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**GRAPHITE FUELS COMBUSTION
OFF-GAS TREATMENT OPTIONS**

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Abstract

Treatment options are identified for each anticipated off-gas component from the burning of graphite spent nuclear fuels, with recommendations for pursuit of the most appropriate options. Flowsheets and mass balances are presented for representative treatment schemes.

Summary

Scenarios for burning bulk graphite and for burning crushed fuel particles from graphite spent nuclear fuels have been considered. Particulate can be removed with sintered metal filters. Subsequent cooling would then condense semi-volatile fission products into or onto a particulate. These particulate would be trapped by a second sintered metal filter or downstream packed bed. A packed bed scrub column can be used to eliminate most of the iodine-129 (^{129}I) and tritium (^3H). A molecular sieve bed is proposed to collect the residual ^{129}I and other tramp radionuclides downstream (Ruthenium, etc.). Krypton-85 (^{85}Kr) can be recovered, if need be, either by cryogenics or by the KALC process (Krypton Adsorption in Liquid Carbon dioxide). Likewise carbon-14 (^{14}C) in the form of carbon dioxide could be collected with a caustic or lime scrub solution and incorporated into a grout, but the ^{14}C poses little concern if released directly to the atmosphere. Sulfur dioxide present will be well below regulatory concern level of 4.0 tons per year and most of it would be removed by the scrubber. Carbon monoxide emissions will depend on the choice of burner and start-up conditions. The regulatory concern level is 100 tons of carbon monoxide per year; should the system exceed that level a catalytic converter in the final packed bed will be provided. Radon (Rn) and its daughters have sufficiently short half-lives (less than two minutes). The holdup time within the off-gas system will eliminate most of them. If necessary, an additional holdup bed can be added before the final High Efficiency Particulate Air (HEPA) filters or additional volume can be added to the molecular sieve bed to limit radon emissions.

The calculated total effective dose equivalent (EDE) at the Idaho National Engineering Laboratory (INEL) boundary from a single release of all the ^3H , ^{14}C , ^{85}Kr , and ^{129}I in the total fuel mass is 0.43 mrem/year. If burning of the fuel were spread out over a period of five years, state permitting would be required, but National Emissions Standard for Hazardous Air Pollutants (NESHAP) permitting would not be necessary. Regardless of the rate of combustion of the fuel, all existing emission requirements can be met with known methods. Exposure to radionuclides will be kept as low as reasonably achievable (ALARA) based on thorough evaluation of the risks and expenses for each available treatment.

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Introduction

One promising option for disposal of graphite matrix spent nuclear fuels is to reduce the quantity of repository waste by burning away the graphite matrix. High Temperature Gas Cooled Reactor (HTGR) spent fuels, such as the Peach Bottom (PB) and Fort St. Vrain (FSV) reactor fuels stored at the INEL, consist of a large volume of contaminated graphite which encases the spent fuel compacts (rods) and fission products. The fuel elements, or blocks, may be burned whole or shredded. If burning for further volume reduction is desired, the fuel rods and the fuel particles within the rods must be crushed in order to expose the carbon within the particle cladding. To minimize the volume of radioactive ash for disposal, two burning operations are needed: one to burn the blocks and one to burn the crushed particles. Off-gas from both burning operations and from the crushing of the particles must be treated to control the emission of radionuclides. Fuel particle breakage results in the greatest release of volatile fission products. Combustion of the bulk graphite blocks releases less than 5% of the volatile fission products to the off-gas because most of the fuel particles still remain intact.

Mechanical disassembly of the rods from the blocks is under separate study for potential application. The disassembly will allow for the burning of rods and particles only. This type of processing will greatly reduce the volume of off-gas to be treated and the subsequent radionuclide release. With successful disassembly and sufficient decontamination the empty graphite blocks may qualify as low-level waste.

Treatment of residual particulate, semi-volatile fission products, ^{14}C , ^3H , ^{129}I , ^{220}Rn and ^{85}Kr are major considerations in evaluating the safety and economic feasibility of the burning and crushing scenarios. The quantities of the radionuclides involved and the technology available to meet current emissions standards and ALARA (as low as reasonably achievable) exposure goals are enumerated below.

Overall Equipment Flowsheets

The burner off-gas flows and quantity of contaminant depend on the processing technique used. Burning the bulk graphite, after crushing the whole fuel element, will produce the most off-gas to be cleaned up and will require the greatest amount of equipment. Processing of the separated compacts (fuel rods) will require less equipment and should result in much less of the contaminants leaving with the exhaust gases.

A representative overall flow scheme is shown as Figure 1. This scheme does not include the mechanical fuel preparation steps which may release some contaminated

off-gas. The recommended treatment of the off-gas is to provide barriers in depth to remove the most concentrated contaminants in order.

Sintered metal filters remove particulate in two stages. The first stage would contain courser filters to recycle elutriated bed material and unburned carbon and the second stage would have a finer pore size (approx. 0.5 micron) for removal of most of the balance of particulate. With proper temperature control most of the semi-volatile fission products should be removed by the sintered metal filters.

The filtered off-gas is then treated by wet scrubbing, which will remove particulate as well as dissolved gases. A favored wet scrubbing system is a subcooled system using a methanol-water mixture and operating at -40°C . This scrub system will be sized to allow a minimum gas residence time of 25 seconds. It should remove the expected particulate, aerosols, and volatiles with various decontamination factors. The two scrubbers shown in Figure 1 have a recirculating scrub solution cleanup system. The solution cleanup will improve the scrubber efficiency by providing a clean scrub solution. Most of the contamination found downstream of batch scrub solution systems is attributable to scrub solution entrainment¹. The smaller scrubber will clean up the more concentrated off-gas stream which contains most of the off-gas radioactive contaminants. The smaller scrubber system effluent will be combined with the larger scrubber stream to achieve greater overall off-gas decontamination.

Downstream of each scrubber a packed bed will remove aerosols and tramp contaminants leaving the scrubbers. The large bed could be used to adsorb radon for decay. Lastly, off-gas will pass through a dual set of HEPA filters before release to a stack.

If fluidized bed burners are used, a recycle blower is recommended to provide for the extra burner fluidization gas volume. This recycle scheme would reduce the volume of gas to be treated downstream and the same size of downstream equipment could be employed as for the non-fluidized bed options. The fluid bed burner systems will have a greater potential for contaminant loading to the off-gas. This difference is not well defined at present.

The primary radioactive contaminant in the off-gas is expected to be ^{85}Kr and most of this will be released when the particle is broken, probably in the particle crusher. The total volume of the crusher gas stream should be small (due to tight ventilation control) and after filtration it will be routed either to the small scrubber or to an optional rare gas removal system. Most of the rare gas present will be non-radioactive fission product xenon.

If the compacts can be easily removed from the fuel block, the bulk graphite system, Primary Burner System of Figure 1, will not be required. A simpler system, such as that shown as Figure 2, will accomplish the task. The barrier scheme of off-gas

treatments is retained for the Combination Compact - Particle System of Figure 2 as for the overall system of Figure 1. As is mentioned later, the series of treatments are not necessary to meet present discharge criteria, but if future requirements dictate, they could be employed.

Material Balances

Material balance calculations were performed on two options, Shaft Burner Block and Combination Compact - Particle. These two options are representative of the possible ranges. The Combination Compact - Particle scheme is practical if the compacts can be separated easily from the bulk graphite. Combining some processing steps may be possible because the matrix material of the compacts is less than one fourth the theoretical density. This may allow a crusher to break up the particles and the matrix simultaneously. This is a preferred option because a burning step could be eliminated. The material balance information is presented in Tables I through VII. Tables III through VII are located in the Appendix. The comparison, between the two representative options in Table I, shows that the amount of off-gas to be cleaned is about seven times as great for the block processing as for the combination system. The emitted radionuclide concentrations appear to be about 7000 times as great as in the combination system. This is mainly because the primary burner system off-gas is treated once in the proposed equipment layout, while the combination system off-gas is treated through two scrubber plus dry packed bed systems. The radionuclide concentrations are very low for either case.

Figure 1 - Overall Graphite Fuels Offgas System

Figure 2 - Combination Compact-Particle Graphite Fuels Offgas System

TABLE I

Comparison Between Whole Block Processing and Compact-Particle Burning

	Average Bulk Burner		Average Compact-Particle	
Carbon Processed - kg/day	477.00		67.50	
Gases Required - kg/day	1,922.18		254.36	
Offgas to DOG Scrubber - SCMM (SCFM)	1.01 38.28		0.14 5.33	
Particulate Collection by Secondary Systems - kg/day	0.069		0.014	
Exit Gas Flow Rate - SCMM (SCFM)	1.01 38.56		0.141 5.36	
Contaminant in Exit Gas	kg/day	curies/day	kg/day	curies/day
H-3	7.79E-15	7.53E-14	1.11E-18	1.07E-17
C-14	4.13E-05	1.84E-07	5.85E-06	2.61E-08
Kr-85	1.09E-03	4.29E-04	1.09E-03	4.29E-04
Ru-106	1.48E-20	4.90E-20	1.95E-22	6.43E-22
I-129	1.82E-11	3.22E-18	2.58E-15	4.56E-22
Cs-137	1.29E-11	1.12E-12	1.83E-15	1.59E-16
Cs-134	5.78E-14	7.48E-14	8.20E-18	1.06E-17
Radionuclides Decayed To	9/23/98			

The parameters used for the material balance calculations are shown as Table II. These can be varied in the Excel spreadsheet as desired to produce mass balances for other conditions. All the inert gaseous species are expected to be released from the opened particles. Some of the elements are considered semi-volatile under the burning conditions. The volatile escape fractions were calculated for the principal semi-volatiles based on equilibrium at 875°C for the expected total element concentration. The primary cesium (and iodine) volatile component is CsI. The ruthenium is not very volatile under the chosen reducing conditions of expected burner operation. This calculation agrees with the literature.² The escape of most other radionuclides from the burners is as particulate through the sintered metal filters.

T A B L E I I
Flowsheet Parameters

FSV Equiv. Blocks /d	4.5
Burn fraction Primary Burner.	0.95
CO fraction in offgas	0.150
Oxygen Fraction of Combustion Gases	0.8
O2 Utilization	0.99
Sintered Metal Filter Efficiency (5.0 micron)	0.98
Sintered Metal Filter Efficiency (0.5 micron)	0.995
Blowback Air and Transfer Gas as Fraction of Flow	0.06
Fraction of Open Particles	0.007
Fraction Particle Carbon Burned	0.25
Burn Fraction Particle Burner	0.99
Ru - Volatile Escape Fraction	1.00E-06
Cs - Volatile Escape Fraction	0.038
I - Volatile Escape Fraction	0.218
Scrubber Decon. Factor	1000
Packed Bed Decon. Factor	1000
CO Combustor efficiency	0.99
CO combustor O2 fraction	1.1

Emission Limitations³

Although current federal regulations are not specific to each volatile radionuclide expected in the off-gas, permitting will be required. The state of Idaho requires extensive permitting for any known release of radionuclides to the environment including as a minimum (for a new source) a Permit to Construct and incorporation into the INEL Operating Permit.

Operations with a source term of greater than 0.1 mrem/yr for all radionuclides (based on the mitigation factors for exposure at the INEL site boundary) must obtain a National Emissions Standard for Hazardous Air Pollutants (NESHAP) Permit. This "Approval to Construct" from the Environmental Protection Agency would be required in addition to the state permits. Unmitigated emission dosage of more than 0.1 mrem/yr will require continuous emission monitoring. The quantity of each radionuclide emitted, in curies, is evaluated to determine the dose to the maximally exposed member of the public. Risks of exposure to individuals and the potential collective dose to the global population need to be evaluated with respect to the cost for each treatment option, to meet the as low as reasonably achievable (ALARA) radiological exposure constraint.

NESHAP Subpart H also limits INEL-wide releases to 10 mrem/yr effective dose equivalent (EDE). Combined simultaneous release of all of the volatile radionuclides (except radon) anticipated in the off-gas would result in an estimated exposure of 0.43 mrem at the INEL boundary due to that release based on mitigating factors and anticipated quantities of each radionuclide. The most probable estimate of ^{14}C from the proposed graphite spent fuel combustion is 130 Ci. This would produce an effective dose equivalent of 0.27 mrem. The contributions from ^3H would be 0.0057 mrem, ^{129}I would be 0.13 mrem, and ^{85}Kr would be 0.024 mrem. See Calculations at the end of this text.

The average radiation exposure to a member of the general public is 364 mrem per year, 200 mrem of which are from natural radon and 100 mrem of which are from other natural sources. Exposure to man-made sources accounts for the remaining 64 mrem per year (53 mrem from medical sources and 10 mrem from consumer products). All other occupational and nuclear energy related exposures combined result in not more than 1 mrem per year.⁴

Radon from a release does not contribute to the estimated EDE at the INEL boundary due to the short half-lives of the radon and its daughter products. The concern for exposure from radon is greatest in the immediate vicinity of the stack. The most stringent applicable exposure limit is an annual average not to exceed 0.02 working level (WL) including background, and never to exceed 0.03 WL. Working level refers

to the amount of alpha emission from decay of radionuclides in one liter of air; one working level contains enough decaying radionuclides to result in the emission of 1.3×10^5 MeV of potential alpha energy. The derived concentration guide provides a comparable limit of 3 pCi/L for ^{220}Rn .⁵ Although the anticipated concentration of radon in the off-gas is 1.07×10^6 pCi/L, radon treatment and appropriate stack design can be implemented to meet the established exposure limits. (See Calculations.)

Realistically the release of graphite fuel burner off-gas will be spread over at least three years. There are 2208 fuel elements from the Fort St. Vrain reactor, and the equivalent of about 305 FSV fuel elements from the Peach Bottom reactor (considering the different size and shape of the PB elements). The total of 2513 equivalent fuel elements processed at $4\frac{1}{2}$ elements per day with 220 operating days per year would take slightly more than $2\frac{1}{2}$ years to complete. Three years are assumed to allow for operating contingencies.

Restrictions have become increasingly more stringent and are continually subject to change. If necessary, extending the duration of the fuel treatment process could reduce annual emissions further. If the conditioning is spread over five years rather than three years, emissions would not require the NESHAP permitting since the release would be below the 0.1 mrem/year EDE.

40 CFR 190 applies specifically to fuel reprocessing but does not govern fuel conditioning for disposal. The distinction between the two is questionable as a basis for radiological release limits; in either case, the more ruptured fuel particles, the more volatile emissions. To meet ALARA, the intent of the reprocessing release limits should be addressed. To compare release of volatile components to the limits for reprocessing, the application of 40 CFR 190 to spent fuel conditioning would add the following constraints. ^{129}I release would be limited to 0.013 Ci and ^{85}Kr to 130,000 Ci based on the 2.6 gigawatt-years of electrical energy produced by the Fort St. Vrain and Peach Bottom fuels.⁶ This would require a 100 fold reduction of the ^{129}I , and a three fold reduction of the ^{85}Kr present in the off-gas.

The iodine-129 release is a legitimate concern, but few countries limit release of krypton-85. The proposed limits are achievable, but the cost of separation and disposal of the krypton may be excessive compared to the small reduction in possible exposure. The effort to keep release related exposures as low as reasonably achievable, should be directed more to controlling the ^{129}I than the ^{85}Kr .

General restrictions on industrial off-gas emissions should not be exceeded, either. Sulfur dioxide and particulate emissions limited in commercial industries will not be significant to this process. Carbon monoxide may require additional consideration depending on the operating conditions.

For a single point of emission, an annual release of less than four tons (3.6 metric tons) of sulfur dioxide (SO_2) is considered below regulatory concern, while between 4.0 and 40 tons/yr requires a state of Idaho permit to construct. The significant emission limit on SO_2 is 40 tons/yr, and beyond that a state permit for prevention of significant deterioration of the environment is required.

The sulfur content in the Fort St. Vrain fuels will be below regulatory concern at the assumed burner operating rate. The original graphite specifications indicate not more than 5000 ppm of sulfur.⁷ Neglecting the sulfur content of the non-graphite fuel components, 86 kg of graphite in each block with an additional 24 kg of carbon in the fuel rods, the maximum sulfur in each block is about 0.55 kg. Burning 4 1/2 blocks per day for 220 operating days in a year produces about 1087 kg of SO_2 per year or 1.087 metric tons (1.198 tons). This allows a large margin for sulfur in any of the non-graphite components within the 4 tons/yr limit. The actual amount of sulfur is believed to be much lower, between 30 and 1500 ppm. A value of 750 ppm was used for the mass balance calculations.

Any particulate released will be well below emission restriction level due to the HEPA filtration necessary for off-gas containing radionuclides.

For a single point of emission, release of less than 10 tons (9.07 metric tons) of carbon monoxide (CO) per year is considered below regulatory concern, while between 10 and 100 tons/yr requires a state of Idaho permit to construct. The significant emission limit on CO is 100 tons/yr, and beyond that a state permit for prevention of significant deterioration of the environment is required.

Choice of burner and possible use of carbon monoxide at burner start up will greatly influence the CO content of the off-gas released. If necessary a catalytic converter could be used, but at the burner operating temperature of 800°C , combustion should go completely to CO_2 if sufficient oxygen is provided.⁸

Treatment Technologies

Methods for removal of many off-gas components have already been developed for the fuel reprocessing industry. General Atomics (GA) Technologies conducted a joint United States/Federal Republic of Germany (GA/FRG) graphite fuel reprocessing study. Drake Engineering Inc. managed initial development and engineering work for a Graphite Fuels Pilot Facility at the Idaho Chemical Processing Plant (ICPP). And the ROVER Process burned unirradiated graphite fuel at the ICPP. Treatment technologies for each off-gas component are reviewed based on the information available to determine the best alternatives for off-gas treatment for the proposed graphite fuel combustion.

Particulate:

The amount of particulate material to be removed will vary depending on the burner and burner efficiency. Release of particulate will be negligible after the required HEPA filtration, but recycle to the burner should minimize filter wastes. Sintered metal filters were used in both GA/FRG pilot plant and ROVER projects. With the advantages of durability and blowback recycle, the price of high efficiency is a large pressure differential.

An electrostatic precipitator followed immediately by a HEPA filter was shown in the original conceptual design for the GA/FRG pilot plant, but its efficiency is not as good as the sintered metal filter for this application.^{9,10} A HEPA filter will be required to remove any fine particulate not otherwise eliminated downstream. But due to replacement and disposal constraints, the HEPA filter should be at the tail end of the off-gas treatment process.

Other options have been considered, but have been discarded for various reasons. Mechanical collectors or cyclones are inefficient (limited to about 90% efficiency), while fabric filters cannot withstand high temperatures and are prone to plugging. Wet scrubbers introduce water, generating large quantities of hazardous wet residue that does not recycle well.¹¹ A scrubber with an organic solvent instead of water has been suggested so that the solvent could be evaporated from the particulate residue, but the solvent generates another effluent.

The preferred option is sintered metal filters, each with blowback system, discharge valve and pneumatic ejector, to allow recycle. With a tall burner, a sintered metal filter can be installed at the top. The temperature of the burner would be maintained high enough that the semi-volatile fission products would pass through and prevent plugging, and low enough to prevent its deterioration.

Semi-Volatile Fission Products:

The quantity of Semi-Volatile Fission Products (SVFP), including ⁸⁸Sr, ⁹⁰Sr, ⁹⁰Y, ⁹⁹Tc, ¹⁰³Ru, ¹⁰⁶Ru, ¹⁰³Rh, ¹⁰⁶Rh, ¹²⁵Sb, ¹³⁴Cs and ¹³⁷Cs, may total as much as 122 kg for the whole inventory of Fort St. Vrain and Peach Bottom fuels. During burning, SVFP are likely to oxidize, vaporize, and react with other fission products present (i.e. iodine). The major components will be Cs₂O, Cs, TcO_x, CsI, ZrI₄, RuO₄, RuO₂, SrI₂ (maybe), SeO₂ and Se. Quantities will depend on equilibrium conditions. An estimate for the Graphite Fuels Pilot Facility stated up to 85 kg/yr of SVFP could be volatilized processing 1760 fuel elements per year. At 4 1/2 fuel elements/day with 220 operating days/year the particle burner is designed for up to 990 fuel elements per year, which may give as much as 48 kg of SVFP per year or 122 kg for all 2517.7 equivalent fuel elements from both Fort St. Vrain and Peach Bottom spent fuels.

The sintered metal filter arrangement recommended for the top portion of the burner to contain particulate may trap a significant portion of the SVFP also. Studies done by Oak Ridge National Laboratory show more than 90% of the off-gas activity was trapped in the two sintered metal filters immediately above their particle burner.¹²

SVFP must be removed immediately so that they do not plate out on the piping and other equipment as the effluent stream cools. Off-gas at the exit of the burner must be maintained at 500°C or higher until the SVFP contact the removal medium. Removal of >99% of the SVFP must be achieved in order to prevent plugging and contamination problems, particularly if sintered metal filters are used downstream.¹³

Brigham Young University (BYU) has been contracted starting in May 1993 to develop a process module for SVFP removal from off-gas. Documentation on the GA/FRG research project and on the Graphite Fuels Pilot Plant Facility for the ICPP indicates that research is needed on SVFP removal.^{9,13}

One proposed technique for SVFP removal uses a rapid cooling mechanism to plate the SVFP onto a readily replaceable or renewable nucleation surface. Multiple stages will probably be required to achieve the necessary efficiency. Monitoring will be needed for periodic removal of accumulated SVFP. Some oxides (like cesium) may require additional filtering or treatment for removal even at lower temperatures.¹³

Another option is use of a cooling vessel in the off-gas line such that the line between the burner and the vessel stays hot enough that the SVFP do not condense until they flow into the vessel.¹² Removal of resulting aerosols will be another consideration.

The researchers at BYU have proposed a cyclone to remove bulk particulate followed by a centrifugal wet scrubber with a nozzle to introduce atomized water (with a poison) to cool the off-gas and coalesce aerosols and fine particles into an aqueous liquid effluent. The wet scrubber was recommended to achieve the desired efficiency. A sintered metal filter may be used between the cyclone and the scrubber as an additional criticality control prior to the introduction of the poisoned water.

Carbon-14:

The 0.029 kg, or 130 Ci (2.08 g/mol), of ¹⁴C is a small constituent of the burner off-gas, contributing just 0.27 mrem to the 10 mrem/yr EDE limit at the INEL boundary (under the worst case assumption that it is all released at one time). ¹⁴C has a half-life of 5730 years; it decays to ¹⁴N which is stable. The total amount of carbon in the off-gas primarily as carbon dioxide will depend on the burner design, but about 110 kg of carbon is present in each of some 2500 fuel elements. Over 270,000 kg of ¹²C will have to be handled along with the 0.029 kg of ¹⁴C.

The GA/FRG research program and the previous work on the Graphite Fuels Pilot Plant Facility separated the carbon dioxide from the off-gas with a hydroxide scrub column. The primary treatment for disposal has been formation of a carbonate grout for shallow land disposal.

Carbonate grout containment requires prior treatment of off-gas to absorb most of the $^{14}\text{CO}_2$ and $^{12}\text{CO}_2$ on an ion exchange resin. The carbon dioxide is then reacted with a grout slurry: $\text{CO}_2 + \text{Ca}(\text{OH})_2 \Rightarrow \text{CaCO}_3 + \text{H}_2\text{O}$. The stoichiometry gives $\sim 2\frac{1}{2}$ times the mass of the original carbon dioxide in the product waste. In practice, carbonation capacities range from 16% to 19% of the grout mass, and for the experimental solid grout samples studied even those values were not attained.¹⁴

Cryogenic separation of $^{14}\text{CO}_2/\text{CO}_2$ from off-gas, by cooling to induce successive phase changes in stream components, is an alternative to the hydroxide scrub column. Cryogenic separation requires reducing the off-gas temperature of 800-900°C to -27°C if the gas can be compressed to 300 psig. This would produce a pure liquid carbon dioxide; at normal atmospheric pressure carbon dioxide desublimates at -90°C or lower producing the solid "dry ice".¹⁵ Unless cryogenic isolation is applied to several components of the off-gas, handling pressurized liquid carbon dioxide or refrigeration sufficient to produce dry ice is not an efficient method for separation of just carbon dioxide.

Pressurized cylinders are not deemed reliable for the long term storage of the $^{14}\text{CO}_2/\text{CO}_2$. The carbon dioxide, once isolated, will require further treatment to establish a stable waste form. However, if the calcium hydroxide can be used in a column to capture the carbon dioxide out of the off-gas, the separating and grouting steps may be integrated to simplify the treatment process.

The desired waste form should contain the radioactive ^{14}C until natural decay eliminates any hazard to the biosphere from that waste. However, prospects for containment for the life of the ^{14}C isotope are not very good, and part of the price of containment includes handling of this additional low-level waste during processing and disposal. Eight to ten half-lives, or 45,840 to 57,300 years, are required to effectively eliminate the ^{14}C .

Leach testing of carbonate grout at room temperature with water for periods of up to 220 days with high flow conditions generated a cumulative release of less than 2% of the total ^{14}C present. Under acidic conditions, leach rates would be higher still.¹⁴ Assuming coating the grout with a sealant would reduce the leach rate to 1% per year, in 200 years only 13.4% of the ^{14}C would remain in the grout and only 1.02% of the original quantity would have decayed. More than 85% of the original ^{14}C would still escape to the environment.

Unless longer term containment can be assured, treatment only delays release to the environment by comparison to the long life of the ^{14}C . In view of the world carbon cycle, the only natural ways to isolate carbon from the earth's interchangeable carbon supply are incorporation in organic shale and rock within the land mass or deposition in the ocean sediment under the deep ocean.

Reaction of barium hydroxide hydrate with the carbon dioxide has been explored as an alternative to the calcium carbonate grouting. The barium carbonate product is a more stable compound than the calcium carbonate, and less volume of waste would be generated. However, barium is a hazardous waste as listed in 40 CFR 261.30, and combined with the carbon-14 the barium carbonate would be a mixed waste for which disposal is more expensive. The barium hydroxide application shows no appreciable advantage over direct release considering the costs of the process and disposal of the waste.¹⁶

Disposing of the carbonate grout in the deep ocean is the most promising treatment. The grout would sink to the ocean bottom rapidly, enabling dilution with more than 90% of the interchangeable carbon reservoir over the life of the grout without any undiluted surface exposure. However, production and handling of the grout present exposure risks prior to disposal which may counteract the benefit of any treatment, and ocean disposal becomes an international political concern.

Direct release of the $^{14}\text{CO}_2$ to the atmosphere is a more practical alternative because the atmosphere will greatly dilute the release and local exposure could be monitored and controlled. The 130 Ci of ^{14}C in this release is small compared to the 4 million Ci of ^{14}C in the atmosphere making treatment expensive compared to the minimal reduction in risk.¹⁷

Tritium:

The off-gas from the combustion of the entire inventory of graphite fuels will contain 0.000623 kg ^3H , or 6001 Ci (0.207 gmol), contributing 0.0057 mrem to the 10 mrem per year EDE limit at the INEL boundary assuming a bulk gaseous release. While this is less than one tenth of a percent of the 34,560,000 Ci present globally from cosmic radiation, release might produce local effects.

Tritium has a 12.3 year half-life; ^3H decays to ^3He which is both stable and inert. About 0.2 kg of tritium is produced each year in the upper atmosphere.¹⁸ Behavior of tritium in the environment depends on chemical form. As hydrogen gas, tritium gas (HT) disperses globally with little biological significance until oxidized to form tritiated water. The mean residence time of tritiated water in the lower atmosphere is about a week before precipitation and dilution with surface water; in some situations it can rain out in a day.

Tritiated water is readily absorbed into biological systems. Small amounts of tritiated hydrocarbons are present, but they are difficult to trace and large samples are required for low-level measurements because samples must be dried to remove free water and then burned before the tritium can be measured.¹⁹ Most of the 0.000623 kg of tritium present in the graphite fuel combustion off-gas will be tritiated water for comparison to the global tritium inventories below.

Inventories of Environmental Tritium¹⁹
(kg)

	<u>Pre 1950</u>	<u>1970</u>	<u>1982</u>
Total oceans	2.3	253	129
Continental waters	1.0	45	18
Atmosphere HTO	0.025	1	0.1
HT	0.0065	1.3	0.5
Hydrocarbons		0.2 (1975)	0.05
Totals	3.6	301	148

Total Bomb Test Releases	~ 600 kg
1 GWe Heavy Water Reactor at equilibrium	~ 4
1 GWe Fusion plant, expected	5-15

The argument for release of tritium is similar to that for ^{14}C . Like the carbon dioxide, the tritiated water vapor will rapidly disperse in air and will not concentrate, in a single body organ or anywhere else. HTO release is more of a health concern than $^{14}\text{CO}_2$ because HTO is more readily absorbed into the body. However, comparing the calculated annual exposure at the INEL boundary from release of the estimated quantities of tritium and carbon-14 present in these graphite fuels, the 0.0057 mrem from tritium is small compared to the 0.27 mrem from the carbon-14.

Unlike $^{14}\text{CO}_2$, tritiated water only needs to be isolated from the environment for 100 to 120 years for all but one thousandth of the original quantity to decay. If a cement similar to the carbonate grout can be used to contain the tritiated water, most of the tritium would decay before being released from the grout.

Some tritium control measure may be merited. Design for the GA/FRG pilot project shows a tritiated moisture adsorber bed of molecular sieve 3A. A decontamination factor of greater than 1000 is possible. Tritium escaping to the environment should be in the liquid form. Advantages of a liquid release over a gaseous emission are twofold: the exposure pathway would be limited to ingestion, not inhalation, and the release could be diluted immediately in a body of water.²⁰

Similar quantities of tritium must be handled in fuel processing plants worldwide, and although much of the tritiated water is scrubbed from the off-gas stream, some does escape to the environment. Most of the tritiated water can be removed with a molecular sieve or desiccant; adsorption on silica gel is one technique.

A wet scrub column will also tend to trap the tritiated water; recycle followed by appropriate disposal or containment would significantly reduce the amount of tritiated water in the off-gas, especially if further condensed by subcooling. This is an attractive possibility since the same scrub column can be used to clean up several other off-gas components at the same time.

The amount of tritium present is well within allowed release levels, but if treatment is desired the wet scrub is the best option. Once isolated as a liquid, tritiated water could even be released deep into a body of water where dilution and slow migration will insulate it from the biosphere until most of it decays away.

Iodine-129:

The off-gas from the combustion of the entire inventory of graphite fuels will contain 6.66 kg ^{129}I , or 1.18 Ci (51.6 gmo). Assuming a bulk release, this constitutes 0.128 mrem toward the 10 mrem per year limit at the INEL boundary. ^{129}I has a 15,700,000 year half-life, after which it decays to ^{129}Xe that is both stable and inert. Only ~40 Ci of ^{129}I are present from natural sources, with an additional 36 Ci introduced between 1945 to 1975 due to nuclear weapons testing. Hundreds of curies of ^{129}I are trapped in the spent nuclear fuels discharged in the U.S. All of the ^{129}I released will effectively remain in the environment forever. Although the risk from exposure to such a long-lived isotope is small, the quantity of ^{129}I anticipated compared to the amount of iodine naturally present in Southeast Idaho. ^{129}I should be contained and isolated from the biosphere as much as possible during conditioning.²¹

Iodine concentrates in the thyroid. In geographical areas deficient in natural iodine, significant short term doses to animals and humans are possible since ^{129}I will be scavenged by the thyroid. Long term doses will be due mostly to food from the mixed layer of the ocean. The ^{129}I will tend to migrate there whether it is originally released to the atmosphere, surface soil, or directly to the mixed ocean.²¹

The GA/FRG research program achieved decontamination factors of better than 1400 with both a silver nitrate impregnated silica gel and a silver-exchanged mordenite bed. In terms of silver use the silica gel was more cost effective. Preliminary integrated tests showed near complete SO_2 adsorption in the silver-exchanged mordenite. Also, in the presence of oxygen, virtually all CO will oxidize to CO_2 . Although SO_2 and CO are not major emission concerns, excessive loading on the iodine adsorber may reduce its effectiveness.⁹ The silver catalyzes the oxidation of CO to CO_2 which overneats

the bed. Platinum forms iodine compounds if used to oxidize the CO prior to iodine removal. A lead mordenite packed bed has been successfully used for iodine adsorption. Decontamination factors of 1000 were achievable and the lead bed did not overheat in the presence of CO.

Several wet scrubbing systems have been considered. Chromic acid has been tested for removal of ^{129}I . Decontamination by a factor of two to three was achieved, complicated by increases in discharge of ruthenium-106 and zirconium-95. Chromic acid is not recommended for this application.²² Caustic scrub solutions achieve decontamination factors of 100, but are unreliable for removal of organic compounds of iodine, and need to be operated at higher temperatures to be most effective. The Mercurex process uses a mercuric nitrate/nitric acid to form mercuric iodine complexes. Subsequent oxidation to the iodate followed by filtration completes the separation, but again the decontamination factor is only about 100. The Iodex process uses 20 M nitric acid, with decontamination factors of better than 10,000 possible; however, this generates a very soluble product that would require further treatment before final storage.

A methanol/water scrub solution has been suggested because of the solubility of sulfur dioxide, nitrogen oxides and stibine as well as the iodine in methanol. Methanol would also effectively dry the off-gas at the projected operation temperature, absorbing most of the tritiated water. The decontamination factor (DF) for such a process is unknown, but merits further investigation since the methanol could be cleaned up and recycled and is easier to handle than strong acids or bases. Scrub solution decontamination factors vary greatly depending on solution cleanup; a DF of 1000 was assumed for the methanol/water scrub because distillation should regenerate nearly pure methanol for recycle. Several successive steps have historically been used for management of iodine in reprocessing plants. The necessary decontamination may require multiple processes to best achieve the required decontamination factor of 1000.

Solid sorbents offer the advantages of a dry, comparatively noncorrosive waste form. Activated charcoal can achieve decontamination factors varying from 10 to 1000 depending on the exact conditions of use but is not used in spent fuel processing due to its flammability and poor durability in extended use. Silver-exchanged zeolites can achieve decontamination factors as high as 100,000, but are inhibited by exposure to acid, steam, H_2S , C_3H_8 , NO_2 and SO_2 . Less expensive silver-exchanged mordenites were developed to be more acid resistant, and decontamination factors of 1000 are possible. But mordenites may be poisoned by some organics and sulfur influences their capacity for iodine sorption. Silver nitrate impregnated silica gel is another option for low temperature, low relative humidity applications. It achieves decontamination factors of 1000, but introduces certain organics and high concentrations of NO_2 , and performance may be influenced by sulfur. Macroreticular resins have performed

similarly in experimental conditions for less expense; but they are not as effective in adsorption of anionic forms of iodine such as I^- and I_3^- .²³

Generally a wet scrub followed by a solid sorbent bed is considered the most effective combination. While nitrogen oxides (NO_x) are a major consideration in reprocessing dissolver off-gas streams, this off-gas will have very little NO_x in it. The methanol-water scrub process should be developed. The cold methanol-water scrub process can be followed by a cold solid sorbent bed. If the methanol-water cold scrub proves unworkable, the hot caustic scrub has been used in reprocessing applications. For the packed bed, a lead mordenite should be used rather than one of the silver catalyzed options to avoid overheating from the oxidation of carbon monoxide.

Radon-220:

Radon gas, primarily radon-220 (^{220}Rn), will also be present in the off-gas. Based on the amounts of ^{232}Th and ^{232}U precursors and a projected 4.5 block per day processing rate of the Fort St. Vrain and Peach Bottom spent fuels, less than 3.0×10^{-9} g, or about 3.0 Ci, of the ^{220}Rn can be expected to enter the off-gas system per day. The concentration of radon in the untreated off-gas is estimated at 1.07×10^6 pCi/L and must be reduced to less than 3 pCi/L prior to possible human exposure to comply with DOE Order 5400.5.⁵ ^{220}Rn rapidly decays to a series of highly radioactive daughters which can be trapped in the off-gas filters. The longer the residence of the radon in the off-gas system, the more the radon decays within the system enabling its daughters to be captured.

Radon isotopes are characterized by a short half-life, 56 seconds for ^{220}Rn . The main concern is the radiation emitted by decaying radon daughters.

Radon-220 Decay Series²⁴

Radon is the only one in the series that is a gas at ambient temperature and pressure; all of the others would be solids if they were present in enough quantity to form a solid. A 10 minute holdup in the off-gas system should give a decontamination factor of about 1000. With 20 minutes of holdup, the factor is over 10^6 . A 10 to 20 foot long column of synthetic mordenite at room temperature may be all that is needed to delay the gas by adsorption until enough decay has occurred. The decay products stick in the packed bed or on the downstream filters and are not released. The radon daughters are not present in great enough quantities to cause plugging problems, but moisture and hydrocarbons erode the efficiency of the mordenite bed. If residence time in the off-gas treatment system does not eliminate enough of the radon such a bed could be added. If moisture loading is a problem, two such holdup beds could be

added in parallel so that one can be regenerated while the other is in use.²⁵ If krypton-85 is to be recovered the radon will be held up long enough by that process that the needed decay will occur.

Krypton-85:

One other large component of the off-gas will be ⁸⁵Kr. About 0.902 kg of ⁸⁵Kr, or 343,000 Ci (10.6 gmol), will be released during the particle crushing and burning operations. After a 10.8 year half-life, ⁸⁵Kr decays to ⁸⁵Rb which is stable.

Krypton is not used or significantly absorbed in biological functions, and is mostly inert. It is soluble in nonpolar solvents like liquid carbon dioxide and freon and particularly in lipids, fats found in living cells. Only about 2.0 Ci of ⁸⁵Kr are naturally present in the earth's atmosphere. The atmosphere is the only reservoir for krypton; once ⁸⁵Kr is dispersed in the atmosphere it remains there until it decays. Although large quantities of ⁸⁵Kr produced in the nuclear fuel cycle have been released in spent fuel reprocessing, the dose from ⁸⁵Kr is still negligible by comparison with natural radiation sources.²⁶ A release of all of the ⁸⁵Kr in the Fort St. Vrain and Peach Bottom spent fuels would result in a 0.02 mrem radiation exposure at the INEL boundary in a year while the average exposure to natural sources results in 300 mrem per year.

Krypton-85 can be recovered along with the stable xenon (and trace amounts of radon) in the off-gas via cryogenic separation. This process was used in the Rare Gas Plant at the Idaho Chemical Processing Plant in the 1960s through the 1980s. The feed gas is cooled and then fed to a distillation column. Liquid nitrogen is added at the top of the column condensing most of the feed gas. When condensate at the bottom of the column contains enough rare gases it is transferred to the reboiler of a fractionating column. The nitrogen and a large part of the oxygen are distilled after each transfer. When enough liquid is prepared, fractionation is performed, isolating the krypton in the first fraction and xenon in the next. The main concern with this process is that radiolysis in the liquid phase in the reboiler can potentially generate ozone in concentrations high enough to result in an explosion.^{27,28}

Oak Ridge National Laboratory developed a process for krypton adsorption in liquid carbon dioxide (KALC process) specifically for removal of krypton from the burner off-gas generated in the reprocessing of HTGR (graphite) fuels. This process is similar to krypton absorption in freon, without the risks of radiolytically produced chlorine gas and potential detriment to the atmospheric ozone layer. The KALC process functions at pressures from 13 to 20 atm and at temperatures of -40°C to -20°C. An absorber-fractionator system absorbs the krypton and more soluble gases (Xe, Rn, etc.) into the liquid carbon dioxide and discharges the less soluble gases (O₂, N₂, CO, etc.) out the stack. A stripper-rectifier system then removes and concentrates the krypton from

that liquid carbon dioxide solution. Xenon will accumulate in the liquid carbon dioxide with recycle and can be recovered in an additional operation or discharged up the stack off the absorber.^{27,29}

The KALC process is the technology best suited to the graphite spent fuel burner off-gas application. The GA/FRG research program original design recommended the KALC process followed by a chromatographic separation of krypton from the carbon dioxide across a molecular sieve.⁹ Cryogenic separation is still a potential alternative, but the KALC process is more promising for this application and its development should be pursued first.

The krypton (and the xenon as well) is a commercially salable byproduct of spent fuel processing. Recovery of these products will depend on the sales, the economics of the recovery process, and the demand for the material. If either is to be recovered the radon will be held up long enough that it will decay. Since krypton is of little significance to biological systems, and market forces do not support its' recovery, preference may be to release it. Storage of gas cylinders for 100 years would allow for decay of all but one thousandth of the ⁸⁵Kr recovered, but the 0.0237 mrem contribution toward the 10 mrem annual limit at the INEL boundary in the event of a bulk release is small compared to the cost of separation and disposal of the krypton.

Conclusions and Recommendations:

The off-gas generated from the burning of the Fort St. Vrain and Peach Bottom fuels can be easily treated to meet the existing effluent criteria. If future criteria require that the emission of ¹⁴C and ⁸⁵Kr be more restricted, then there are treatments that can be applied to meet any reasonable criteria. Calculated concentrations of other projected contaminants exiting the proposed flowsheet treatments are extremely low, well below any reasonable future criteria. The concentrations indicated are low enough that some of the treatment is unnecessary and future cost savings will result from elimination of them from the flowsheet. Two scrubber-packed bed systems may not be needed. One may do the job. One sintered metal filtration system may also be adequate.

Further work should be concentrated on verifying the contaminant removal efficiency of the selected alternatives. The contract with BYU should provide the information needed on the semi-volatile fission products. There is enough information available in the literature on the use of packed beds. This information needs further review to determine applicability and more accurately project the operational efficiency. There is much literature information on scrubbing of radioactive off-gas as well, but not much on the combination of continuous cleanup and subcooling. Laboratory investigations should be conducted on the proposed subcooled scrubbing system because of the possible high decontamination factor and its potential industrial use.

Laboratory studies should be done at the ICPP on the subcooled scrubber and packed bed systems. A bench scale scrubber and packed bed system could be fabricated out of existing equipment.

Appendix
Material Balance Tables and
Calculations

TABLE III
Shaft Furnace - Whole Block
(Fluidized Bed with CO₂ Recycle)

	Min.	Avg.	Max.
To Primary Burner			
kg/day			
Carbon	391.5	477.	511.65
Combustion Gases	1232.074	1501.328	1610.283
Actinides	38.516	47.473	50.608
Silicon	14.774	18.	19.308
Sulfur	0.294	0.358	0.384
Mixed F.P.	2.222	2.739	2.92
Filter Blowback	73.924	90.08	96.617
&Transfer Gas			
To Particle Crusher			
kg/day			
Carbon	55.401	67.5	72.403
Combustion Gases	0.	0.	0.
Actinides	38.246	47.141	50.254
Silicon	14.774	18.	19.308
Sulfur	0.139	0.169	0.181
Mixed F.P.	2.207	2.72	2.9

TABLE IV

Shaft Furnace - Whole Block (Cont.)
(Fluidized Bed with CO2 Recycle)

	Min.	Avg.	Max.
To Particle Burner			
kg/day			
Carbon	55.398	67.497	72.4
Combustion Gases	185.203	225.829	242.13
Actinides	38.246	47.141	50.254
Silicon	14.478	17.64	18.921
Sulfur	0.138	0.169	0.181
Mixed F.P.	1.911	2.356	2.511
Filter Blowback	11.112	13.55	14.528
& Transfer Gas			

To Particle Proc. Scrubber

Carbon Dioxide	172.658	210.364	225.646
Carbon Monoxide	19.389	23.624	25.34
Particulate	0.014	0.017	0.018
Oxygen	3.538	4.315	4.626
Nitrogen	11.236	13.701	14.689
Mixed F. P.	0.296	0.365	0.389
SO2	0.277	0.337	0.362
Rare gases	0.296	0.364	0.388
I+F. P. Particulate	3.90E-04	4.82E-04	5.13E-04
H-3	1.85E-07	1.11E-06	2.38E-06
C-14	8.46E-06	4.13E-05	6.91E-05
Kr-85	1.77E-04	1.08E-03	2.55E-03
Ru-106	3.05E-15	2.10E-12	2.84E-09
I-129	4.32E-04	2.58E-03	6.68E-03
Cs-137	3.61E-04	1.83E-03	3.27E-03
Cs-134	2.42E-08	8.20E-06	1.04E-04
Actinides	4.79E-07	5.90E-07	6.29E-07

TABLE V

Shaft Furnace - Whole Block (Cont.)

	Min.	Avg.	Max.
To DOG Scrubber kg/day			
Carbon Dioxide	1220.166	1486.639	1594.631
Carbon Monoxide	137.024	166.949	179.076
Particulate	0.043	0.052	0.056
Oxygen	38.474	47.027	50.357
Nitrogen	85.119	103.821	111.298
Mixed F. P.	0.296	0.366	0.391
SO2	0.31	0.378	0.405
Rare gases	0.296	0.364	0.388
I+F. P. Particulate	3.83E-03	4.74E-03	5.08E-03
H-3	1.30E-09	7.79E-09	1.67E-08
C-14	8.46E-06	4.13E-05	6.91E-05
Kr-85	1.78E-04	1.09E-03	2.57E-03
Ru-106	2.15E-17	1.48E-14	2.00E-11
I-129	3.04E-06	1.82E-05	4.71E-05
Cs-137	2.55E-06	1.29E-05	2.30E-05
Cs-134	1.70E-10	5.78E-08	7.34E-07
Actinides	1.91E-04	2.36E-04	2.51E-04
CO Combustor O2	86.129	104.939	112.562
Exit Gas Stream			
kg/day			
Carbon Dioxide	1433.337	1746.364	1873.223
Carbon Monoxide	1.37	1.669	1.791
Particulate	4.30E-08	5.24E-08	5.62E-08
Oxygen	47.087	57.521	61.613
Nitrogen	85.119	103.821	111.298
SO2	3.10E-07	3.78E-07	4.05E-07
Rare gases	2.96E-01	3.64E-01	3.88E-01
I+F. P. Particulate	3.83E-09	4.74E-09	5.08E-09
H-3	1.30E-15	7.79E-15	1.67E-14
C-14	8.46E-06	4.13E-05	6.91E-05
Kr-85	1.78E-04	1.09E-03	2.57E-03
Ru-106	2.15E-23	1.48E-20	2.00E-17
I-129	3.04E-12	1.82E-11	4.71E-11
Cs-137	2.55E-12	1.29E-11	2.30E-11
Cs-134	1.70E-16	5.78E-14	7.34E-13
Actinides	1.91E-10	2.36E-10	2.51E-10

TABLE VI
Combination Compact-Particle Burner
Dual Scrubber

To Combination System

kg/day	Min.	Avg.	Max.
Carbon	55.401	67.5	72.403
Combustion Gases	185.301	225.95	242.259
Actinides	38.516	47.473	50.608
Silicon	14.774	18.	19.308
Sulfur	0.139	0.169	0.181
Mixed F.P.	2.222	2.739	2.92
Filter Blowback & Transfer Gas	11.118	13.557	14.536

To Primary Scrubber

Carbon Dioxide	172.666	210.375	225.657
Carbon Monoxide	19.39	23.625	25.341
Particulate	0.011	0.014	0.015
Oxygen	3.54	4.317	4.629
Nitrogen	11.242	13.708	14.697
Mixed F. P.	0.296	0.364	0.388
SO ₂	0.277	0.338	0.362
Rare gases	0.296	0.364	0.388
I + F. P. Particulate	8.91E-06	1.11E-05	1.18E-05
H-3	1.85E-07	1.11E-06	2.38E-06
C-14	1.20E-06	5.85E-06	9.78E-06
Kr-85	1.77E-04	1.08E-03	2.55E-03
Ru-106	3.06E-15	1.95E-10	1.22E-10
I-129	4.32E-04	2.58E-03	6.68E-03
Cs-137	2.48E-07	1.83E-03	3.25E-07
Cs-134	4.46E-12	8.20E-06	2.74E-09
Actinides	3.85E-07	4.75E-07	5.06E-07

TABLE VII

Compact-Particle Burner (cont.)

	Min.	Avg.	Max.
To DOG Scrubber kg/day			
Carbon Dioxide	172.666	210.375	225.657
Carbon Monoxide	19.390	23.625	25.341
Particulate	1.11E-08	1.36E-08	1.45E-08
Oxygen	3.540	4.317	4.629
Nitrogen	11.242	13.708	14.697
Mixed F. P.	0.296	0.365	0.391
SO2	2.77E-07	3.38E-07	3.62E-07
Rare gases	0.296	0.364	0.388
I+F. P. Particulate	6.54E-10	2.85E-09	6.97E-09
H-3	1.85E-13	1.11E-12	2.38E-12
C-14	1.20E-06	5.85E-06	9.78E-06
Kr-85	1.78E-04	1.09E-03	2.57E-03
Ru-106	3.06E-21	1.95E-16	1.22E-16
I-129	4.32E-10	2.58E-09	6.68E-09
Cs-137	2.48E-13	1.83E-09	3.25E-13
Cs-134	4.46E-18	8.20E-12	2.74E-15
Actinides	3.85E-13	4.75E-13	5.06E-13
CO Combustor O2	12.188	14.85	15.929
Exit Gas Stream			
kg/day			
Carbon Dioxide	202.832	247.129	265.081
Carbon Monoxide	0.194	0.236	0.253
Particulate	1.11E-14	1.36E-14	0.000
Oxygen	4.759	5.802	6.221
Nitrogen	11.242	13.708	14.697
SO2	2.77E-13	3.38E-13	0.000
Rare gases	0.296	0.364	0.388
I+F. P. Particulate	6.54E-16	2.85E-15	6.97E-15
H-3	1.85E-19	1.11E-18	2.38E-18
C-14	1.20E-06	5.85E-06	9.78E-06
Kr-85	1.78E-04	1.09E-03	2.57E-03
Ru-106	3.06E-27	1.95E-22	1.22E-22
I-129	4.32E-16	2.58E-15	6.68E-15
Cs-137	2.48E-19	1.83E-15	3.25E-19
Cs-134	4.46E-24	8.20E-18	2.74E-21
Actinides	3.85E-19	4.75E-19	5.06E-19

Calculations

Estimation of exposure at INEL site boundary based on release of volatile off-gas components:

$$130 \text{ Ci } ^{14}\text{C} \times 2.1 \times 10^{-3} \text{ mrem/Ci } ^{14}\text{C} = 0.27 \text{ mrem from } ^{14}\text{C}$$

$$22,000 \text{ Ci } ^3\text{H} \times 2.6 \times 10^{-7} \text{ mrem/Ci } ^3\text{H} = 0.0057 \text{ mrem from } ^3\text{H}$$

$$1.18 \text{ Ci } ^{129}\text{I} \times 1.09 \times 10^{-1} \text{ mrem/Ci } ^{129}\text{I} = 0.128 \text{ mrem from } ^{129}\text{I}$$

$$343,000 \text{ Ci } ^{85}\text{Kr} \times 6.89 \times 10^{-8} \text{ mrem/Ci } ^{85}\text{Kr} = 0.0237 \text{ mrem from } ^{85}\text{Kr}$$

The sum of the exposure from these components is 0.43 mrem.

Any influence from ^{220}Rn has been neglected because of the small physical quantity present and its short half-life; the radon decay daughters are expected to plate out or stick to filters within the off-gas treatment system. Mitigating factors are not available for ^{220}Rn since its release does not significantly influence the dose at the site boundary.

The concentration of radon in the off-gas stream was calculated as follows:

$$1.15 \text{ SCMM} \times 1000 \text{ L/m}^3 \times 1440 \text{ min/day} + 4.5 \text{ blocks/day} \times 2513 \text{ blocks} \\ = 924,000,000 \text{ L of Off-Gas}$$

$$1600 \text{ Ci } ^{220}\text{Rn} \times 10^{12} \text{ pCi/Ci} + 924,000,000 \text{ L} = 1.73 \times 10^8 \text{ pCi/L}$$

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