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Nuclear power growth spurs interest in fuel plant wastes

Off-gas treatment available for use where meteorological conditions preclude safe atmospheric dispersal

In the production of nuclear power, radioactive waste products are produced which require some treatment processing. The problem of waste management of the fission products is encountered primarily in the spent fuel reprocessing step of the nuclear fuel cycle. Under present practice, the fission product noble gases are released with the gaseous waste and discharged to the atmosphere where they are diluted to low enough concentrations at which exposure to the general population is negligible.

Many authors dealing with the disposal of radioactive gaseous wastes to the atmosphere have presumed that, eventually, the discharge of fission product noble gases would be prohibited during the reprocessing of nuclear fuel. If a 6 tonne (1000 kg.) per day uranium fuel processing plant were located on the Oak Ridge reservation, the estimated Kr-85 released from a 100 meter stack would expose the population seven miles downwind to about 120 millirem (mREM) per year. This dose is 70% of the maximum allowable whole body dose of 170 millirem (mREM) per year for the general population in unrestricted areas.

The potential radiation hazard connected with release of Kr-85 from the expanding fuel cycle industry has been studied in detail. The steady and continuous release of Kr-85 from a reprocessing plant is a different exposure condition than the sudden short-term release of fission products postulated for an accident at a reactor site. One of the main problems in studying this nuclear waste management is estimating the rate of atmospheric dilution of the relatively concentrated off-gas from fuel reprocessing plants. The major variables here are:

- Rate at which the waste is generated (the size of the reprocessing plant).
- Stack height.
- Local meteorology.
- Nature of the terrain.
- Time interval on which the dose is calculated.
- Downwind distance of the perimeter fence around the plant.

Eventually, the gases will be diluted in the atmosphere to negligible levels. However, if the off-gas comes in contact with the public for appreciable periods of time when it is above allowable concentrations, then special provisions may be required to make the operation safe.

Kr-85 generation, 1970–2000

The sudden spurt in the purchase of nuclear power reactors has increased the projections of the rate of waste radioactive gas generation in the next 15–30 years. The annual generation rate, annual release rate, and net accumulated inventory of Kr-85 have been projected for the years 1980 and 2000. The inventory of Kr-85 between 1970–2000 was calculated with the CURIE portion of the Radiological Safety Analysis Computer Code (RSAC). The CURIE code assumes that only U-235 atoms are fissioned. Twenty thousand megawatts (MWe) of installed power were used for 1970, and straight line interpolations were used for 1980 and from 1980–2000.

If Kr-85 is released from the spent reactor fuel approximately three years after the fuel is charged to the reactor, the annual release of Kr-85 in 1980 will be that amount produced in 1977, or approximately 48 megacuries (MCi.). Thus, the total amount of Kr-85 generated by the year 1980 will be 380 MCi., and this will grow to 6610 MCi. by the year 2000. However, the net accumulated inventory is 357 MCi. for 1980, and 4550 MCi. for 2000, because Kr-85 decays with a 10.6 year half-life.

Assuming that the fuel, 2.2% enriched U-235, is irradiated to 21,000 megawatt-days (MWD) per metric ton of uranium, and that the reactor operates at 80% load factor and 32% thermal efficiency, one tonne of spent uranium is produced annually for each 23 MWe of installed power. In 1980, therefore, an amount of fuel equivalent to 120,000 MWe will be reprocessed, equivalent to 5210 tonnes of uranium. Similarly, the fuel to be reprocessed in the year 2000 is the byproduct of 647,000 MWe of installed capacity, equivalent to 28,100 tonnes of uranium. If the reprocessing plants operate 300 days per year, then the average plant rate is 17.4 tonnes of uranium per day in 1980 and 93.5 tonnes per day in 2000. The processing rate in tonnes of uranium is less definite and less important than the curies (ci.) of Kr-85 released.

Dispersal of Kr-85 into the atmosphere, of course, will depend on the production rate at any reprocessing site, not on the total Kr-85 released in the U.S. Assuming that 9200 ci. of Kr-85 is produced per tonne of uranium, plants of 3, 6, and 15 tonnes per day capacity will release 27,600, 55,200 and 138,000 ci. Kr-85 per day, respectively. A one tonne per day reprocessing plant now is in operation, and a five tonne per day plant is under design. Our calculations are based on Kr-85 releases that bracket plant capacities of interest.

These calculations assume that only U-235 would be burned in light water reactors (LWR) of the present boiling water or pressurized water types. Calculations for Pu-239 content and for the change in Kr-85 generation with reactor type were made in order to estimate long-term uncertainties in the total generation of Kr-85. Since LWR will be the major reactor design for many years, the assumptions of Kr-85 generation probably will be valid to within 25% until the year 2000. In any event, the Kr-85 generation and release data should be adequate for the calculations on dispersal of waste treatment presented.

Siting of reprocessing plants

The principal problem in disposing of Kr-85 to the atmosphere is dilution of relatively concentrated off-gas to a safe concentration without overexposure to the populace. Kr-85 is essentially a beta radiation emitter, and its effect on man is by direct, whole-body exposure. The dilution of plant off-gas by the atmosphere is accomplished at varying rates by diffusion and mixing effects of the weather. For the case of the reprocessing plant, plans must be made for a steady release, day after day, of tens of thousands of ci. of Kr-85 per day.

Variations in the extremes of weather become very important in assessing the exposure conditions. The Atomic Energy Commission limitations for the release of radioactive gases are based on three different periods of exposure: one hour, one week, and one year. The conclusions on gaseous dispersion for one hour under the worst regime are generally limiting, since nearly every probable plant location experiences several hours of every conceivable weather regime. The seven-day limiting exposure period has a good probability of combining several regimes of weather, and usually will be less critical than the one-hour limit. For a one-year exposure, the specific weather conditions of a plant site must be used, since average weather conditions apply to the longer exposure periods.

The meteorological characteristics important in dispersing Kr-85, or any stack gas, are wind direction, wind velocity (especially calm conditions, i.e., mean wind speed = 0.22 m/sec.) fraction of time wind is in a sector or quandrant, and the stability class (regime) involved. These data should be taken at the height of the stack. Any additional dispersal capability required at a site will have to depend on such variables as nature of the terrain, taller stacks, longer downwind distances to the perimeter fence, and, possibly, the addition of separation processes to remove the necessary fraction of Kr-85 from the off-gas. The contribution of effluents from nearby nuclear industries also must be considered in any calculation of an integrated dose to the populace.

Maximum, ground-level concentration is affected significantly by the release height, and varies inversely as the square of the release height. Other factors—including effluent velocity and gas temperature—also influence the effective stack height.

Changes in terrain can affect plume concentration calculations markedly. A downwash can occur on the leeward side of a prominent ridge, or the plume can be trapped in a major valley circulation pattern. Another effect is noted when the plume travels over a large body of water whose temperature differs from that of the adjacent land mass. All of these effects should be considered for any specific chemical reprocessing plant site.

Dose criteria

Currently acceptable dose limits require a prospective reprocessing licensee to demonstrate that he will not subject a general population to radiation equivalent to more than 2 mREM per hour, 100 mREM per week, or 170 mREM per year, whichever occurs first. In most cases, the weekly dose is not a problem, since week-long periods of persistently bad diffusion with low windspeeds seldom occur. However, the hourly limit is easily exceeded when, for example, calm weather sets in and persists for an unreasonable length of time. For example, persistence curves for the site for National Reactor Testing Station (NRTS) illustrate that a calm could persist for a 10-hour period with a 1% probability and a 4–5 hour period with a 10% probability.

Kr-85 generation from nuclear power reactors in the U.S. (1967 estimate)

Year	1970	1980	2000
Installed nuclear power, MWe.	20,000	150,000	734,000[a]
Annual generation of Kr-85, MCi.	8	64	315
Annual Kr-85 release (3 yr lag) MCi.		48	278
Net accumulated Kr-85 inventory, MCi.		357	4,550

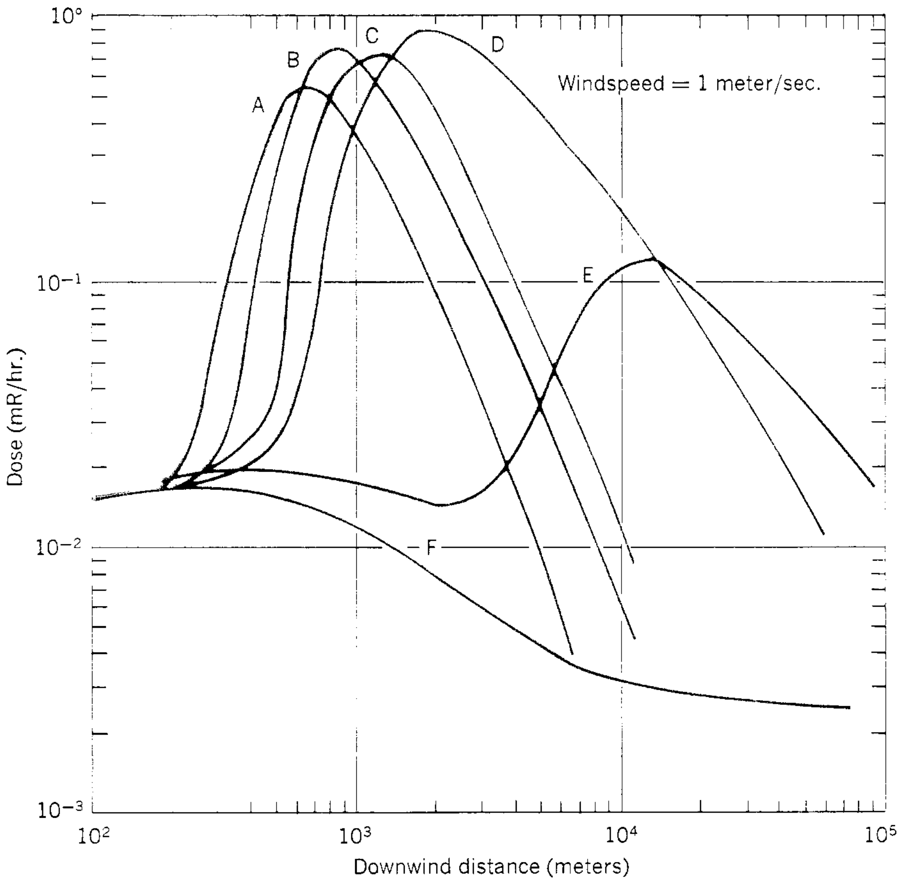
a Source: Atomic Energy Commission (1962).

Four site examples

The annual dose for a 22.5° sector has been calculated from the wind roses for four sites, for a prominent wind direction: NRTS; Indian Point, N. Y.; Oak Ridge National Laboratory, Tenn.; and Argonne National Laboratory (ANL).

The downwind distance beyond which the Kr-85 dose drops below 170 mREM per year has been calculated for various plant sizes at these sites.

Dose rates for various weather stability classes can be calculated and are composed of a minor gamma contribution calculated with the RSAC code and a dominating beta contribution proportional to the ground-level concentration. The stability classes follow the conventional meteorological range of strong lapse conditions for Class A to strong inversion for Class F. For the hourly dose rate curves, such as this set calculated for a 26,700 Curie-per-day Kr-85 release from a 100 meter stack, the general Gaussian diffusion equation holds. (To obtain dose rate other conditions, divide by appropriate wind speed.) The annual dose curves used were based on a crosswind-averaged form of the diffusion equation, applicable to 22.5° sectors. The Markee diffusion parameters were utilized for these calculations. Because these parameters apply to a smooth, sagebrush-covered terrain with unlimited vertical mixing, the dose rate values for stability Classes A and B will be low compared to actual values for those geographic locations where vertical mixed layer depths are characteristically shallow



Plants releasing 26,700 ci. per day would have a perimeter limit of 2000-3000 m. with a 100 meter stack, and with the exception for ORNL, no perimeter limit with a 200 meter stack. Doses calculated with this method compare favorably with corresponding reported values for the NRTS and ORNL sites. The dose curves will tend to underestimate ground level doses under those stability classes where mixing depths may be shallow. Mixing depths tend to decrease with a drop in the altitude of the site.

In the event that a plume travels over a considerable body of water before reaching a population center, the dose estimates could be low by a factor of 3. Although channeling of winds due to topography will show up in corresponding wind roses, the downwash effect on the leeward side of a prominent ridge will not. No quantitative value can be assigned to this effect.

Low velocities and atmospheric temperatures are typical of the plume emerging from the stacks of chemical reprocessing plants; hence, the effective stack height will be nearly the same as the actual stack height.

Excess release of Kr-85

Thus, the data show that a plant of small size can be located in an area of favorable meteorology with a stack of suitable height such that the atmospheric release would be unrestricted. If Kr-85 exposures cannot be reduced below permissible quantities, as in unfavorable weather at a large plant site, alternative choices of action might include:

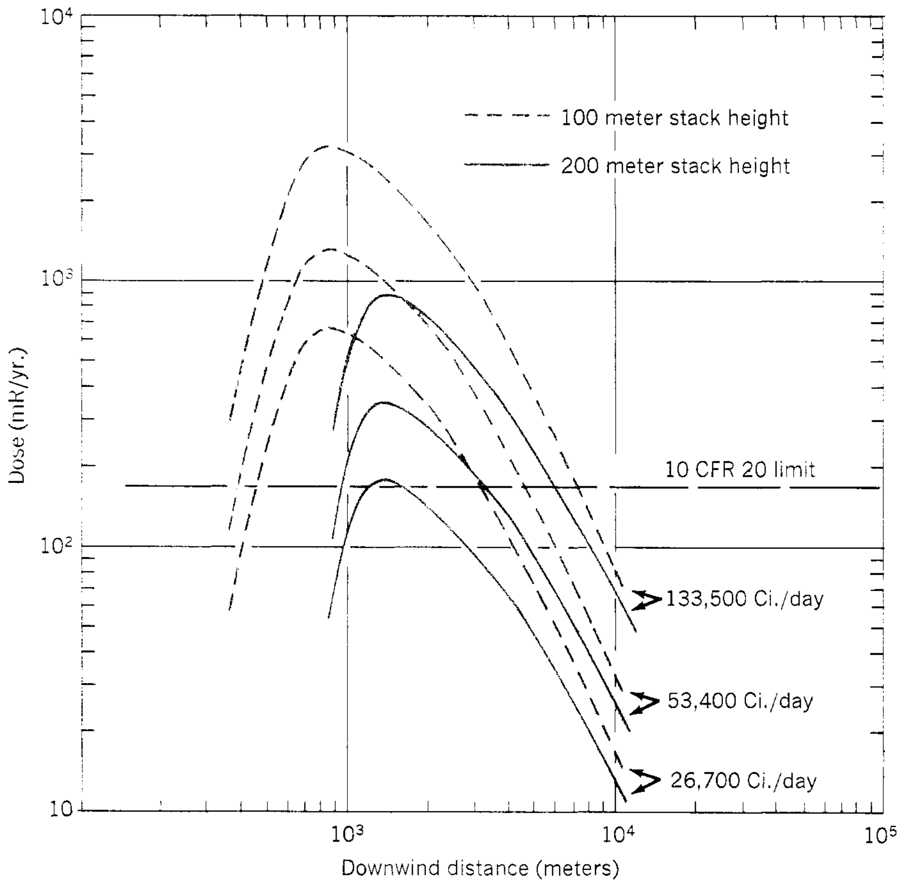
- Closing the plant during periods of restrictive meteorological conditions.
- Storing the plant off-gas during restrictive meteorological conditions.
- Disposing of off-gas to porous underground media for storage.
- Separating Kr-85 from the majority of the off-gas and storing it for future dispersal or for decay. Such action would be necessary only after the advantages of a taller stack, or of increasing the downwind distance to the plant perimeter, had been explored. A detailed technical and economic study of alternatives is necessary before making such a choice.

Approximately 99.9% of the Kr-85 generated in nuclear fuels will be released during the head-end fuel preparation step in the reprocessing of the spent nuclear fuel. Normally, fuel is stored from three to four months prior to reprocessing, which allows all radioactive noble gas isotopes except Kr-85 to decay. The Purex process utilizes nitric acid for the dissolution of uranium or uranium dioxide fuel. The off-gas from the dissolution step is composed of oxides of nitrogen, noble gases, steam, hydrogen, and great quantities of diluent air; its volume has been reported as 43,200 cubic feet per tonne of uranium. The average composition of the off-gas is 1.3% NO_2, 7.6% NO, 5.6% H_2O, 14.4% O_2, and 71.7% N_2. The Kr-85 content is 9200 ± 2300 ci. per tonne of uranium.

In general, temporary closure of the reprocessing plant to avoid the release of Kr-85 during stagnant air conditions would be unduly expensive.

Plant size, stack height determine perimeter dose limits

Annual dose rates from continuous release of Kr-85 from plants of various sizes at four different sites have been calculated. Perimeter limits for the sites are determined from curves such as the one shown, which is for the tower shielding facility at the Oak Ridge National Laboratory. Here, the prominent wind direction is from the southwest 22½° sector; original data for the curves were taken at the 96 meter level. The table summarizes the result for the four sites, and presents the downwind distance, in meters, at which the annual dose rate is 170 mREM



Downwind distance (m.) to reach average annual dose of 170 milliREM

Site	Wind direction	100 m. stack height, Kr-85 Release, Ci. day: 26,700	53,400	133,000	200 m. stack height, Kr-85 Release, Ci. day: 26,700	53,400	133,000
NRTS[a]	NE	2200	3500 m.	5800 m.	Unlimited	Unlimited	4000 m.
Indian Pt., N.Y.	S	2100	3400	6000	Unlimited	Unlimited	4400
ORNL (TSP)[b]	SW	3200	4800	7400	1500	3200	6100
ANL[c]	SSW	Unlimited	1500	2700	Unlimited	Unlimited	Unlimited
Hourly limiting case for Class D Calm[d]	ANY	4900	8800	10,600	Unlimited	6200	10,400

[a] National Reactor Testing Station, Idaho.
[b] Oak Ridge National Laboratory, Tenn.
[c] Argonne National Laboratory, Ill.
[d] Mean windspeed 0.22 m./sec.

These plants are designed to run continuously, and daily overhead costs are high. A 3 tonne per day plant will lose $90,000 revenue for each day of down time, an amount equivalent to $30,000 per ton of processed uranium. When weeks of unfavorable weather are encountered, other means of reducing Kr-85 release are necessary.

The possibility of storing all of the off-gas from the dissolution step during stagnant weather conditions depends on the duration of the storage period. If only a few days at a time are involved, the use of a gas-holder at a slightly subatmospheric pressure or compressing the gas into cylinders might be economically feasible. However the storage volume required for a 3 tonne per day plant is approximately 60,000 s.c.f. per day, which would require 1,800,000 cubic feet of storage at atmospheric pressure for a 30 day period, or a proportionally smaller volume at higher pressures. Since this gas will be very corrosive because of the formation of nitric acid from residual NO_2 gas, stainless steel will be needed for the construction of the gas holder. Together with shielding for radioactive Kr-85, the gas holder will be a relatively expensive item, costing more than $1 million. The amortization of this tank plus operating costs must be compared with alternative methods of treatment.

The underground storage of radioactive gases has not been demonstrated, and much development work is needed. But the disposal of off-gas containing radioactive noble gases to porous underground media for long-term storage is being considered at the NRTS, ORNL, Pacific Northwest Laboratories, and Harvard School of Public Health. Capital costs of $228,500 and $347,500 were estimated for storage wells 1000 feet and 4500 feet deep, respectively. Operating costs, of

Several processes available for Kr-85 off-gas treatment

Process	Kr recovered	Development status	Advantages	Disadvantages	Capital cost ($1000's)	Operating cost per metric ton of uranium (dollars)
Room temperature adsorption on charcoal beds or molecular sieves	99%	Bench scale work completed; scale-up feasible	•Simple operation; •Accepts dilute feed gas	•Large-volume adsorber beds needed; charcoal can ignite; •Strong oxidizing gases must be removed prior to adsorption	$1000	$170
Low temperature adsorption on charcoal beds or silica gel	99%	Development completed; plant in operation	•Small-volume beds; •Uses dilute feed gas	•Charcoal can ignite; •Oxidizing gases, CO_2 and H_2O must be removed; •High consumption of liquid nitrogen; •Adsorbers must withstand high pressure; •High operating cost	1000	150
Cryogenic distillation	98%	Developed and operated on a significant scale	•Low capital and operating cost	•Explosion hazard in forming and concentrating ozone	800	100
Liquid extraction	98%	Bench scale completed; large scale demonstration needed	•Uses Freon-12 with low refrigeration costs; •No explosion hazard; •Might eliminate pretreatment	•Absorber column operates at 200 p.s.i.g.; •Volume of extractant is large if operated at 15 p.s.i.g.	800	100
Clathrate precipitation	Unknown	Laboratory studies only; no engineering	•Kr-85 is collected as a storable solid	•Needs concentrated feed gas; •Crystallization step slow	n.a.	n.a.
Permselective membranes	99%	Bench scale work only; engineering tests needed		•Membranes sensitive to chemicals; •High power costs	1500	200
Thermal diffusion		Little pertinent data available			n.a.	n.a.

All of the processes for the removal of Kr-85 from off-gases from nuclear fuel reprocessing plants require some pretreatment of the dissolver gas. The cost data presented are for a plant which processes three tonnes per day of uranium. Comparative costs for alternatives to off-gas treatment for the control of radioactive stack emissions are available. For instance, disposal of the gases to ground reservoirs would require a capital cost of $345,000 for a plant of the same size, and operating costs would range between $75-100 per tonne of uranium. Facilities for the storage of the off-gas during meteorological conditions unfavorable for safe atmospheric dispersal would cost $1 million and operating costs could run to $100 per tonne of uranium, depending on the length of storage time. The most drastic alternative, shut-down of the plant during unfavorable conditions, would involve a loss of $15,000-30,000 per tonne of uranium capacity for the duration of the shutdown

$2.00 per 1000 cubic feet (STP) of gas stored add an additional $75–100 to the cost of processing one tonne of spent uranium. Capital costs for a shallow well with high (1000 c.f.m) flow rates have been estimated at $210,000. Thus, underground storage of Kr-85 could be competitive with storage in gas holders.

Another action that might be required is the separation and recovery of Kr-85 from the off-gas. Several processes of this type have been demonstrated on a plant scale, and others are in the development stage. Separation processes offer the opportunity to recover Kr-85 for commercial use as well as to prevent its dispersal to the environs; other treatment methods lose control of the Kr-85 by disposing it to the atmosphere or the ground.

Several recent literature surveys update the removal of radioactive noble gases from off-gas streams. No practical process is available for the removal of xenon and krypton from the high-velocity off-gas stream (millions of c.f.m.) from a gas-cooled reactor, but several processes have been developed and installed for low velocity streams such as from fuel reprocessing plants.

Methods of Kr-85 removal from gas streams can be classified into the following processes:

- Room temperature adsorption on charcoal, silica gel, or molecular sieves.
- Low temperature adsorption on charcoal or molecular sieves.
- Cryogenic distillation and scrubbing.
- Preferential extraction by liquids.
- Trap as clathrates.
- Permselective silicone rubber membranes in a diffusion cascade.
- Thermal diffusion.

Comparison of off-gas treatment

One basis for comparing these processes is their efficiency in recovering Kr-85, at the p.p.m. concentration level, in dissolver off-gas during the recovery of spent nuclear fuel.

All processes except the clathrate process can remove more than 90% of Kr-85. In actual practice, the recovery might be considerably lower than the reported 98–99%. For instance, a variable gas flow to the gas separation plant probably would contribute to lower krypton recovery.

All these processes require some gas pretreatment to remove oxides of nitrogen, CO_2, hydrocarbons, and H_2. In some cases, even O_2 must be removed.

The disadvantages for some of the processes have been overcome in certain cases by suitable design or process operation. For example, at the Idaho Chemical Processing Plant, the cryogenic distillation process has been demonstrated and found to be superior to the low temperature carbon bed process. Comparison of the liquid extraction and permselective membrane processes with the other processes are difficult, since neither has been developed nor demonstrated to the same degree as the first three. However, the liquid extraction process looks very promising as a conventional chemical engineering operation, and should prove competitive with the cryogenic distillation process. The operating pressure of the Freon-12 absorber is 14 atmospheres, comparable to the 10 atmospheres used in the permselective membrane process.

Cost comparisons

The final criteria for choosing an off-gas treatment process are the capital equipment cost and contribution to the unit cost of recovering uranium and plutonium from spent nuclear fuel. Unfortunately, at this time, sufficient information is not available to make a detailed cost comparison. But the approximate cost of various waste treatment procedures can be compared with the cost of increasing the stack height. For example, recent estimates of \$800,000 for a 400 foot stack and \$2 million for a 700 foot stack have been given. Thus, if a plant were to be built with a 400 foot stack, and, upon examining the dose curves, it was found that a 700 foot stack would be necessary to bring the exposures to a desired level, the extra cost of \$1.2 million could be balanced against one of the alternative treatment methods.

In any event, the incremental cost of treating the Kr-85 waste by any one of the above methods should not add more than \$200 per tonne to the cost of reprocessing fuel, which is approximately 1.0% of the total reprocessing cost. This increment is a negligible factor in the cost of nuclear power; however, the need for off-gas treatment is recognized as an important consideration during the planning for a reprocessing plant to avoid time-consuming plant modifications after the plant is built and in operation.

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