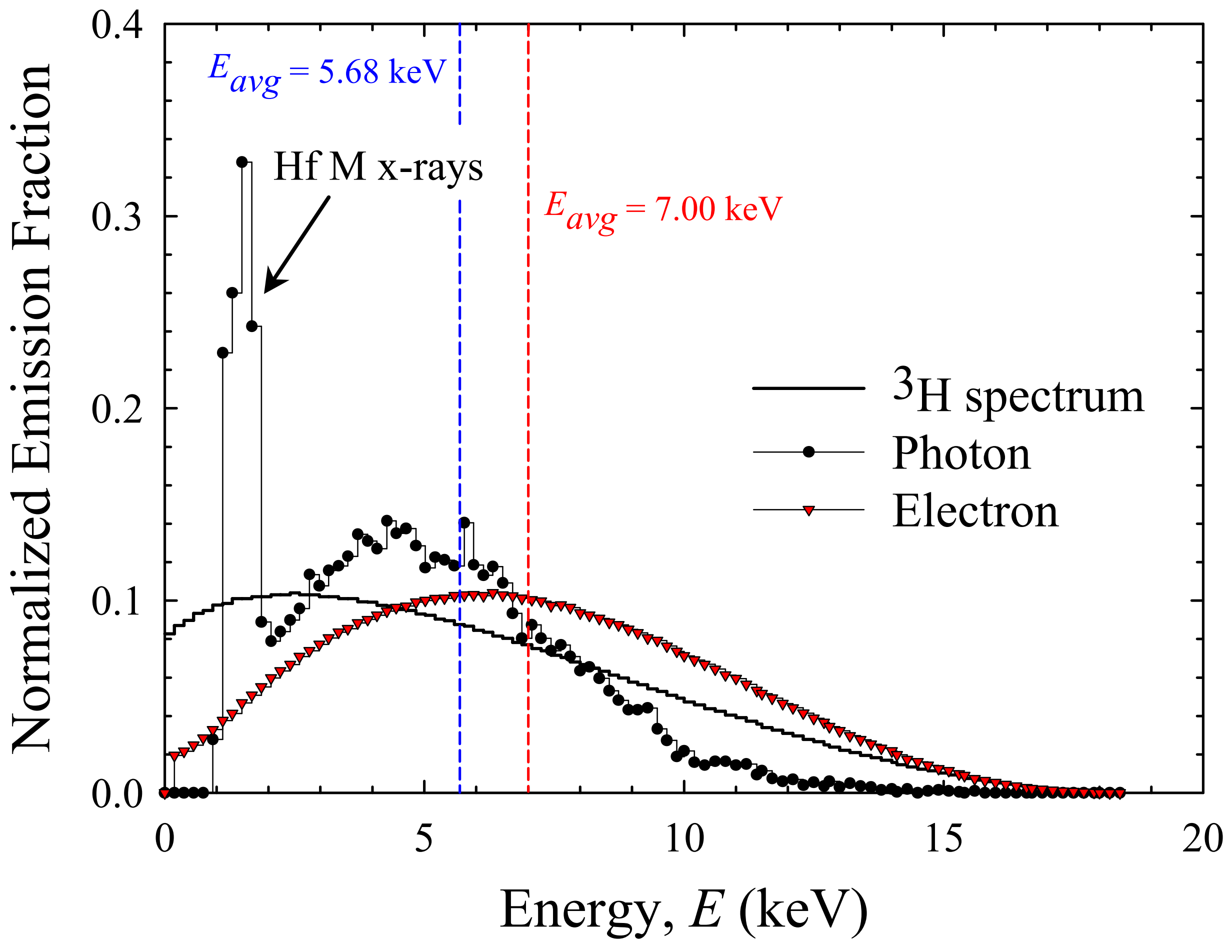
- The fraction of the radiation that escapes
  - decreases with increasing density
  - decreases with increasing atomic number

Normalized Electron Emission Spectra



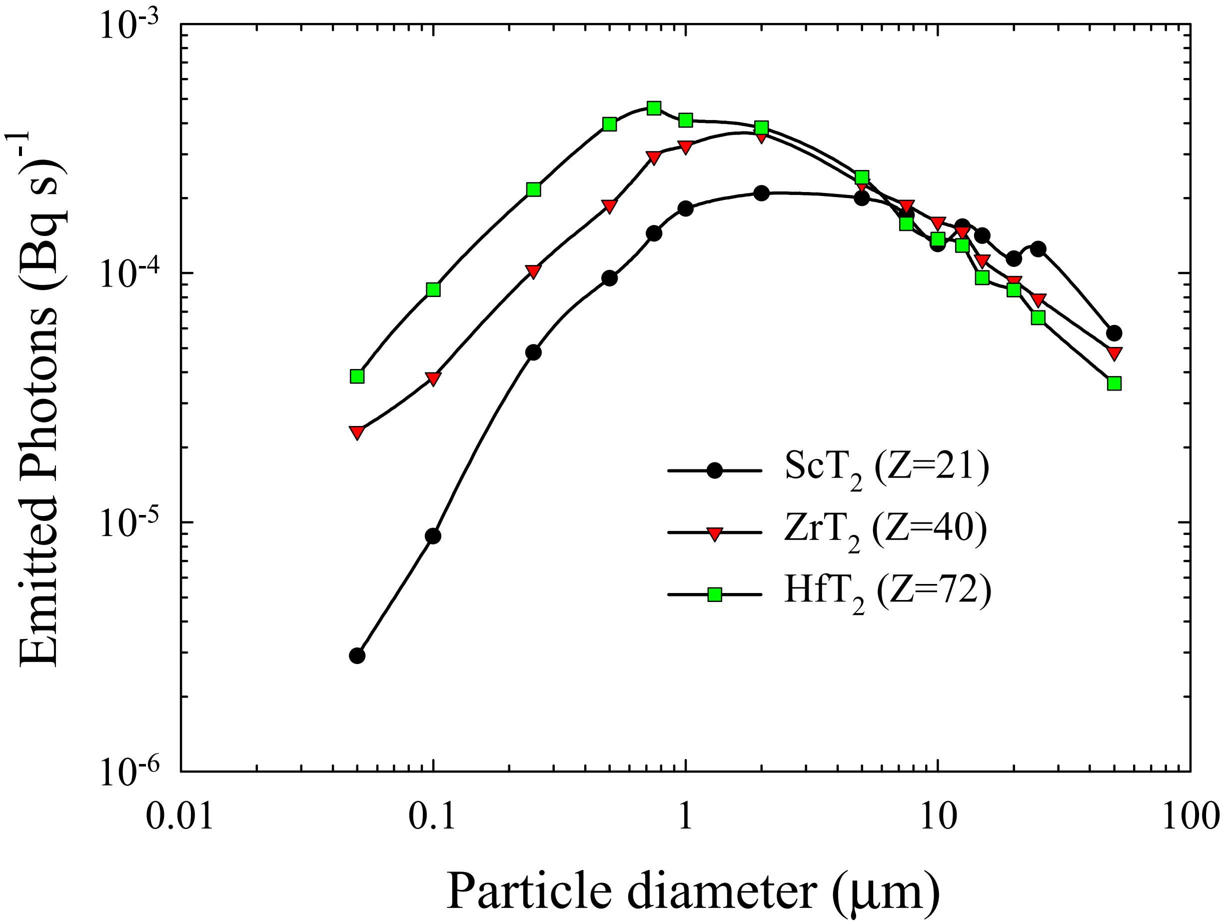
- The emitted electron spectrum tends to "harden" as the particle size increases

Electron and Photon Emission Spectra



- 5 µm HfT_2 particle

Photons Emitted from Tritide Surface

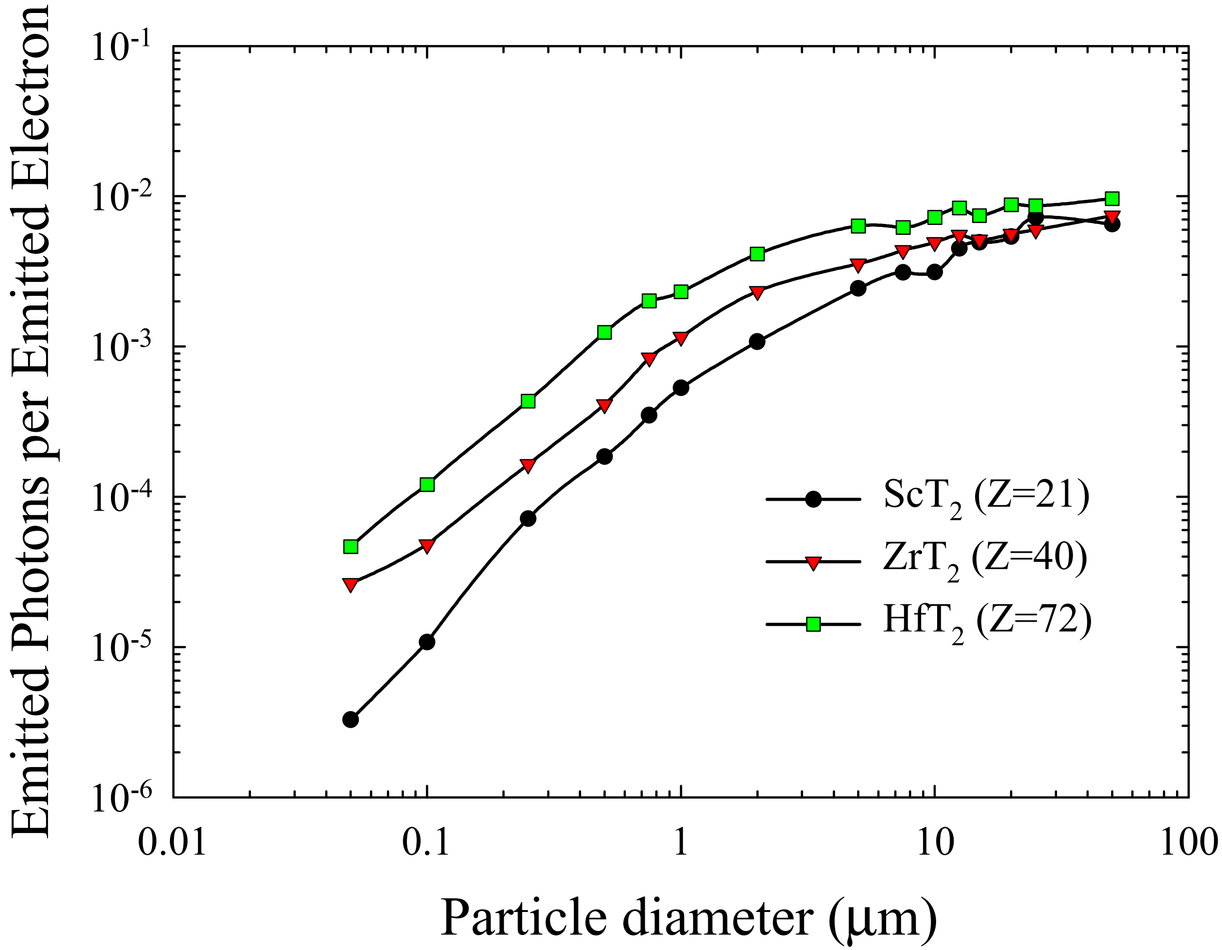


Bremsstrahlung increases

- linearly with particle size below 1 µm
- atomic number for particle sizes below 1 µm

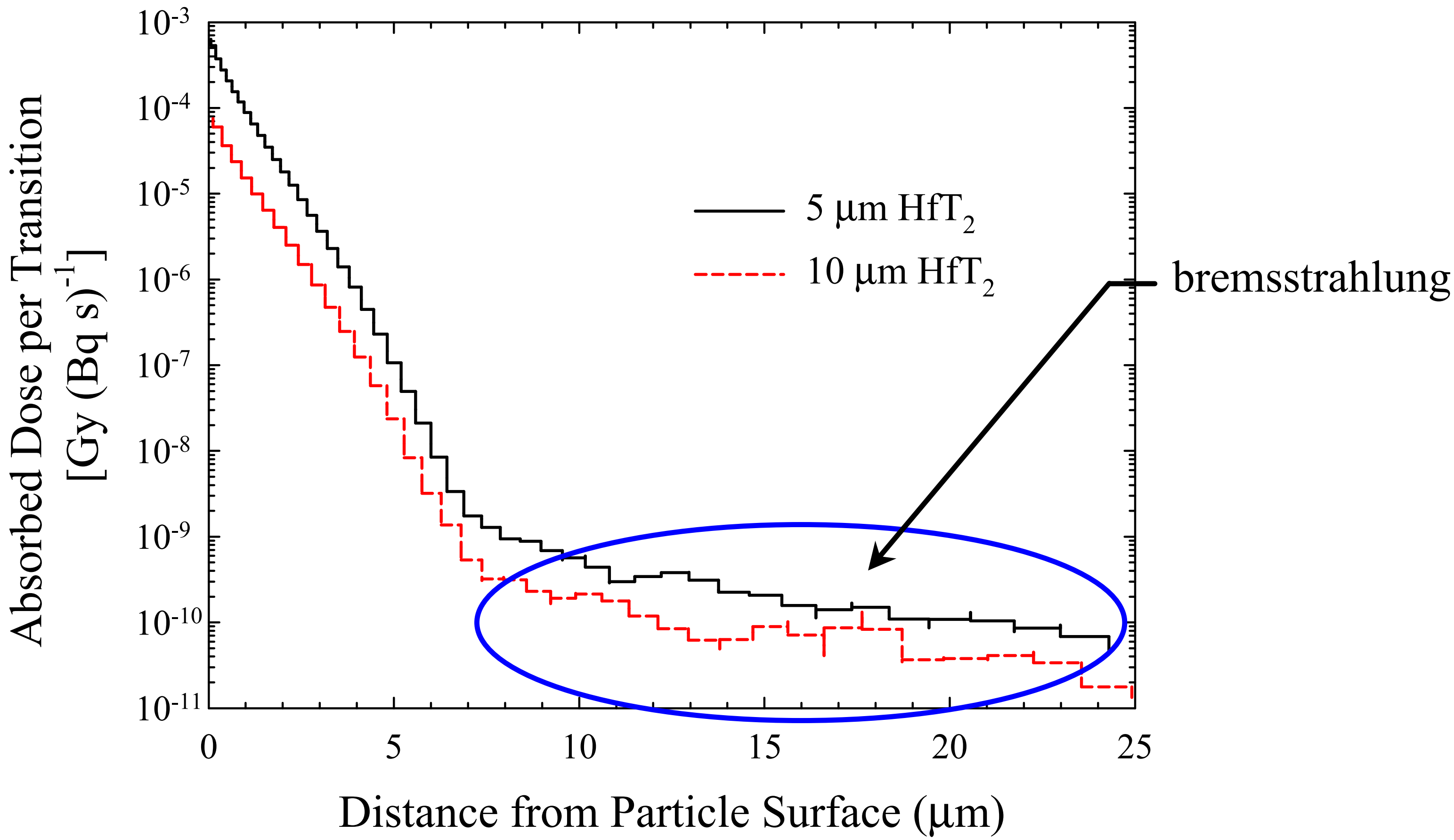
In larger particles, self-absorption overtakes bremsstrahlung production, and drops linearly with particle size

Photons Emitted from Tritide Surface



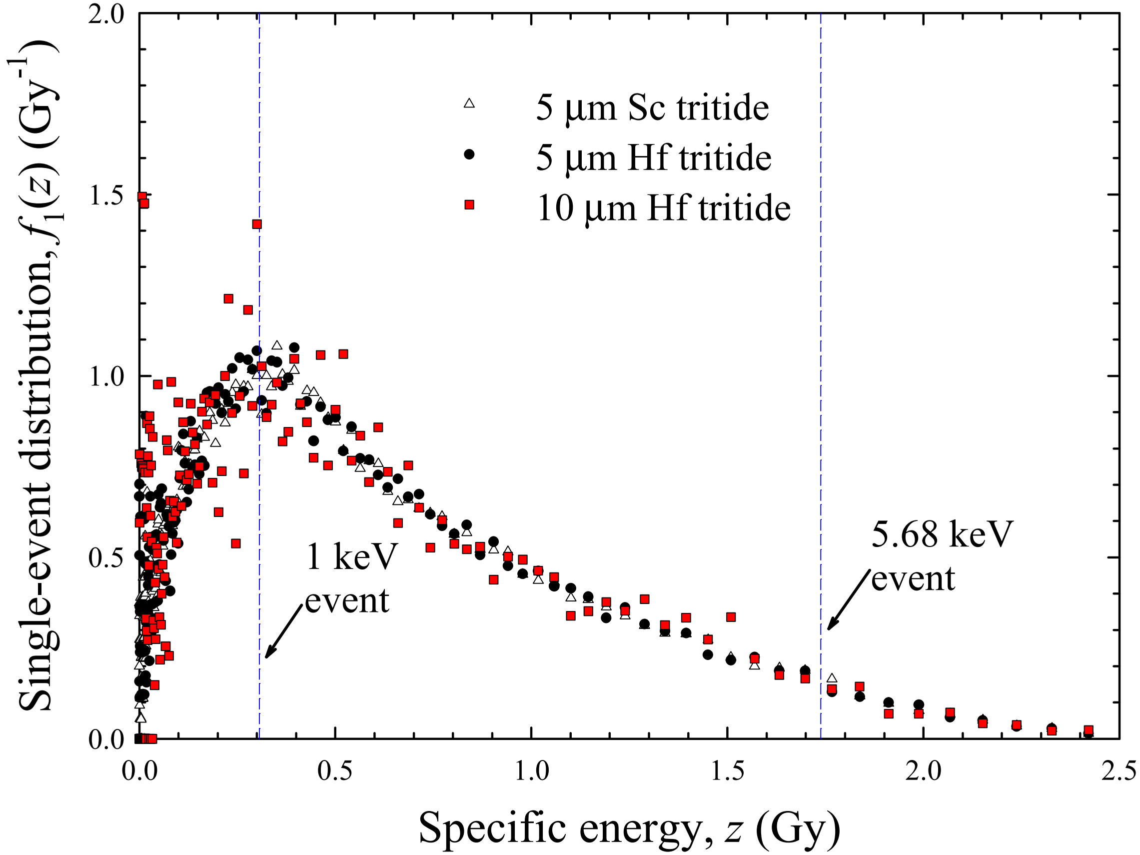
Relative number of emitted bremsstrahlung photons increases as the particle size and atomic number increases

Radial Dose Distribution (Hf)



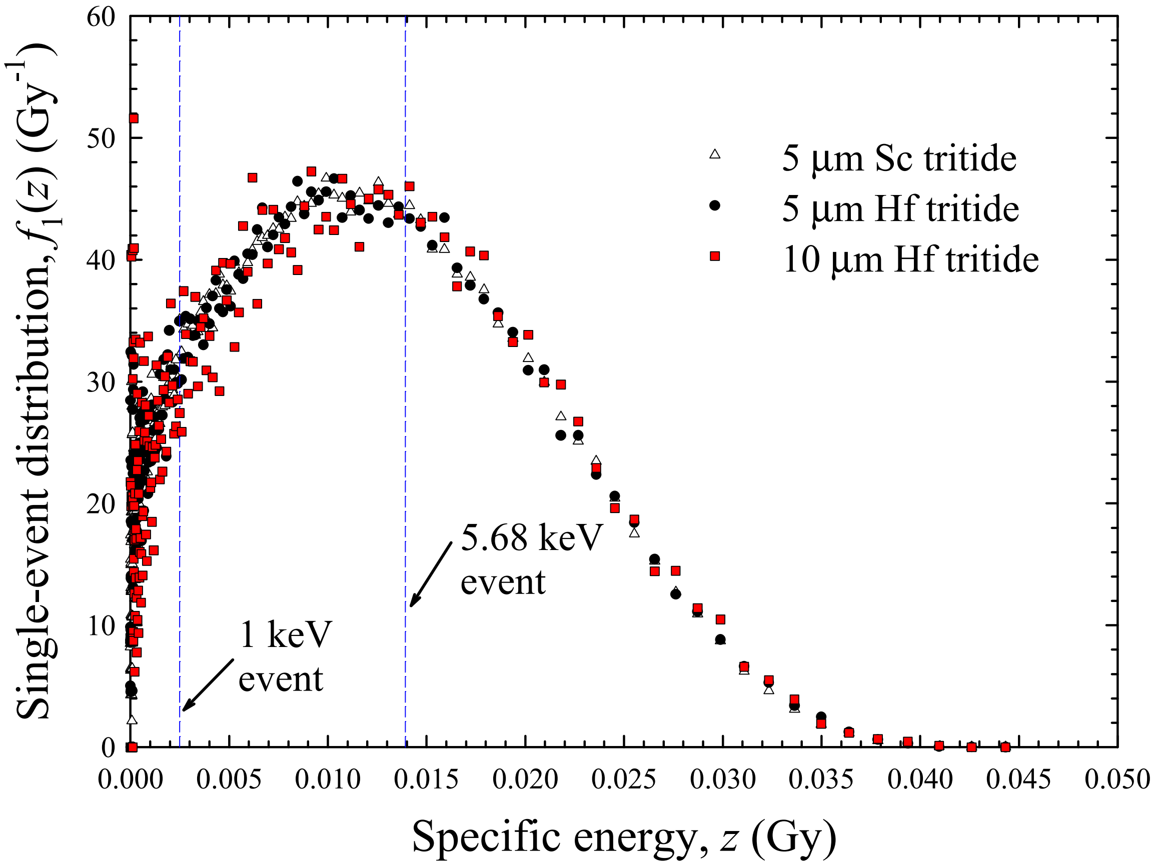
- Bremsstrahlung contributes a small fraction to the dose
- But bremsstrahlung is the only type of radiation that contributes to dose for distances greater than about 7 μm

Single-Event Distribution (1 um site)



- The energy imparted peaks at an event size of about 1 keV

Single-Event Distribution (5 um site)



- Energy imparted peaks at an event size of about 5 keV

Hf Tritide Microdosimetry (1 um site)

Site distance from particle surface (µm)	Energy imparted (keV)	Lineal energy (keV/µm)	Specific energy (mGy)	Probability site "hit" per decay
0.00	2.38	3.57	728.43	3.36E-04
0.50	2.41	3.62	737.69	1.34E-04
1.00	2.39	3.59	732.25	5.82E-05
1.50	2.43	3.64	743.36	2.60E-05
2.00	2.55	3.83	780.46	1.15E-05
2.50	2.56	3.84	782.68	5.38E-06
3.00	2.55	3.82	780.07	2.24E-06
3.50	2.60	3.90	795.59	9.47E-07
4.00	2.13	3.20	651.94	2.99E-07

Single-event distribution mean energy imparted, lineal energy, and specific energy for a 1 µm site (5 µm Hf tritide particle, r = 11.5 g cm^{-3}). Data are based on PENELOPE calculations for 8×10^6 particle tracks.

Hf Tritide Microdosimetry (5 um site)

Site distance from particle surface (μm)	Energy imparted (keV)	Lineal energy (keV/μm)	Specific energy (mGy)	Probability site "hit" per Bq-s
0.00	5.61	1.68	13.73	1.75E-03
0.50	5.71	1.71	13.97	7.69E-04
1.00	5.71	1.71	13.99	3.55E-04
1.50	5.65	1.70	13.84	1.66E-04
2.00	5.48	1.64	13.41	7.71E-05
2.50	5.37	1.61	13.16	3.44E-05
3.00	5.08	1.52	12.43	1.52E-05
4.50	3.45	1.04	8.45	9.47E-07
5.00	3.13	0.94	7.66	3.75E-07
7.50	2.74	0.82	6.70	1.18E-07
10.00	3.07	0.92	7.52	1.97E-08

Single-event distribution mean energy imparted, lineal energy, and specific energy for a 5 μm site (5 μm Hf tritide particle, $r = 11.5$ g cm^{-3}). Data are based on PENELOPE calculations for 5×10^6 particle tracks.

Sc Tritide Microdosimetry (1 um site)

Site distance from particle surface (µm)	Energy imparted (keV)	Lineal energy (keV/µm)	Specific energy (mGy)	Probability site "hit" per decay
0.00	2.40	3.59	732.92	1.02E-03
0.50	2.43	3.65	744.10	4.01E-04
1.00	2.46	3.69	752.38	1.72E-04
1.50	2.48	3.72	758.63	7.64E-05
2.00	2.51	3.77	769.33	3.39E-05
2.50	2.47	3.70	755.37	1.51E-05
3.00	2.57	3.85	785.65	6.16E-06
3.50	2.86	4.29	875.03	2.41E-06
4.00	2.94	4.40	898.52	8.90E-07
4.50	2.96	4.43	904.42	3.66E-07
7.00	0.00	0.00	0.00	0.00E+00

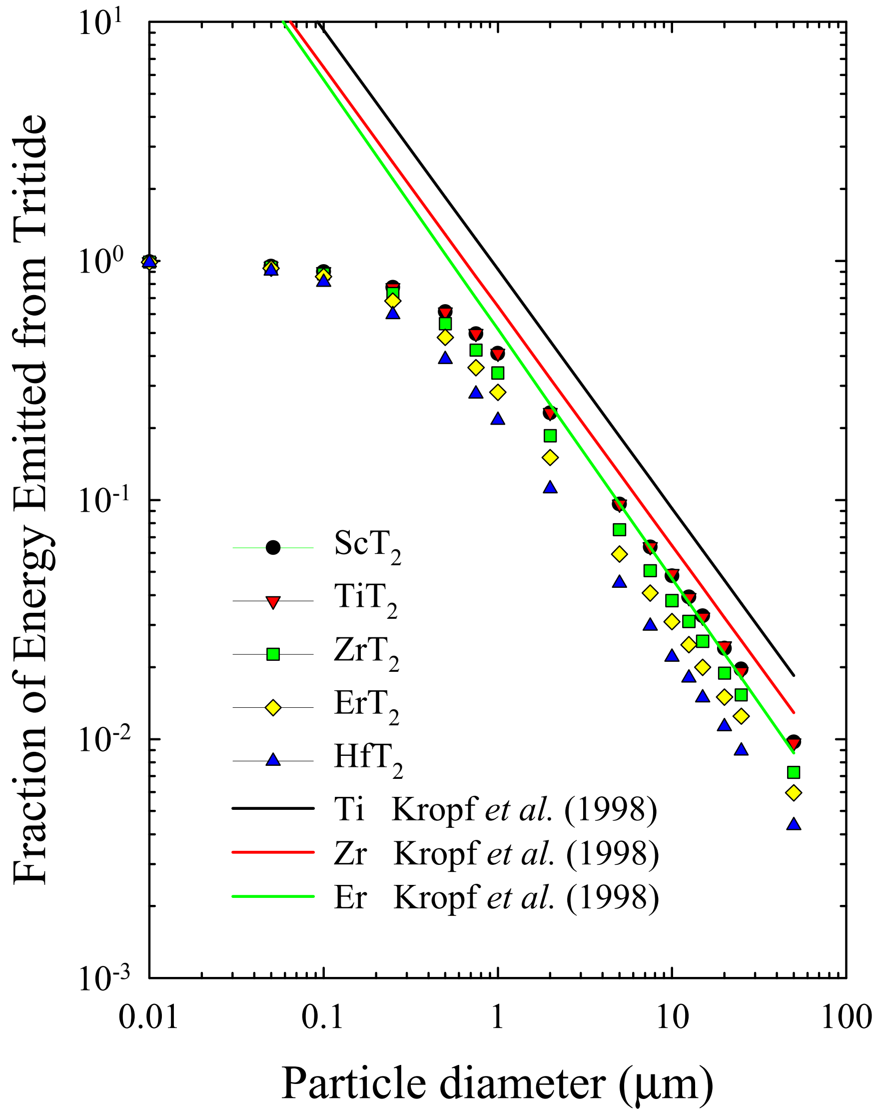
Single-event distribution mean energy imparted, lineal energy, and specific energy for a 1 µm site (5 µm Sc tritide particle, $r = 2.989$ g cm^{-3}). Data are based on PENELOPE calculations for 5.7×10^6 particle tracks.

Sc Tritide Microdosimetry (5 um site)

Site distance from particle surface (µm)	Energy imparted (keV)	Lineal energy (keV/µm)	Specific energy (mGy)	Probability site "hit" per decay
0.00	5.58	1.67	13.65	5.27E-03
0.50	5.66	1.70	13.87	2.30E-03
1.00	5.65	1.69	13.82	1.05E-03
1.50	5.56	1.67	13.60	4.82E-04
2.00	5.42	1.63	13.28	2.18E-04
2.50	5.37	1.61	13.14	9.48E-05
3.00	5.25	1.57	12.85	4.01E-05
4.50	3.76	1.13	9.21	2.71E-06
5.00	2.95	0.89	7.22	1.08E-06
7.50	2.22	0.66	5.43	3.87E-07
10.00	2.08	0.62	5.09	1.38E-07

Single-event distribution mean energy imparted, lineal energy, and specific energy for a 5 µm site (5 µm Sc tritide particle, $r = 2.989$ g cm^{-3}). Data are based on PENELOPE calculations for 3.6×10^6 particle tracks.

Energy Emitted from Various Tritides



Kropf *et al.* (1998):

Energy emission fraction = bd^{-q}

	b	q
Ti:	0.923	1.00
Zr:	0.645	1.00
Er:	0.518	1.043

- Over-estimates amount of energy emitted from particle.
- Dose estimates based on Kropf *et al.* formula are too high by at least a factor of 2 or 3.

Effect of Composition

	Fraction of Energy Emitted from Tritide				Ratio of Emission Fractions	
Diameter (mm)	**Sc**	**ScT_2**	**Hf**	**HfT_2**	**$Sc:ScT_2$**	**$Hf:HfT_2$**
0.01	0.9934	0.9925	0.9817	0.9805	1.0009	1.0012
0.10	0.9301	0.9233	0.7972	0.7899	1.0073	1.0092
1.00	0.5162	0.4935	0.1976	0.1916	1.0460	1.0312
5.00	0.1361	0.1262	0.0405	0.0393	1.0784	1.0303
10.00	0.0688	0.0639	0.0205	0.0197	1.0779	1.0425

2.989 g cm^3 used for both Sc tritides; 13.31 g cm^3 used for both Hf tritides.

- Composition of the particle affects the emitted energy fraction by less that 4% for Hf and less than 8% for Sc.
- Effects of tritide composition increases as the atomic number decreases.

Effect of Density (ScT_2)

	Fraction of Energy Emitted from Sc Tritide			
Diameter (mm)	Minimum	Best Estimate	Maximum	% Difference (Max. - Min.)
0.01	0.9920	0.9903	0.9925	0.06%
0.10	0.9181	0.9010	0.9234	0.59%
1.00	0.4719	0.4107	0.4934	5.25%
5.00	0.1187	0.0963	0.1267	8.32%
10.00	0.0599	0.0485	0.0643	9.02%
Density (g cm^3)	3.211	3.100	2.989	7.16%

Effect of Density (TiT_2)

Diameter (mm)	Fraction of Energy Emitted from Ti Tritide Minimum	Best Estimate	Maximum	% Difference (Max. - Min.)
0.01	0.9890	0.9903	0.9915	0.25%
0.10	0.8893	0.9010	0.9127	2.60%
1.00	0.3762	0.4106	0.4494	17.83%
5.00	0.0852	0.0962	0.1099	25.68%
10.00	0.0437	0.0490	0.0561	25.44%
Density (g cm^3)	4.540	4.030	3.520	25.31%

Effect of Density (ZrT_2)

	Fraction of Energy Emitted from Zr Tritide			
Diameter (mm)	Minimum	Best Estimate	Maximum	% Difference (Max. - Min.)
0.01	0.9878	0.9891	0.9905	0.26%
0.10	0.8696	0.8826	0.8955	2.94%
1.00	0.3125	0.3409	0.3745	18.22%
5.00	0.0685	0.0762	0.0852	22.03%
10.00	0.0345	0.0383	0.0436	23.59%
Density (g cm^3)	6.506	5.830	5.154	23.19%

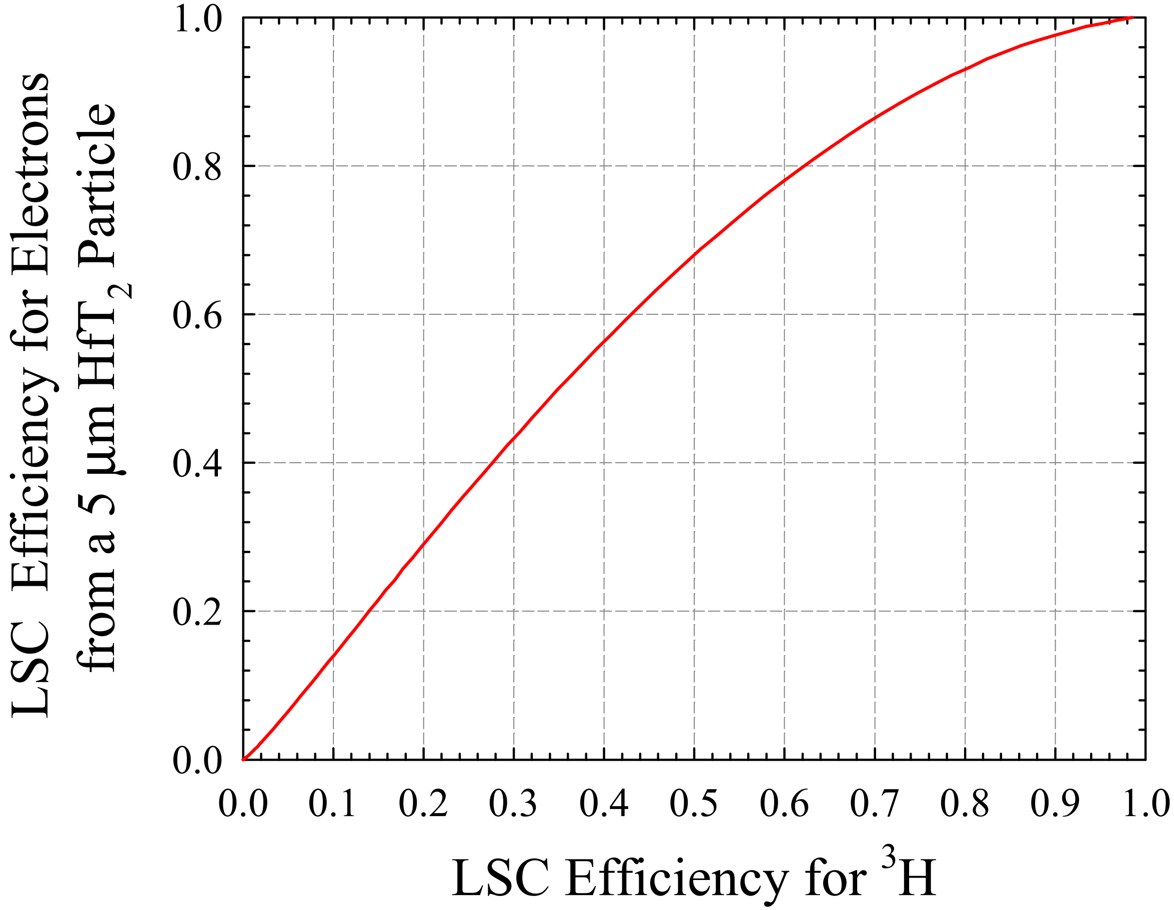
Effect of Density (ErT_2)

	Fraction of Energy Emitted from Er Tritide			
Diameter (mm)	**Minimum**	**Best Estimate**	**Maximum**	**% Difference (Max. - Min.)**
0.01	0.9866	0.9878	0.9882	0.16%
0.10	0.8499	0.8605	0.8656	1.82%
1.00	0.2642	0.2810	0.2927	10.13%
5.00	0.0562	0.0607	0.0636	12.03%
10.00	0.0286	0.0308	0.0320	11.03%
Density (g cm^3)	9.066	8.560	8.054	11.82%

Effect of Density (HfT_2)

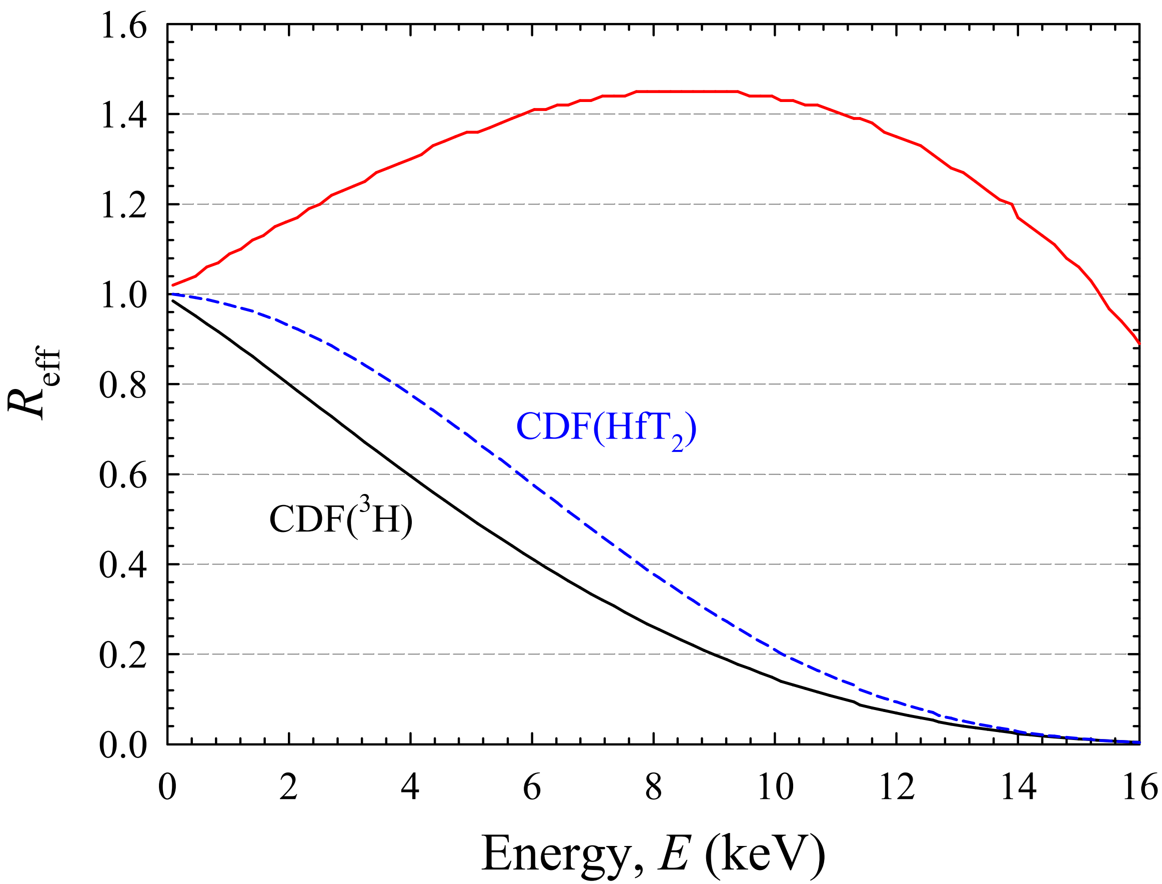
	Fraction of Energy Emitted from Hf Tritide			
Diameter (mm)	Minimum	Best Estimate	Maximum	% Difference (Max. - Min.)
0.01	0.9805	0.9831	0.9856	0.52%
0.10	0.7895	0.8137	0.8383	5.99%
1.00	0.1913	0.2155	0.2455	25.16%
5.00	0.0394	0.0449	0.0519	27.89%
10.00	0.0200	0.0227	0.0265	28.85%
Density (g cm^3)	13.31	11.67	10.03	28.11%

Liquid Scintillation Counting Efficiency



Liquid scintillation counting (LSC) efficiency for 5 μm HfT_2 versus the LSC for ^3H. The LSC is calculated based on the assumption that all electrons above a fixed cutoff energy are detected in the scintillation counter.

Liquid Scintillation Counting Efficiency



$$R_{eff} = \frac{(1 - CDF(HfT_2))}{(1 - CDF(^3H))}$$

- R_{eff} is the ratio of counting efficiency for the tritide particle to the counting efficiency of pure ^3H
- **"Observed activity" differs from "apparent activity!"**

Conclusions

- Radiation emitted by Sc, Ti, Zr, Er, and Hf tritides can be considered low-LET radiation
- Uncertainties associated with the composition and density of the tritides are on the order of 20 to 30%
- Electron emission spectrum tends to harden as the particle diameter increases
- Bremsstrahlung production initial increases with particle diameter, reaches a maximum for diameters from about 0.8 and 2.0 μm, and then decreases
  - Impacts calibration of air samplers that measure bremsstrahlung
- The differences between "observed" and "apparent" activity needs to be considered
- Activity median aerodynamic diameter (AMAD) is not appropriate for particles with significant self-absorption

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Supplemental information

http://www.pnl.gov/eshs/pub/MetalTritide/