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Nuclear Fuel Cycle Facility Accident Analysis Handbook

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3.4 Source Term Estimations for Inadvertent Nuclear Criticality Excursions

A nuclear criticality accident is defined as the release of energy as a result of inadvertently producing a self-sustaining or divergent neutron chain reaction. In fuel cycle facilities, a criticality accident may occur following: (1) transfer or leakage of fissile material from a geometrically favorable container to a container with unfavorable geometry; (2) introduction of excess fissile material into a container or area; (3) over-concentration of a solution; (4) failure to maintain adequate quantities of neutron absorbing materials; (5) precipitation of fissile solids; (6) introduction of neutron moderators into a system or area, and (7) alteration of system or area geometry.

A nuclear criticality differs from other fuel cycle facility accidents in that radioactive material is generated in the event and short-lived fission products not normally encountered are present. Nuclear criticality events may be terminated by control systems or by thermal expansion, loss of fissile material or moderator mass, change in density, or mixing caused by the rapid release of energy.

Criticality accident hazards include direct exposure to prompt neutron and gamma radiation produced in the criticality event, release of gaseous radioactive material, and expulsion or entrainment of radioactive aerosols. Direct exposures in the immediate vicinity of the event can be substantial, but off-site impacts are generally below detectable levels.

For nuclear criticality safety (NCS) evaluations, systems are categorized as solutions; moderated/reflected solids; bare, dry solids; and large storage arrays. The total amount of energy released in the event is generally small, and physical damage to ventilation and gas clean-up systems is not expected to occur. Mixing within the confining building and normal function of off-gas systems delays and mitigates the consequences of the event. The time during which a criticality occurs and the severity of the event depend in a complicated manner on the quantities, physical and chemical form, and concentrations of the fissile material and on the size, configuration, moderation, reflection, and neutron absorption characteristics of the system.

NRC requirements for NCS at fuel cycle facilities include adherence to the double-contingency principle in process design and operation, thus reducing the probability of an accident to a small value. Because of application of the double contingency principle, criticality accident scenarios involve multiple equipment or control system failures and are of such complexity that generally applicable analytical tools are not available. Thus, the nature and magnitude of possible accidents are assessed individually and conservative analyses are used to evaluate the adequacy of NCS protection systems. The balance of this section discusses the magnitude of possible criticality events at fuel cycle facilities and presents a representative method of source term estimation using a solution criticality at a uranium fuel fabrication facility as an example case. Other aspects of accident analysis for nuclear criticality events are discussed in a sample problem presented in Section 9 of Appendix D.

3.4.1 Fission Yields

The amounts of energy and radionuclides produced in nuclear criticality accidents are proportional to the total number of fissions occurring in the event. Thus, estimation of the total number of fissions provides a necessary basis for estimation of accident source terms. Estimates of the total number of fissions have been based on review of criticality accidents (Stratton 1989 and Frolov, et al 1995), reactor excursion experiments (Nyer, et al 1965), and other experiments. Reviews of these data and empirical models developed primarily for solution systems (Olson, et al. 1974) provide perspective on estimates of total

number of fissions and energy generation rates. Descriptive information and data reported in the literature for a variety of systems are summarized in Tables 3-8, 3-9, 3-10, 3-11, 3-12, and 3-13. These data and estimates are discussed below for the four major types of reactive systems. Mathematical models capable of estimation of criticality accident progression and termination require coupled representation of hydrodynamic and neutronic phenomena. Although such models have been developed for power reactor system, models capable of estimating energy generation and total number of fissions have not been developed for the diverse systems encountered at fuel cycle facilities.

3.4.1.1 Solution Systems

Solutions are of principal concern for NCS evaluations at fuel cycle facilities because liquids are mobile and can be formed into geometrically unfavorable shapes with inadvertent moderation. Data for accidents that occurred in the U.S. are presented in Tables 3-8 and 3-9. Data for accidents that occurred in the former Soviet Union are presented in Table 3-13. Of the 13 accidents listed in Tables 3-8 and 3-9 that involve solutions, 8 occurred in process facilities whereas 5 occurred in nuclear criticality experimental facilities. Inadvertent transfers to non-favorable geometry vessels and failure to follow or properly interpret procedures were the most common causes of the accidents. The total number of fissions for the accidents ranged from 1×10^{15} to 4×10^{19} and an initial burst was generally followed by a plateau period characterized by a lesser and declining fission rate. Because of the duration of the plateau period, the major portion of energy release occurred during this time. The highest fission yield (4×10^{19}) occurred when a relatively large volume containing approximately 70 times the critical mass (i.e., 34.8 kg of U-235) was transferred to a vessel of unfavorable geometry positioned above concrete, which reflects neutrons. Of the 12 accidents listed in Table 3-13, 11 involve aqueous or organic solutions. The accidents generally had an initial burst followed by repeated smaller bursts extending over periods of time of the order of hours. Of these events, the largest had a yield of 7.9×10^{17} fissions and involved approximately 2.8 kg (6 lb) of highly enriched uranium.

3.4.1.2 Fully Moderated and Reflected Solids

Estimates of peak fission rate and total number of fissions for an accidental nuclear criticality in a moderated, reflected solid system may be derived from data from accidents and from experiments with light- and heavy-water-moderated reactors. Criticality accident data are reported in Tables 3-8(b) and 3-11 for uranium and plutonium elements of various shapes with water or graphite moderation. The reactor excursion data reported in Table 3-10 is for uranium-aluminum and UO_2 stainless steel clad fuels. The total number of fissions for the relevant accidental criticalities reported in Table 3-8(a) ranges from 1×10^{15} to 1×10^{17} while the total number of fissions for reactor excursions is bounded by 5×10^{18} for the power levels reported in Table 3-10. Criticality events in moderated, reflected solid systems were characterized by an initial burst with little or no plateau period.

3.4.1.3 Powder Systems

Limited experimental data exist for powder systems of the type found in fuel cycle facilities. Data from accidents presented in Tables 3-8(c), 3-11, and 3-12, for systems that may serve as surrogates for powder system, are bounded by a total number of fissions that is less than 2×10^{17} .

Table 3-8. Summary of Known Accidental Criticality Excursions (1945 to 1974)
(a) Solution Systems, (b) Metal Systems, and (c) Moderated Foil and Powder Systems
(Olsen, et al. 1974)

No.	Date	Location	Title for Ref.	Fissionable Material	Arrangement	Initial Prompt Critical Burst, Fissions	Duration	Total Fissions	Cause	Physical Damage
(a) Solution Systems										
SE 1	12/49	LASL, NM	Water Boiler	U(93)O ₂ (NO ₃) ₂ (- 1 kg U235; 13.6 l)	Sphere, graphite-reflected	- 3x10 ¹⁵ (barely over prompt critical)	Not known	3-4x10 ¹⁶	Control rods withdrawn too fast	None
SE 2	11/16/51	Hanford works - Richland, WA	P-11	PuO ₂ (NO ₃) ₂ (1.15 kg Pu; 63.8 l)	Sphere, 93 percent full-reflected	8x10 ¹⁶	Single burst	8x10 ¹⁶	Too high fuel addition	None
SE 3	5/26/54	ORNL, TN	Spider	UO ₂ F ₂ (18.3 kg U235; 55.4 l)	Cylindrical annulus, unreflected	5x10 ¹⁶	Not known	1x10 ¹⁷	Shift of poison	None
SE 4	2/1/56	ORNL, TN	Scram blade	UO ₂ F ₂ (27.7 kg U235; 58.9 l)	Cylinder, unreflected	1.6x10 ¹⁷	Single burst	1.6x10 ¹⁷	Geometry change	Warping of bottom of cylinder
SE 5	1/30/68	ORNL, TN	U-233	UO ₂ (NO ₃) ₂ - 1 kg U233; 5.8 l)	Sphere, water-reflected	1.0x10 ¹⁶	Single burst	1.1x10 ¹⁶	Air in line	None
P 1	6/16/58	ORNL, TN - Y-12 Processing Plant	Y-12	UO ₂ (NO ₃) ₂ (2.5 kg U235; 56 l)	Cylinder, concrete-reflected below	- 1.1x10 ¹⁶	13 min.	1.3x10 ¹⁸	Valve leaked or left open	None (loss: \$1000)
P 2	12/30/58	LASL, NM - Pu Processing Plant	Agitator	PuO ₂ (NO ₃) ₂ (3.27 kg Pu; 168 l)	Cylinder, water-reflected below	1.5x10 ¹⁷	Single burst	1.5x10 ¹⁷	Procedure not followed	None
P 3	10/16/59	Idaho Reactor Testing Area, Chemical Processing Plant	1F-1 (siphon)	UO ₂ (NO ₃) ₂ (34.5 kg U235; - 800 l)	Cylinder, concrete-reflected below	- 10 ¹⁷	Not known	- 4x10 ¹⁹	Sparge gauge plugged	None (loss: \$62,000)

Table 3-8. Summary of Known Accidental Criticality Excursions (1945 to 1974)
(a) Solution Systems, (b) Metal Systems, and (c) Moderated Foil and Powder Systems
(Olsen, et al. 1974) (Continued)

No.	Date	Location	Title for Ref.	Fissionable Material	Arrangement	Initial Prompt Critical Burst, Fissions	Duration	Total Fissions	Cause	Physical Damage
P 4	1/25/61	Idaho Reactor Testing Area, Chemical Processing Plant	IF-11 (air lift)	UO ₂ (NO ₃) ₂ (8 kg U235; 40 l)	Cylinder	No estimate	Not known	6x10 ¹⁷	Instruction misinterpreted	None (loss: \$6000)
P 5	4/7/62	Hanford works - Richland, WA	Recuplex	Pu complex (1.5 kg Pu)	Cylinder, unreflected	- 10 ¹⁶	37 hrs	8x10 ¹⁷	Valve leaked or opened	None (loss: \$1000)
P 6	7/24/64	Wood River Junction, RI - scrap recovery facility	Wood River	UO ₂ (NO ₃) ₂ (2.64 kg U235)	Cylinder, unreflected	1.1x10 ¹⁷	Not known	1.3x10 ¹⁷	Procedure not followed	None
P 7	8/24/70	Windscale Works, England	Windscale	Pu complex (~ 2.5 kg Pu; ~ 100 l)	Cylinder	No estimate	5-10 sec	1x10 ¹⁵	Pu accumulated in organic	None
(b) Metal Systems										
ME 1	6/4/45	LASL, NM	Metal Cubes	83% U235 enriched U metal 1/2 in. cubes (35.4 kg)	Array of cubes; water-reflected	- 3x10 ¹⁵	Not known (perhaps 3 bursts)	- 3x10 ¹⁶	Water leaked into array	None
ME 2	8/21/45	LASL, NM	Dragon	Delta phase Pu metal (6.2 kg)	Sphere reflected by Be	- 1.8x10 ¹⁵ (10 cents over)	< 1 sec	- 1x10 ¹⁶	Dropped reflector block	None
ME 3	5/21/46	LASL, NM	Screw-driver	Delta phase Pu metal	Sphere reflected by WC	- 1.8x10 ¹⁴	Not known	- 3x10 ¹⁵	Screwdriver holding reflector away from Pu slipped	None
ME 4	2/1/51	LASL, NM	Aquarium machine	Two cylinders U(93) metal (24.4 kg and 38.5 kg)	Side by side in water tank	- 6x10 ¹⁵	Not known (perhaps several bursts)	1x10 ¹⁷	Went critical during practice scram	Slight oxidation

Table 3-8. Summary of Known Accidental Criticality Excursions (1945 to 1974)
 (a) Solution Systems, (b) Metal Systems, and (c) Moderated Foil and Powder Systems
 (Olsen, et al. 1974) (Continued)

No.	Date	Location	Title for Ref.	Fissionable Material	Arrangement	Initial Prompt Critical Burst, Fissions	Duration	Total Fissions	Cause	Physical Damage
ME 5	4/18/52	LASL, NM	Jamima	U(93) metal (92.4 kg)	Cylinder, unreflected	$\sim 1 \times 10^{15}$ (21 cents over)	< 1 sec	1.5×10^{16}	Computation error	None
ME 6	2/3/54	LASL, NM	Godiva I	U(93) metal (53 kg)	Sphere, unreflected	5.6×10^{16}	Single burst	5.6×10^{16}	Assembled too rapidly	Slight warping of pieces (loss: \$600)
ME 7	2/12/57	LASL, NM	Godiva II	U(93) metal (54 kg)	Sphere, unreflected	1.2×10^{17} (21 cents over)	Single burst	1.2×10^{17}	Graphite fell against assembly	Warping oxidation; near melting close to center (loss: \$2100)
ME 8	6/17/60	LASL, NM	9-inch cylinder	U(93) metal (48 kg)	Cylinder, graphite-reflected	$\sim 1 \times 10^{15}$	Not known	6×10^{16}	Error in addition estimate	Trivial
ME 9	11/10/61	ORNL, TN	U-Paraffin	U(93) metal (~ 75 kg)	Cylinder, paraffin-reflected	$\sim 1 \times 10^{15}$	Not known	$\sim 1 \times 10^{16}$	Error in addition estimate	None
ME 10	3/26/63	LARL, CA	LRL	U(93) metal (47 kg)	Cylinder, Be-reflected	1×10^{17}	Not known	3.8×10^{17}	Ram caught reflector; lifted; fell	Metal melted and some burned; contamination (loss: \$95,000)
ME 11	5/28/65	WSMR, NM	U-Mo Alloy	U(93)-1.0% Mo (96 kg)	Cylinder, unreflected	1.5×10^{17}	Not known	1.5×10^{17}	Incorrect operation	Assembly bolts broken, minor damage to coating

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Table 3-8. Summary of Known Accidental Criticality Excursions (1945 to 1974)
(a) Solution Systems, (b) Metal Systems, and (c) Moderated Foil and Powder Systems
(Olsen, et al. 1974) (Continued)

No.	Date	Location	Title for Ref.	Fissionable Material	Arrangement	Initial Prompt Critical Burst, Fissions	Duration	Total Fissions	Cause	Physical Damage
(c) Moderated Foil and Powder Systems										
H 1	2/11/45	LASL, NM	UH ₃ - Styrex blocks	U(93)H ₁₀ pressed in Styrex (UC ₂ H ₁₀)	Assembly of blocks	- 6x10 ¹⁵	Single burst	- 6x10 ¹⁵	Excess reactivity addition	Cubes swollen and blistered
H 2	7/3/56	LASL, NM	Honey-comb	U(93) metal foils sandwiched with carbon	Cylinder, Be-reflected	Not known	Not known	3.2x10 ¹⁶	Assembled too rapidly	None
H 3	12/11/62	LASL, NM	ZEPO	U(93) metal foils sandwiched with carbon	Cylinder, C- and Be-reflected	3x10 ¹⁶ (12 cents prompt critical)	Single burst	3x10 ¹⁶	Excess fuel addition	None

Table 3-9. Accidents in Processing Plants (Paxton 1980)

Date	Plant	Total Fissions	First Pulse	Doses (Rads)	Notes
6/16/58	Y-12	1.3×10^{18}	$\sim 7 \times 10^{16}$	365, 339, 327, 270, 236, 69, 69, 23	^{235}U solution washed into drum
12/30/58	LASL	1.5×10^{17}	1.5×10^{17}	~ 4400 (fatal), 135, 35	Pu concentrated in solvent layer
10/16/59	Idaho CPP	4×10^{19}	$\sim 10^{17}$	50 R, 32 R, mostly beta	^{235}U solution siphoned into tank
1/2/61	Idaho CPP	6×10^{17}	6×10^{17}	None	^{235}U solution forced into cylinder by air
4/7/62	RECUPLE X	8.2×10^{17}	$\sim 10^{16}$	87, 33, 16	Pu solution in sump sucked into tank
7/24/64	Wood River Junction	1.3×10^{17}	$\sim 10^{17}$	10,000 (fatal), two 60-100	^{235}U solution poured into tank
8/24/70	Windscale	10^{15}	$\sim 10^{15}$	Negligible	Pu concentrated in trapped solvent
10/17/78	Idaho CPP	3×10^{18}	Unknown	None	^{235}U buildup in diluted scrub solution

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**Table 3-10. Destructive Power Excursion Summary
(Nyer, Bright, and McWhorter 1965)**

Reactor	Reactivity Addition, Δk	λ , s ⁻¹	Peak Power, MW	Energy Release, MWs	Maximum Temp. °C	Maximum energy density, W-cm ³	Maximum pressure, psi	Remarks
BORAX I [28]	3.1	384	≤ 19,000	135	≤ 1,800	≤ 6,500	6,000 - 10,000	Destroyed core, vessel, and some associated equipment. Small fission-product release. Steam explosion proposed as cause.
SL-1 [29]	3.0	280	~ 19,000	133	> 2,075	> 7,300	10,000	Destroyed core, bulged vessel, local fission-product contamination. 10% fission-product release. Steam explosion - minor contribution from metal-H ₂ O reaction.
SPERT-I [30] D 12/25	2.6	200	1,130	11	585	2,000	7	Melted > 0.5% of core.
	2.7	218	1,270	19	680	2,300	8	Melted ~ 3% of core.
	3.55	313	2,250	31	1,360	4,600	≤ 4,000	Melted ~ 35% of core. Destroyed core and associated equipment, bulged tank. ~ 4% fission-product release. Probable steam explosion - Al ₂ O ₃ analysis indicates ~ 3.5 MWs energy release from metal-H ₂ O reaction.
SPERT I [31] oxide core	2.6	455	17,400	155	1,800	2,200	70	
	3.3	645	35,000	155	1,800	2,200	130	Two fuel rods ruptured. Discoloration and/or deformation of 25% of fuel rods. Negligible fission-product release.
SNAPTRAN-3 [32]	3.5	1,400	~ 20,000	50	> 2,500	7,100	~ 4,000	Burst pressure vessel. All fuel rods ruptured, ~ half of fuel reduced to powder form. Negligible fission-product release.

Table 3-11. Inhomogenous Water-Moderated Systems
(Stratton 1967)

Date	Location	Active Material	Geometry	Total Fissions	Cause	Physical Damage
6/4/45	Los Alamos, New Mexico	35.4 kg U - 83% ²³⁵ U ½ in cubes	Pseudosphere, water- reflected	$- 3 \times 10^{16}$	Water seeping between blocks	None
2/1/51	LASL, New Mexico	2 cylinders U 24.4 and 38.5 kg 93% ²³⁵ U	2 cylinders, water- reflected	10^{17}	Scram increased reactivity	Slight oxidation
7/6/52	ANL, Illinois	6.8 kg ²³⁵ U oxide particles in plastic	Inhomogeneous cylinder water- reflected	1.22×10^{17}	Manual withdrawal of central safety rod	Plastic destroyed
12/12/52	Chalk River, Canada	Normal U	Rods, D ₂ O-moderated, graphite-reflected	1.2×10^{20}	Safety circuits failed; control rod misoperation	Core ruined
7/22/54	Reactor Testing Area, Idaho Falls, Idaho	U-A1 plates, A1 clad	Inhomogeneous cylinder, water- reflected	4.68×10^{18}	Estimate of expected excursion too low	Reactor destroyed
10/15/58	Vinca, Yugoslavia	3996 kg Normal U	Rods D ₂ O-moderated, unreflected	2.5×10^{18}	Too much D ₂ O added in final step of experiment	None
3/15/60	Sucloy, France	2.2 tons UO ₂ , 1.5% enriched	Canned UO ₂ rods in water	3×10^{18}	Control rod withdrawn	None
1/1/61	Reactor Testing Area, Idaho Falls, Idaho	U-A1 plate, A1 clad	Inhomogeneous cylinder, water- moderated	4.4×10^{18}	Quick manual withdrawal of control rod	Reactor destroyed, building contaminated
12/30/65	Mol, Belgium	1.2×10^4 g UO ₂ , 7% enriched	Canned UO ₂ rods in H ₂ O-D ₂ O	4.3×10^{17}	Manual withdrawal of control rod	None

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Table 3-12. Miscellaneous Systems
(Stratton 1967)

Date	Location	Active Material	Geometry	Total Fissions	Cause	Physical Damage
2/11/45	Los Alamos, New Mexico	UH, pressed in styrex	Cylinder	$\sim 6 \times 10^{15}$	Reflector added and/or source too large	UH ₃ -styrex cubes swollen and blistered
1953	USSR	Pu(NO ₃) ₄	Block tank	2.5×10^{17}	Transfer to unsafe geometry	None
—	Idaho Reactor Testing Area	½ in ²³⁵ U rods	Cylinder, rods NaK cooled	4.7×10^{17}	Incorrect scram used	Core molten
7/3/56	LASL, New Mexico	58 kg U 93% ²³⁵ U, 2- and 5-mil foils	Cylinder	3.2×10^{16}	Change of k from previous assembly too large	None
11/18/56	Reactor Testing Area, Idaho Falls, Idaho	²³⁵ U Ni-Cr elements, ZrH-moderated	Cylinder, prototype aircraft engine	2.5×10^{19}	Incorrect wiring in ion chamber circuit	Every fuel cartridge melted
12/11/62	LASL, New Mexico	²³⁵ U foils in graphite	Cylinder, graphitic and Be-reflected	3×10^{16}	Inadequate communication between work crews	None

Table 3-13. Nuclear Criticality Accidents at Russian Industrial Facilities (Frolov, et al. 1995)

Date	Plant	Total Fissions	Notes
3/15/53	Mayak Enterprise	2.5×10^{17}	Transfer of Pu solution into unsafe geometry vessel.
4/21/57	Mayak Enterprise	2×10^{17}	Unmonitored accumulation of uranium oxalate precipitate.
1/2/58	Mayak Enterprise	2.3×10^{17}	Workers tip vessel, creating unsafe solution geometry.
12/5/60	Mayak Enterprise	1×10^{17}	Faulty plutonium mass analysis, accumulation of solution and precipitate in unsafe geometry vessel.
8/14/61	Siberian Chemical Combine	1×10^{16}	Unmonitored accumulation of uranium hexafluoride in oil vessel.
9/7/62	Mayak Enterprise	2×10^{17}	Unmonitored addition of Pu scrap to dissolver vessel.
1/30/63	Siberian Chemical Combine	7.9×10^{17}	Error in measuring uranium solution concentration, transfer to unsafe geometry vessel.
12/13/65	Siberian Chemical Combine	2×10^{17}	Unmonitored accumulation of uranium solution in unsafe geometry vessel.
11/13/65	Electrostal Fuel Fabrication Plant	1×10^{15}	Unmonitored accumulation of UO_2 slurry in holding vessel.
12/16/65	Mayak Enterprise	7×10^{17}	Faulty accounting, loading of uranium solid into a dissolver with unsafe geometry.
12/10/68	Mayak Enterprise	6×10^{16}	Unmonitored concentration in extractant, transfer to unsafe geometry vessel.
12/13/78	Siberian Chemical Combine	3×10^{15}	Unmonitored transfer of Pu metal into storage container.

3.4.1.4 Large Storage Arrays

Limited experimental data exists for large storage array systems of the type found in fuel cycle facilities. Use of the reactor excursion data assumes development of a flooded condition and a bounding estimate of total number of fissions of 5×10^{18} . As in the case of moderated, reflected metal systems, the event is expected to be characterized by a single burst with little or no plateau period.

3.4.2 Nuclear Criticality Accident Source Term Estimates

Estimation of the potential impacts of a nuclear criticality accident involves estimation of the total amounts of radioactive material and energy generated in the event and consideration of the fraction of this material and energy that escapes from the facility. This section presents a method for estimation of criticality accident source terms using a uranium solution criticality as an example. The method is representative of the technical approach that may be applied to evaluation of generic criticality events. Methods for estimation of airborne release fractions and respirable fractions for release modes characterizing criticality accidents are also discussed.

Estimation of criticality accident source terms begins with development of a conceptualization of the physical system that retains the important characteristics of the system, but which allows application of mathematical models and analytical tools to predict the behavior of the system. The analysis aims to bound potential impacts rather than predict system behavior in great detail. To this end, the historically observed behavior of solution criticalities as involving an initial burst followed by an extended plateau is adopted as describing the event. The plateau period is represented as comprising a series of pulses each with total number of fissions less than the initial burst. Following reported accident events, the initial burst is taken to be 1×10^{18} fissions, with 47 subsequent bursts each involving 1.92×10^{17} fissions, for a total of 1×10^{19} fissions. Each burst lasts 5 seconds and is separated from the preceding burst by 10 minutes, yielding a total accident duration of 8 hours. The example consists of a tank containing a solution of $\text{UO}_2(\text{NO}_3)_2$ enriched to 4 per cent in the U-235 isotope. The concentration of uranium is taken to be 400 g/l ($\sim 25 \text{ lb/ft}^3$) and the total volume is 400 l ($\sim 0.011 \text{ ft}^3$). The actual geometry of the system is approximated by a sphere and the physical state is represented as a homogeneous liquid mixture.

For the example, the analysis may be separated into estimation of fission product generation rates and estimation of gamma ray and neutron currents escaping the system. The fission product generation rates are estimated using the ORIGEN2 computer code (ORNL 1989). ORIGEN2 is a point-depletion and radioactive decay computer code that can be used to simulate nuclear reactor processes. The fission product yields for important isotopes predicted using the code are presented in Table 3-14.

Because ORIGEN2 does not provide output data on prompt gamma and neutron generation rates, a supporting model is needed. The gamma ray generation rate and energy distribution are adopted from power reactor operating data. The prompt photon production rate is reported as 7.03 photons per fission, with the energy spectra presented in Table 3-15. For the example case, a total of 7×10^{19} prompt photons would be produced over the 8-hour period. The total production could be conservatively adopted as the release rate or the source could be represented as uniformly distributed through the spherical, aqueous system (with a radius of 45.7 cm (18 in)) and a shielding code used to estimate the rate and energy distribution of photons escaping the system.

A similar approach could be used to evaluate escape of prompt neutrons. For U-235, approximately 2.4 prompt neutrons are produced for each fission, yielding a total of 2.4×10^{19} prompt neutrons for the entire

Table 3-14. Radioactivity Generated in a Uranium Solution Criticality Accident

Nuclide	Half-life	Radioactivity, Bq (Ci)			Average Decay Energy (MeV)	
		0-0.5 hr	0.5-8 hr	Total	γ	β
Kr-83m	1.8 hr	7.4E11* (20)	4.6E12 (130)	5.4E12 (150)	2.7E-3	2.9E-2
Kr-85m	4.5 hr	4.5E11 (12)	2.8E12 (77)	3.3E12 (89)	1.6E-1	26E-1
Kr-85	10.7 hr	6.7E4 (1.8E-6)	4.1E5 (1.1E-5)	4.8E5 (1.3E-5)	2.2E-3	2.5E-1
Kr-87	76.3 min	5.5E12 (150)	3.4E13 (920)	4.0E13 (1070)	7.9E-1	1.3E0
Kr-88	2.8 hr	3.4E12 (91)	2.1E13 (570)	2.4E13 (660)	1.9E0	3.6E-1
Kr-89	3.2 min	2.3E14 (6300)	1.5E15 (40000)	1.7E15 (46000)	1.6E0	1.3E0

Nuclide	Half-life	Radioactivity, Bq (Ci)			Average Decay Energy (MeV)	
		0-0.5 hr	0.5-8 hr	Total	γ	β
Sr-91	9.5 hr	1.6E12 (44)	1.0E13 (280)	1.2E13 (320)	6.9E-1	6.6E-1
Sr-92	2.7 hr	5.9E12 (160)	3.7E13 (990)	4.2E13 (1200)	1.3E0	2.0E-1
Ru-106	368 day	1.2E8 (3.3E-3)	7.4E8 (2.0E-2)	7.4E8 (2.0E-2)	-	1.0E-2
Cs-137	30.0 yr	6.3E7 (1.7E-3)	3.7E8 (1.0E-2)	3.7E8 (1.0E-2)	-	1.9E-1
Ba-139	82.7 min	1.3E13 (340)	7.8E13 (2100)	9.1E13 (2400)	4.3E-2	9.0E-1
Ba-140	12.7 day	5.5E10 (1.5)	3.5E11 (9.5)	4.1E11 (11)	1.8E-1	3.1E-1
Ce-143	33.0 hr	5.2E11 (14)	3.2E12 (87)	3.7E12 (100)	2.8E-1	4.3E-1

Table 3-14. Radioactivity Generated in a Uranium Solution Criticality Accident (Continued)

Nuclide	Half-life	Radioactivity, Bq (Ci)			Average Decay Energy (MeV)	
		0-0.5 hr	0.5-8 hr	Total	γ	β
Xe-133	5.2 day	1.4E7 (3.7E-4)	8.5E7 (2.3E-3)	1.0E8 (2.7E-3)	4.6E-2	1.4E-1
Xe-133m	2.2 day	9.6E7 (2.6E-3)	6.3E8 (1.7E-2)	7.0E8 (1.9E-2)	4.1E-2	1.9E-1
Xe-135	9.1 hr	2.7E10 (7.3E-1)	1.7E11 (4.5)	1.9E11 (5.2)	2.5E-1	3.2E-1
Xe-135m	15.3 min	1.7E12 (45)	1.0E13 (280)	1.2E13 (330)	4.3E-1	9.8E-2
Xe-137	3.8 min	1.2E14 (3300)	7.6E14 (21000)	8.8E14 (24000)	1.6E-1	1.8E0
Xe-138	14.2 min	5.2E13 (1400)	3.2E14 (8700)	3.7E14 (10000)	1.1E0	6.7E-1

Nuclide	Half-life	Radioactivity, Bq (Ci)			Average Decay Energy (MeV)	
		0-0.5 hr	0.5-8 hr	Total	γ	β
I-131	8.0 day	3.7E10 (1.0)	2.3E11 (6.3)	2.7E11 (7.3)	3.8E-1	1.9E-1
I-132	2.3 hr	5.2E12 (140)	3.2E13 (880)	3.8E13 (1,000)	2.2E0	4.9E-1
I-133	20.8 hr	8.6E11 (23)	5.4E12 (150)	6.2E12 (170)	6.0E-1	4.1E-1
I-134	52.6 min	2.1E13 (570)	1.3E14 (3,600)	1.5E14 (4,200)	2.6E0	6.2E-1
I-135	6.6 hr	2.6E12 (69)	1.6E13 (430)	1.9E13 (500)	1.6E0	3.6E-1

^a exponential notation, 7.4E11 = 7.4x10¹¹

Table 3-15. Prompt Fission Gamma-Ray Spectra*

E (MEV)	N(E) γ's/fission
0.5	3.1
1.0	1.9
1.5	0.84
2.0	0.55
2.5	0.29
3.0	0.15
3.5	0.062
4.0	0.065
4.5	0.024
5.0	0.019
5.5	0.017
6.0	0.007
6.5	0.004

*from ANL 1963

Table 3-16. Prompt Neutron Energy Spectrum

Energy (MeV)	Fraction of Neutrons
0.0-0.2	0.038
0.2-0.4	0.061
0.4-0.6	0.069
0.6-0.8	0.071
0.8-1.0	0.071
1.0-1.2	0.068
1.2-1.4	0.064
1.4-1.6	0.059
1.6-1.8	0.055
1.8-2.0	0.050
2.0-2.2	0.046
2.2-2.4	0.041
2.4-2.6	0.037
2.6-2.8	0.032
2.8-3.0	0.030
3.0-3.2	0.026
3.2-3.4	0.023
3.4-3.6	0.020
3.6-3.8	0.018
3.8-4.0	0.016
4.0-5.0	0.054
5.0-6.0	0.027
6.0-8.0	0.019
8.0-10.0	0.004
>10.0	0.001

criticality event. The energy distribution reported (LaMarsh 1983) for these neutrons is summarized in Table 3-16. As in the case of the prompt gamma rays, the prompt neutron source could conservatively be used as the released source or the source could be uniformly distributed through the spherical system, with the rate and energy distribution of neutrons escaping the system estimated using a shielding code.

In the case of neutrons, a third analytical approach could be applied. A single-group thermal diffusion model could be used to model the neutron flux, with the escape fraction estimated as the integral over the surface of the system of the product of neutron diffusivity and gradient of neutron flux. This approach yields a total escape of 2.7×10^{17} neutrons during the 8 hours of the criticality event. Application of these concepts is further illustrated in the sample problem discussed in Section 9 of Appendix D.

3.4.3 Estimation of ARF and RF for Nuclear Criticality Accidents

Estimation of the airborne source term resulting from an inadvertent nuclear criticality (INC) is based upon the same 5-factor formula used for other events. The evaluation of the various components may have expanded definitions. Some considerations in the evaluation of the five components are:

MAR. The MAR is the radionuclide inventory that can be at risk during the postulated event. Unlike the MAR for most events, where the MAR is a fixed value (typically the entire radionuclide inventory within some fixed physical boundary), the MAR for an INC depends on the fissile/fissionable materials involved and the total fission yield. As discussed previously, the inventory can be estimated by use of a computer code such as ORIGEN2, which evaluates the fission product (FP) inventory generated by a specific fissile material (i.e., ^{233}U , ^{235}U , ^{239}Pu) during a nuclear excursion. In some instances (e.g., acid solutions of SNF and other materials that contain a mixture of fissile and fissionable materials), the other fissile and fissionable materials can fission (depending on duration of excursion and neutronics of the situation) and generate their own spectrum of FPs. Typically, the duration of the excursion is short, and contributions from this source are ignored, but it should be borne in mind that even code values are approximations, with some level of uncertainty depending on the conditions.

DR. The DR reflects the fraction of the MAR that is actually impacted by the specific event postulated. As an example, a fire may range from a small fire with a limited amount of fuel or oxygen availability affecting only a fraction of the MAR, to a large fire that affects all of the contents within the physical boundaries that prevent its propagation. For INCs, the DR is typically assumed to be 1.0.

ARF. Because of the various physical forms of the radionuclides (gases, vapor, non-volatile) and the matrix (solutions, metal, powders, irradiated compacted ceramic oxides [SNF] and cermets), there is more than a single ARF for the airborne release resulting from an INC. The ARFs for various physical forms of the radionuclides and matrix are discussed in greater detail below.

RF. The RF is defined as the fraction of the ARF that is in the particle size range D_{AED} 10 micrometers and less. Since noncondensable gases and vapors (materials airborne in the gaseous state) are not particles, the RF applies only to materials that are made airborne as particles (non-volatile compounds in solid or liquid form).

LPF. There are many LPFs possible during the transport of materials made airborne during an event from the point of origination to release from the facility-atmosphere interface. The LPFs may be expressed as a single value that represents the summation of all LPFs, or that represents the single LPF phenomenon considered. In most event analyses, the LPF represents phenomena that occur after the radionuclides are made airborne in some confinement. In the case of INCs, there is one category of matrix (large storage arrays), with one class of materials, clad, (i.e., SNF, "pits"), where an LPF may exist at the point of origination.

For SNF, the clad is designed for operation at high temperatures and pressure found in nuclear reactors, and those that have not failed in this environment have a reasonable expectation of resisting the temperature and additional internal pressure generated by an INC. No airborne release is anticipated from intact, clad, SNF in an INC. The SNFs that have already failed are assumed to have released their "gap" inventory of FPs generated by irradiation in the reactor, and the same fraction of the INC-generated inventory would be released.

"Pits" (weapons-grade plutonium hollow metal spheres encased into a welded and tested stainless clad with and without a small-diameter tube leading into the internal cavity) also offer some degree of resistance to airborne release of the event-generated FPs. If the calculated metal temperature induced by the energy from the INC exceeds the melting point of the weld material (~ 423 K 302 °F), the encased Pu would be exposed to air. At these temperatures, air oxidation of Pu is slow. If the temperatures exceed the ignition temperature (the temperature at which a self-sustained oxidation reaction is initiated, the metal temperature would increase to 1273 to 1373 K (1832 to 2012 °F), failing the stainless steel clad); all of the metal would be assumed to be oxidized; and the ARF and RF for Pu metal under thermal stress (ARF 5E-4 and RF 0.5) would also apply.

3.4.3.1 Rules-of-Thumb for Total Fissions during INC

Because of the complexity of the phenomenon and the many factors that should be considered to obtain an estimate of the total fissions generated by a specific event, "rules-of-thumb" provide users with a quick, bounding assumption for total fissions and other phenomena that may affect the total release of radionuclides.

a. Liquid Systems

1. Large systems involving greater than 380 ℓ (100 gal). The reaction terminates by evaporation of 100 liters (26 gal) of liquid: 1×10^{19} total fissions (1×10^{18} initial burst [0.5 seconds], followed by 47 bursts of 1.9×10^{17} fissions at 10 min intervals). The fission product generated is dependent on the initial fissile materials involved and should be estimated by a code such as ORIGEN2 or equivalent. The aerosol generated by the boil-off of water suspends 5×10^{-4} of the salt content of the liquid evaporated (100 liters (26 gal) required to terminate the reaction).

2. Small systems involving less than 380 ℓ (100 gal). The reaction terminates by eruption of a portion of the liquid (release from a free-fall spill of liquid and suspension of a fraction of the liquid): 1×10^{18} total fissions in a 0.5-s burst. Reaction is terminated by eruption of liquid, and a free-fall spill release value should be applied under this assumption. The fission products generated are dependent on the initial fissile materials involved and should be estimated by a code, such as ORIGEN2 or equivalent.

For any liquid system, the assumptions for fission product release are:

- All the noble gases generated are released
- Most of the volatile iodine isotopes are retained in the liquid and 25 percent is released.

b. Solid Metal Systems - 1×10^{18} total fissions. Fission product generated depending on initial fissile materials involved and should be estimated by a code such as ORIGEN2 or equivalent. For release assumption, see text.

c. Powder Systems - 1×10^{17} total fissions. Fission product generated depending on initial fissile materials involved and should be estimated by a code such as ORIGEN2 or equivalent. For release assumptions, see text.

d. Large Storage Arrays - 1×10^{20} total fissions. Fission product generated depending on initial fissile materials involved and should be estimated by a code such as ORIGEN2 or equivalent.

Technical Basis

The value for the solid metal system and powder systems is based upon a value that bounds most of the reported data for inadvertent nuclear criticalities that have occurred and are listed in Tables 3-8 - "Summary of Known Accidental Criticality Excursions (a. Solution Systems, b. Metal Systems, and c. Moderated Foil and Powder Systems (1945 to 1974)"; 3-9 - "Accidents in Processing Plants"; 3-10 - "Destructive Power Excursion Summary"; 3-11 - "Inhomogeneous Water-Moderated Systems"; 3-12 - "Miscellaneous Systems"; and 3-13 - "Nuclear Criticality Accidents at Russian Industrial Facilities." All but one of the inadvertent nuclear criticalities that have occurred in processing facilities have involved solutions. Solids are more readily controlled because of their fixed physical configuration. Likewise, powders tend to flow and much more than the minimum critical mass stated in the standards (ANSI 1983) and other relevant documents (Clayton 1979; Thomas 1978) is required for criticality in the conditions and geometries encountered in processing plants. McLaughlin (1991) presented a table of "Criticality Accident Fission Yields" that was proposed for estimation of safe exclusion areas within work places (Table 3-17 "Criticality Accident Fission Yields"). The values are larger in many cases than those cited here.

The total fission value for large storage arrays is based upon the data presented of the events that have occurred, consideration of the yield from reactor excursions, and Woodcock's estimates shown in McLaughlin's table. No estimates are made for the fission product release.

3.4.3.2 Scenario Assumptions and ARFs and RFs

Solution Systems

1. Scenario Assumptions. As discussed in Subsection 3.4.2.1, "Nuclear Criticality Accident Source Term Estimation Methods," the assumed total fission yield from the event in a spherical system 45.7 cm (18 in) in radius is an initial burst of 1×10^{18} fissions, followed by 47 5-sec bursts of 1.92×10^{17} fissions each 10 minutes, for a total of 1×10^{19} total fissions. The volume of a sphere with a radius of 45.7 cm is approximately 400 liters (~ 100 gal). The "rule-of-thumb" for the total fission yield is based on the estimated total fission from the INCs that have occurred in solutions and provides a reasonable upper bound value for events other than those that occur in very large volumes that greatly exceed the critical mass.

2. ARFs and RFs. The estimated inventories from an INC involving ^{235}U in solution is shown in Table 3-14, "Radioactivity Generated in a Uranium Solution Criticality Accident." The physical forms of radionuclides generated (shown in Table 3-14) are those typically involved (although the inventories differ) and include noncondensable gas (noble gases), volatiles (iodine in heated, acid solution), and non-volatiles (all remaining radionuclides that are dissolved in the acid solution). Thus, ARFs are required for the noble gases and iodine and ARFs and RFs are required for the non-volatile materials generated and present in the solution (the fissile material and other non-volatiles present in dissolved SNF).

- Noble gases - All noble gases present are assumed to be released during the generation and subsequent boiling of the liquid. The ARF assigned is 1E+0 (unity).

**Table 3-17. Criticality Accident Fission Yields
(McLaughlin 1991)**

System	Initial Burst Yield (Fissions)	Total Yield (Fissions)
Solutions under 100 gal (0.38 m ³)	1x10 ¹⁷	3x10 ¹⁸
Solutions over 100 gal (0.38 m ³)	1x10 ¹⁸	3x10 ¹⁹
Liquid/powder	3x10 ²⁰	3x10 ²⁰
Liquid/metal pieces	3x10 ¹⁸	1x10 ¹⁹
Solid uranium	3x10 ¹⁹	3x10 ¹⁹
Solid plutonium	1x10 ¹⁸	1x10 ¹⁸
Large storage arrays below prompt critical	None	1x10 ¹⁹
Large storage arrays above prompt critical	3x10 ²²	3x10 ²²

- Iodine - All iodine isotopes are assumed to be quantitatively released during the boiling of the acid solution. This includes all iodine isotopes that may already be in solution (e.g., ¹²⁹I and ¹³¹I in dissolved SNF). The ARF assigned is 1E+0 (unity). Iodine is a highly reactive chemical material that will react with materials (structural, airborne inert particles, engineered exhaust treatment components such as filters and absorbers) encountered along pathway to release. The typical assumption is that an upper bound of 0.25 of the iodine released from the liquid escapes the facility and is released into the atmosphere.
- Non-Volatile - The suspension mechanism for non-volatiles in solution is the formation and suspension of liquid droplets from film-break up. Films are formed by the generation of bubbles of gas or vapor as they rise to the surface thinning the upper film of the bubble until the bubbles burst.

The experimentally measured upper bound value for the ARF is 2E-3, with no measured RF. Since the boiling is assumed to reduce the volume of solution to a level that is no longer a critical configuration, not all the liquid is evaporated in the process. For small volumes, there is a reasonable expectation that the initial burst of 1x10¹⁸ fissions should result in a sufficient loss of volume to terminate the criticality excursion. Thus, it is assumed that less than 25 percent of the volume is evaporated and that the applicable ARF is [2E-3][0.25] or 5E-4 with an RF of 1.0.

The ARF and RF values are applicable to all non-volatile compounds in the liquid, including the fissile material and non-volatiles present in the reacting solution and all the INC-generated FPs. As shown in Table 3-14, for an INC involving ²³⁵U, the highest inventory of non-volatile FP generated by the excursion is 1200 Ci of ⁹²Sr.

Metal Systems

Scenario Assumptions. An INC involving a metal would result in FPs formed throughout the metal matrix. Unless energy levels are sufficient to soften the metal, only the FPs on the metal surface are likely to be released. Restrepo (1992) reviewed the literature on FP release from failed fuel elements heated to a temperature that resulted in fuel slumping. He divided the elements into categories that appear to behave similarly under these conditions. The ARFs derived are shown in Table 3-18, "Release Fractions for Various Chemical Classes from Heated Spent Fuel". The values represent the release estimated for unclad SNF heated to temperatures exceeding those anticipated for INCs and are easily upper-bound values for these materials from INCs.

Powder Systems

1. Scenario Assumptions. As with a metal matrix, an INC involving fissile materials in powder form would result in the formation of FPs within the matrix of the material involved. The tendency is to favor formation near the surface region where the neutron flux is greater. With the larger surface area per mass of powder than for a metal, the values applicable to metal are not relevant.
2. ARFs and RFs. The surface to mass ratio for a powder is dependent on both its particle size distribution and shape. Both are unknown and the upper-bound values are conservatively assumed to be:

Noble gases - ARF 1E+0, RF NA

Iodine - ARF 1E+0, RF NA

Non-volatile - [based on the suspension during the heating of an chemically, non-reactive compound from (DOE 1994, p. 4-57)] ARF 6E-3, RF 0.01.

Large Storage Arrays

1. *Scenario Assumptions.* This category of material represents a very large inventory of fissile materials storage in a geometrically favorable configuration. It is impacted by an event that defeats the system to produce a geometrically unfavorable configuration.

There are two relevant classes of materials:

1. Metal Clad Material - SNF, "pits"
2. Unclad - metal, powders, etc.

The anticipated behavior of the two classes of Large Storage Array materials was discussed in Subsection 3.4.3, LPE. The amount released to the ambient atmosphere is the product of the ARF & RF, if applicable, and the LPF.

2. *ARFs and RFs for INCs Involving Large Storage Arrays.* For the unclad materials, the ARFs and RFs for the material forms are applicable.

For clad materials, the integrity of the clad during and following the excursion determines the release. The amount of material released through the clad may be considered LPF_i (Leak Path Factor, initial - the fraction of material released within the clad that penetrates the clad).

There is more than one form of SNF. The common forms are a compacted, sintered, ceramic oxide pellet clad in zircaloy or stainless steel (commercial nuclear fuel); uranium metal clad in aluminum or zircaloy; enriched uranium metal-aluminum alloy; cermets, etc. There has been experimental measurements of FP release during fuel failure from heating, and metal alloy and cermet targets. The derivation of ARFs from ceramic oxide fuel heated to slumping has been covered in Subsection 3.4.3.1.b ("Metals"), and these are applicable for this class of clad material.

Table 3-18. Release Fraction for Various Chemical Classes from Heated Spent Fuel (Restrepo 1991)

Group No.	Group Name	Rep. Ele.	Elements in Group	ARF
1	Noble Gases	Xe	Xe, Kr, He, Ne, Ar, Rn, H	5E-1
2	Alkali Metals	Cs	Cs, Rb, Li, K, Fr, Na	2E-1
3	Alkali Earths	Ba	Ba, Sr, Mg, Ca, Ra, Be	3E-2
4	Halogens	I	I, F, Cl, Br, At	5E-2
5	Chalogens	Te	Te, S, Se, O, Po, N	7E-2
6	Platinoids	Ru	Ru, Rh, Pd, Os, Ir, Pt, Au, Ni	2E-3
7	Transition Metals	Mo	Mo, V, Cr, Fe, Co, Mn, Nb, Tc	3E-2
8	Tetravalent	Ce	Ce, Ti, Zr, Hf, Th, Pa, U, Np, Pu	4E-4
9	Trivalent	La	La, Al, Sc, Y, Ac, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Am, Bk, Cf	6E-4
10	Main Group I	Cd	Cd, Hg, Zn, As, Sb, Pd, Tl, Bi	4E-3
11	Main Group II	Sn	Sn, Ca, In, Ag	4E-3
12	Boron	B	B, Si, P, C	6E-4

The experimental data for release from metals and cermets were analyzed in DOE (December 1994). The ARFs are divided into "instantaneous" (recovered during a 2-min collection period) and "total" (recovered during a 60-min collection period). Only values for three elemental forms were given:

	"Instantaneous"	"Total"
Cesium	0.06	0.09
Iodine	0.8	0.9
Tellurium	0.00	0.007

Although the other non-volatile radionuclides are not specifically listed, the values for tellurium, which can be volatile under some conditions, can be applied as upper bounds.

3.5 Chemicals

The purpose of this section is to describe how to calculate the characteristics of accidental releases of hazardous chemicals such as uranium hexafluoride, hydrogen fluoride, ammonia, chlorine, and other chemicals commonly found at nuclear fuel cycle facilities. The characteristics of the chemical source term include rate of release, temperature, momentum, orientation, height, and aerosol content. These quantities are required for atmospheric dispersion modeling, which is discussed in Chapter 5 of this Handbook.

3.5.1 Identification of a Representative Range of Source Terms

No single atmospheric dispersion model or source term will suffice for the range of scenarios possible at the fuel cycle facilities. At the same time, it may not be necessary to develop or use highly sophisticated models that take into account every possible detail about the source term. Certain aspects of the source term need not be fully quantified, if they can be shown to have little or no impact on the final risk estimate. Hence, the selection of the appropriate source term is as much an art as a science. This section provides guidance on the selection of the appropriate source term for the types of accident scenarios involving chemicals possible at the fuel cycle facilities by providing a broad overview of the range of source terms. For modeling details of each type of source term described herein, the reader is directed to Appendix B of this Handbook.

3.5.1.1 Liquid below Its Boiling Point

If spilled accidentally, liquids with boiling points that are well above ambient temperature will form a pool on the ground and evaporate slowly at a rate determined by the ambient temperature, the windspeed and the area of the spill. No aerosolization is expected in such a spillage. Hydrofluoric acid (normal boiling point 293 K (68 °F)) is an example of a liquid that can be spilled below its boiling point if the spill occurs from a storage tank at an ambient temperature that is less than 293 K (68 °F).

If the spill initially occurs from a severed line, then Bernoulli's equation can be used to estimate the flow rate onto the ground (see equation B.1 in Appendix B). Bernoulli's equation takes into account the influence of static liquid pressure head, and the influence of vapor pressure above the liquid level.

After a spill, the methods developed for estimating the evaporation rate of slowly evaporating liquids from pools can be used. These methods assume that heat transfer from the underlying surface of the pool is rapid, and is not the limiting factor for evaporation. The rate of evaporation is limited by the rate of mass transfer across a stagnant film of air at the surface of the pool. The method for predicting the evaporation rate is described in Section B.2.1, equations B.2, B.3, and B.4 of Appendix B. An example of a calculation of an evaporating pool of HF at a temperature of 280 K (44 °F) is given in Sample Problem No. 5.

3.5.1.2 Refrigerated Liquid in a Vessel

The method used to estimate the release rate of a liquid below its boiling point (Bernoulli's equation), can also be used to estimate the release rate of a refrigerated liquid. Examples of refrigerated liquids include freon and ammonia. In the present context, refrigerated liquids have boiling points that are well below