# ENVIRONMENTAL ANALYSIS OF THE URANIUM FUEL CYCLE

## PART III - Nuclear Fuel Reprocessing

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U.S. ENVIRONMENTAL PROTECTION AGENCY
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# FOREWORD

The generation of electricity by light-water-cooled nuclear power reactors using enriched uranium for fuel is experiencing rapid growth in the United States. This increase in nuclear power reactors will require similar growth in the other activities that must exist to support these reactors. These activities, the sum total of which comprises the uranium fuel cycle, can be conveniently separated into three parts: 1) the operations of milling, conversion, enrichment, fuel fabrication and transportation that convert mined uranium ore into reactor fuel, 2) the light-water-cooled reactor that burns this fuel, and 3) the reprocessing of spent fuel after it leaves the reactor.

This report is one part of a three-part analysis of the impact of the various operations within the uranium fuel cycle. The complete analysis comprises three reports: The Fuel Supply (Part I), Light-Water Reactors (Part II), and Fuel Reprocessing (Part III). High-level waste disposal operations have not been included in this analysis since these have no planned discharges to the environment. Similarly, accidents, although of potential environmental risk significance, have also not been included. Other fuel cycles such as plutonium recycle, plutonium, and thorium have been excluded. Insofar as uranium may be used in high-temperature gas-cooled reactors, this use has also been excluded.

The principal purposes of the analysis are to project what effects the total uranium fuel cycle may have on public health and to indicate where, when, and how standards limiting environmental releases could be effectively applied to mitigate these effects. The growth of nuclear energy has been managed so that environmental contamination is minimal at the present time; however, the projected growth of this industry and its anticipated releases of radioactivity to the environment warrant a careful examination of potential health effects. Considerable emphasis has been placed on the long-term health consequences of radioactivity releases from the various operations, especially in terms of expected persistence in the environment and for any regional, national or world-wide migration that may occur. It is believed that these perspectives are important in judging the potential impact of radiation-related activities and should be used in public policy decisions for their control.

Comments on this analysis would be appreciated. These should be sent to the Director, Criteria and Standards Division of the Office of Radiation Programs.

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# PART III - NUCLEAR FUEL REPROCESSING

## INTRODUCTION

Economic analyses performed both by the AEC and by commercial investors have concluded that the economical generation of electric power by nuclear plants requires that valuable isotopes of uranium and plutonium be recovered from spent reactor fuels for re-use in new fuel elements. Recovery of uranium and plutonium involves mechanical chopping of spent fuel elements into small pieces and placing them in an acid dissolver to separate the spent fuel from its metal cladding prior to chemical separation of useful isotopes from waste products by some adaptation of the Purex solvent extraction process. This operation results in the controlled release of fission products and other radioactive waste materials which accumulate in the elements during burnup in the power reactor. Since the quantities of these waste materials are large, considerable care is taken to assure that the fuel elements maintain integrity through the cycle in the reactor. In essence, therefore, the fuel reprocessing step breaks this carefully constructed barrier and, as a consequence, represents the main source of radioactivity from the nuclear power industry which could potentially enter the environment.

Many complex technological, environmental, and biological factors are involved in judging the impact of radioactivity on the environment. It is the purpose of this analysis to examine these factors with respect to fuel reprocessing requirements over the next several decades in terms of the potential public health and environmental risks involved.

The main objectives of this review are twofold: (1) to determine the population health risks and the cost effectiveness of waste controls relating to these risks for the present and future nuclear fuel reprocessing industry, and (2) to document the data base and techniques for assessing the environmental impact of the fuel reprocessing industry. A number of considerations are involved in accomplishing these objectives. These include forecasts of fuel reprocessing requirements through the year 2020, a detailed analysis of effluent control systems, the environmental transport of radionuclides, and considerations of the resultant doses and health effects. The data base and techniques used for these analyses are presented in detail in the appendixes of this report.

These studies are based on the performance of a hypothetical reprocessing plant having characteristics typical of plants now under construction. This model plant was used to evaluate potential reductions in health effects for various controls that limit the release of radionuclides to the environment. Control system costs were considered as well as their relation to total plant costs. These analyses were then expanded to project the impact of all the nuclear fuel reprocessing operations expected in the United States up to the year 2020. Doses and world wide health effects have been estimated on the basis of projected nuclear power production and anticipated radioactive effluent control techniques. Only the contribution from United States reprocessing facilities and only the most significant radioisotopes, in terms of total effect and persistence in the environment, have been considered in this analysis. Major changes in normal operating procedures, such as shorter cooling time (storage between fuel discharge from the reactor and initiation of reprocessing) should significantly affect local and regional doses but would not appreciably affect national and world impact.

Three reprocessing plants, with an estimated combined annual capacity of 2,700 metric tons of uranium (MTU), are expected to be in operation in the United States in the next several years. The fuel reprocessing industry is expected to increase along with nuclear power growth to about 50 to 60 plants with reprocessing capacity of about 80,000 MTU per year projected by the year 2020.

Up to the present, only one commercial facility has been operational, but is now shut down for modification. This is the Nuclear Fuel Services plant in West Valley, New York, with a capacity of 1 tonne of uranium per day. Processing of spent nuclear fuel was initiated in 1966 and continued with sporadic interruptions until December 1971 when the plant was shut down to permit expansion of processing capability which will increase plant capacity to 3 tonnes per day. The plant is located on a 3,300 acre tract owned by the State of New York in Ashford Township, Cattaraugus County, New York (Ref. 1). Buffalo, New York, is 26 miles from the plant and several of its southern suburbs are within a 25 mile radius. Considerable dairy farming and other agricultural activities are conducted close to the site. New York State conducts a comprehensive monitoring program around the facility including daily raw milk surveillance (Ref. 2 and 3). Extensive studies have been conducted at this facility by EPA and its predecessor organizations (Ref. 4, 5, 6, and 7).

Two other plants are under construction. The Midwest Fuel Recovery Plant is a tonne per day plant located at Morris, Illinois, adjacent to the Dresden Nuclear Power Station on privately owned property (Ref. 8). The General Electric Company, which has received a permit to store spent fuel and process unirradiated fuel, owns and operates the plant. Operations with irradiated fuel from nuclear power reactors will probably be started during the summer or fall of 1973.

The closest population center is Joliet, Illinois, 14 miles from the plant. Part of the city of Chicago is included within a 50-mile radius of the plant, thus significantly increasing the local population of concern. Offsite concentrations must be evaluated in terms of a multiple source since the three Dresden reactors are in close proximity.

The Barnwell Nuclear Fuel Plant is designed to reprocess 5 tonnes of uranium fuel per day (1,500 tonne per year), and is located adjacent to the Savannah River Laboratories (SRL) in Barnwell County, South Carolina, on privately owned property (Ref. 9). Construction of this facility was begun during the spring of 1971, and the projected completion date is 1975.

Augusta, Georgia, which is 31 miles from Barnwell, is the nearest population center. The population within 50 miles of the facility is about 500,000 or a factor of 10 below the corresponding population at Midwest. Additional site characteristics of these three facilities are presented in table 1.

## TABLE 1

## Site Characteristics of Nuclear Fuel Reprocessing Plants

| | NFS | MIDWEST | BARNWELL |
|---|---|---|---|
| Capacity | | | |
| MTU/day | 1 (3) | 1 | 5 |
| MTU/year | 300 (900) | 300 | 1,500 |
| Population within | | | |
| 10 miles | 21,409 | 25,000 | 7,100 |
| 25 miles | 293,711 | 303,460 | 65,209 |
| 50 miles | -- | 6,300,000 | 601,084 |
| Population Center (10 CFR 100) | Buffalo<br>(532,759)<br>26 miles | Joliet<br>(67,000)<br>14 miles | Augusta<br>(70,600)<br>31 miles |
| Maximum Off-site Annual Average $\chi/Q$ (sec/m$^3$) | $1x10^{-6}$ (1)<br>($2.2x10^{-7}$) (2) | $2.8x10^{-8}$ (3)<br>($1.1x10^{-7}$) (2) | $5.7x10^{-8}$ |

(1) Technical Specifications for NFS (Ref. 1)
(2) Adapted from ORNL-4451 (Ref. 11)
(3) Dresden stack use assumed

The Western Interstate Nuclear Board has been assessing the need for a fuel reprocessing facility in the Western region. It has concluded that such a facility will be required in the late 1970's but may be profitable during the middle 1970's (Ref. 10).

## EFFLUENT CONTROL TECHNOLOGY

Current practices and effluent control measures in the nuclear fuel reprocessing industry are not totally indicative of the workload and performance to be expected in new plants coming online during the period discussed in this report (1970-2020). Because newer designs will differ substantially from the first commercial facility, Nuclear Fuel Services, presently available performance data are of limited use in predicting environmental releases. The two newer plants, the Midwest Fuel Recovery Plant at Morris, Illinois, and the Barnwell Nuclear Fuel Plant at Barnwell, South Carolina, are far enough along in their design and construction to provide some idea of what the cost and expected performance characteristics of new plants may be. Both of these plants will utilize the recycling of liquid wastes so that in theory the only radioactive liquid discharges will be from spills, accidents, or leaks. The tritium present in the fuel will be released as a stack gas. This approach is in contrast to the NFS facility which uses a system designed to release low-level radioactive liquid wastes into the environment.

Waste control systems under construction for use in fuel reprocessing plants and their costs are described in appendix B. The design of NFS and the two plants under construction calls for the release of radioactive krypton-85 as stack gas, although the owners of

Midwest and Barnwell have indicated a willingness to add krypton removal equipment when it is considered practical (Ref. B-38 and B-39). Removal efficiencies of at least 99% are expected for such systems, although an efficiency of 99.9% has been claimed by some vendors (Ref. 12). Other effluent control systems to be incorporated in these plants will include iodine removal systems, which are expected to retain more than 99.9% of the iodine-129 and iodine-131, and particulate filters which should reduce releases of the actinides (particulate forms) by a factor of $10^3$ greater than the decontamination which is effected by the chemical separation process. While the operating experience at NFS confirms the effectiveness of particulate filters (Ref B-41), the anticipated efficiency of silver zeolite which provides backup to the aqueous scrubber iodine removal systems has not been verified in operational situations. The experience at NFS with a different but modern iodine recovery system has not been as good as orginally predicted (Ref. B-40); this equipment is now being replaced with a silver zeolite system. The radionuclide releases used in this report are estimates based on current knowledge of the capability of waste control technology. Technological innovation in waste control systems may reduce projected releases, while maintenance problems in aging equipment may result in increased environmental contamination.

## RADIOLOGICAL IMPACT OF A REPESENTATIVE PLANT

The assessment of the local, regional, or worldwide radiological health impact that may result from operation of a particular source or distribution of sources of radioactive pollutants is dependent upon a number of assumptions. The calculational models used are generally categorized as follows:

1. Source term models.
2. Environmental transport models.
3. Dose models
4. Risk models.

Source term modeling includes assessments of pollutant generation rates, inventories, and physical and chemical characteristics and release rates to the hydrologic or atmospheric carrier.

The environmental transport model permits an estimate of the media (air, water, food chains, etc.) concentrations at a particular point in the space-time continuum. These models considered all important pathways from the source to the receiver.

The dose model allows an estimate of the energy deposition and biological effectiveness in a biological system that results from exposure to the media concentrations. It considers such variables as ingestion rates, in-vivo distributions and biological half-life, and energy deposition in critical organs.

The risk model provides for estimation of biological effect resulting from doses due to ionizing radiation.

A useful approach to evaluating the potential environmental impact of fuel reprocessing plants and related control costs is to consider a single plant which is representative of current technology and design.

Such a representative plant has been assumed to have a capacity of 5 tonnes per day, or an annual capacity of 1,500 tonnes over an operating lifetime of 40 years. The fuel mix to be processed was assumed to consist of equal amounts of uranium and plutonium fuel. Of the residual waste product and fissionable material in the fuel, only the following specific radionuclides are considered in this analysis: krypton-85, tritium, iodine-129, iodine-131, plutonium-239, and other actinides. The release of other nuclides into the environment is anticipated to be less than one part in $10^8$ and will not produce health effects comparable to those produced by the nuclides considered here (Ref. D-8), although the situation for ruthenium is unclear at present.

Projected amounts of the radionuclides in spent fuel are given in table A.2 through A.4, appendix A. Liquid radioactive releases are assumed to be insignificant, and all environmental releases are assumed to be via the air pathway. The population within 80 km of the model plant was estimated to be $1.5 \times 10^6$ people by the year 1980 and was projected to double during a 40-year plant operating life (see appendix D). Using these assumptions, annual dose rates from the various nuclides were calculated (appendix D) for individuals located 3 kilometers from the plant, and both annual and total population doses, measured in person-rem, were determined for: (1) the population within 80 km of the plant, (2) the total United States population excluding those persons residing within the 80 km zone, and (3) the world population excluding the United States. These doses are listed in table 2. For tritium, most of the United States population dose results from

## TABLE 2

### Projections of Average Annual Population Dose from a 5 MT/Day Nuclear Fuel Reprocessing Plant

| Radionuclide Source [1] | Critical Organ | Average Organ Dose from One Year's Release for 1980 Startup<br>mrem/yr. @ 3 km | man-rem/yr. Regional | man-rem/yr. U. S. | man-rem/yr. World (less U.S.) |
|---|---|---|---|---|---|
| Kr-85 | Whole-body | 0.38 | 24 | 520 | 8,100 |
| | Lung | 0.75 | 47 | 1,000 | 16,000 |
| | Skin | 13 | 790 | 17,000 | 270,000 |
| | Gonads | 0.50 | 14 | 300 | 4,700 |
| H-3 | Whole-body | 3.2 | 200 | 3,700 | 1,100 |
| | Gonads | 3.2 | 100 | 1,800 | 570 |
| I-129 | Thyroid-infant | 1.4 | 2.3 | 2 | - |
| | Thyroid-adult | .4 | 27 | 85 | - |
| I-131 | Thyroid-infant | 13 | 20 | - | - |
| | Thyroid-adult | .8 | 53 | - | - |
| Actinides | Lung | 1 | 64 | 400 | - |

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[1] Decontamination factors used are 1.0 for Kr-85 and H-3, 1000 for the iodines and $10^9$ for the actinides.

nationally deposited tritium rather than from the worldwide distribution of tritium. However, most of the United States population dose from krypton-85 is due to the worldwide distribution of this isotope.

Local and regional population doses could vary considerably due to differences in the population distributions and meteorological conditions around specific plants as well as variations in efficiency of effluent control equipment. However, estimated national doses are probably conservative. The projected world doses are probably correct within a factor of five unless decontamination factors change by large amounts.

The estimated health effects (in terms of cancer induction and/or mortality) for a single 5 MTU/day reprocessing plant operating over 40 years are given in table 3. These values are based on the population doses in table 2 and dose-effect conversion factors presented in appendix C. The dose-effect conversion factors are based on a recent study performed by the National Academy of Sciences and use the assumption of a linear non-threshold dose-effect relationship. This assumption is considered prudent for decision making. The health effects were projected on the basis of the total dose irreversibly committed by environmental releases, and include the effects of extremely long-lived radionuclides for the first 100 years. The individual risk calculation assumes that an individual resides 40 years at a location 3 kilmeters from a reprocessing plant and also obtains his food and water from this location.

## TABLE 3

## Projections of Total Health Impact from a 5 tonne/Day Nuclear Fuel Reprocessing Plant

| Radionuclide | Critical Organ | Estimated Induced Health Effects for 40 Years of Operation | | | | |
|---|---|---|---|---|---|---|
| | | Individual @ 3 km | Regional Population | United States Population | World Population | Total |
| Kr-85 | Whole-body | $6.0\times10^{-6}$ | 0.38 | 8.3 | 130 | 140[1] |
| | Lung | $1.5\times10^{-6}$ | 0.095 | 2.1 | 32 | 34[2] |
| | Skin | $1.5\times10^{-6}$ | 0.095 | 2.0 | 32 | 34[3] |
| | Gonads[5] | $6.0\times10^{-6}$ | 0.17 | 3.6 | 57 | 61[2] |
| H-3 | Whole-body | $5.2\times10^{-5}$ | 3.3 | 59 | 18 | 80[1] |
| | Gonads[5] | $5.2\times10^{-5}$ | 1.2 | 22 | 6.9 | 30[2] |
| I-129 | Thyroid-infant | $7.4\times10^{-7}$ | .012 | .024 | - | .04[4] |
| | Thyroid-adult | | .02 | .12 | - | .14[4] |
| I-131 | Thyroid-infant | $4.5\times10^{-6}$ | .12 | - | - | .12[4] |
| | Thyroid-adult | | .04 | - | - | .04[4] |
| Actinides | Lung | $2.0\times10^{-6}$ | 0.13 | 0.4 | - | 0.5[2] |

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1. 50% Mortality
2. Very high Mortality
3. Low or Zero Mortality
4. Probably less than 25% Mortality
5. Genetic effects only.

About two-thirds of the health effects are estimated to accrue to the world population outside the United States, but for this group, the individual dose rates are extremely small (much less that one millirem per year). The regional population group incurs about 25% of the total effects, but receives larger individual doses than experienced by the world population group.

## CUMULATIVE RADIOLOGICAL IMPACT OF THE INDUSTRY

In addition to determining the impact of individual plants, the radiological impact of the entire United States nuclear fuel reprocessing industry was assessed. This total industry impact assessment considered the total accumulation of the long-lived isotopes and the health effects which can be attributed to them. A determination was made of the total environmental buildup of long-lived environmental radionuclides resulting from operations of the United States industry through the year 2020. These accumulations are shown in figure 1 and represent the estimated cumulative environmental inventories of tritium, krypton-85, iodine-129, and plutonium-239. These inventories determine the magnitudes of doses and of future health effects resulting from such cumulative environmental contamination.

Health effects attributable to the presence of these radioisotopes in the environment were considered in terms of the cumulative health effects that will be caused in the future due to release of these isotopes during one year of operation of the entire industry, as well as for the accumulated inventories shown in figure 1.

Figures 2 through 5 represent the estimated health effects that will be committed by the environmental buildup of certain long-lived radionuclides if the industry is allowed to operate through a given year at the plant decontamination factors typical of current design (1.0 for

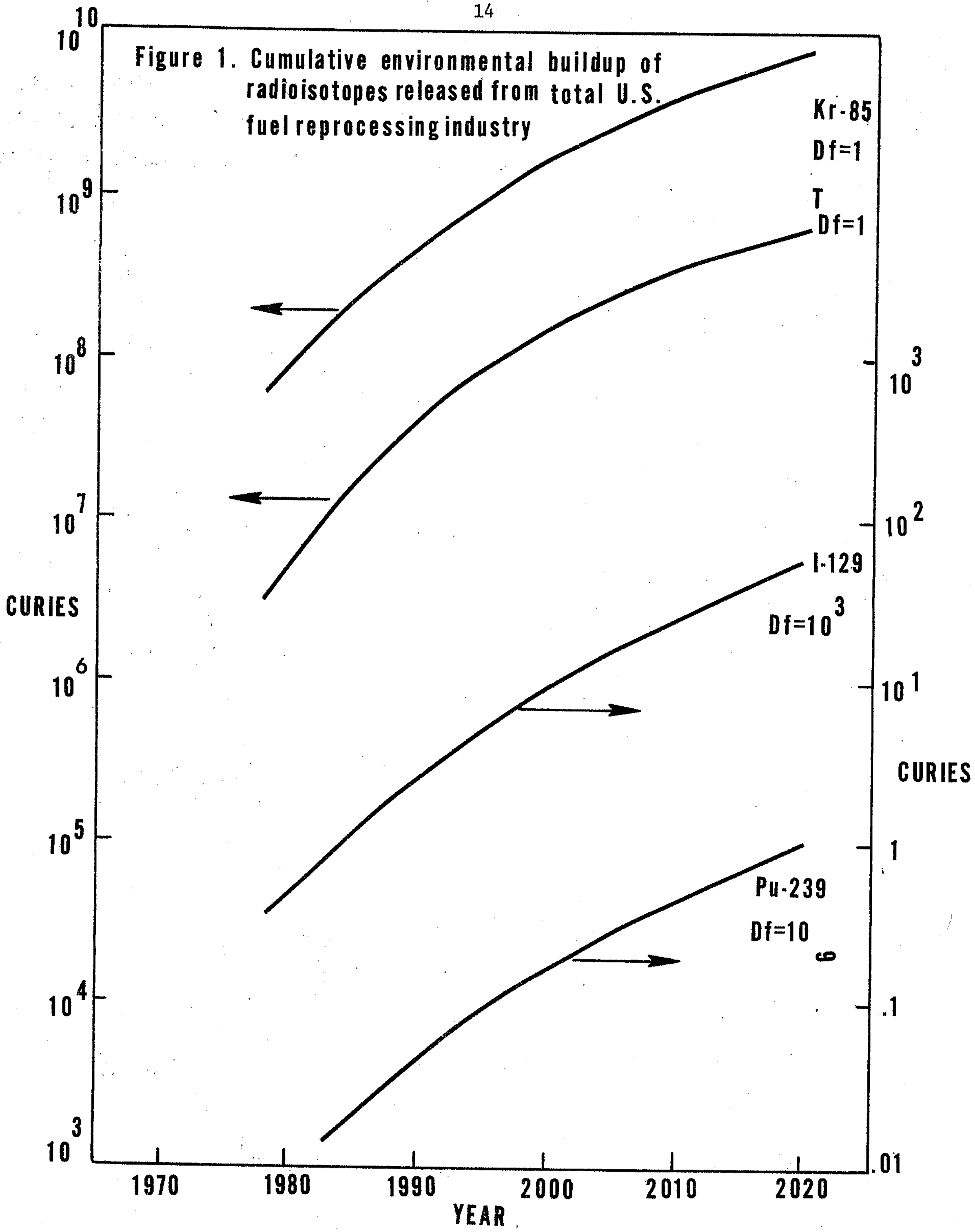


Figure 1. Cumulative environmental buildup of radioisotopes released from total U.S. fuel reprocessing industry

Figure 2. Estimated past and future health effects committed by tritium releases from the United States fuel reprocessing industry

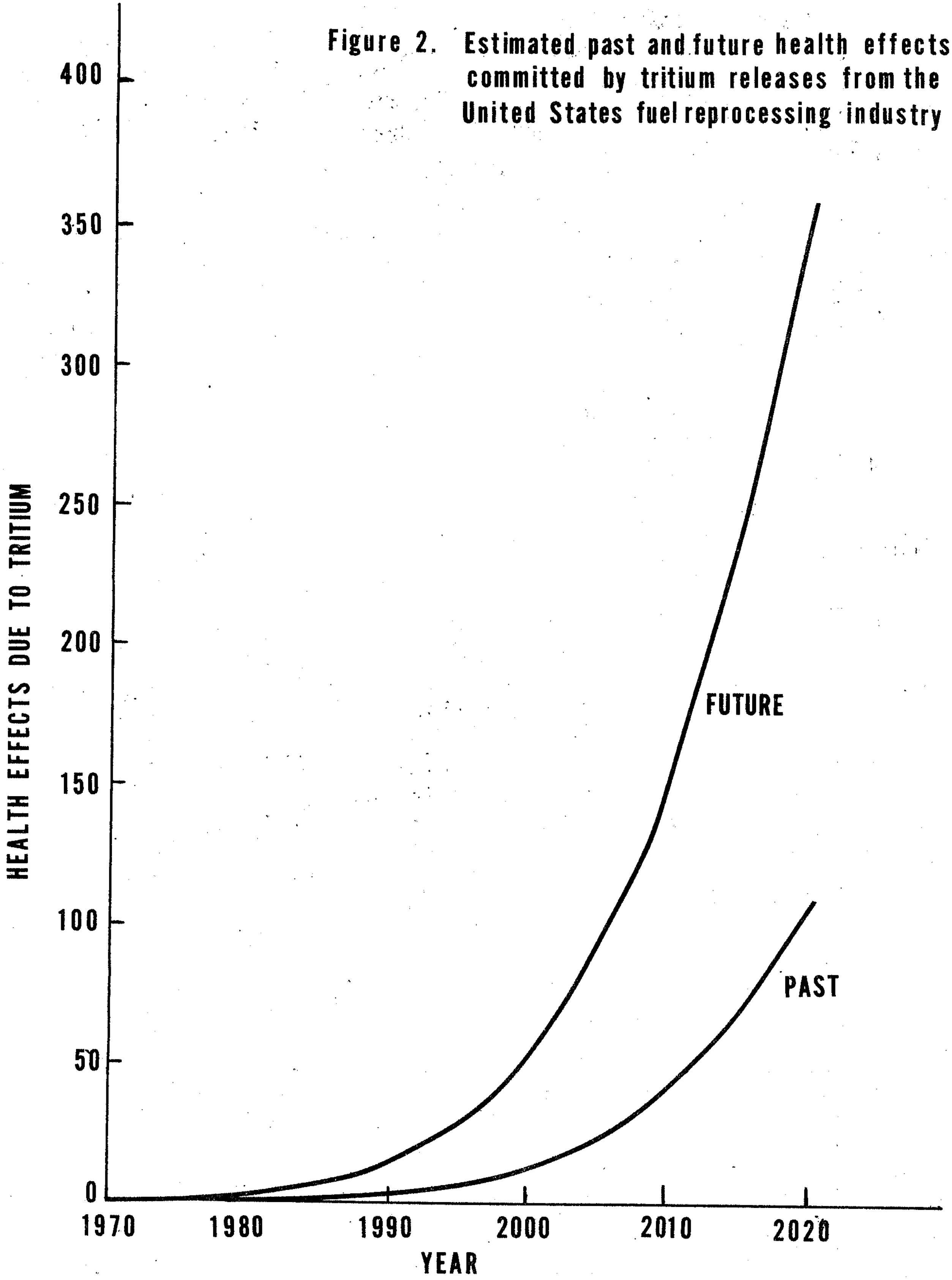


Figure 3. Estimated past and future health effects committed by krypton-85 releases from the United States fuel reprocessing industry

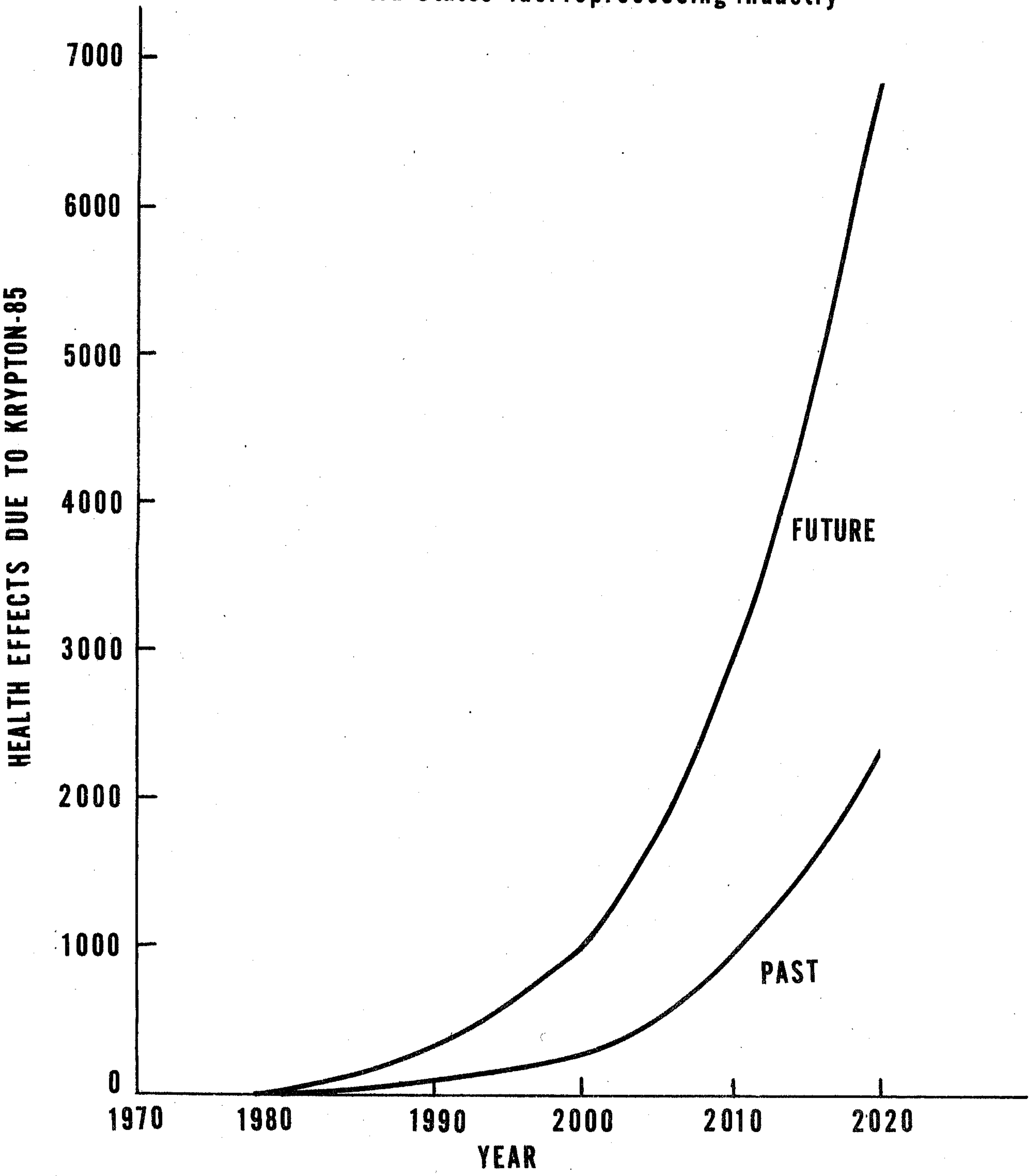


Figure 4. Estimated past and future health effects committed by iodine-129 releases from the United States fuel reprocessing industry

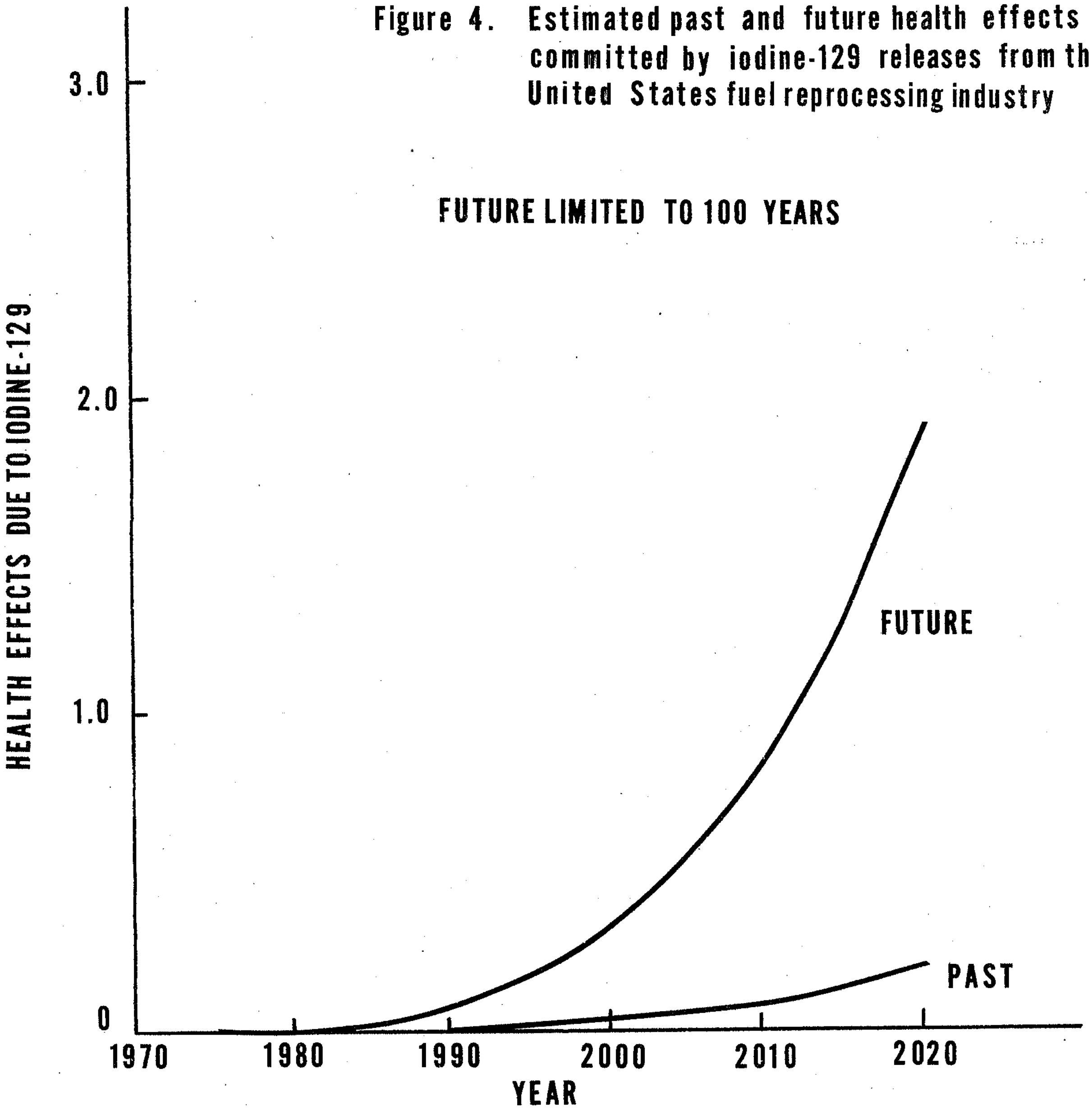
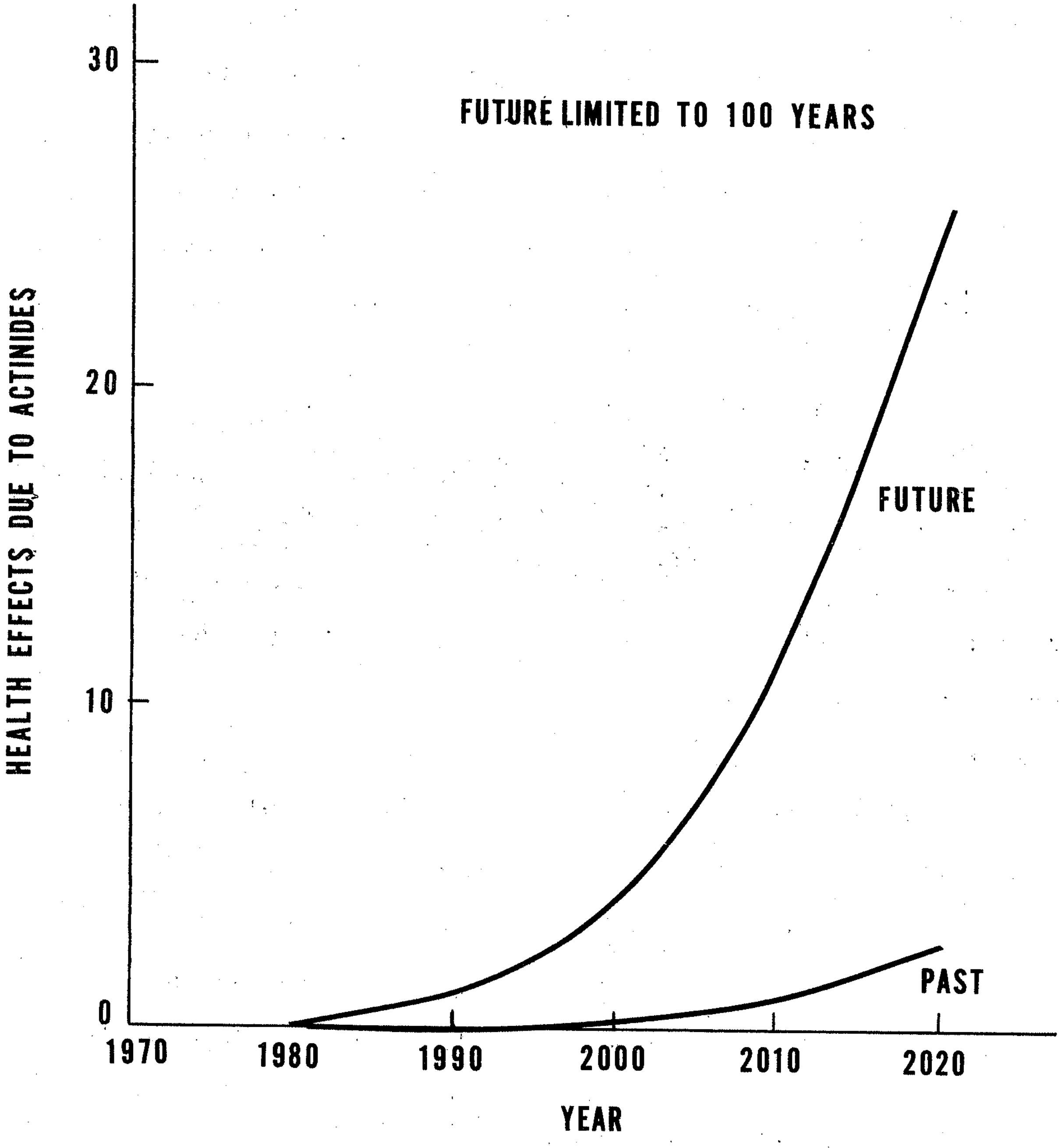


Figure 5. Estimated past and future health effects committed by actinide releases from the United States fuel reprocessing industry



tritium and krypton-85, $10^3$ for iodine-129 and $10^9$ for the actinides). For example, in figure 2 for tritium, if all tritium is released from fuel reprocessing plants with a decontamination factor of 1.0, then by the year 1990, the graph shows that three estimated health effects will have been committed by exposures received prior to 1990 and nine estimated health effects will be caused by exposures beyond 1990 from the quantity of tritium already in the environment in 1990. By the year 2020, 105 health effects will have been committed by past exposures to tritium and 250 estimated health effects will be caused by future exposures to past releases.

Future health effects for tritium and krypton-85 are estimated assuming the complete decay of the quantities present. For iodine-129 and the actinides ($^{238}$Pu, $^{239}$Pu, $^{240}$Pu, $^{241}$Pu, $^{241}$Am, $^{244}$Cm) only the first hundred years of exposure beyond the time of interest are considered for estimating health effects. The calculational techniques used are described in appendix D.

These curves demonstrate a rapid change in the environmental impact of the fuel reprocessing industry, especially after the 1980's. They indicate that if reprocessing plants continue to release radioactive material, especially krypton-85 and tritium, at current levels of emission (which are well below current regulations), an environmental burden of radioactive material will accrue which presumably could result in a significant number of avoidable health effects. Iodine-129 and the actinides have extremely long half-lives, and could impose additional health risks for future generations if they remain in the biosphere beyond the 100 years assumed for this analysis.

## ECONOMICS OF HEALTH EFFECTS REDUCTION AND EFFLUENT CONTROL

The following economic considerations apply to a single representative 5 tonne/day fuel reprocessing plant. Industry totals, as applicable, are obtained by multiplying by the number of projected plants (appendix A).

The fractional cost of fuel reprocessing in terms of total energy cost was derived. At present, the value of plutonium is approximately \$7-8/gram while the value of uranium, in the form of oxide, is about \$.02/gram. For a typical 1,000 MW(e) nuclear power plant, costs of all aspects of the fuel cycle from mining through waste disposal represent about 18% of the total costs of power (Ref. D-16). The reprocessing portion alone represents about 7% of the total costs of the fuel, or 1.3% of the cost of power generation (Ref. D-16). The capital investment for the Allied-Gulf Company's Barnwell Fuel Recovery Plant (5 tonne/day capacity for $UO_2$ and $UO_2$ + $PuO_2$ fuels) is approximately \$80 million. A plant of this size is capable of processing fuel from approximately 40 to 50 power reactors, each of which cost \$300 to 400 million. Since the only reason for fuel reprocessing is to support electric power generation by reactors, it is apparent that neither reprocessing nor substantial emission control cost could significantly alter the cost of power production.

Operating costs for a 5 tonne/day plant have been estimated to be about \$13 million per year when operating at full capacity. Of this, approximately $3 million is for labor. State and local taxes, insurance and interim replacement can give rise to an estimated additional annual expense of about $2.4 million. The capital costs associated with environmental studies, research, and construction is reported to approach 30% of the capital costs of the plant, much of which has already been incorporated in plants appropriate for reprocessing fuels used in the uranium cycle. For the plutonium cycle, the level of investment for environmental considerations is anticipated to be about twice that for processing of uranium-cycle fuel. Present effluent treatment capital costs are estimated to be approximately 6% of the total capital costs.

Table 4 summarizes, for a model plant, the estimated health effects to the world's population attributable to 40 years of operation and the total costs of waste control systems for the four isotopes considered in detail by this analysis. These total costs were computed assuming an effective interest rate of 24%, a debt lifetime of 20 years and the operating costs of the system over the projected life of 40 years. A 7.5% discount factor was used to estimate the present worth of these costs. From these values, the cost effectiveness of the individual component systems for control of emissions have been calculated. The health effects reduction as a function of system cost is displayed in figure 6. From these data, the overall cost effectiveness of combined systems is generated and is displayed in figure 7 for health effects to the world's population.

TABLE 4

Summary of Health Effects and Costs of Emission Controls

for a 5 tonne/yr Fuel Reprocessing Plant

| Radionuclide | Removal System | Total Cost (present worth) | Health Effects Averted | Δ Effects/$\$10^6$ |
|---|---|---|---|---|
| Krypton | Cryogenic Distillation | $ 11.5 million | 270[a] | 23.5 |
| | Cryogenic Adsorption | 12.1 million | | 22.3 |
| | Freon Adsorption | 6.4 million | | 42.2 |
| Tritium | Voloxidation (est) | 31 million | 110[b] | 3.5 |
| Iodine | Caustic Scrubber | 3.5 million | 303[c] | 86.6 |
| | Silver Zeolite | 1.85 million | 34 | 18.7 |
| Actinides | HEPA Filters | 1 million | 530[d] | 530 |
| | Sand Filter and HEPA FILTER | 2.1 million | | 260 |

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a ∿ 60% mortality
b ∿ 63% mortality
c Probably less than 25% mortality
d ∿ 100% mortality

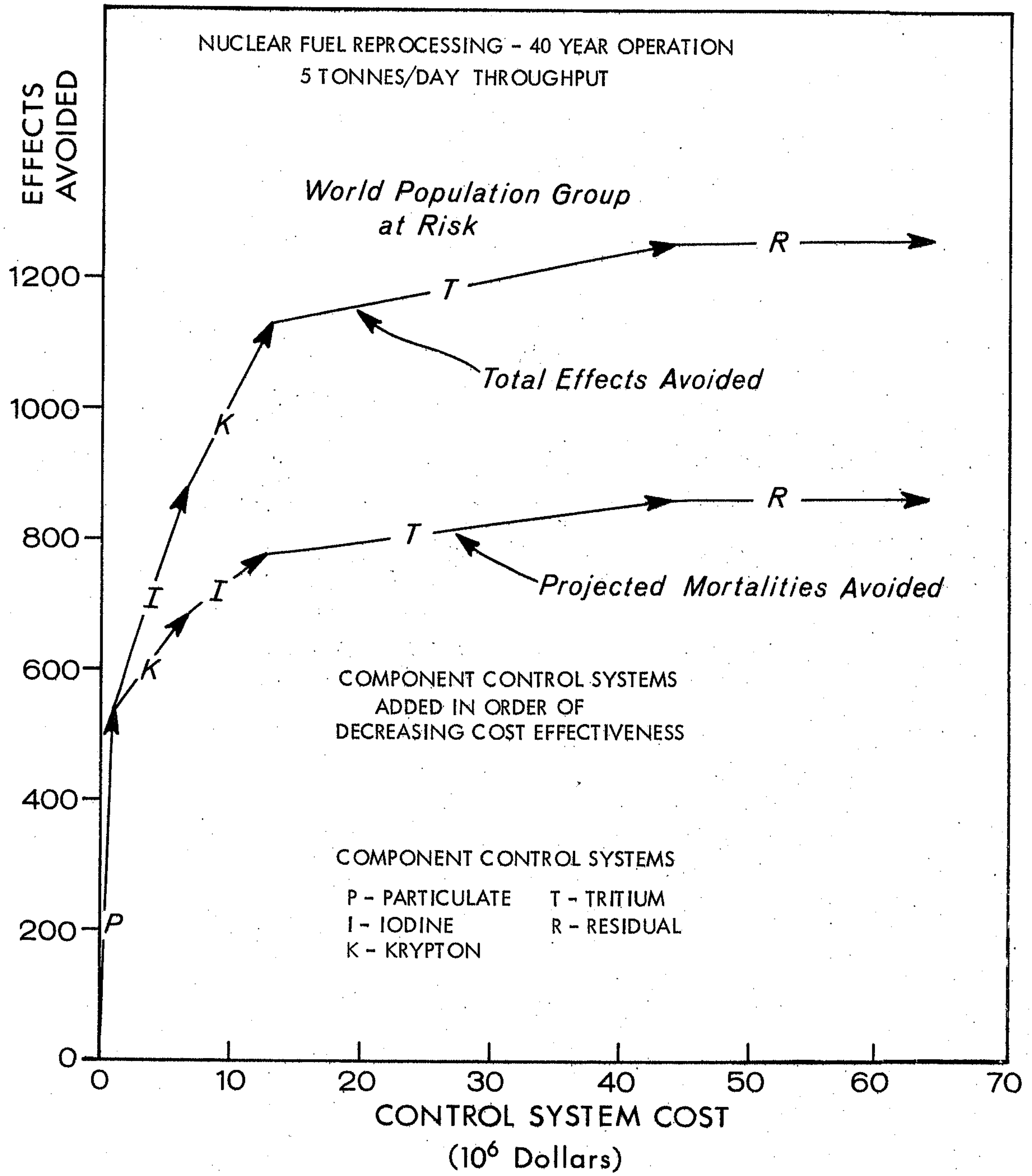

FIGURE 6. RISK REDUCTION vs CONTROL SYSTEM COST

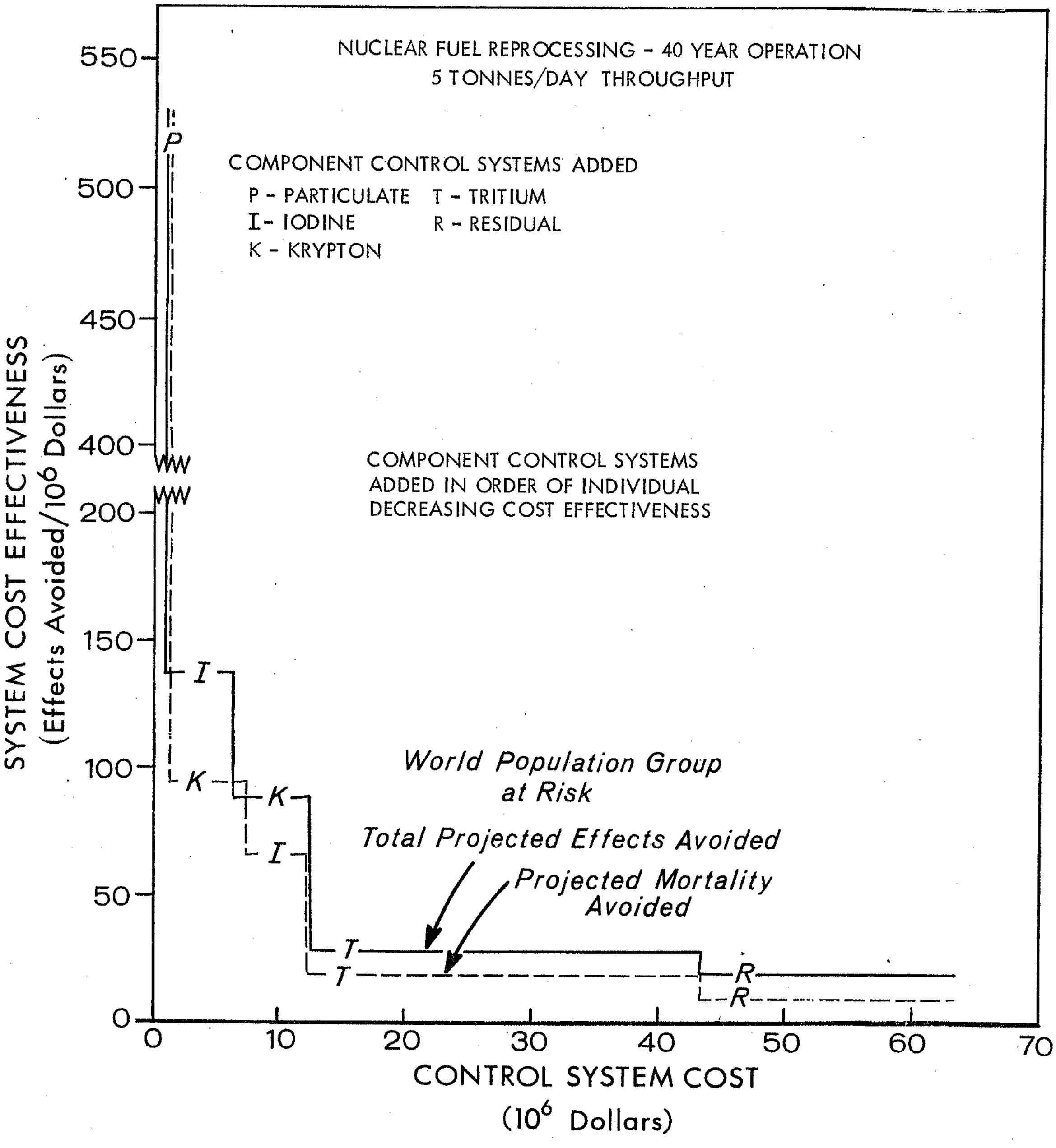

FIGURE 7 - CONTROL SYSTEM COST EFFECTIVENESS vs CONTROL SYSTEM COST

Certain waste product removal systems are already incorporated in present plant designs. These include provisions for filtration of particulates, including the actinides, and for reduction of iodine emissions. As is shown by the figures, krypton removal systems are almost equally as cost effective as control systems for iodine which are currently incorporated into plant designs when viewed in terms of the worldwide population. Tritium removal systems are less cost effective, on the same basis.

Some of these conclusions may be modified as the underlying bases are changed. For example, if the calculations are based on mortality rather than total health effects, the relative cost effectiveness of iodine and krypton removal systems are interchanged. Similarly, if the calculation is based on regional effects rather than world effects, the cost effectiveness of all systems except that for control of actinides is sharply reduced. These results are described in more detail in appendix D.

## SUMMARY AND CONCLUSIONS

The foregoing analysis of the potential environmental impact of the fuel reprocessing industry, and of the feasibility of minimizing this, has involved consideration of:

1. projection of nuclear power demand and associated spent fuel inventories expected to be reprocessed,

2. the present and potentially available technology for fission product removal including estimates of costs based on initial cost and operating expense,

3. the distribution throughout the environment of certain radionuclides released during normal operation and the resultant doses to the regional, national, and world populations, and

4. estimates of the statistical relationships of population exposure for specific radionuclides and the number of health effects expected to be associated with these.

The major conclusions derived from these considerations are as follows:

1. the fuel reprocessing industry, as an intergral part of the entire nuclear power industry, can be treated as a separate entity for purposes of evaluating its contribution to the overall release of radionuclides to the environment,

2. the quantites of certain radionuclides which may be released, if industry growth projections are substantially correct, are large. Consequently, the reprocessing step represents the point in the nuclear fuel cycle where the consequences of release of the long-lived isotopes, such as tritium, krypton-85, iodine-129, and plutonium 239 should be carefully considered,

3. the consequences of the buildup of very long-lived radionuclides (such as iodine-129 and plutonium-239) from the fuel reprocessing industry can produce cumulative and irreversible environmental levels which can be projected to cause adverse health effects on a national and worldwide scale. The consequences of the release of the shorter-lived isotopes, while possibly more severe on a short-term scale, are more easily reversed by effective control techniques,

4. control technology exists to reduce emissions of these materials. During the near-term future, all the tritium and krypton-85 produced during the fission process will be released to the environment during the reprocessing stage. Some control currently is applied for iodine and plutonium releases. Control technology to achieve confinement of krypton is essentially developed and some reduction for nearly all radioisotopes would appear feasible on a long-term basis,

5. removal of plutonium and other actinides is the most cost effective method of health risk reduction. Next in order of effectiveness are systems for removal of krypton and iodine which are about equal from a cost effectiveness standpoint. Tritium control technology is least cost effective at the present time, but future developments should be pursued to alleviate this problem,

6. available data allows an estimate of the incremental additional cost to the cost of nuclear production of electricity from the imposition of waste control systems at fuel reprocessing facilities to be about 0.1 percent of the total present cost to the producer of electricity,

7. evaluation of the total environmental impact of radioactive effluents requires a consideration of dose commitments beyond those delivered immediately in the site area, and, because of the long life of those materials in the biosphere, must include the exposures to national and world populations, beyond those delivered in the year of release, and

8. although the estimated health impact from the operation of a single 5 tonne/day reprocessing facility is relatively small, extrapolation of the industry as a whole over the next 50 years of operation indicates that long-term cumulative effects may be quite large.

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# APPENDIX A

# SPENT NUCLEAR FUEL RADIOACTIVITY FORECASTS

## I. INTRODUCTION

As a starting point in the assessment of the radiological impact of nuclear fuel reprocessing plants on the general population, an estimate of the total quantity of radioactive materials present in spent fuels produced by nuclear electric power generation must be obtained.

These estimates are based primarily on the projected electric power demand and on the fraction of that demand expected to be safisfied by nuclear plants. To some extent, they are also contingent on the projected number of various reactor types resulting in differing amounts of the various radionuclides produced. The annual reprocessing radioactive waste and product inventories for specific nuclides are forecast through the year 2020.

## II. SIZE AND SCOPE OF THE U.S. INDUSTRY

Electrical power demand forecasts for the United States, and the fractional amount expected to be produced by the nuclear industry through the year 2020 as used in this study, are summarized in table A.1 (Ref. A-1 and A-2).

The isotopic composition of nuclear fuels is expected to change over the next several decades. Presently uranium-235 is the most widely used reactor fuel material. However, the expected increase in energy demand, coupled with the inefficient utilization of the fissionable material by reactors as currently designed, would deplete the available low-cost natural uranium resources, which contain only about 0.7% uranium-235, by the

## TABLE A.1

## Estimated U.S. Fuel Reprocessing Requirements

(Adapted from References A-1 and A-2)

| Year | Nuclear electric generation:GW(e) | Tonnes of fuel discharged annually* | | | | | Number of 5 tonne/day reprocessing plants required |
|---|---|---|---|---|---|---|---|
| | | LWR-U | LWR-PU | LMFBR | HTGR | TOTAL | |
| 1970 | 2.6 | 25 | 0 | 0 | 0 | 25 | 1 |
| 1975 | 40 | 700 | 90 | 0 | 0 | 790 | 1 |
| 1980 | 110 | 1900 | 500 | 1 | 1 | 2,400 | 2 |
| 1985 | 220 | 2700 | 2600 | 0 | 100 | 5,400 | 4 |
| 1990 | 420 | 3700 | 3800 | 480 | 2,420 | 10,400 | 7 |
| 1995 | 650 | 4100 | 4100 | 2,600 | 6,600 | 17,400 | 12 |
| 2000 | 1000 | 3700 | 3800 | 11,500 | 7,800 | 26,800 | 18 |
| 2005 | 1360 | 3700 | 3700 | 20,600 | 10,000 | 38,000 | 26 |
| 2010 | 1780 | 4300 | 4400 | 32,800 | 10,000 | 51,500 | 35 |
| 2015 | 2220 | 5300 | 5400 | 43,800 | 10,000 | 64,500 | 43 |
| 2020 | 2700 | 6100 | 6100 | 58,000 | 8,800 | 79,000 | 53 |

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[*] Burnup: 33 GWd(t)/tonne and 0.35 thermal efficiency.

end of the century. Other fissionable materials, plutonium-239 and -241 and uranium-233, are then increasingly expected to be used to meet long-term power requirements since these can be produced as byproducts of reactors generating electric power; i.e., plutonium-239, -241 from uranium-238 and uranium-233 from thorium-232.

The types of reactors are also expected to be altered as technology advances. Currently, in the United States, most nuclear power plants use light-water-cooled reactors (LWR) of two types, pressurized water reactors (PWR) and boiling water reactors (BWR). These are fueled with natural uranium slightly enriched to give an isotopic composition of approximately 3% uranium-235 and 97% uranium-238. In the future, many of these LWR systems are expected to be partially fueled with recycled plutonium instead of uranium-235.

Light water reactors are inefficient producers of plutonium; the ratio of the amount of fissionable plutonium produced to the amount of fuel consumed (the conversion ratio) is about 1:3. More efficient reactor types, namely converter reactors which produce nearly as much fuel as is used, and breeder reactors, which produce more fuel than is used, are expected to become a significant part of the nuclear power industry after the year 1985. Examples of these two reactor types most likely to be used are the high temperature gas-cooled converter reactor (HTGR) fueled with uranium-233 and uranium-235, and the liquid metal fast breeder reactor (LMFBR) fueled with plutonium.

Knowledge of the relative numbers of each type of reactor is important primarily for determination of the total quantities of the actinides produced per unit of electric power generation. Fission yields for most of the other elements of interest remain nearly constant for all fissionable materials of interest. For this study, a mix of reactor types as given in a study by the AEC (Ref. A-1) was used. Power capacity values were converted to power generation values by using a 64% load factor (percent of maximum available power utilized). These were then converted to tonnes of fuel discharged in any given year by using data on power generated two years earlier, a thermal efficiency of 0.35, and a burnup of 33 GWd per tonne of fuel:

$$\text{tonne of fuel discharged per year} = \left(\text{gigawatt (GW)} \text{ power capacity}\right) \times \left(0.64\ \frac{\text{power generated}}{\text{power capacity}}\right) \times \left(\frac{365\ \text{days}}{\text{year}}\right) \times \left(\frac{1}{0.35}\ \frac{\text{Thermal power (t)}}{\text{electrical power (e)}}\right) \times \left(\frac{1}{33}\ \frac{\text{tonne fuel}}{\text{GW days (t)}}\right).$$

A summary of spent fuel projections by reactor type and total amounts is given in table A.1. The number of fuel reprocessing plants necessary to service the nuclear power industry was estimated by using these spent fuel projections and the assumption that each fuel reprocessing plant will handle 5 tonne of spent fuel per day (equivalent to 1500 tonne/year). These numbers were also summarized in table A.1 for 5-year increments.

The uncertainty in the above total nuclear electric power generation values and total spent fuel discharge values after 1980 is estimated to be less than a factor of two. The distribution by reactor types may have a somewhat larger uncertainty.

## III. QUANTITIES OF RADIONUCLIDES IN SPENT REACTOR FUEL

There are three types of radioactive material present in spent reactor fuel: fission products, activation products, and actinide isotopes. The quantities of specific radionuclides present are primarily determined by fuel type, amount of burnup, and time of cooling (time between removal from the reactor and reprocessing).

Tables A.2 and A.3 show quantities of the potentially significant fission product and activation radionuclides present in one tonne of spent fuel with 33 GW(t)-days burnup and 150 days cooling time. These values (adapted from Ref. A-2) are considered reasonably representative of all nuclear fuel types. There is indication that cooling times shorter than 150 days may be used for some fuel cycles in the future, since quicker recycling of the recovered fuel produces an economic benefit. This would significantly increase the amounts of shorter-lived radionuclides in the fuel and available for release, but would not affect the long-lived fission product inventories.

The amounts of actinides estimated to be present in uranium fuels and plutonium-recycle fuels are given in

## TABLE A.2

### Representative Quantities of Potentially Significant Fission Products In Spent Reactor Fuels

(Adapted from Reference A-2)

| Isotope | Half-life (Years) | Curies per tonne | Grams per tonne | Release state | Notes |
|---|---|---|---|---|---|
| H -3 | 12.3 | 800 | 0.083 | Gas | 95% released as HTO |
| Kr -85 | 10.7 | 10,500 | 27 | Gas | |
| Tc -99 | 2.13x10$^{5}$ | 15 | 880 | Semivolatile | Oxide b.p. 200°C |
| Ru -103 | 0.11 | 180,000 | 5.7 | Semivolatile | Tetroxide b.p. .80°C |
| 106 | 1.01 | 820,000 | 240 | Semivolatile | $^{169}$Rh + $^{106}$Rh daughters |
| Te -125m | 0.16 | 6,500 | 0.36 | Semivolatile | Oxide b.p. 750°C |
| 127m | 0.30 | 25,000 | 2.7 | Semivolatile | Oxide b.p.$^{127}$Te daughter |
| 129m | 0.09 | 13,000 | 0.42 | Semivolatile | Oxide b.p.$^{129}$Te daughter |
| I -129 | 17x10$^{6}$ | 0.04 | 250 | Volatile | b.p. 184°C |
| 131 | 0.02 | 2.0 | 0.01 | Volatile | b.p. 184°C |
| Cs -134 | 2.05 | 100,000 | 77 | Semivolatile | Oxide b.p. 750°C |
| 135 | 3x10$^{6}$ | 1.2 | 1400 | Semivolatile | Oxide b.p. |
| 137 | 30.2 | 106,000 | 1200 | Semivolatile | Oxide b.p. 750°C$^{137m}$Ba daughter |
| Sr -89 | 0.14 | 100,000 | 3.5 | Solid | |
| 90 | 28.9 | 60,000 | 430 | Solid | $^{90}$Y daughter |
| Y -91 | 0.16 | 190,000 | 7.8 | Solid | |
| Zr -93 | 0.95x10$^{6}$ | 2 | 490 | Solid | |
| -95 | 0.18 | 400,000 | 19 | Solid | $^{95m}$Nb + $^{95}$Nb daughters |
| Nb -95 | 0.10 | 800,000 | 21 | Solid | |
| Sb -125 | 2.73 | 13,000 | 12 | Solid | |
| Ce -141 | 0.09 | 80,000 | 2.8 | Solid | |
| 144 | 0.78 | 800,000 | 250 | Solid | $^{144}$Pr + $^{144}$Nd daughters |
| Pm -147 | 2.62 | 200,000 | 220 | Solid | |
| Eu -155 | 5.0 | 40,000 | 87 | Solid | |

Burnup = 33 GWd(t)/tonne
Cooling Time = 150 days

## TABLE A.3

## Representative Quantities of Potentially Significant Activation Products in Spent Reactor Fuels

(Adapted from Reference A-2)

| Isotope | Half-life (years) | Curies per tonne | Grams per tonne | Release stage |
|---|---|---|---|---|
| Mn-54 | 0.86 | 30,000 | 3.9 | Solid |
| Fe-55 | 2.7 | 20,000 | 8.3 | Solid |
| Fe-59 | 0.12 | 500 | .01 | Solid |
| Co-58 | 0.20 | 30,000 | 1.0 | Solid |
| Co-60 | 5.26 | 2.000 | 1.8 | Solid |

Based on 33 GWd(t) burnup/tonne

150 days cooling time

table A.4 (Ref. A-2 to A-8). It was assumed that all fuels (including those used in HTGR's) other than uranium-235 fuels, can be considered equivalent to plutonium-recycle fuels. This introduces an uncertainty in these values proportional to the deviation from this assumption.

Based on the amounts of spent fuel to be processed, and on the estimated quantities of radionuclides per tonne of spent fuel, the projected annual quantities of several of the most significant radionuclides in fuel to be processed were calculated and are presented in table A.5.

Based upon plant decontamination factors appropriate for control systems presently incorporated in fuel reprocessing facilities [$10^9$, $10^3$, 1 and 1 for airborne pollutants (actinides, iodines, krypton, and tritium, respectively)], the annual inventories shown in table A.5 can be used to project the buildup of long-lived radionuclides in the environment as a result of fuel reprocessing. The estimated buildup of tritium, krypton-85, iodine-129, and plutonium-239 are shown in figures A.1 through A.4. Estimates of the environmental levels for time periods beyond 2020 by extrapolation of these curves is inappropriate. Detailed discussion of the status of system decontamination factors is presented in appendix B.

TABLE A.4

Representative Quantities of Actinides Present in Spent Reactor Fuels

(Adapted from References A-2 to A-8)

| Isotope | Half-life (years) | Uranium fuels Ci/tonne | Uranium fuels g/tonne | Pu-recycle fuel Ci/tonne | Pu-recycle fuel g/tonne |
|---|---|---|---|---|---|
| U-235 | $710 \times 10^6$ | <1 | 8,000 | <1 | 3,000 |
| 236 | $24 \times 10^6$ | <1 | 4,000 | <1 | 1,500 |
| 238 | $4510 \times 10^8$ | <1 | 950,000 | <1 | 950,000 |
| Np-237 | $2 \times 10^6$ | <1 | 600 | <1 | 200 |
| Pu-238 | 86 | 4,000 | 230 | 6,000 | 340 |
| 239 | 24,400 | 500 | 8,100 | 750 | 12,000 |
| 240 | 6,580 | 650 | 2,900 | 1,000 | 4,400 |
| 241 | 13 | 150,000 | 1,300 | 300,000 | 2,600 |
| 242 | 379,000 | 2 | 510 | 5 | 1,300 |
| Am-241 | 458 | 750 | 230 | 2,000 | 620 |
| 243 | 7,800 | 20 | 100 | 200 | 1,000 |
| Cm-242 | 0.45 | 35,000 | 10 | 250,000 | 75 |
| 244 | 17.6 | 2,000 | 25 | 25,000 | 300 |
| TOTAL (excluding uranium) | | 193,000 | 14,000 | 585,000 | 23,000 |

Cooling time = 150 days

Burnup = 33 GWd(t)/tonne

## TABLE A.5

Estimated Annual Inventories of Selected Nuclides in Spent Reactor Fuels[a]
(Curies)

| Year | Fuel Discharge (MT) | Tritium | Krypton-85 | Iodine-129 | Plutonium-239 | Plutonium-241 |
|---|---|---|---|---|---|---|
| 1970 | 25 | $2.0x10^{4}$ | $2.6x10^{5}$ | 1.0 | $1.9x10^{4}$ | $7.5x10^{6}$ |
| 1975 | 790 | $6.3x10^{5}$ | $8.3x10^{6}$ | $3.2x10^{1}$ | $5.9x10^{5}$ | $2.4x10^{8}$ |
| 1980 | 2,400 | $1.9x10^{6}$ | $2.5x10^{7}$ | $9.6x10^{1}$ | $1.8x10^{6}$ | $7.2x10^{8}$ |
| 1985 | 5,400 | $4.3x10^{6}$ | $5.7x10^{7}$ | $2.2x10^{2}$ | $4.2x10^{6}$ | $1.6x10^{9}$ |
| 1990 | 10,400 | $8.3x10^{6}$ | $1.1x10^{8}$ | $4.2x10^{2}$ | $7.8x10^{6}$ | $3.1x10^{9}$ |
| 1995 | 17,400 | $1.4x10^{7}$ | $1.8x10^{8}$ | $7.0x10^{2}$ | $1.3x10^{7}$ | $5.2x10^{9}$ |
| 2000 | 26,800 | $2.1x10^{7}$ | $2.8x10^{8}$ | $1.1x10^{3}$ | $2.0x10^{7}$ | $8.0x10^{9}$ |
| 2005 | 38,000 | $3.0x10^{7}$ | $4.0x10^{8}$ | $1.5x10^{3}$ | $2.9x10^{7}$ | $1.1x10^{10}$ |
| 2010 | 51,500 | $41.1x10^{7}$ | $5.4x10^{8}$ | $2.1x10^{3}$ | $3.9x10^{7}$ | $1.5x10^{10}$ |
| 2015 | 64,500 | $5.2x10^{7}$ | $6.8x10^{8}$ | $2.6x10^{3}$ | $4.8x10^{7}$ | $1.9x10^{10}$ |
| 2020 | 79,000 | $6.3x10^{7}$ | $8.3x10^{8}$ | $3.2x10^{3}$ | $5.9x10^{7}$ | $2.3x10^{10}$ |

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[a] Based on plutonium-recycle fuel and reactor type distribution in table A.1 (33 GWd(t)/tonne burnup and 150 days cooling period.)

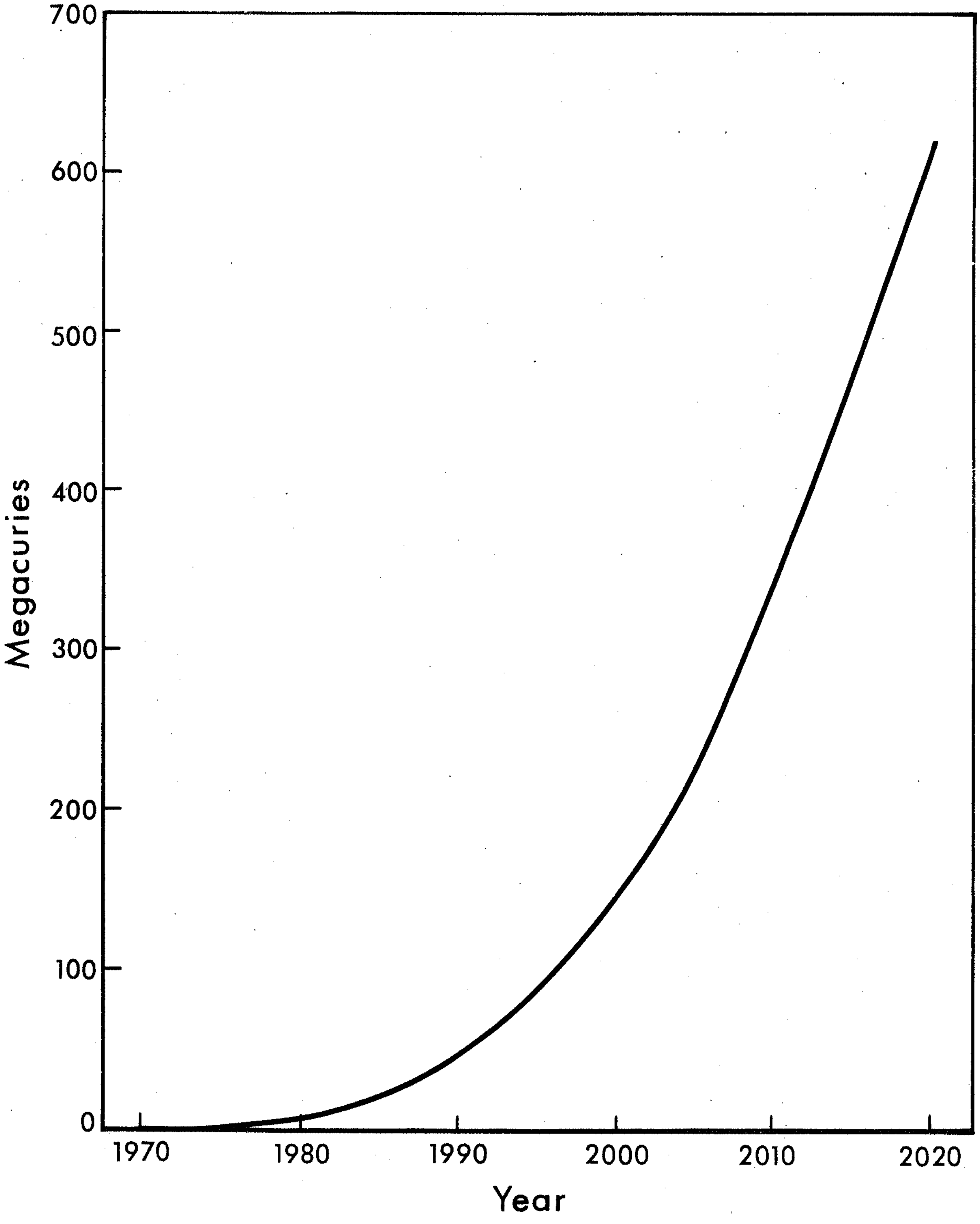

Figure A.1 Estimated Cumulative Environmental Build-up of Tritium from the Fuel Reprocessing Industry in the United States using a Decontamination Factor of 1.

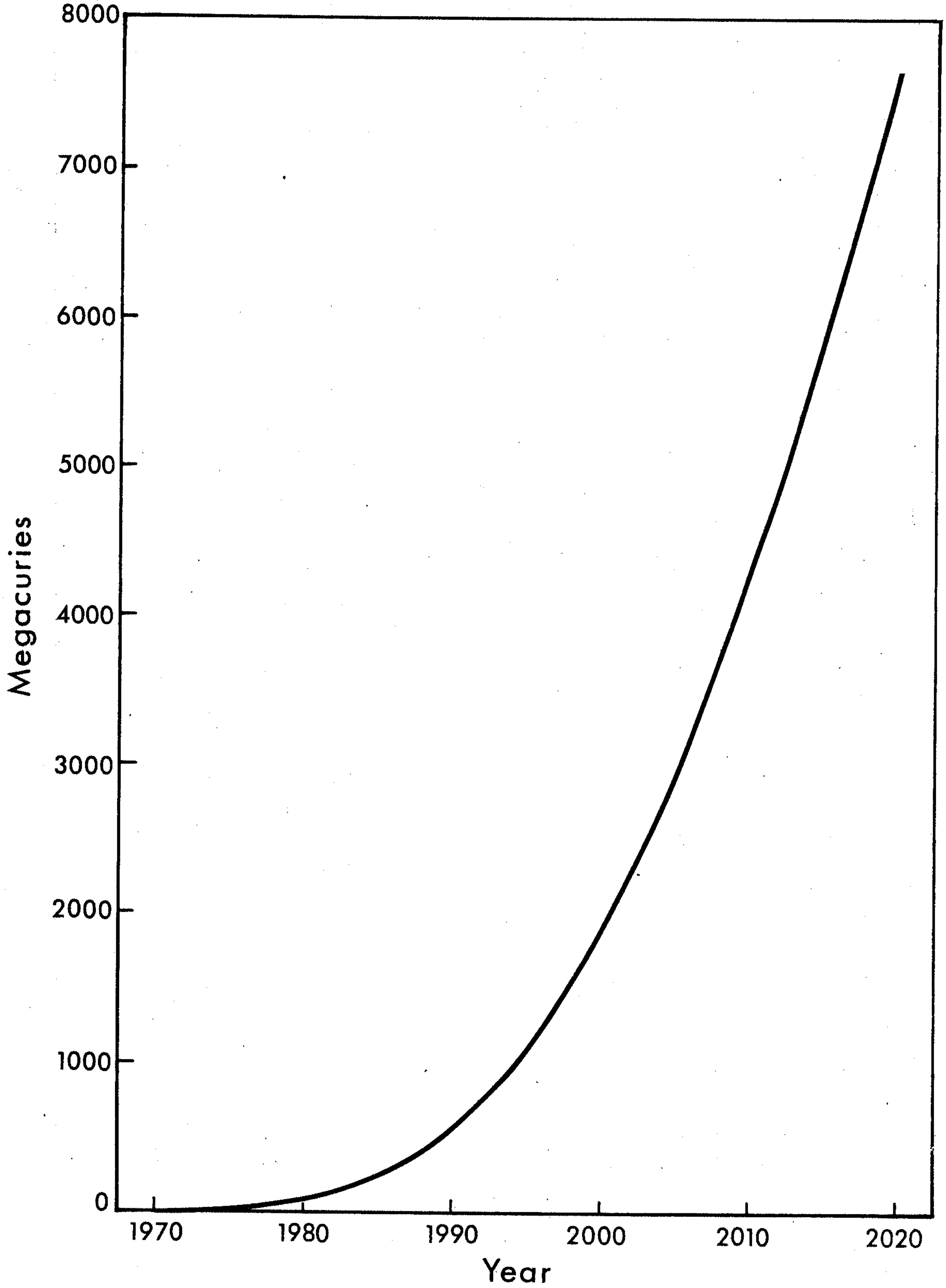

Figure A.2 Estimated Cumulative Environmental Build-up of Krypton-85 from the Fuel Reprocessing Industry in the United States using a Decontamination Factor of 1.

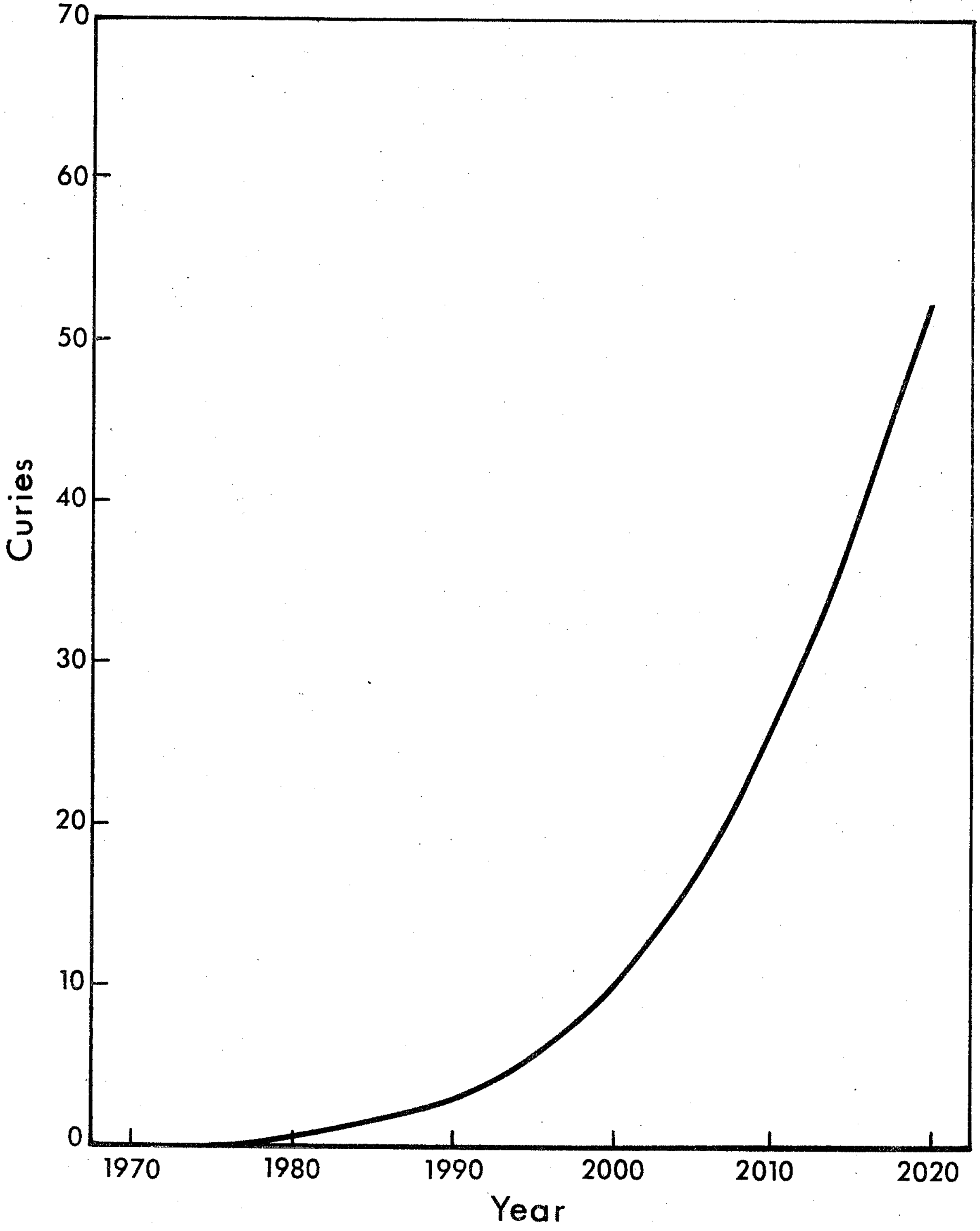

Figure A.3 Estimated Cumulative Environmental Build-up of Iodine-129 from the Fuel Reprocessing Industry in the United States using a Decontamination Factor of 1000.

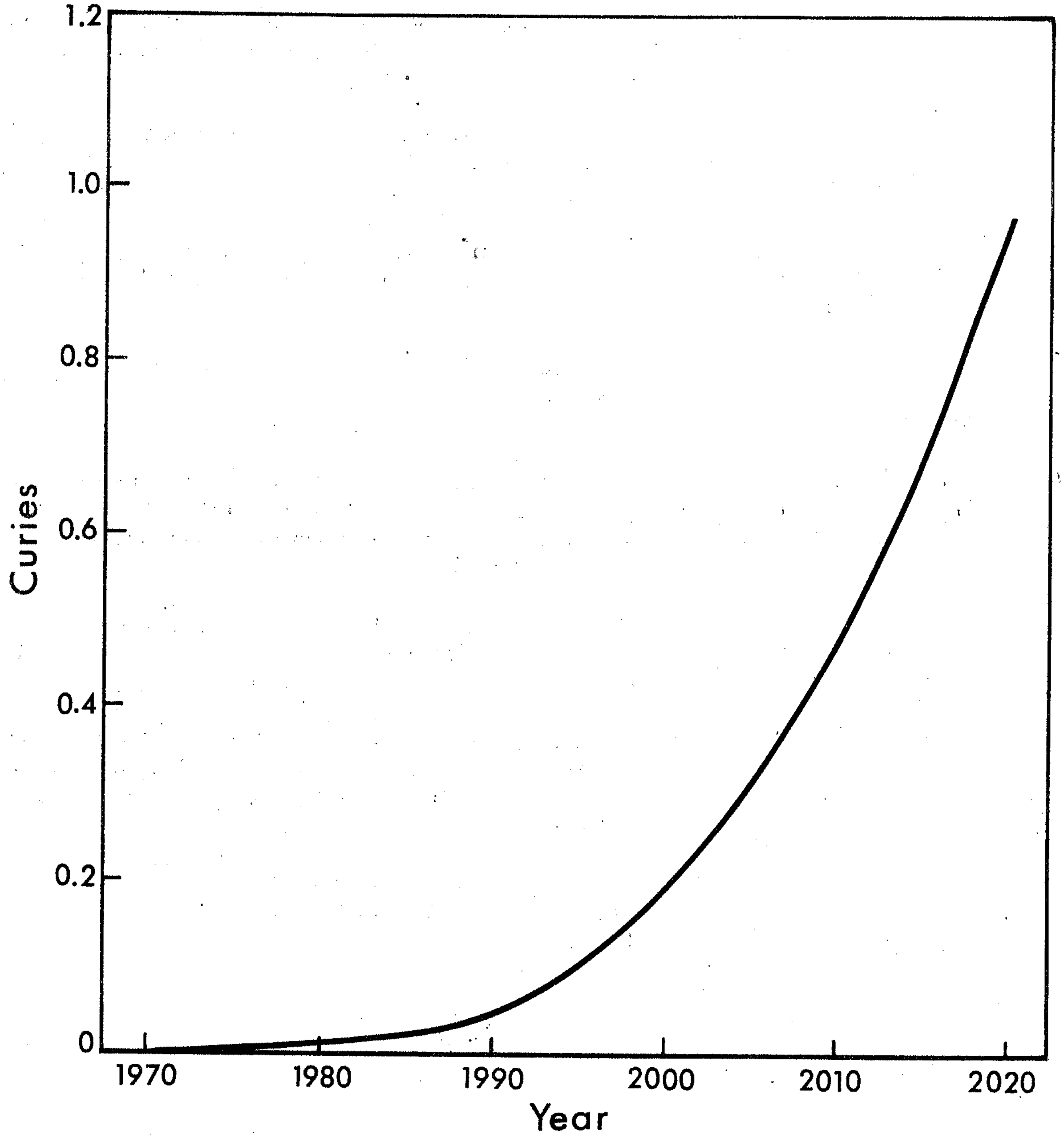

Figure A.4 Estimated Cumlulative Environmental Build-up of Plutonium-239 from the Fuel Reprocessing Industry in the United States using a Decontamination Factor of $10^9$.

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# APPENDIX B

# FUEL REPROCESSING FACILITIES

## I. INTRODUCTION

This appendix provides a basic description of the processes used at a fuel reprocessing plant and the control systems for reducing the radioactive discharges. There are three commercial nuclear fuel reprocessing plants capable of operation or under construction in the United States. Table B.1 summarizes general information for these three facilities. Currently, these plants are not processing irradiated fuel. The first commerical plant was Nuclear Fuel Services, Inc. (NFS) which is shut down for expansion. The Midwest plant is processing unirradiated fuel, and operation with spent fuel is anticipated for the summer of 1973. The Barnwell plant is under construction, and operations are expected to begin in 1974 or 1975.

## II. GENERAL DESCRIPTION OF PROCESS

Fuel reprocessing plants are essentially complex chemical plants, the complexity being compounded by the fact that the materials being processed are highly radioactive. The specific process used to separate the spent fuel element into the product streams and waste stream is dependent upon the particular type of reactor fuel being serviced.

All three present facilities use a shear (chop) and nitric acid leach method to separate the light water reactor (LWR) spent fuel from the metal cladding. Following this step, the Purex process with tributyl phosphate (TBP) as the solvent is used to extract the uranium and plutonium from the fission product waste in column contactors (Ref. B-1, B-2, B-3, and B-4). The uranium and plutonium are separated and further purified by various means, including ion

## TABLE B.1

## General Information for Commercial U.S. Nuclear Fuel Reprocessing Plants

| Plant | Location | Owner | Capacity (MTU[a]/year) | Type of Process |
|---|---|---|---|---|
| Nuclear Fuel Services, Inc. | West Valley, New York | Getty Oil Co. | 300 expanding to 600 to 900 | Purex |
| Midwest Fuel Recovery Plant | Morris, Illinois | General Electric Co. | 300 | Purex<br>Aquafluor process |
| Barnwell Nuclear Fuel Plant | Barnwell, South Carolina | Allied-Gulf Nuclear Services, Inc. | 1500 | Purex |

[a] MTU = metric tons uranium

exchange, scrubbing, evaporation, etc. The design recovery rate is 99.5% for uranium and plutonium.

Plants designed for processing of spent fuel elements from LWR systems could be used to process elements from LMFBR as long as the facilities are derated (handle smaller quantities of fuel) to avoid criticality problems and not exceed constraints on effluents (Ref. B-14).

The processing of HTGR spent elements requires a different "head end" processing system since such fuel elements may require the burning of the graphite which contains the coated fuel particles. The decontamination of the off-gas stream resulting from the burning operation requires development of "head end" processes unique to the HTGR processing facility (Ref. B-15). The Thorex process (Ref. B-3) will be used for the separation of uranium and thorium from the fission product wastes in spent HTGR fuels (Ref. B-15).

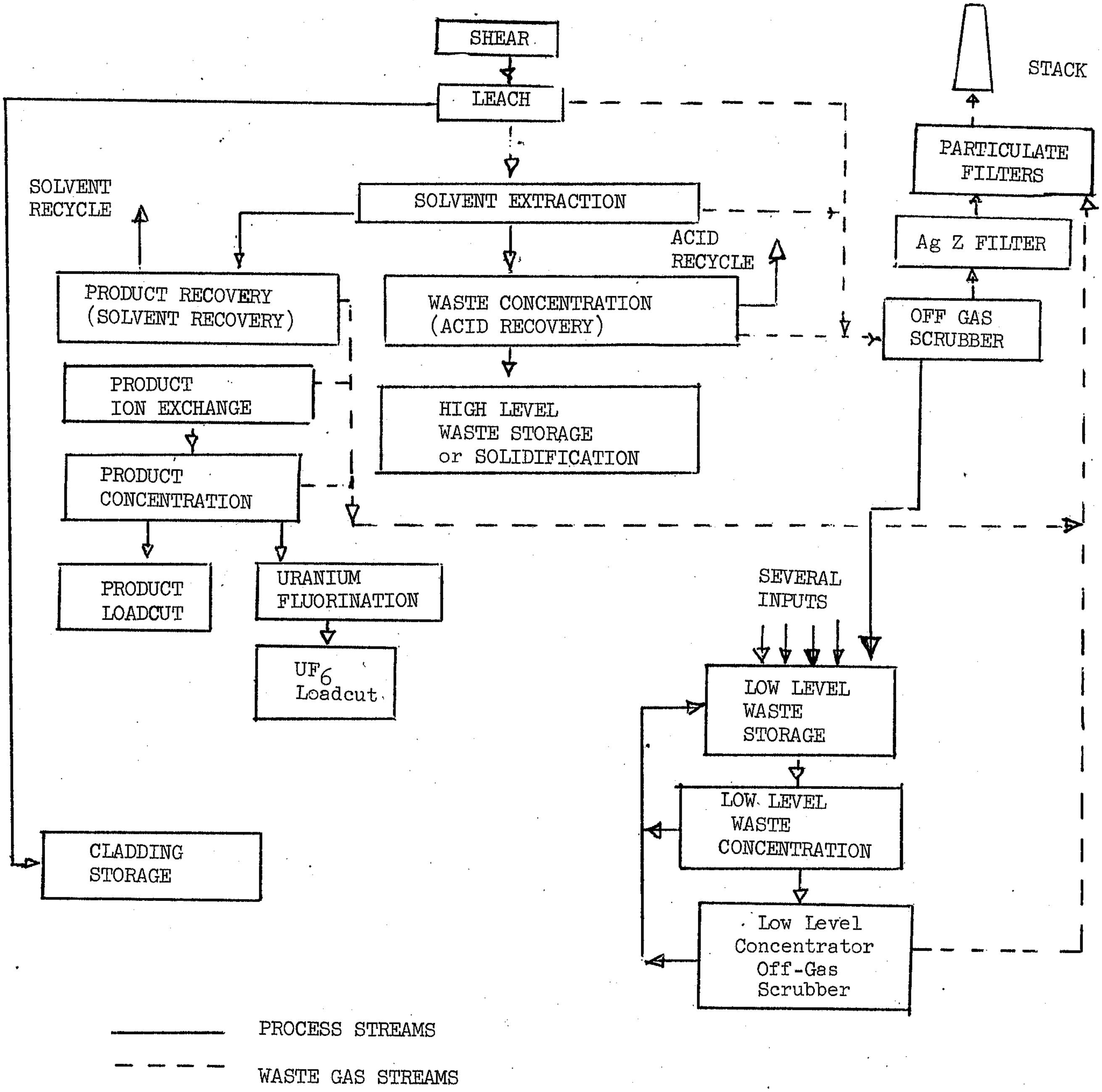
## III. PROCESS FLOW

A simplified summary of the aqueous processing steps in plants under construction or in operation is displayed in figure B.1. The main process steps will be summarized here to indicate sources and handling of radioactive waste liquids and gases.

1. Cask Unloading and Decontamination -- Cask wash water is discharged to low-level waste system.

2. Fuel Pool Storage -- Special provisions are made for leaking fuel elements storage and resulting contaminated water and off gases. Prevention of criticality is a major design factor. Inventory control and accountability are important operating parameters.

Figure B.1 Typical process flow schematic
Midwest Nuclear Fuel Reprocessing Plant



3. Fuel Transfer and Mechanical Processing -- NFS and Midwest disassemble fuel elements before shearing the individual fuel rods. Barnwell will shear the entire element and thus is essentially limited to reprocessing fuel from light water power reactors because of the geometries involved. Lengths of sheared fuel rods range from about 1.25 to 7.5 cm (1/2 to 3 inches). Very little of the krypton-85 and tritium is released during the shearing process. Goode (Ref. B-5) reported that less than 1% of the krypton-85 is released during this step and Cochran et al. (Ref. B-6) confirmed this conclusion based on field studies at an operating plant.

4. Fuel Dissolution -- In this step the spent fuel is leached from the sheared cladding in nitric acid as a preparatory step to the chemical separation processes. The leached hulls are analyzed for plutonium content and returned for further leaching if necessary. Over 99% of the krypton-85 (Ref. B-5, B-6) and about 6% (Ref. B-7) of the tritium is released into the off-gas system by this operation. In addition to the noble gases, a large fraction of the halogens are also released to the off gas in this step. Most of the radionuclide particulates present in the off gas result from this step (Ref. B-8), although this may not be the case at Midwest where large quantities of particulates will be added from the high-level waste solidification process. Since NFS dissolves fuel on a batch basis, essentially all the krypton-85 in the batch is discharged within a 3- to 4-hour period. Present NFS Technical Specifications limit fuel dissolution to 2 metric tons per day. Midwest and Barnwell will dissolve on a semicontinuous basis. Waste gases are processed

separately through the dissolver off gas system (DOG) at NFS and Barnwell, since most of the radionuclides in the waste gases are generated in this step at these facilities. Midwest has designed their gaseous waste treatment system somewhat differently and waste gases are not segregated by source.

5. Chemical Separation and Purification -- The nitric acid feed from the dissolver which contains the uranium, plutonium, and fission products is counterflowed in a contactor column with TBP and nitric acid. The uranium and plutonium are preferentially dissolved in the TBP, and the fission products are retained in the nitric acid. Separation of the plutonium and uranium in the organic solvent is achieved by reducing the plutonium to its trivalent state where it can be stripped with a nitric acid scrubbing process in a contactor column. The uranium is subsequently also stripped from the TBP which is then recycled. The plutonium and uranium are then purified through a series of processes. Midwest has designed a calcining system for conversion of the uranium to UF for shipment directly to an enrichment facility. NFS and Barnwell will ship uranyl nitrate in tank trucks to conversion facilities. Plutonium will be stored and shipped in the nitrate form in critically safe containers.

6. Recovery of Solvent and Acid -- The TBP solvent and the acid are recovered and recycled. The fission products are concentrated in the evaporator bottoms of the high-level waste concentrator system and the acid overheads are recovered. Additional fractions of the semivolatiles and the halogens are discharged to the waste gas system in this step, along with some particulates.

7. High Level Waste -- At NFS and Barnwell high-level liquid waste will be stored in stainless steel tanks which are located underground and externally cooled. These tanks are placed on concrete saucers to permit monitoring for leaks and inside concrete vaults which provide a secondary containment. These acidic wastes are not neutralized prior to storage to simplify future solidification. At the Midwest facility, which uses a fluidized bed calcination process for solidification, high-level wastes are solidified immediately after separation. This process has been used on a production scale with intermediate level waste since 1963 at the Idaho Chemical Processing Plant (ICPP) (Ref. B-9). The solidified waste will be sealed and stored in a cooling pool. The off gases from this process will contain some radioactive particulates (Ref. B-8, B-9), especially the semivolatile fission products such as ruthenium.

8. Low Level Waste -- The low-level liquid waste stream consists of wastes collected from sources throughout the plant. Midwest and Barnwell have been designed to completely eliminate the discharge of low-level liquid waste by adding an evaporator to the system to process the final low-level liquid waste stream (it is discharged to the environment at NFS). The evaporator overheads are discharged through the stack and theoretically contain all remaining tritium from the fuel. The bottoms are solidified and are currently shipped to privately operated waste burial areas on site.

In the gaseous stream, almost all of the krypton-85, and varying fractions of tritium and other volatile nuclides, such as iodine, are released to the dissolver off gas (DOG) during the dissolution process. Treatment of this waste stream is complicated since it contains varying concentrations of the oxides of nitrogen. NFS and Barnwell are designed to have a separate waste system for the DOG.

The various process tanks have vents for off gases which are contaminated with volatile fission products. These are routed to the vessel off-gas system (VOG). Midwest will combine the DOG and VOG systems since they have additional sources of airborne wastes from the calcining of the high level liquid waste.

The gaseous waste treatment system at all three facilities basically consists of a caustic scrubber, followed by a silver zeolite absorber and then final filtration through a high efficiency particulate air (HEPA) filter. NFS has incorporated both an acid scrubber and a caustic scrubber in their system. The scrubbers and silver zeolite adsorption systems are installed to collect the iodine in the off-gas stream. Midwest and Barnwell have installed or plan to install in series two independent particulate filtration systems which are isolated to avoid dual failure. Two HEPA filter sytems are to be installed at Barnwell. Midwest has installed a HEPA filter system in the off-gas stream and a sand bed filter system for final filtration of both the off-gas stream and the plant ventilation air.

## IV. CONTROL POINTS FOR EFFLUENTS

The cladding on the fuel normally provides the primary barrier (or containment) for preventing the release of fission product wastes. This barrier must necessarily be destroyed in the nuclear fuel reprocessing plant in order to recover the fissile and fertile material for reuse. This fact, in addition to the very large quantities of fission products present, requires that effluent control procedures must be incorporated at all processing steps. In practice, control of the effluents from each process can be achieved through the use of a common collecting system such as the vessel off-gas header system which collects the off gases from several processing vessels. The use of such common systems has definite economic advantage. Fewer control systems are required and a higher degree of reliability for control of discharges is possible since there are fewer components subject to failure, better quality equipment can be installed.

An example of a common collection system is the low-level liquid radioactive waste system. Sources feeding this system include, but are not limited to: cask decontamination water, leakage from the fuel storage pool and waste storage tanks, laboratory wastes, laundry wastes, high-level waste equipment drains, and floor drains. This waste is collected and processed through an evaporator where the overheads can be condensed and recycled, discharged, stored, or handled in a combination of these options. NFS, Inc. has chosen the method of discharge of the condensed overheads through a system of settling lagoons to a public waterway. Midwest has chosen a combination of treatments where the evaporator overheads are condensed and recycled to the maximum extent possible. Low-level liquid wastes which cannot be recycled, such as air scrubbing wastes, are collected in a low-level waste vault which is maintained at a constant volume by use of a second evaporator. The overheads of this evaporator are not condensed but discharged through a second air cleaning system and the sand filter to the stack.

In general, air cleaning systems follow the same principle. Contaminated air is collected from various processes in a common header and then treated to remove contaminants. Reprocessing plants have sufficient chemical contaminants in some off-gas streams to cause problems, such as overloading of various air cleaning systems. Therefore, the off-gas streams are frequently segregated by source, especially for initial treatments.

Specific effluent control points and the principal contaminants are: (1) Dissolver Off Gas -- noble gases (krypton-85 and xenon-133), halogens (iodine-129 and iodine-131), tritium, and particulates. (2) Solvent Extraction and Purification Off Gas -- particulates. (3) Solvent Recovery Off Gas -- mixed fission products. (4) Acid Recovery Off Gas (High-Level Waste Concentration) -- halogens and other volatile species. (5) High-Level Waste Solidification -- ruthenium and other potential volatile radionuclides including technetium, cesium, selenium, and tellurium (Ref. B-8). (6) Low-Level Waste Treatment -- tritium, strontium, ruthenium, cesium, and other longer-lived radionuclides (Ref. B-10).

Investigations are being conducted into modifying spent fuel reprocessing systems to provide more positive control of the effluents. ORNL is presently directing efforts to the design of equipment and process flow to obtain "near zero release" processing of short-cooled LMFBR fuel (Ref. B-11). The design includes a new "head-end" processing step (voloxidation) that is designed to release

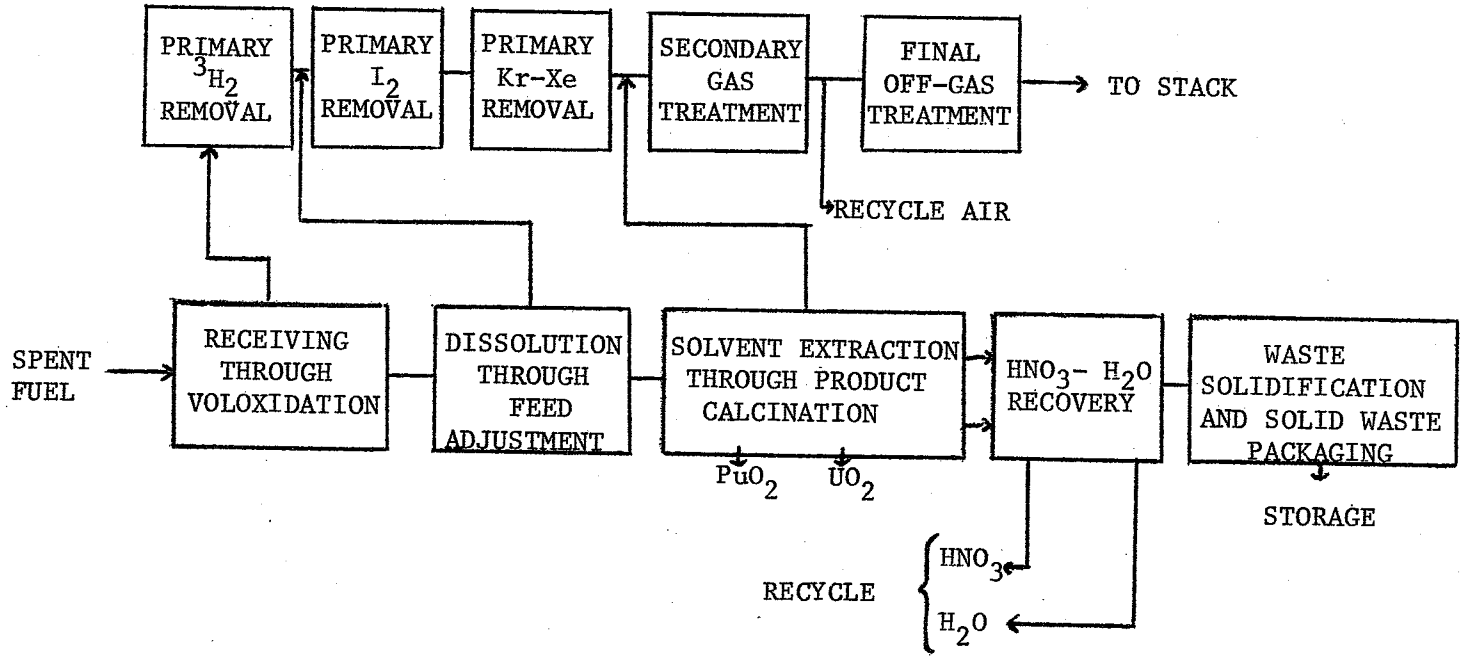


Figure B.2 Schematic diagram showing the steps required for the "zero release" reprocessing concept (B.11) tritium from LMFBR fuel and deactivate sodium prior to aqueous processing. The tritium, krypton, and iodine are evolved during the head end operations (which include voloxidation and dissolution) and vented to their respective primary removal systems. The off gas from the dissolution cell and from the process equipment beyond dissolution and feed adjustment is subjected to a secondary off-gas treatment system which includes filters for particulates and scrubbers for the oxides of nitrogen, halogens, and ruthenium. Argonne National Laboratory is also developing an alternate "head-end" pyrochemical process for decladding of LMFBR fuel which potentially may produce an improved method for control of the effluents (Ref. B-12, B-13).

## V. SOLID WASTES

The solid wastes resulting from the recovery of uranium and plutonium can be categorized as high-level solidified wastes, spent fuel cladding wastes, and low and intermediate level wastes. Federal regulations (Ref. B-16) require that high-level wastes, generally interpreted as self-heating, be solidified within five years of processing. In addition to the radiation exposure protection which must be provided for these wastes, cooling must also be provided. The spent fuel cladding wastes contain residual amounts of the fuel in addition to the activated metallic radionuclides making up the cladding itself. There are many sources of low and intermediate level wastes: air filters, spent resins, silver zeolite, evaporator bottoms, sand filters, etc. It appears the greatest problem presented by these wastes is the presence of long-lived radionuclides of health significance such as the alpha-emitting transuranics and iodine-129. These long-lived components will be present in all solid wastes from reprocessing plants.

## VI. DISCHARGE CONTROL OPTIONS

In general, the fuel reprocessing industry has incorporated the most advanced technology into their waste treatment systems. For example, the control system for iodine, one of the limiting radionuclides in the local environs of a reprocessing plant, (Ref. B-17) will use silver zeolite technology which has only recently been developed. It should be noted that a control method is not available for tritium and only the one control system is planned for iodine.

Most of the radioactive discharges to the environment from reprocessing plants will be in the gaseous waste effluent. The two newer plants have designed their processing systems to eliminate the discharge of liquid radioactive material. However, neither of these facilities has been operated with irradiated fuel. Thus, a decision on whether it is preferable to discharge radioactive waste to the atmosphere only or to discharge via both the liquid and gaseous pathways should be postponed until operating experience with both methods is obtained. Since most of the current efforts to reduce discharges have been directed toward gaseous effluents, the methods discussed in this section are limited to the control of airborne discharges. However, liquid waste discharges may require additional investigation in the future.

The radioactive pollutants that are most likely to be released from normally operating reprocessing plants are krypton, tritium, iodine, and the actinides. Other fission products such as strontium, ruthenium, and cesium, and induced activities in the fuel element cladding can also be released. In addition to the anticipated normal discharges (gaseous waste stream), miscellaneous airborne releases can occur because of the complexity of the various processing operations and the unproven reliability of some of the control systems. Such releases may not be detected by monitoring of the gaseous waste stream (stack effluent). Inplant air monitoring for contamination control can, however, indicate these possible pollutants. A summary of the characteristics of gaseous waste control systems for the isotopes of major concern is presented in table B.2. It is seen that the long-term operational reliability of control systems is unproven with the exception of the HEPA (high efficiency particulate air) and sand filter systems.

## VII. KRYPTON-85 CONTROL

Up to the present, krypton-85 and other noble gases have been released directly to the atmosphere from nuclear reactors and fuel reprocessing plants. With no off-gas treatment for noble gases about 10,000 Ci of krypton-85 is estimated to be discharged per metric ton of spent fuel processed, assuming a burnup of 33,000 megawatt days per metric ton.

Several methods have been suggested to limit such releases. The processes are classified as ambient temperature adsorption, cryogenic adsorption, cryogenic distillation, selective absorption, permaselective membranes, and clathrate precipitation.

## TABLE B.2

### Control System Data for Nuclear Fuel Reprocessing (LWR + Recycle Fuels)

| Isotope | Control System | Reliability | System Decon Factor | Projected Release Rate | COSTS | |
|---|---|---|---|---|---|---|
| | | | | | Capital/plant | Operation/yr |
| Krypton-85 | (a) None | NA[1] | 1 | $10^4$ Ci/MTU | NA | NA |
| | (b) Cryogenic distillation | Good | $10^3$ | 10 Ci/MTU | $3x$10^6$ | $1x$10^5$ |
| | (c) Cryogenic adsorption (charcoal) | Unproven | $10^2$ | $10^2$ Ci/MTU | $3x$10^6$ | $1.5x$10^5$ |
| | (d) Freon adsorption | Unproven | $10^2$ | $10^2$ Ci/MTU | $1.5x$10^6$ | $1x$10^5$ |
| Tritium | (a) None available | NA | 1 | 800 Ci/MTU | NA | NA |
| | (b) Voloxidation | Unproven | $10^2$ | 8 Ci/MTU | Unknown | Unknown |
| Iodine-131 | (a) None | NA | 1 | 2.0 Ci/MTU | NA | NA |
| | (b) Scrubber + AgZ | Unproven | $10^3$ | 0.02 Ci/MTU | $1.2x$10^6$ | Unknown |
| Iodine-129 | (a) None | NA | 1 | 0.04 Ci/MTU | NA | NA |
| | (b) Scrubber + AgZ | Unproven | $10^3$ | 0.0004 Ci/MTU | $1.2x$10^6$ | Unknown |
| Actinides | (a) None | NA | $10^5$-$10^6$ | 0.6-6 Ci/MTU | NA | NA |
| | (b) Pre-filter + 2 HEPA's | Good | $10^9$ | 6x$10^{-4}$ Ci/MTU | $1.0x$10^5$ | $5.0x$10^4$ |
| | (c) Pre-filter + HEPA + Sand Filter | Excellent | $10^9$ | 6x$10^{-4}$ Ci/MTU | $3.5x$10^5$ | $7.0x$10^4$ |

Notes:

1) NA indicates not applicable
2) Iodine-131 content is estimated for fuel with 33,000 MWd/MTU burnup and 150 day cooling

The processes have been previously reviewed by several authors (Ref. B-18, B-19, B-20, B-21, and B-22) with regard to development status, advantages and disadvantages, cost, and efficiency. Kirk (Ref. B-18) has prepared a comprehensive review of the radiation hazard from krypton-85. The conclusions reached in these reviews indicate that at present only cryogenic distillation, selective absorption processes, and cryogenic adsorption are worthy of consideration for control of krypton-85 discharges from reprocessing plants. (See table B.3 from Ref. B-19.) Systems based upon both the cryogenic adsorption and cryogenic distillation processes have been designed for and are being installed at light-water-reactors for extension of holdup times for gaseous effluents containing noble gas radionuclides. The selective absorption process has been developed for application to reactor systems, but requires further development to be applicable to fuel reprocessing plants (Ref. B-20).

The cost of krypton collection systems is highly dependent on the design of the dissolution process. Essentially all of the krypton present in the spent fuel is released during this dissolving or leaching process. To minimize the costs, it is necessary to minimize the total volume of off gases from this process since all of the off gas must be treated to remove the krypton. Therefore, in a new plant the cost of a krypton collection system would probably be significantly less than installation of such a system in an operating plant where no effort was made to minimize the total off gas from the leaching process. The costs presented are typical of costs for a

## TABLE B.3

## COMPARISON OF PROCESSES FOR THE REMOVAL OF $^{85}$Kr FROM DISSOLVER OFF-GAS FROM A FUEL-REPROCESSING PLANT

| Process | Kr recovery (%) | Development status | Advantages | Disadvantages |
|---|---|---|---|---|
| 1. Room-temperature charcoal beds or molecular sieves | 99 | Bench scale completed; scale-up feasible. | Simple operation; accepts dilute feed gas | Large-volume adsorber beds; charcoal can ignite; strong oxidizing gases must be removed prior to adsorption |
| 2. Low-temperature charcoal beds or silica gel | 99 | Development completed; plant operated | Small-volume beds; uses dilute feed gas | Charcoal can ignite; oxidizing gases, $CO_2$ and $H_2O$ must be removed; large consumption of liquid nitrogen: adsorbers must withstand high pressure; high operating cost |
| 3. Cryogenic distillation | 98 | Developed and operated on a significant scale | Low capital cost and low operating cost | Explosion hazard in forming and concentrating ozone |
| 4. Liquid extraction | 99 | Bench scale completed; demonstration needed for large scale | Using Freon-12: low refrigeration costs; low solvent costs; no explosion hazard; might eliminate pre-treatment | The absorber column operates at 200 lb/in$^2$ (gauge); the volume of extractant is large if operated at 15 lb/in$^2$ (gauge) |
| 5. Clathrate precipitation Kryptonates | Unknown | Laboratory studies only; no engineering | $^{85}$Kr is collected as a solid;storable | Needs concentrated feed gas; crystallization step slow |
| 6. Perm-selective membranes | 99 | Bench-scale work; need engineering tests | | Membranes sensitive to chemicals; high power costs |
| 7. Thermal diffusion | | Little pertinent data | | |
| 8. Electrostatic diffusion | | Limited; technical feasibility not proven | | Poor economics for disposal of dilute $^{85}$Kr waste |

**This Table is reprinted with the kind permission of Dr. C. M. Slansky who presented it in a paper published in Atomic Energy Review (See reference B-19)**

krypton control system in a new facility. At the Midwest facility an effort was made to prevent large scale dilution of the off gas.

Cryogenic adsorption systems remove krypton from process gas streams by adsorption in refrigerated activated charcoal beds until the bed capacity is reached, followed by desorption into a purging gas while heating the beds. This was demonstrated on a large scale at the Idaho Chemical Processing Plant (ICPP) more than 15 years ago. The disadvantages of this process are high refrigeration costs, fire hazard potential, explosion potential due to hydrocarbons, nitrogen oxides and ozone, and impurity plugging of the adsorbers. An overall recovery fraction of 31% was obtained for krypton although with design changes and modifications in operating procedures, a recovery fraction of the order of 99% could be achieved. The system can be considered but may not be the best for application to fuel reprocessing plants. It may have more potential for application to interim holdup for effluent gases in reactors.

Experience has been gained for this method in the development of the HTGR and a decontamination factor of 10 appears to be achievable under most operating conditions. Operation of the adsorber beds at cryogenic temperatures helps overcome the problem of an occasional abrupt release of adsorbed contaminants which has been experienced with ambient temperature absorbed beds. Assigning a decontamination factor to ambient temperature systems is questionable since experience has shown that under adverse conditions it is possible to experience a negative decontamination factor for interim periods.

Total capital costs, including installation, for a cryogenic adsorption system are estimated at 3 million dollars based on general estimates for use of systems at reactors. Slansky (Ref. B-19) estimated one million dollars for capital costs which appears low. An annual operating cost of $150,000 is reasonable.

Cryogenic distillation, which is based upon separation of gases due to differences in their relative volatilities at low temperatures, has been demonstrated and operated on a significant scale at ICPP for removal of krypton and xenon from an off-gas stream (Ref. B-23 and B-24). Recovery of krypton and xenon in a form suitable for bottling in gas cylinders is possible with this process. An additional advantage is the lower capital and operating cost of this cryogenic system as compared to cryogenic adsorption. The cryogenic distillation process entails some potential for explosion. In spite of the explosion potential, cryogenic distillation is considered to be one of the two most promising processes for noble gas control at reprocessing plants.

Considerable experience has been gained in operating these systems in liquified gas (or air) plants. While decontamination factors across the cryogenic stage of $10^3$ have been estimated, small leaks can occur in a system. Estimates of the recovery factor for an overall system range from 98% to 99.99%. Decontamination factors of $10^3$ (99.9% recovery) should be attainable with this system and guarantees of such performance have been submitted to the General Electric Co. with bids for installation of a cryogenic distillation system at their Midwest plant (Ref. B-26).

The General Electric Co. received bids ranging from 0.75 to 1.5 million dollars for the equipment needed to install a cryogenic distillation system in their Midwest plant (Ref. B-26). It is estimated that installation costs would match equipment costs, thus producing a total capital cost of about 3 million dollars for this system. Annual operating costs of $100,000, postulated by Slansky (Ref. B-19), are probably a good estimate. Xenon is also recovered by this system and if kept separate from the krypton, has a potential market value.

Selective absorption (liquid extraction) depends upon the relative solubilities of gases in the solvent used -- Freon-12 being the typical solvent under consideration. Krypton and xenon are selectively absorbed in this solvent while other materials pass through. The solvent is then processed to recover the krypton and xenon which can be stored while the solvent is recycled. Bench-scale studies have been completed (Ref. B-20 and B-25) and, as indicated previously, commercial systems for application to reactors have been developed. Because of the anticipated explosion hazard associated with the cryogenic systems, management personnel of commercial fuel reprocessing facilities (NFS, Barnwell) have indicated a preference for fluorocarbon selective absorption systems of this type.

Major disadvantages of such a system include radiation degradation of the solvent, and a requirement for pretreatment of gas streams to increase system tolerance to impurities. Babcock and Wilcox indicates that these problems can be circumvented with good engineering design. Decontamination factors of 100 to 1000 are expected for such systems (Ref. B-20).

The cost of a selective absorption system for installation at a reactor is estimated at one million dollars. Assuming a slightly greater capacity system for a reprocessing plant results in a total installed cost of $1.5 million. Annual operating costs of $100,000 predicted by Slansky (Ref. B-19) also appear reasonable.

## VIII. TRITIUM CONTROL

Processes available for tritium control have recently been reviewed (Ref. B-27). The techniques considered include: chemical exchange, distillation, electrolysis, diffusion and centrifugation, radiolysis, adsorption and chromography, solvent extraction, and molecular excitation. At present the information on these techniques is inadequate to project the technical or economic feasibility of retaining tritium.

Voloxidation is a new method of tritium control that looks promising (Ref. B-11). In this process the fuel pins, just after shearing, are heated to approximately 650° C in a stream of air or oxygen. Tritiated water is generated which should be relatively free of ordinary water and consequently occupy a much smaller volume than tritium wastes from presently planned or operating reprocessing plants. This process may collect approximately 99% of the tritium which is present in the unprocessed fuel. Since the voloxidation process requires a major change in the head-end design of fuel reprocessing plants, it would probably be impractical to back-fit existing or planned facilities.

The projected release rate of tritium from a facility having no tritium control systems is 88 Ci/MTU. Releases may be either by the air or water pathways depending on plant design. With the voloxidation head-end process, tritium decontamination factors of 100 are anticipated. Cost information on the voloxidation process is not presently available; however, General Electric considered three other methods of tritium recovery and/or disposal in connection with their Midwest plant (Ref. B-26). Removal from off gas was stated to cost approximately $10 million with no process or technical feasibility defined. Deep-well disposal of tritiated water was reported to run between $400,000 and $500.000. Finally, off site shipment cost was estimated to be between $250,000 and $350,000 with no estimate of feasibility or safety associated with the packaging and transport.

## IX. RADIOIODINE CONTROL

A variety of processes have been developed and used for collection of radioactive iodine. These processes include (1) wet collection -- aqueous scrubbing (reactive sprays, towers, wet filters); (2) adsorption (charcoal, activated charcoal, silver zeolite metallic filters); and (3) filtration (high efficiency particulate -- HEPA, sand deep-bed fiber glass).

The particular treatment process to be selected depends upon the collection efficiency desired, quantity and chemical species of iodine involved, stream characteristics, i.e., flow rates, temperature, humidity, etc. The actual overall removal efficiency of a system is dependent upon the off-gas flow paths as well as the characteristics of the treatment techniques.

Wet Collection Techniques have been used to quantitatively retain iodine in the liquid phase by adding mercury salts during fuel dissolution at the Savannah River Plant. The resulting Hg-I complex is solvent extracted and the solvent is washed in a solvent scrubber to remove any remaining iodine. Disposal of the solid Hg-I complex must be effected. However, there are no AEC guidelines for disposal of such waste at present.

A mercuric iodate precipitate can result from scrubbing the off gas with a mixture of 8 to 14 molar nitric acid and 0.2 to 0.4 molar $Hg(NO_3)_2$ (mercuric nitrate). Solid iodine can be obtained by use of concentrated 17 to 19 molar nitric acid at room temperature. The latter systems can be considered capable of removing massive concentrations of elemental iodine and trace quantities of other forms. Decontamination factors of $10^4$ for all forms of iodine are reputed to be theoretically feasible in either packed or bubble-packed scrubbers at operating throughput rates considered appropriate for fuel reprocessing (Ref. B-28). If the off gas contains high concentrations of NO some loss in the decontamination factor occurs. Alkaline solutions (NaOH, $NaHCO_3$, $NaCO_3$) are also used in scrubber columns and have reported decontamination factors of 10 to 20. In packed columns, the separation efficiency is a function of bed height, fiber drag coefficient, fiber diameter and fiber volume fraction.

An operational facility (Eurochemi Fuel Reprocessing Plant) which uses two scrubbers (NaOH for low iodine concentrations and $NaHCO_3$ or $Hg(NO_3)_2$ for high iodine concentrations) reports decontamination factors of 500 for gaseous iodine and 2,500 for iodine in aerosols. The chemical form of iodine which emanates from these scrubbers is predominantly organic (Ref. B-29) although traces of hypoiodous acid have also been reported.

Reactive sprays, hydrazine and thiosulfate, have been studied for application to iodine and methyl iodide washout from reactor containment atmospheres with decontamination factors of 2,000 and 100 reported respectively. Such systems have not been applied to fuel reprocessing plants. Their performance is found to be a function of relative humidity, temperature, drop size, solution pH, and concentration.

<u>Charcoal Adsorption</u> has been used for more than 10 years in reactor and fuel reprocessing off-gas systems. The iodine removal efficiencies of activated charcoal that have been reported cover a broad range (50-99.99%). The actual decontamination factor is dependent upon the forms of iodine, concentration, stream humidity and flow velocity, and charcoal impregnant.

Silver zeolite ($AgNO_3$ impregnated in an alumina-silica molecular sieve) is reputed to be superior to impregnated charcoal for the adsorption of methyl iodide (approximately 20 times that for impregnated charcoal under dry air conditions)(Ref. B-30). The principal design consideration is the effective residence time in the bed which is related to the face velocity. If the silver zeolite (AgZ) beds are designed to permit a mean residence time of about 0.5 seconds, efficiencies are greater than 99.9% under the most adverse conditions of humidity and chemical form of the iodine (Ref. B-30 and

B-31). Removal efficiencies can be optimized through selection of the zeolite material with its characteristic alumina-silica ratio. However, this ratio largely determines the resistance of the AgZ to acid vapors and thus the effective life of the bed. In general, high acid resistance results in lower removal efficiencies.

Although the efficiency of the AgZ is acceptable, consideration must be given to the loading characteristics of AgZ in terms of the total iodine cleanup system. The AgZ beds will adsorb all halogens and probably cannot be used when HCl is used for fuel dissolution. In addition, there is a considerable amount of iodine (iodine-129 and -127) present in the off gas, and this may load the AgZ at unacceptably high rates. Current technology employs the alkaline scrubber to remove a large fraction of this iodine which is theoretically elemental in composition. Work is continuing on the development of other metallic zeolites which may eventually replace the scrubbers. Lead zeolite currently appears to be the most attractive of these more economical and less efficient systems.

The lifetime of the AgZ system is improved by introduction of an oxidizing catalyst upstream of the sorbent (Ref. B-28). The use of AgZ systems has been shown to be feasible and practicable, but full scale operation has yet to be accomplished. Adsorption of iodine on metallic filters (copper, steel or aluminum, silver-coated copper) has resulted in recovery in the range of 97.4 to 99.9%. Silver-coated silica gel has a reported decontamination factor of 10 for iodine (elemental).

Filtration is primarily designed for control of particulates. It cannot be seriously considered as a primary technique for iodine collection, since it depends upon sorption of iodine to particulates which are then trapped by the filter. In any event, all off-gas streams from fuel reprocessing plants will be filtered through sand, deep-bed fiber glass or high efficiency particulate air filters in addition to the iodine control systems. The iodine collected by particulate filters will experience desorption at a rate which is dependent on stream conditions. While this desorption rate may be significant for the short-lived iodine-131 discharge, it will have a negligible effect on the iodine-129 discharge. Therefore, the removal efficiency for filtration of iodine is negligible.

Control Systems Evaluation. It is estimated (Ref. B-32) that much of the discharged iodine will be in an organic form and that most of this iodine will interact with atmospheric particles and settle out within 10 to 20 miles of the discharge point. Thus, the discharged iodine appears to be principally a local problem, although long-term environmental transport must be considered because of iodine-129 with its long half-life.

LWR spent fuel with a burnup of 33,000 megawatt days per metric ton will contain about 2 Ci of iodine-131 per metric ton after 150 days cooling. In addition to being dependent on the irradiation history of the spent fuel, the iodine-131 content decreases rapidly with the cooling period because of its 8 day half-life. The iodine-129 content will be about 0.04 Ci per metric ton of spent fuel.

An overall system for iodine collection in fuel reprocessing plants will probably consist of a combination of wet collectors (liquids and off-gas scrubbers) followed by a catalytic decomposition system, an adsorption system, and a filtration system. The overall efficiency will be dependent upon the detailed design of the off-gas flow system. For example, if the system is assembled in the manner of the Midwest facility, the off-gas stream bearing the iodine is first processed through the caustic scrubber (eff. = 90%) and then through the AgZ bed. The gas stream is then routed to the sand filter and stack. The overall removal efficiency for this treatment should be greater than 99.9%. However, the scrubber solution, theoretically containing 90% of the iodine, is routed to the low-level-waste storage vault. The low-level waste in the vault is routinely evaporated for volume reduction, opening a way for a large fraction of the iodine to be revolatilized. This second off-gas stream is processed through a second scrubber but no AgZ bed. Therefore, about 10% of the iodine routed to the low-level vault can be discharged to the sand filter and the stack. The buildup of the long-lived iodine-129 in the waste storage vault must also be considered in this system since it is reasonable to assume that the iodine-129 discharge will increase with the inventory buildup.

The development work previously discussed may produce more effective iodine cleaning systems for reprocessing plant off gases (Ref. B-11, B-12, and B-13). However, even with present technology it is difficult to speculate what the cleaning efficiencies of actual installed systems will be. Once these systems are in operation their performance can be monitored and documented. In the interim the use of an overall decontamination factor of 100 to 1,000 appears acceptable.

Based on experimental evidence it is reasonable to assume that the wet scrubber/AgZ system can be designed to achieve a minimum decontamination factor of 1,000 under most conditions. However, as discussed previously, the performance of the off-gas iodine cleaning system may not be the controlling factor in determining total iodine discharges from a reprocessing plant. In particular, the system for handling or processing the scrubber solutions must minimally provide the same cleaning efficiency as the off-gas stream since a large fraction of the iodine is expected to be in the scrubber solutions. Therefore, it can be concluded that while the efficiency of currently planned iodine removal systems will be a minimum of 99.9%, the total iodine waste handling system introduces a large uncertainty.

Meager data are available regarding the costs for iodine removal systems. The costs are dependent upon the volume of off gas which requires processing as well as the total quantity of iodine to be removed. The following estimates have been adjusted to 1970 dollars using the Marshall and Stevens Equipment Index.

| Wet Collection | Capital Costs* | Operating Costs* |
|---|---|---|
| a. Spray tower | $310-620/1,000 cfm | Unknown |
| b. Packed towers | $620-1,240/1,000 cfm | $510-1,020/1,000 cfm |
| c. Solvent extraction | Unknown | Unknown |
| Absorbing Systems: | | |
| a. Charcoal | $40,000/bed | Unknown |
| b. Charcoal + catalytic combiner (KRB) | $750,000-$10 | Unknown |
| c. Ag zeolite | $400,000/bed | Unknown |
| Complete System Estimate: | | |
| 5 MTU/day throughput | | |
| a. Caustic scrubbers | $\$7.5 \times 10^5$ | $\$7.5 \times 10^4$ |

* At an average plant the total air volume moved is approximately 60,000 cfm.

The costs listed above are for the caustic scrubber systems only since the quantities of AgZ needed will not be known until operating experience is gained to determine the extent of the parasitic loading.

The alternative of using holdup for control of iodine-131 from fuel reprocessing plants is applicable only in conjunction with a removal system that will extract the long-lived radioiodine, iodine-129. Estimates of the cost involved for holdup are as follows:

| Given: | | |
|---|---|---|
| | plutonium price | \$7/g |
| | annual interest rate | 10% |
| | LWR fuel plutonium content | 16* kg/MTU |
| | LMFBR fuel plutonium content | 86 kg/MT |
| | time increment (1 month) | 30 days |

$$\frac{16\ \text{kg}}{\text{MTU}} \times \frac{7\ \$}{\text{g}} \times \frac{0.10}{\text{year}} \times \frac{1\ \text{year}}{12\ \text{month}} = \$930/\text{tonne month}$$

Example: Barnwell will have a capacity of 1,500 MTU/year and will process at 150 days cooling time. If the cooling time is increased to 300 days to permit the iodine-131 to decay by a factor of about $10^6$, the cost will be:

$$\frac{930\ \$}{\text{MTU month}} \times \frac{1{,}500\ \text{MTU}}{\text{year}} \times 5\ \text{months} = \frac{\$6{,}975{,}000}{\text{year}}$$

For each additional 30 days of holdup the cost would be:

$$\frac{930\ \$}{\text{MTU month}} \times \frac{1{,}500\ \text{MTU}}{\text{year}} \times 1\ \text{month} = \frac{\$1{,}395{,}000}{\text{year}}$$

For LMFBR fuel cooled for 150 days, the cost would be approximately 10 times greater.

*Based on 2/3 $UO_2$ fuel and 1/3 mixed oxide fuel. (See table A-4)

## X. PARTICULATE CONTROL

Radioactive particulates associated with fuel reprocessing will be found in both the liquid and the gaseous effluent streams. The particulates arise due to the various operations (shearing through waste solidification) and as a consequence can include a variety of radioisotopes. They will also be in a variety of chemical and physical forms. Easily condensable vapors should be considered as a source of particulates bearing radioactivity. Most of the radioactive wastes available for discharges are attached to particulates. Included in this category are the isotopes in the actinide series, many of which are highly radiotoxic since they decay by alpha emission. Other radionuclides which potentially can contribute significantly to the waste gas stream particulate makeup include the volatile isotopes, such as ruthenium-106, cesium-134, and the longer-lived isotopes, such as strontium-90 and cesium-137.

The particulate effluent from reprocessing plants will for the most part be soluble and will quite probably be in the nitrate form. The particulate effluents from the solidification process will probably be oxides and thus insoluble. Ruthenium may be an exception, especially from the solidification process, and may be complexed as an organic.

Probably the most acceptable theoretical estimates of discharged radionuclides were made by Oak Ridge scientists (Ref. B-8). These estimates appear conservative, however, as indicated by the gaseous discharges measured at the NFS facility (Ref. B-17). An annual discharge of 1.0 to 10 Ci of beta-gamma fission products, excluding tritium, noble gases and the iodines, currently is the most reasonable estimate. The routine annual gaseous discharge of actinides at the USAEC Rocky Flats Plutonium Recovery Facility (Ref. B-37) averaged 2.4 millicuries from 1953 to 1970. Fuel reprocessing plants may be expected to have higher discharges (see table B.2).

Control Techniques - Particulates are generally collected from gaseous waste streams through the use of inertial separators (cyclone, or gravity settling), filtration (fabric, glassfil, sandbeds, HEPA), precipitation (electric, thermal) sonic agglomeration, and liquid scrubbing. The specific system used is dependent upon the type of source and efficiency of control desired.

Filtration - Because of the concern regarding the inhalation of radioactive particulate material, high efficiency filtration systems for removal of particulates from air have received the primary emphasis in the nuclear industry and are widely used (Ref. B-32). Particulate filters and materials of a variety of types are available. The materials include cellulose-asbestos, glass, glass-asbestos, plastic fibers and ceramics. Filters are generally classified as panel (viscous impingement) filters which are coated with a tacky substance to increase particle adherence, and extended medium dry-type filters which are called "bag" or "sock" filters. Panel filters are designated Group I and have a low efficiency for small particulates. Bag filters are designated as Group II and Group III (medium and high efficiency, respectively). HEPA (High Efficiency Particulate Air) filters are classified as a special group. Sand filters (Ref. B-33) in general are designed for a specific application, usually for use in cleaning corrosive materials from air streams. Their use in the nuclear field has been limited, but is expected to increase since they can be considered relatively fail-safe. Deep-bed sand filters have been employed at Hanford and Savannah River for reprocessing plant gaseous effluent filtration. The Midwest facility has also installed a deep-bed sand filter for final filtration.

The parameters affecting filter performance, in addition to face velocity characteristics are: resistance, geometrical size, pressure drop, penetration, collection efficiency, dust and holding capacity, particle sizes, loading, humidity, temperature, and chemical resistance.

Liquid Scrubbing - This method has been used in cleaning the off-gas stream from solidification processes (Ref. B-34 and B-35). Scrubbers are used as initial cleaning devices and the scrubber solutions can be recycled to the process streams. Experience is limited to the Idaho Chemical Processing Plant and the prototype work conducted under the Waste Solidification Engineering Prototype program conducted by Battelle at Hanford.

Control Systems Evaluation - The various methods of particulate control may be grouped in the order of increasing efficiency as follows:

Fair

Group I panel filters

Good

a) Group II bag filters

b) Deep-bed sand or fiberglass filters

c) Scrubbers

Better

Group III bag filters

Best

HEPA filters

The anticipated performance of fibrous filters can vary from 2 to 99.97% retention for 0.3 micrometer particulates and up to 100% for 10 micrometer size. HEPA filters have consistently demonstrated penetrations less than $0.3 \times 10^{-3}$ (99.97% efficiency). However, the overall system efficiency is highly dependent on filter integrity, proper installation, operating conditions (particularly air flow rates and loading), and aging characteristics. Adequate in-place testing is necessary since improper installation or filter damage in shipment or installation could result in leakages up to 30%.

A typical filter installation consists of a prefilter (roughing filter) followed by two HEPA filters in series or by a HEPA filter and a sand filter. The prefilter generally has a rating of greater than 75% (Group III for 1.0 micrometer particles) and offers a considerable cost savings by reducing mass loadings on the more costly HEPA filters. The HEPA filters are rated at a minimum efficiency of 99.97% for 0.3 micrometer particulates. The reported efficiency of deep-bed sand filters for submicrometer particles is greater than 99%. Thus, the overall decontamination factor for the filter themselves should be on the order of $10^5$. However, the efficiency of the system is also dependent upon factors such as leak tightness and aging characteristics. The minimally acceptable in-place test results would probably be in the order of 99.97% for a typical HEPA system (it is emphasized this is a system test). The system would probably be assigned a credit of 99% to 99.9%, the actual credit depending on the testing frequency.

Capital and operating costs are highly dependent on volume flow rates. Based on Silverman's estimates (Ref. B-36), a HEPA filter would have a capital cost between $200-800/1000 cfm and annual operating costs between $100-450/1000 cfm depending upon specifications, corrosion resistance, and other operating conditions. A pre-filter for reducing the HEPA loading would cost between $100-300/1000 cfm for capital cost and $15-25/1000 cfm for annual operation. Using approximate costs and a total flow of 60,000 cfm the system costs would be:

| | HEPA | Pre-filter |
|---|---|---|
| Capital cost (X60) | $48,000 | $12,000 |
| Annual operating costs (X60) | $18,000 | $ 1,200 |
| 40-year operating costs (present worth @10%) | $176,000 | $11,700 |
| Total costs (1960 prices) | $224,000 | $23,700 |
| Conversion to 1970 price - (Miller-Stevens index 1.28) | $287,000 | $30,400 |

The costs of a glass-fiber or sand deep-bed filters were reported by Silverman (Ref. B-36) to be between $2,860-5,500 capital costs per 100 cfm with associated operating cost between $400 and $800 per 100 cfm (based on only depreciation and air-moving costs). Taking a representative tidal flow of 60,000 cfm the cost of the deep-bed system would be:

| | |
|---|---|
| Capital cost ($3,500 x 60) | $210,000 |
| Annual operating cost ($800 x60) | $ 48,000 |
| 40-year operating costs<br>(present worth @10% discount) | $470,000 |
| Total cost (1960 prices) | $680,000 |
| Conversion to 1970 prices Total | $870,000 |

The reported capital cost for the sand-bed filter, discharge stack, and associated equipment is $400,000 for the Midwest facility (Ref. B-33). The corrected capital cost to 1970 for a deep-bed sand filter is $269,000 which agrees well with the estimate for Midwest.

The total filter installation and 40-year operating costs at reprocessing plants are:

| | 1970 Present Worth @10% |
|---|---|
| Prefilter + HEPA filter + Deep-bed sand filter | $1,187,000 |
| Prefilter + HEPA filter + HEPA filter | $ 604,000 |

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# APPENDIX C

# RADIOLOGICAL DOSE AND HEALTH IMPACT CONVERSION FACTORS

## I. INTRODUCTION

Radioactive materials released into the environment from nuclear fuel reprocessing become dispersed in the surrounding media (air, water, etc.) and ultimately may produce health effects in man. The impact of a given radionuclide release on the population surrounding a source is assessed here in terms of three factors: (1) a dilution factor to calculate the concentration of the released activity in the medium of interest, (2) a medium concentration to dose conversion factor, and (3) a risk factor which relates the likelihood of a given biological effect to an absorbed dose of one rad.[1] These factors are discussed below for tritium, krypton-85, the radioiodines, and alpha-emitting transuranics (such as plutonium).

## II. PATHWAYS

Releases of radionuclides from a nuclear fuel reprocessing plant can occur by venting through an exhaust stack to the atmosphere or drainage to a nearby waterway. The principal pathway of concern in assessing the health impact due to nuclear fuel reprocessing plant operations is the atmospheric pathway because such releases can become dispersed in any direction and lead directly to radiation exposure to man. The significance of the water pathway is expected to be quite small for future reprocessing plants because presently proposed designs do not plan on any releases to waterways.

Atmospheric dispersion of radioactivity has been discussed by a number of authors (Ref. C-1, C-2) and for the case of interest here, a

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[1] Rad - The unit of energy imparted to matter by ionizing radiation and equal to .01 J/kg in any medium.

gaussian plume diffusion model is usually assumed to be the best choice. At distances relatively close to a source (2-3 km) this model can predict the air concentration of krypton-85, for example, within a factor of two or three. Its applicability at longer distances depends upon the local weather conditions at the time of radioactive release and the topography. For unstable atmospheric conditions, it may be reasonably accurate as far as 10 km from a source. Since the average radionuclide concentrations in air around sites have been calculated for distances as far as 80 km (50 miles) from the source, it is obvious that the validity of the atmospheric transport model used is an important limitation. However, the point here is to examine the general case and provide an overall index of health risk; the risk from a particular plant will depend on the details of the local meteorological situation. For worldwide distribution of gases, uniform dispersion was assumed in determining air concentrations.

An important radionuclide pathway for man is the direct contamination of foodstuffs - particularly milk. For iodine-131, this pathway has been studied extensively by several authors including Garner (Ref. C-3) and Bryant (Ref. C-5, C-5). Long-term buildup of the isotope iodine-129 may be important due to its half-life of $17 \times 10^6$ years but appreciable buildup has not been documented in the literature. The short-term behavior of iodine-129 has been considered, however (Ref. C-6), and health risk estimates are given for both iodine-129 and iodine-131.

## III. MEDIA CONCENTRATION TO DOSE CONVERSION FACTORS

Organ or total body dose estimates are critically sensitive to assumptions concerning the route of uptake, the amount of radioactivity inhaled or ingested daily, the fraction of activity retained in the organ of interest, and the residence time of the activity in various parts of the body. The final necessary elements entering into the dose computations are the physical considerations of organ mass and radionuclide distribution within the organ. In the present state of the art, the complexities of the radionuclide distribution within organs are nearly always circumvented by assuming a uniform depositon. Information concerning the other inputs is based mainly on empirical evidence, largely gathered from fallout studies and medical investigations. In order to reduce the number of variables to be considered in dose calculations, the International Commission on Radiological Protection (ICRP) has postulated a "standard man;" i.e., a model system having standardized biological parameters based on either average values or best estimates as listed in the scientific literature. The standard man is a hypothetical adult industrial worker and it is not clear to what extent parameters so defined are applicable to an environmentally exposed population.

For particular radionuclides, the sensitivity of certain age groups may be the limiting factor. In the case of iodine-131, the Federal Radiation Council (Ref. C-7) has defined children as the most sensitive population group and, therefore, the biological parameters used in these media to dose conversion factors are not based on standard man. Rather, models appropriate for children's thyroid glands and thyroid metabolism have been used. For the other radionuclides considered here, little is known concerning differences between adults and children. Such differences are seldom considered in the literature. This being the case, the conversion factors listed in the subsequent sections, while adequate, must be considered only as first order approximations and not as definitive dose estimates from environmentally distributed radionuclides.

Media concentration-to-dose conversion factors are defined below for krypton-85, tritium, iodine 131, iodine-129, and some of the actinides. Other radionuclides are not considered likely to cause significant environmental exposures of the population based on the control technologies discussed in appendix B.

## A. Range of Expected Doses from Krypton-85 Exposure

Since krypton-85 is not metabolized, exposure to humans is limited to external beta and gamma rays and, to a much lesser extent, the dose due to krypton-85 dissolved in body fluids. The health risk from krypton-85 is further limited by the fact that 99% of the decay energy is dissipated by beta rays which have no potential for deep penetration.

Four target organs are considered for these dose and risk estimates: total body, gonad, lung, and skin. In each case it can be shown that only one type of exposure need be considered, the other contributing an insignificant fraction of the dose.

Kirk (Ref. C-8) has recently reviewed the literature on krypton-85 dose and established relationships between the concentration in air of krypton-85 and various organ doses. A review of these results show which radiations and source locations are important. For the whole body, dose and risk estimates can be based on a consideration of external photon exposures, i.e., gamma rays and bremsstrahlung. For genetic risk calculations the gonadal dose, in the case of males, is from exposure from external photons; while for females, the whole body dose estimate can be used. Dose estimates for the lung are based on internal beta dose plus the total body gamma-ray dose. Skin dose and risk estimates are based on the dose delivered by external beta radiation after making an appropriate allowance (0.25) for the shielding provided by clothing and the nonviable epithelium.

## B. Range of Expected Doses from Tritium Exposure

Dose estimates from tritium exposure have been based on the assumption that the isotope is contained in body water (Ref. C-9). However, chronic exposure to environmental tritium has been shown to lead to incorporation into organic molecules from which tritium is lost at a slower rate than from body water (Ref. C-10, C-11). If it is assumed that, under equilibrium conditions, all body hydrogen (7.0 kg in standard man) is uniformly labelled, a sustained concentration of 1 μCi/1 body water would lead to a body burden of 63 μCi, as opposed to 43 μCi if, as in the ICRP model only, distribution in body water alone is considered. Evans (Ref. C-10) found that tritium was not, in fact, uniformly distributed through deer tissues and, assuming that his observed factors are applicable to man, has calculated that the body burden carried by standard man at a sustained concentration in body water of 1 μCi/1 would be 60 μCi, i.e., higher by a factor of 1.4 than that based on the ICRP model. While Evans' factor has been adopted in some dose calculations, a factor of 1.5 (63/43), although only marginally different, may be a more appropriate value to use for calculations in man and, therefore, it is used here.

Since it is apparent that, under chronic exposure conditions, tritium may become incorporated into the genetic material (DNA), it has been suggested that the relative biological effectiveness of tritium in terms of genetic effects may be greater than unity as a result of DNA degradation from transmutation and recoil processes in addition to that due to absorbed energy from ionization processes due to beta emissions. However, from both experimental (Ref. C-12) and theoretical (Ref. C-13) considerations, it has been concluded that it is the absorbed dose to mammalian cell nuclei from incorporated internuclear tritium which determines quantitatively the degree of effect (Ref. C-24). The assumption made in these calculations that the appropriate value for quality factor for tritium dose equivalent estimation is 1.0 as recently adopted by the National Council on Radiation Protection and Measurements (NCRP) (Ref. C-14).

A sustained concentration of 1 μCi tritium per liter of body water would thus be equivalent to a specific activity (assuming uniform labelling of all body hydrogen) of $9 \times 10^{-3}$ μCi tritium/g hydrogen, and would deliver an annual dose to body tissues of approximately 100 mrem.

The concentration of tritium in body water resulting from exposure to tritium in air is obtained by diluting the daily intake of tritium by inhalation into the 43 liters of body water with a biological half-life of 12 days. This amount of tritium is doubled to account for absorption of tritium through the skin. This leads to an annual dose of $1.7 \times 10^{6}$ rem[2] for an air concentration of 1 μCi tritium/cm$^3$.

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[2] In the case of beta and gamma rays emitted by fuel reprocessing plant effluents, the quality factor is one and the dose equivalent in rems is identical to the dose in rads. Where the effects of such effluents are considered in this report, doses are expressed in rem units and biological effects are presented on a per rem basis.
REM - The rem represents that quantity of radiation that is equivalent--in biological damage of a specified sort - to 1 rad of 250 kVp x rays.

## C. Range of Expected Doses from Iodine-129 and Iodine-131 Exposures

Atmospheric releases of iodine from fuel reprocessing may result in an accumulation of iodine-129 and iodine-131 in the thyroid glands of persons living in the surrounding area. The pathway potentially most hazardous to man for isotopes of iodine is the grass-cow-milk chain, particularly in cases where the milk is not diluted with uncontaminated supplies. Direct deposition on foliage will be the only significant route of contamination of edible herbage for iodine-131 and is likely to be the most important for iodine-129, at least over the duration of plant operation.

Because of the long half-life of iodine-129, recycling through the soil should be considered. In organically rich soils, the iodine will be strongly bounded to the soil, but it will be leached rather rapidly from other types of soils. In any case, plants will incorporate iodine-129 in ratio proportional to the amount of natural iodine-127 available. The actual amount of iodine-129 incorporated will depend on the location of the reprocessing plant, and the specific activity of the iodine-129 (curies of iodine-129 per gram of iodine) in each component of the terrestrial pathway will change as a function of time as build-up in the soil increases. At any given time the specific activity in the ecological chain will be somewhat less that the specific activity of the iodine-129 in the air. In many cases the specific activity will be much less because of the large stable iodine reservoir in soils and other parts of the terrestrial pathway.

When considering the exposure of individuals to iodine isotopes via the grass-cow-milk chain, the population potentially at greatest risk is young children consuming fresh milk (Ref. C-15, C-16). From the data of Durbin, et al. (Ref. C-17) the average daily intake of whole cows milk by U.S. children over the first year of life is about 760 ml. Appropriate representative data to define the relationship between the amount of iodine ingested by a 6-month-old child and its concentration in the critical organ, the thyroid gland, are (Ref. C-6); thyroid weight, 1.8 g; fraction of ingested iodine reaching critical organ, 0.35; and biological half-life of iodine in thyroid, 23 d. Equivalent data for adults, appropriate to the calculation of average population doses, are: daily milk consumption, 500 ml (Ref. C-17); thyroid weight, 20 g; fraction ingested reaching critical organ, 0.3; and biological half-life in thyroid, 138 d (Ref. C-6). Use of these values yields an annual dose to the adult thyroid of 0.29 mrem for iodine-131 or 1.9 mrem for iodine-129 for a concentration of 1 pCi/l of each isotope. The corresponding annual doses to the thyroid of children whose daily consumption of milk during the first year of life contains 1 Ci/l of the respective radionuclides are 4.3 mrem for iodine-131 and 6.3 mrem for iodine-129. The media conversion factors presented in table C.1 are also derived from considerations discussed in references C-6 and C-7, an assumed grazing area for a dairy cow of 80 $m^2$ per day and an iodine deposition velocity of 0.5 cm/s (Ref. C-5).

TABLE C.1

Milk Concentrations of Iodine-131 and Iodine-129 from Given Input Concentration and Corresponding Doses

| Input Concentration of Respective Nuclide | Milk Concentrations | |
|---|---|---|
| | Iodine-131 | Iodine-129 |
| 1 pCi/m$^2$ (ground surface) | 0.20 pCi/l | 0.28 pCi/l |
| 1 pCi/m$^3$ (air) | $6.2 \times 10^2$ pCi/l | $2.4 \times 10^3$ pCi/l |
| | Annual Dose to Child Thyroid | |
| | Iodine-131 | Iodine-129 |
| 1 pCi/m$^3$ (air) | 2.7 rem | 15 rem |
| | Annual Dose to Adult Thyroid | |
| | Iodine-131 | Iodine-129 |
| 1 pCi/m$^3$ (air) | .18 rem | 4.6 rem |

Estimates of the specific activity (μCi iodine-129/g total iodine) in the thyroid gland corresponding to an annual dose of 1 rem are, for an adult, 2.3, and for a 6-month child, 4.1 (Ref. C-6). Adoption of a value of 0.44 rem/yr as the dose delivered to a thyroid gland containing 1 μCi iodine-129/m$^3$ total iodine would thus appear to be a reasonable estimate for all cases. The mean concentration of stable iodine in the atmosphere is given as 0.2 μg/m$^3$ (Ref. C-18). Using this value, it can be shown that an air concentration of 1 pCi iodine-129/m$^3$ would lead to an annual thyroid dose of 1.8 rem. In areas where the atmospheric concentration of stable iodine-127 is low, the dose could be up to 40 times higher; this upper limit on the dose is set by the amount of stable iodine-127 released in fuel reprocessing. Thus, for adults the higher value presented in table C.1 of 4.6 rem/yr per $pCi/m^3$ of iodine-129 in air is selected for use in this study.

## D. Range of Expected Doses from Plutonium-239 and Other Actinide Exposures

The potential risks from inhalation of plutonium-239 depend on whether the plutonium is in a soluble or an insoluble form. Present experience indicates that, in the case of fuel reprocessing, the plutonium will be present in the environment in a relatively insoluble form and the present dose estimates are based on this assumption. There is also evidence that a considerable fraction of plutonium-239 inhaled in insoluble form is translocated, largely to the bronchial and mediastinal lymph nodes (Ref. C-19). Since the risk to be pulmonary region depends upon both the amount of plutonium in the organ and its microdistribution, the region containing the largest amount of plutonium may not be the region at greatest risk. Particularly, since the relative sensitivity of the various cell types encountered has not been established, the dose to the lung from inhaled particules is calculated on the basis of an average dose to the entire pulmonary region for this report. In the case of alpha emitters, such averaging is obviously inappropriate if there are only a few particles present. ICRP Publication No. 6 (Ref. C-20) recognizes this and states, "in the case of the lung, an estimate of the dose equivalent to the critical tissue determined merely by the product of quality factor and mean dose may be greatly in error, but further experimental evidence is needed before a better estimate can be made."

In this report, dose estimates are based on the new ICRP lung model (Ref. C-21, C-22). The biological half-life of material in the lung (pulmonary region) is assumed to be 1,000 days. Using this model, sustained exposure to an air concentration of 1 pCi/m$^3$ of plutonium-239 in insoluble form would lead to a dose rate of 12 rem per year in the pulmonary region. It is assumed that the risk to this region is representative of the total risk to the lung.

Media-dose conversion factors for other actinide radionuclides are related to the plutonium-239 conversion factor by taking into account the effective energy absorbed per disintegration of each radionuclide and the physical half-life as given in ICRP Publication Nos. 2 and 6 (Ref C-9, C-20). Table C.2 gives the conversion factors used in this study for several actinides relative to plutonium-239.

TABLE C.2

Actinide Air-Dose Conversion Factors
Relative to Plutonium-239

| Radionuclide | Relative Conversion Factor[a] |
|---|---|
| Pu-238 | 1 |
| Pu-239 | 1 |
| Pu-240 | 1 |
| Pu-241 | 0.001 |
| Am-241 | 0.25 |
| Cm-242 | 0.17 |
| Cm-244 | 0.33 |

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[a] Plutonium-239 Conversion Factor - (12x10$^6$ rem/yr)/(1 μCi/m$^3$).

## E. Summary

Table C.3 summarizes the media-dose conversion factors presented in this section. Conversion factors are expressed in terms of rem/yr resulting from continuous exposure to concentrations expressed in $\mu$Ci/cm$^3$ of air.

TABLE C.3

Summary of Media-Dose Conversion Factors

| Radionuclide | Critical Organ | Conversion Factor (rem/yr)/($\mu$Ci/cm$^3$ air) |
|---|---|---|
| Kr-85 | Whole body | $1.5\text{x}10^4$ |
| | Gonads (female) | $1.5\text{x}10^4$ |
| | Gonads (male) | $2.0\text{x}10^4$ |
| | Lung | $3.0\text{x}10^4$ |
| | Skin | $50.0\text{x}10^4$ |
| H-3 | Whole body | $1.7\text{x}10^6$ or<br>100 (rem/yr)/($\mu$Ci/cm$^3$ water) |
| I-129 | Infant thyroid | $15.0\text{x}10^{12}$ |
| | Adult thyroid | $4.6\text{x}10^{12}$ |
| I-131 | Infant thyroid | $2.7\text{x}10^{12}$ |
| | Adult thyroid | $0.18\text{x}10^{12}$ |
| Pu-239 | Lung [a] | $12.0\text{x}10^{12}$ |

[a] See paragraph IV D regarding consequences of soluble form of plutonium.

## IV. DOSE-RISK CONVERSION FACTORS

Assumptions made in predicting radiation-induced health effects from nuclear fuel reprocessing are given in this section. Consistent with recommendations made in the recent (November 1972) National

Academy of Sciences Committee on Biological Effects of Ionizing Radiation (BEIR) report (Ref. C-23), the health risks presented in this report are based on an assumed linear relationship between absorbed dose and biological effects and that any increased risk is in addition to that produced by natural radiation; i.e., no threshold exists. It is further assumed that health effects that have been observed at dose rates much greater than those likely to be encountered around fuel reprocessing plants are indicative of radiation effects at lower dose rates. Only insofar as any biological repair of radiation damage from low dose rate radiation is neglected do the BEIR health risk estimates represent upper limits of risk. In most cases the risk estimates are based on relatively large doses where cell killing may have influenced the probability of delayed effects being observed. The BEIR risk estimates used in this report are neither upper nor lower estimates of risk, but simply the "best available."

As the BEIR report points out, a nonthreshold linear relationship hypothesis is not in itself sufficient for the prediction of health risk. It is also necessary to assume that all members of the exposed population have equal sensitivity to the radiation insult so that the expression of health risk is independent of how individual exposures are distributed. This requirement is not wholly satisfied. As documented in the BEIR report, differences in sensitivity do exist; for example, children are more radiosensitive than adults. There are two considerations, however, which help validate the application of available mortality data to a consideration of health effects from fuel reprocessing. Some of these data (those taken from Hiroshima and Nagasaki) reflect, to a limited extent, exposure of a relatively heterogeneous population. More importantly, even though the number of health effects will be dependent on the exact makeup of the populations at risk, the relative order of importance of the various pathways of exposure will not be very sensitive to the population characteristics near a given fuel reprocessing plant. Finally, it should be pointed out that the health risk estimates made here assume that the expected radiation effects are independent of other environmental stresses, which may be either unique to the population surrounding fuel reprocessing facilities or unique to the exposed groups considered in the BEIR report.

The numerical risk estimates used in this appendix are primarily from the BEIR report. What must be emphasized is that though these numbers may be used as the best available for the purpose of risk-cost-benefit analyses, they cannot be used to accurately predict the number of casualities. For a given dose equivalent, the BEIR report estimates a range for the health impact per million exposed persons. For example, the BEIR results from a study of the major sources of cancer mortality data yield an absolute risk[3] estimate of 54-123 deaths annually per $10^6$ persons per rem for a 27-year followup period. Depending on the details of the risk model used, the BEIR Committee's relative risk[4] estimate is

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[3] Absolute risk estimates are based on the reported number of cancer deaths per rad that have been observed in exposed population groups, e.g., Hiroshima, Nagasaki, etc.

[4] Relative risk estimates are based on the percentage increase of the ambient cancer mortality per rem.

160-450 deaths per $10^6$ persons per rem. It is seen that the precision of these estimates is at best about a factor of 3-4, even when applied to sample populations studied on the basis of the same dose rates. The application of the BEIR risk estimates to exposures at lower dose rates and to population groups more heterogenous than those studied increases the uncertainty in the risk estimates. Considering the limitations of presently available data and the lack of an accepted theory of radiocarcinogenesis, emphasis should be placed on the difference in risk estimates between the various procedures and countermeasures discussed in this report rather than on the absolute numbers. Where the absolute numbers must be used for risk-cost-benefit balancing, it should be remembered that these risk estimates are likely to be revised as new information becomes available. Notwithstanding these disclaimers, it is also pertinent to note that we are in a better position to evaluate the true risks and the accompanying uncertainties from low levels of radiation than from low concentrations of other environmental pollutants which might affect populations in the vicinity of a fuel reprocessing plant.

## A. Dose-Risk Conversion Factors for Krypton-85

### 1. Total Body Dose-Risk

The BEIR report calculates the cancer mortality risk (including leukemia mortality) from whole body radiation by two quite different models. The absolute risk model predicts about 100 cancer deaths per $10^6$ person-rem while the relative risk model predicts between 160 and 450. An average cancer mortality of 300 annually per $10^6$ person-rem would seem to be an appropriate mean for the relative risk model. The average of the absolute and relative risk models is 200, which is close to the estimates of cancer mortality risk listed as "most likely" by the Committee. Cancer mortality is not a measure of the total cancer risk, which the Committee states is about twice that of the yearly mortality.

The computation of health risk from continuous krypton-85 total body exposure used in this report is the one appropriate for total body irradiation.

Estimated Cancer Risk from Total Body Irradiation

Cancer mortality = 200 deaths per year for $10^6$ person-rem annual exposure. Total cancers = 400 cancers per year for $10^6$ person-rem annual exposure.

2. Gonadal Dose-Risk

The range of the risk estimates for genetic effects set forth in the BEIR report is so large that such risks are better considered on a relative basis for different exposure situations than in terms of absolute numbers. The range of uncertainty for the "doubling dose" (the dose required to double the natural mutation rate) is 10-fold (from 20 to 200 rad); and because of the additional uncertainties in (1) the fraction of presently observed genetic effects due to background radiation, and (2) the fraction of deleterious mutations eliminated per generation, the overall uncertainty is about a factor of 25. The total number of individuals showing very serious genetic effects such as congenital anomalies, constitutional and degenerative diseases, etc., is estimated at somewhere between 1,800 and 45,000 per generation per rad of continuous exposure; i.e., 60-1,500 per year if a 30-year generation time is assumed. This level of effect will not be reached until after several generations of exposure; the risk to the first generation postexposure is about a factor of 5 less.

The authors of the BEIR report reject the notion of "genetic death" as a measure of radiation risk. Their risk analysis is in terms of early and delayed effects observed post partum and not early abortion, stillbirths or reduced fecundity. Because of the seriousness of the genetic effects considered here, e.g., mongolism, the emotional and financial stress would be somewhat similar to death impact. Indeed, many of the effects described are those which lead directly to infant mortality (fetal mortality is excluded). For the purposes of this report this class of genetic effects will be considered on the same basis as mortality data.

### Estimated Serious Genetic Risk from Continuous Gonadal Irradiation

Total risk = 300 effects per year for $10^6$ person-rem annual exposure.

Less serious genetic effects have also been considered by the BEIR Committee. These have been quantified under the categorgy "unspecified ill health." The Committee states that a continuous exposure of one rem per year would lead to an increase in ill health of between 3 and 30 percent.

### 3. Lung Dose-Risk

Due to the insufficient data for the younger age groups, estimates of lung cancer mortality in the BEIR report are only for that fraction of the population of age 10 or more. For the risk estimate made below, it is assumed that the fractional abundance for lung tumors is the same for those irradiated at less than 10 years of age as it is for those over 10[5]. On an absolute risk basis lung cancer mortality is about 26 deaths per annum per $10^6$ persons irradiated continuously at a dose rate of 1 rem per year. This is a minimum value. The BEIR report states that the absolute risk estimates may be too low because observation times for exposed persons are still relatively short compared to the long latency period for lung cancer. Furthermore, lung cancer risks calculated on the basis of relative risk would be larger. For the risk estimate made here, it is assumed that the ratio of absolute risk to the average relative risk is at least a factor of 0.5, i.e., the same ratio as in the case of total body irradiation discussed above.

Estimated Lung Cancer Risk
from Continuous Lung Irradiation

Lung cancer mortality = 50 deaths per year for $10^6$ person-rem annual exposure.

### 4. Skin Dose-Risk

There is no doubt that the dose to the skin delivered by krypton-85 exceeds by about two orders of magnitude the insult to other organs. However, epidemiological evidence of any real risk from such

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[5] An absolute risk estimate is not very sensitive to the inclusions of this assumption since lung cancer incidence is very small in the young.

insults <u>at the dose levels considered here</u> is nonexistent. This is not to say that the linear dose-effect assumption does not hold for skin cancer but rather that the BEIR Committee found that from the extensive evidence they examined "numerical estimates of risk at low dose levels would not seem to be warranted." The authors of this report concurred with the BEIR report. However, rather than defining a zero risk per rad for any radiation insult from krypton-85, an upper limit of risk is proposed. It should be noted that skin cancers are rarely fatal and usually not very debilitating. The estimated upper risk for continuous exposure is:

Skin cancer - upper limit = 3 skin cancers[6] per year for $10^6$ person-rem annual exposure.

## B. Dose-Risk Conversion Factors for Tritium

### 1. Total Body Dose-Risk

The somatic dose-effects from tritium are not expected to be unique. Risk estimates for total body irradiation are based on the same information reviewed in Section A for krypton-85 total body exposure.

### 2. Gonadal Dose-Risk

The genetic risk from tritium per unit gonadal dose is expected to be the same as for the beta and gamma radiation from other isotopes. Some experiments with bacteria (Ref. C-24) have shown that the location of a tritium atom on a particular DNA base can enhance the mutation rate.

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[6] Assuming 30 years at risk exposure and that the incidence of skin cancer will be 10% of all radiation-produced cancers except leukemia, breast, lung, G.I. tract, and bone cancers.

However, if it is assumed that tritium labeling is a random phenomenon, the percentage for such locations that are specifically labeled will be extremely small at the exposure levels considered here. Therefore, the risk estimates for gonadal irradiation from krypton-85 listed in section A are also appropriate for estimating the genetic risk from tritium exposures.

## C. Dose-Risk Conversion Factors for Iodine-129 and Iodine-131

Iodine is concentrated in the human thyroid. Therefore, the insult from radioiodines is important only for the thyroid. The dose to other organs is orders of magnitude less. Two health effects follow high level exposures of thyroid tissue to ionizing radiation: benign neoplasms and thyroid cancer. Though the former are a more common radiation effect, only the risk from cancer is considered here.

While children are particularly sensitive to radiation damage to their thyroid glands, thyroid cancer is not usually a deadly disease for persons in younger age groups but mortality approaches 25% in persons well past middle age. It is not presently known if the radiation-induced cancers which are more frequent for persons irradiated early in life will follow the same patterns of late mortality.

The BEIR report provides risk estimates only for morbidity (not mortality) and only for persons under 9 years of age, i.e., 1.6-9.3 cancers per $10^6$ person-rem years. From the Hiroshima data and other studies it would appear that for persons over 20 years old the radiation-induced thyroid cancer incidence is much lower and may approach zero.

Since information in the BEIR report is not sufficient in itself to estimate the cancer incidence from continuous exposure, tentative risk estimates for this study are also based on risk estimates described in ICRP Publication No. 8 (Ref. C-25) as well as the mean of the BEIR Committee's various estimates of incidence per rem. Infants and fetuses, composing approximately 2.5 percent of the population, are, of course, the most sensitive group. For this group about 150 thyroid cancers may accrue annually per 10  person-rem annual exposure. For the approximately 40% of the populations that is in the 1-19 year age group it is assumed that the incidence is a factor of about 4 less, and that for the balance of the population, it is a factor of 30 less.

### Estimated Thyroid Cancer Risk from Radioiodines

Morbidity for less than age 1:
150 cases per year for $10^6$ person-rem annual exposure.

Morbidity for less than age 20:
35 cases per year for $10^6$ person-rem annual exposure.

Morbidity for more than age 20:
5 cases per year for $10^6$ person-rem annual exposure.

It is unlikely that the annual mortality from this cancer would be much larger than 25% of the rate of incidence. As for other radiation effects, a true measure of the risk from thyroid cancers would be life shortening, but insufficient mortality data prevents such an approach.

## D. Dose-Risk Conversion Factors for Plutonium and other Actinides

The lung cancer mortality risk discussed in Section A is the best available information on the consequences of lung exposure. While it is based on mortality information from miners exposed to alpha emissions from particulates as well as more conventional dose-risk data, it is probably not really adequate for describing the risk from inhaled plutonium. There is good evidence that a fraction of such particulates are cleared from the lungs and relocated into the respiratory lymph nodes. The organ dose received by a lymph node in this case is not really known, but is probably on the order of 50 times the average dose to lung tissue. The ICRP does not consider these highly irradiated nodes to be the organ at maximum risk and preliminary results of animal experiments would tend to confirm their judgment. However, it is not a settled question.

Even if the lung is the critical organ for such exposures, there is little reason to believe that the average lung dose, presently used in health-risk analyses, is really relevant to estimating the risk from air-borne particles. Estimates of the actual dose from discrete sources of alpha radiation are subject to large variability simply because little is known about the volume over which the energy deposition takes place. Even though as many as $4 \times 10^6$ particles (0.2 μm diameter) of plutonium-239 are required to deliver an average energy deposition of 1 rad to the lung, the dose is not evenly distributed; only about 0.2% of the lung volume absorbs the emitted energy. Health risk estimates based on dosimetry are probably unwarranted under these circumstances and use of a body burden approach to health-risk assessments would appear to be a more likely route to success. Unfortunately, experiments allowing this approach are not yet complete. Therefore, the lung cancer risk estimate for exposure to actinides for purposes of this study is as given in Section A for uniform lung exposure.

The dose conversion factor for a soluble form of plutonium will differ from that presented for insoluble plutonium for a given air concentration. However, the associated risk (expressed in effects induced per unit air concentration) resulting from the soluble form is expected to be the same order as for the insoluble form as analyzed here.

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# APPENDIX D

## Radiation and Economic Impact Analysis Techniques

## I. INTRODUCTION

This appendix presents the calculational techniques used in arriving at radiation dose and health risk estimates for the general population due to nuclear fuel reprocessing and the general approach taken to assess the economic impact of effluent controls on the industry. The dose and risk estimates are made for individuals living at 3 kilometers from a reprocessing plant, the regional population within 80 kilometers (50 miles) of a reprocessing plant, for the total United States population (including that of the regional zone), and for the world population (excluding the U.S. population). The dose and risk estimates are made for a representative single five tonne per day plant and for the projected industry as a whole. Only releases via the atmospheric route are considered since future plants are not expected to release significant quantities of radioactive material in liquid effluents. One present facility has significant liquid releases and this case should be considered on an individual basis.

## II. ESTIMATED REGIONAL POPULATION EFFECTS

The regional population around a reprocessing plant may be exposed to higher radioactive material concentrations due to releases from the plant than is the total U.S. population. Thus, this regional population group is considered as a special case. The total regional population dose received by a specific body organ (organ i) exposure to a specific radionuclide (radionuclide j) is estimated by using the following equation:

$$D_{ij} = Q_j \quad \left(\frac{\chi}{Q}\right)_{(3\ \mathrm{km})} \quad F_{ij} \quad C \quad P_R \quad \text{person-rem/yr}$$

Where:

$Q_j$= Release rate of radionuclide j from the fuel reprocessing plant in curies released per second (Ci/s). The average annual release rate is determined by using the curie/tonne values in appendix A, the plant decontamination factors in appendix B, and the size of the plant in tonne/day capacity. For this evaluation, the capacity of a single plant is taken as 5 tonne/day which is equivalent to 1500 tonne/year. It is assumed that a plant operates at full capacity.

$\left(\frac{\chi}{Q}\right)_{(3\ \mathrm{km})}$ = The meteorological dispersion factor, i.e., the radionuclide concentration in air, $\chi$, 3 km from the plant per unit of release rate from the plant [($\mu$Ci/cm$^3$)/(Ci released/s)]. A distance of 3 km is chosen as a reference point since the air concentration at this point is generally not significantly affected by the stack height of the plant. Using the methods referenced in appendix C, $\chi$/Q values have been calculated for many sites by many people. In this evaluation, a value for $(\chi/Q)_{3km)}$ of 5 X $10^{-8}$ ( Ci/cm$^3$)/(Ci/s) is considered to be representative of a typical plant. This value is assumed to be uniform in all directions around the plant. Actual plant values may vary from the above value by as much as factors of five or ten.

$F_{ij}$ = The pathway dependent dose conversion factor which gives the dose to organ i due to a medium (air, food, or water) concentration of radionuclide j [(rem/year)/( Ci/cm$^3$)]. For example, iodine exposure of the thyroid gland by inhalation and milk ingestion would have two separate dose conversion factors. The dose conversion factors used in this study are given in appendix C. For radionuclides with long retention times in the body, e.g., the actinides, the conversion factor represents the equilibrium dose rate resulting from a continuous factor represents the equilibrium dose rate resulting from a continuous constant intake for several years.

C = The regional dilution and population distribution correction factor. It is a ratio of the average individual dose within 80 km of the plant to the average individual dose at 3 km. It takes into account increased dilution as the radionuclides are transported further from the plant and an uneven distribution of population around the plant. It can be calculated theoretically by assuming a population distribution (Ref. D-1) or it can be determined from population dose calculations around real plant sites. For this study, the results of dose calculations for about 50 reactor sites (Ref. D-2) were analyzed and a value of 0.028 (rem/person within 80 km)/ rem/person at 3 km) was obtained. Theoretical results are similar. This value was assumed to be the same for all nuclides. Fuel reprocessing plants are assumed to be located at sites with population characteristics similar to those at reactor sites. Individual plant correction factors may vary by as much as a factor of five from the average value given above. The distance of 80 km (50 miles) was chosen as a cut-off on regional calculations since the distance is large enough to include any nearby large population center yet small enough so that the area affected can be considered a local area.

$P_R$ = The populations within 80 km of the fuel reprocessing plant site. The population values of the above-mentioned 50 reactor sites, taken primarily from environmental reports, lead to an average population around a site of $1.5 \times 10^6$ people in 1980. Population sizes around individual plants can vary from this by a factor of three. The doubling time of this population is assumed to be about 40 years. For purposes of characterizing age-specific factors, 2.5% of the population is considered to be under 1 year old, 45% between 1 and 20 years old, and the remainder over 20. One-half of the population is used for genetically significant dose calculations because of the child-bearing potential.

Using the above factors, the dose equation becomes:

$$D_{ij} = (Q_j \text{ Ci/s}) \times (5 \times 10^{-8} \frac{\mu\text{Ci/cm}^3}{\text{Ci/s}})$$

$$\times (F_{ij} \frac{\text{rem/yr}}{\mu\text{Ci/cm}^3}) \times (0.028) \times (1.5 \times 10^6 \text{ people})$$

$$\times (1.5 \text{ to take into account population growth})$$

$$= 0.0032\ Q_j\ F_{ij} \text{ person-rem/year}$$

This represents the average annual population dose for a 40 year period of constant emissions during constant plant operation. The equation can be modified further by considering that:

$$Q_j = \frac{A_j \times 1500}{3.15 \times 10^7 \times d_j} \text{ Ci/s}$$

where:

Aj = Curies of radionuclide j per tonne of fuel reprocessed (appendix A)

dj = Plant system decontamination factor for radionuclide j (appendix B) and 1500 tonne are processed per year at a 5 tonne per day capacity, and there are $3.17 \times 10^7$ seconds per year).

Therefore:

$$D_{ij} = 1.5 \times 10^{-7} \frac{A_j}{d_j} F_{ij} \qquad \text{average person-rem/year}$$

The individual dose rate at 3 km is estimated by taking out the population-related factors of the above equation (C, $P_R$, and population growth). This results in:

$$D_{ij} = 2.4 \times 10^{-9} \frac{A_j}{d_j} F_{ij} \qquad \text{average millirem/year}$$

The regional health risk due to doses to organ i, H , is determined by multiplying the total dose values for organ i by the health risk conversion factor, $J_i$ (health risks/person-rem), for organ i, Therefore:

$$H_i = J_i \sum_j D_{ij}$$

where the $J_i$ values are those given in appendix C. The total regional health risk for one plant, H, is:

$$H_{REG.} = \sum_i H_i = \sum_i \left(J_i \sum_j D_{ij}\right)$$

The total annual fuel reprocessing industry effect on the regional populations is obtained by multiplying the effect for one plant by the number of operating plants in any given year (appendix A).

## III. ESTIMATED EFFECTS ON THE UNITED STATES POPULATION

Several radionuclides released in the gaseous effluents from a fuel reprocessing plant spread from the local region to all or part of the total United States land area before being diluted around the world. The method of estimating effects to the U.S. population depends on the radionuclide being considered. The radionuclides considered in this study are tritium, krypton-85, iodine-129, and the actinides. Iodine-131 is considered to be only a regional problem because of its short half-life and the rate of local deposition assumed. Krypton-85 and tritium expose the U.S. population only briefly before subsequent dispersion throughout the world, while iodine-129 and the actinides are assumed to build up only on U.S. soil. In fact, the impact of these latter two may be restricted to a region.

### A. Tritium

It is assumed that most of the tritium released as a gaseous effluent from the reprocessing plant will enter into the hydrologic cycle. The released tritium is assumed to fall out or rain out over the Eastern United States [$3.7 \times 10^6$ km$^2$ ($1.5 \times 10^6$ mi$^2$)] and becomes diluted by the annual rainfall [40 inches (average 100 cm)] (Ref. D-3) over this area. With some further dilution by uncontaminated water, this then becomes the water concentration to which the population of the Eastern United States (80% of the total U.S. population) along with their animals and crops are exposed.

The total population dose is then given by:

$$D_{ij} = (C_j \frac{\mu Ci}{ml}) \times f \times (F_{ij} \frac{rem/yr}{\mu Ci/ml}) \times 0.80\ P_u \text{ person-rem/yr}$$

where:

$C_j$ = The water concentration of tritium determined by diluting the yearly input, $Q_j$ (Ci/s) x 3.15 x $10^7$ s/yr, of tritium to the environment by the average annual rainfall over one-half of the United States.

f = A factor to take into account dilution of tritium by uncontaminated water from deep artesian wells and the fact that not all tritium will fall out over the Eastern United States. Some, perhaps a large portion, will fallout over the Atlantic Ocean and will be diluted in a larger volume. For this evaluation f = 0.5 is used although it is only an estimate.

$F_{ij}$ = Dose conversion factor from appendix C [100 (rem/yr)/(μCi/ml) for this case.]

$P_u$ = Population of the United States. The population growth of the U.S. is estimated as in figure D.1. One-half $P_u$ is used for genetically significant dose calculations.

Tritium concentration, $C_j$, is related to the environmental input by:

$$C_j = (Q_j \frac{\text{Ci}}{\text{s}} \times 10^6 \frac{\mu\text{Ci}}{\text{Ci}} \times \frac{3.15 \times 10^7 \text{ s/yr}}{(1.5\text{x}10^6 \text{ mi}^2)\text{x}(1.61\text{x}10^5 \text{ cm/mi})^2\text{x}(40 \text{ in})\text{x}(2.54 \text{ cm/in})}$$

$$= 8.0 \times 10^{-6} \text{ Q } \mu\text{Ci/ml}$$

$$= 2.5 \times 10^{-13} \frac{A_j}{d_j} \text{ N } \mu\text{Ci/ml}$$

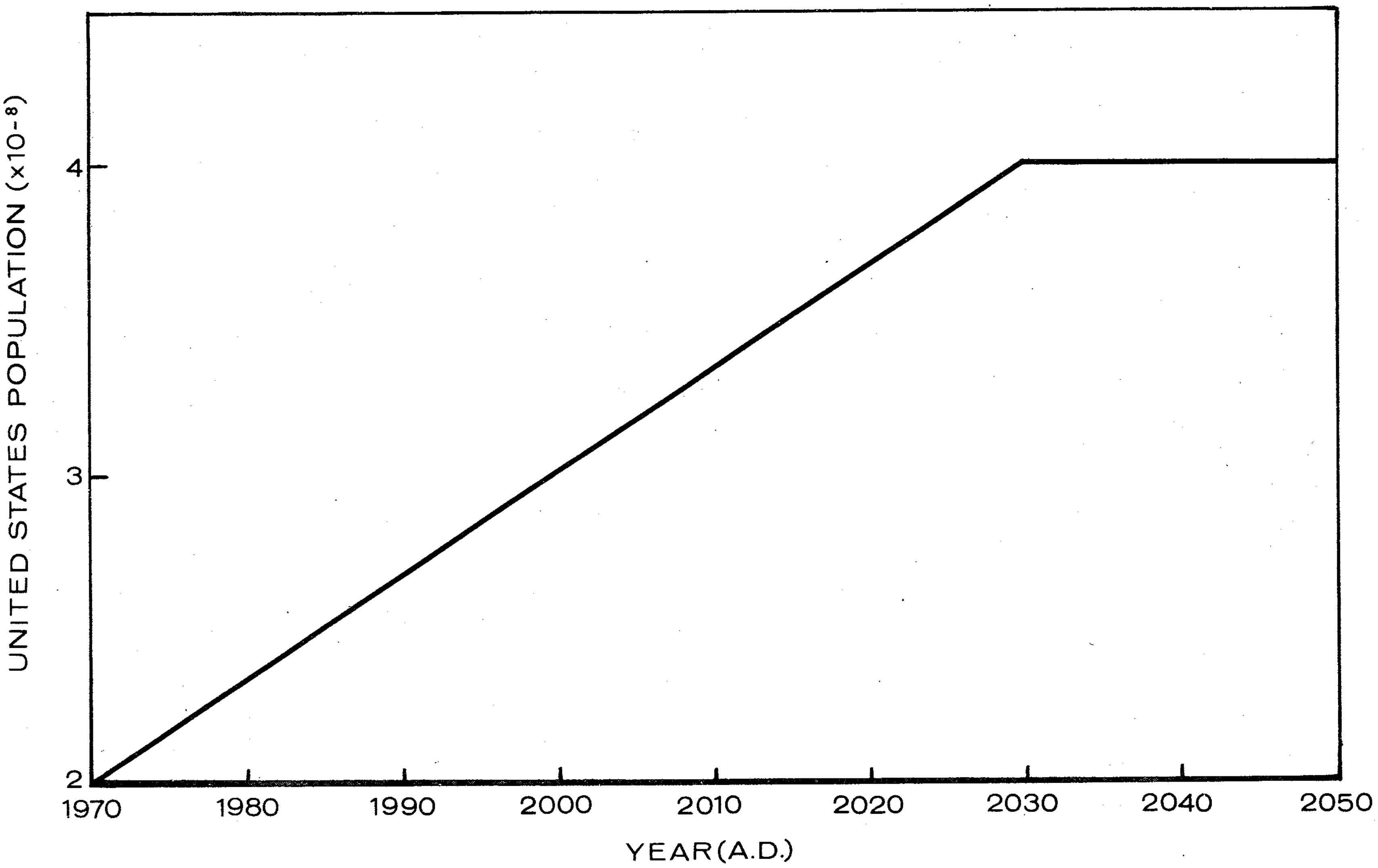


Figure D.1 PROJECTED TOTAL UNITED STATES POPULATION where:

$A_j$ = Curies/tonne - from appendix A.

$D_j$ = System decontamination factor from appendix B.

N = Number of tonnes processed per year. (1500 tonne/yr for a single plant and as in appendix A for the total industry.)

Therefore:

$$D_{ij} = 2.5 \times 10^{-13} \times 0.80\ f\ N \frac{A_j}{d_j}\ F_{ij}\ \ P_u \quad \text{person-rem/yr to U. S. A. population from the fallout of tritium}$$

B. Krypton-85

Part of the population of the Eastern United States is exposed to air concentrations of krypton-85 as it passes from the fuel reprocessing plant to the Atlantic Ocean on its first pass around the world. The dose from this exposure pathway is taken from a study recently performed at the National Oceanic and Atmospheric Administration (Reference D-4). For a plant in Morris, Illinois, releasing one curie of krypton-85 per year, the population-weighted concentration on its first pass over the Eastern United States to the Atlantic Ocean is $2.5 \times 10^{-16}$ person-Ci/cm$^3$. For purposes of this evaluation, this value is considered adequate to use for all plants. This value is then multiplied by total annual releases and dose conversion factors to obtain dose values.

$$D_{ij} = [2.5\text{x}10^{-16} \frac{\text{person-}\frac{\text{Ci}}{\text{cm}^3}}{\text{Ci}}] \text{ x } 10^6 \frac{\mu\text{Ci}}{\text{Ci}} \text{ x N x } \frac{A_j}{d_j} \text{ x } F_{ij} \text{ x } 1.5$$

where N, $A_j$, $d_j$, and $F_{ij}$ are as defined previously. The 1.5 factor is to account for population growth based on a doubling time of 40 years.

Therefore:

$$\boxed{D_{ij} = 3.8 \text{ x } 10^{-10} \text{ N } \frac{A_j}{d_j} F_{ij}}$$

person-rem/yr to U. S. population from release of krypton-85

C. Iodine-129

As a first approximation, all of the iodine-129 release is assumed to fall out over the Eastern United States [3.75 x $10^6$ km$^2$ (1.5 x $10^6$ mi)$^2$] and uniformly mix with the stable iodine in the soil to a depth of 20 cm. This then becomes the specific activity of iodine in the diet to which all persons in this part of the country are exposed. Because of its long half-life (1.6 x $10^7$ years), iodine-129 will build up on the soil and expose the population long after it has been released to the environment. The movement of iodine-129 in the biosphere is not well documented at the present time. However, even with the assumptions that must be made, the first estimate obtained is considered reasonable.

The specific activity of iodine-129 in the soil at time t, curies $^{129}\mathrm{I}/\mathrm{gr}\ ^{127}\mathrm{I}$, is:

$$\text{Specific Activity} = \frac{\sum\limits_{o} \dfrac{N(t') \cdot A_j}{d_j} \cdot f \cdot \dfrac{1}{1.5 \times 10^{6}\ \mathrm{mi}^2 \times (1.6 \times 10^{5}\ \mathrm{cm/mi})^2}}{4 \times 10^{-6}\ \dfrac{\mathrm{g}\ ^{127}\mathrm{I}}{\mathrm{g\ soil}} \cdot 1.5\ \dfrac{\mathrm{g\ soil}}{\mathrm{cm}^3} \times 20\ \mathrm{cm\ depth}}$$

$$= 2.2 \times 10^{-13} \sum_{o}^{t} \frac{N(t')\ A_j}{d_j}\ f\ \frac{\mathrm{Ci}\ ^{129}\mathrm{I}}{\mathrm{g}\ ^{127}\mathrm{I}}$$

where f is the fraction of iodine-129 release that stays in the soil, and it also includes dilution by iodine taken in from other sources, and the other factors are as defined previously. For purposes of this evaluation, f is taken as 0.5. This value is only a rough estimate, however, and may be considerably in error. Likewise, the value of $4 \times 10^{-6}$ g $^{127}\mathrm{I}$/g soil is taken from only one Ref. (D-5) and may not be representative of the Eastern United States. These numbers can easily be changed when better values are determined. Using the dose conversion factor given in appendix C, the annual thyroid population dose rate as a function of time, t, is determined from:

$$D_{ij} = \left(\text{specific activity}\ \frac{\mathrm{Ci}^{129}\mathrm{I}}{\mathrm{g}^{127}\mathrm{I}} \times \left(4.4\mathrm{x}10^{5}\ \frac{\mathrm{rem/yr}}{\mathrm{Ci}^{129}\mathrm{I}/\mathrm{g}^{127}\mathrm{I}}\right)\right.$$

$$\times\ (0.80\ P_u(t))$$

$$= 7.7 \times 10^{-8} P_u(t) \sum_{o}^{t} N(t')\ \frac{A_j}{d_j}\ f\ \ \text{thyroid person-rem/yr}$$

This is the annual population dose (thyroid) due to the total amount of iodine-129 in the environment at the time of calculation. For purposes of determining health risk, the iodine-129 is assumed to expose the population for 100 years beyond the time of release.

D. Actinides

The actinides are assumed to build up in the Eastern United States in the same manner as iodine-129 with the principal exposure pathway believed to be resuspension of the material and inhalation. The annual population dose to the lungs from the buildup of actinides in the environment is:

$$D_{ij} = \frac{\sum_{t'=0}^{t} 0.8\, P_u(t)\, N(t')\, \frac{A_j}{d_j}\, f\, R_s\, F_{ij}}{3.8 \times 10^6 \ \text{km}^2}$$

where $R_s$ is the resuspension factor in terms of $\text{Ci/m}^3$ air per $\text{Ci/m}^2$ on the ground. Based on calculations using fallout data and data from around Rocky Flats (Reference D-6), a value of $R_s = 10^{-6}$ is used in this study. The uncertainity of this value may be of the order of several orders of magnitude. The fraction of actinides released that remains on the soil is taken as 0.5 for this study. Therefore:

$$\boxed{D_{ij} = 2.1 \times 10^{-19} P_u(t) f \sum_{0}^{t} N(t') \sum_{j} \frac{A_j F_{ij}}{d_j}} \qquad \frac{\text{person-rem}}{\text{year}}$$

The dose calculation is performed for plutonium-239 and the total of all the long-lived actinides is about 10 times the plutonium-239 dose for long-term exposure. This results from a consideration of $A_j$ and $F_{ij}$ values in appendixes A and C. For purposes of determining health risk, the actinides are assumed to expose the population for 100 years beyond the time of release.

The health risks from all of these pathways are estimated by:

$$\boxed{H_{US} = \sum_{i,j} D_{ij} J_i}$$

Admittedly, the pathway models used in this section for the determination of environmental effects to the United States population are uncertain and unproven. They are presented as a first order approximation of these effects and the points of uncertainty, especially relative to environmental behavior, are indicative of research needs already identified in these areas.

## IV. EFFECTS ON THE WORLD POPULATION OUTSIDE THE U.S.

Two radionuclides released in fuel reprocessing are expected to result in doses to the world population - krypton-85 and tritium.

### A. Krypton-85

The worldwide dose due to krypton-85 exposure can be estimated by diluting the output from one year of fuel reprocessing into the world's atmosphere ($5.14 \times 10^{21}$ g; sea level air density = 0.00129 g/cm$^3$) (Ref. D-7), and then determining population dose while it decays away. The total yearly dose rate at any instant in time is a combination of contributions from all previous environmental releases of krypton-85. The total population dose to be received by a specific body organ (organ i) over the total time of decay from a one year's input of krypton-85 is given by:

$$D_{i_j}(\text{decay}) = \left(\frac{NA_j}{d_j} \times 10^6 \ \frac{\mu Ci}{yr}\right) \times \left(\frac{0.00129 \ g/cm^3}{5.14 \times 10^{21} \ g}\right) \times \left(F_{i_j} \ \frac{rem/yr}{\mu Ci/cm^3}\right) \times$$

$$\int P_w(t') \times e^{-\lambda t'} dt'$$

where:

$\lambda$ = Decay constant for krypton-85 (0.0645/yr)

$P_w(t)'$ = World population at time t; (t=0 at time of release). The world population is taken as $3.56 \times 10^9$ people in 1970 with an annual growth rate of 1.9%/yr (Ref. D-8). For genetically significant dose calculations, one-half the world population is used. Five percent of the world effect is subtracted to account for the United States contribution to world population dose. The other factors are as defined previously.

Therefore:

$$\boxed{D_{ij}\ (\text{decay}) = 1.9 \times 10^{-8}\ \frac{NA_j}{d_j}\ F_{ij}\ e^{0.019t}}$$

total world person-rem committed by one year's release (less U. S. dose).

where t´ is measured from 1970 to the time of release.

The annual dose rate at time t from the buildup of krypton-85 in the atmosphere from releases is given by:

$$D_{ij}(\text{buildup}) = \sum_{o}^{t} \frac{N(t')A_j}{d_j} \times 10^6 \times e^{-0.0645(t-t')}$$

$$\times \Big(\frac{0.00129\ \text{g/cm}^3}{5.14 \times 10^{21}\ \text{g}} \times F_{ij} \times P_w(t)$$

where t=t´=0 in 1970.

Thus: $$D_{ij}(\text{buildup}) = 8.5 \times 10^{-10}\ \frac{A_j}{d_j}\ F_{ij} e^{-0.0455t} \times \sum_{o}^{t} N(t')e^{+0.0645t'}$$

person-rem/yr to the world's population (less U.S. dose). For genetically significant doses use one-half this value.

To estimate future doses due to the environmental buildup of krypton-85, the above annual dose can be multiplied by $e^{-0.0645T}$ $e^{0.019T}$ and then integrated from T=0 to T=∞. This accounts for radioactive decay and for population growth.

B. Tritium

The worldwide dose due to tritium exposure is estimated by diluting the tritium release into the circulating waters of only the Northern Hemisphere (one-half of $2.7 \times 10^{19}$ 1) (Ref. D-7) and assuming that the Northern Hemisphere's population (80% of the world population) (Ref. D-8) is exposed to the resulting concentration. The U.S. contribution (about 7%) is subtracted out.

The total population dose for organ i that will be received over the total time of decay from a one year's input of tritium is given by:

$$D_{ij}(\text{decay}) = (\frac{NA_j}{d_j} \times 10^6 \frac{\mu Ci}{yr}) \times (\frac{2}{2.7 \times 10^{19} \times 10^3 \text{ ml}})$$

$$\times (F_{ij} \frac{\text{rem/yr}}{\mu Ci/ml}) \times \int P_w(t') \, e^{-0.0562t'} dt'$$

$$\boxed{D_{ij}(\text{decay}) = 5.3 \times 10^{-6} \frac{NA_j}{d_j} F_{ij} \; e^{0.019t}}$$

world person-rem committed

by one year's release of tritium (less U. S. dose) where t is measured from 1970 to the time of release.

The annual dose rate at time t from the buildup of U. S. nuclear power generated tritium released to the environment is given by:

$$D_{ij}(\text{buildup}) = \sum_{o}^{t} \frac{N(t')A_j}{d_j} \times 10^6 \; e^{-0.0562(t-t')}$$

$$\times (\frac{2}{2.7 \times 10^{22} \text{ ml}}) \times F_{ij} \times P_w(t)$$

where: $t = t\infty = 0$ in 1970.

$$D_{ij}(\text{buildup}) = 0.20 \times 10^{-6} \frac{A_j}{d_j} F_{ij} e^{-0.0372t} \sum_{o}^{t} N(t') e^{+0.0562t'}$$

person-rem/yr to world's population from U. S. reactor produced tritium (less U. S. dose). For genetically significant doses use one-half this value.

Again, to evaluate future doses due to the environmental buildup of tritium, multiply by $e^{-0.0562T} e^{0.019T}$ and integrate from $T=o$ to $T=\infty$ to account for population growth and decay.

The health effects resulting from exposure to tritium and krypton-85 are determined by:

$$H = \sum_{ij} D_{ij} J_i$$

(Some radionuclide fuel content and system decontamination factors used in these calculations are given in table D.1)

## Table D.1

## Factors Used in the Assessment of Environmental Radiological Impact of Fuel Reprocessing

| | Activity in Fuel (Ci/MT) | Plant Decontamination Factor |
|---|---|---|
| H-3 | 800.00 | 1.0 |
| Kr-85 | 10,500.00 | 1.0 |
| I-129 | 0.04 | 1000 |
| I-131 | 2.00 | 1000 |
| Actinides | (See Appendix A Table A.4) | $10^9$ |

## V. ECONOMIC ANALYSES

Annual costs should include considerations of debt service, depreciation, and Federal taxes. The first two are dependent on the assumed plant life, salvage value, investment capital and debt. An example of the total annual costs for a 5 tonne/day plant is presented in table D-2 (Ref. D-9). In this regard, the total economic impact upon the investors of the inclusion of a particular control system requires consideration of the cost of money invested over the life of the plant. This, of course, will be somewhat offset by the increased value of the product over the same period.

Several methods are available for presenting the cost ascribable solely to control systems. These methods are described as annual cost, present worth, and total commitment.

In order to obtain estimates of these values, it is necessary to specify first costs (P), interest rate (i), debt lifetime (n) plant life and estimated salvage value (L). In addition a discount factor of 7.5% was used to estimate present worth. For the present analysis, the lifetime is considered to be 40 years, the salvage value is considered negligible, and the effective interest rate is assumed to be either 10% or 24%. These estimates of costs are based upon the assumption that the control systems are add-on facilities and not directly required for processing.

The annual costs are considered to be the sum of capital recovery costs and the annual operating expenses (A.O.E.). Annual costs therefore are estimated from the following equation:

$$\text{Annual cost} = P \times \text{crf} + \text{A.O.E.}$$

TABLE D.2

Estimate of the Economics of a 5 MTU/Day Reprocessing Plant[a]

(Note that "000" is omitted from all money values)

| Year | | Fuel Processed, Metric Tons/Year | Operating Costs | Debt Service | Depreciation[b] | Other Fixed Costs[c] | Total Costs | Gross Receipts at $27.50/kg | Before-Tax Income | Federal Tax[d] | Cash Flow | Obligation |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1971 | -2 | | | | | | | | | | (18,200) | -19,660 |
| 1972 | -1 | | | | | | | | | | (18,200) | -42,470 |
| 1973 | 0 | | | | | | | | | | (18,200) | -68,870 |
| 1974 | 1 | 290 | 7,708 | 2,567 | 9,750 | 2,430 | 22,455 | 7,975 | (14,480) | | (4,730) | -85,370 |
| 1975 | 2 | 690 | 9,975 | 2,567 | 9,130 | 2,430 | 24,102 | 19,000 | (5,120) | | 4,028 | -94,342 |
| 1976 | 3 | 1,200 | 12,070 | 2,567 | 8,420 | 2,430 | 25,487 | 33,000 | 7,513 | | 15,933 | -90,939 |
| 1977 | 4 | 1,320 | 12,520 | 2,567 | 7,800 | 2,430 | 25,317 | 36,300 | 10,983 | | 18,783 | -83,685 |
| 1978 | 5 | 1,500 | 13,215 | 2,567 | 7,170 | 2,430 | 25,382 | 41,250 | 15,868 | 7,090 | 15,948 | -78,568 |
| 1979 | 6 | 1,500 | 13,215 | 2,567 | 6,500 | 2,430 | 24,712 | 41,250 | 16,538 | 7,940 | 15,098 | -73,590 |
| 1980 | 7 | 1,500 | 13,215 | 2,567 | 5,840 | 2,430 | 24,052 | 41,250 | 17,198 | 8,250 | 14,788 | -68,202 |
| 1981 | 8 | 1,500 | 13,215 | 2,567 | 5,220 | 2,430 | 23,432 | 41,250 | 17,818 | 8,560 | 14,478 | -62,324 |
| 1982 | 9 | 1,500 | 13,215 | 2,567 | 4,550 | 2,430 | 22,762 | 41,250 | 18,468 | 8,860 | 14,158 | -55,856 |
| 1983 | 10 | 1,500 | 13,215 | 2,567 | 3,500 | 2,430 | 22,112 | 41,250 | 19,138 | 9,200 | 13,838 | -48,748 |
| 1984 | 11 | 1,500 | 13,215 | 2,567 | 3,270 | 2,430 | 21,482 | 41,250 | 19,768 | 9,480 | 13,558 | -40,820 |
| 1985 | 12 | 1,500 | 13,215 | 2,567 | 2,570 | 2,430 | 20,782 | 41,250 | 20,468 | 9,820 | 13,218 | -32,022 |
| 1986 | 13 | 1,500 | 13,215 | 2,567 | 1,950 | 2,430 | 20,162 | 41,250 | 21,088 | 10,100 | 12,938 | -22,134 |
| 1987 | 14 | 1,500 | 13,215 | 2,567 | 1,330 | 2,430 | 19,542 | 41,250 | 21,708 | 10,400 | 12,638 | -11,016 |
| 1988 | 15 | 1,500 | 13,215 | 2,567 | 620 | 2,430 | 18,832 | 41,250 | 22,418 | 10,780 | 12,258 | +1,242 |

---

[a] 70 percent equity at 16 percent; 30 percent debt at 7 percent.

[b] Sum-of-year digits.

[c] State and local taxes (2.51 percent), insurance (0.25 percent), and interim replacement (0.35 percent).

[d] At 48 percent.

From - Lecture VII - Nuclear Fuel Processing, G.G. Eichhole, 1972 Short Course at Georgia Tech., re: Nuclear Fuels-Management and Economics where P and A.O.E. are obtained from the estimates presented in table B-2. The capital recovery factor (crf) for 40 years at 10% is equal to .105 (Ref. D-10).

Annual costs can be converted to present worth of all disbursements for lifetime, n. This is accomplished by multiplication of the annual costs by the present worth factor (pwf) for the appropriate lifetime and interest rate and by adding the result to the first cost: PW = A.C. (pwf) + P.

The present worth factor for 40 years at 10% is 9.779 (Ref. D-10).

The total control system cost can also be estimated over the life of the plant. Assuming that the principal cost is repaid within 10 years at an interest rate of 10%, the total cost over 40 years is estimated to be as follows:

$$\text{T.C.} = (\text{P x crf}\,{}^{i}_{10}) \text{ x } 10 + \text{A.O.E. x } 40$$

where: $\text{crf}\,{}^{10}_{10} = .163$

The system cost presented in table D-3 is in terms of total cost for both 10% interest with 10 yr debt life and 24% interest with 20 yr debt life. In addition, the present worth for the latter parameters is presented. These figures do not consider the loss of opportunity costs.

The costs to the investors in time and money associated with litigation because of suits by opponents are not presently known, but could be substantial. Introduction of expensive control systems during the initial construction stage and early use might be cost-effective in an overall view even if the system might not be cost-effective on the basis of risk reduction alone.

## TABLE D.3

## CONTROL SYSTEM COSTS

| Isotope | System | First Cost ($10^6$ Dollar) | 10% int. 10 yr. debt 40 yr. life Total Cost ($10^6$ Dollar) | 24% eff. int. 20 yr. debt 40 yr. life Total Cost ($10^6$ Dollar) | 24% eff. int. 20 yr. debt 7.5% discount 40 yr. life P. W. ($10^6$ Dollar) |
|---|---|---|---|---|---|
| Krypton-85 | Cryogenic Distillation | 3 | 8.9 | 19 | 11.5 |
| | Cryogenic Adsorption | 3 | 11.1 | 21 | 12.1 |
| | Freon-Adsorption | 1.5 | 6.4 | 11 | 6.4 |
| Iodines | Caustic Scrubber and AgZ Adsorption | 1.2 | 6 | 10 | 5.4 * |
| Actinides | HEPA Filter (2) | .1 | 2.2 | 2.5 | 1 |
| | HEPA + Sand | .35 | 3.3 | 4.5 | 2.1 |
| Tritium | Voloxidation | 10 | 20 | 54 | 31 |

* The present worth of a caustic scrubber alone is estimated to be $\$3.85 \times 10^6$.

Beyond the normal costs considered, there is also the matter that failure to introduce control systems for effluents that could have worldwide distribution (tritium and krypton-85) could be costly to the Federal government in terms of foreign policy expenditures. The magnitude of these costs is unclear at present, however.

The impact of fuel reprocessing plants upon the economy, e.g., on industries which depend upon the economy, e.g., on industries which depend upon the use of radiosensitive materials has been estimated to be negligible as long as the media concentration of radioisotopes are lower than the constraining levels set for humans by ICRP (Ref. D-11, D-12).

Cost Effectiveness Considerations. The effectiveness of risk reduction of the previously described isotope specific control (appendix B) is dependent upon: (1) the group at risk which is considered, and (2) the specific health effects used as a risk index. The rate of decrease in number of health effects avoided by sequential addition of the individual effluent control systems is dependent upon the order in which the systems are incorporated. The number of health effects avoided by incorporation of the individual systems in the order of decreasing unit cost-effectiveness is presented as a function of the cumulative system control cost in figures D-2 and D-3. The individual component for control of particulates, iodines, krypton, and tritium were assumed to have decontamination factors $10^3$, $10^3$, $10^2$, and $10^2$ with present worth costs of 1, 5.4, 6.4, and 31 million dollars respectively. A system capable of removing the residual pollutants was assumed to be available with a cost of 20 million dollars also. Figure D-2 displays the reduction of the projected risks of mortality (cancer induction and genetically coupled) as a function of cumulative system costs for the regional, Eastern United States, and worldwide population groups. The health risks considered and presented in figure D-3 include nonfatal cancer induction as well. The figures show that in every instance, the particulate control system is the most cost-effective. The iodine control system is the next most cost-effective, except for the situation which considers the estimated mortalities to the world population which are avoided. When considering the world population group, the iodine and krypton control systems appear to be practically equally cost-effective for risk reduction. In consideration of the regional and United States population groups, tritium control appears the next most cost-effective to the particulate and iodine systems although krypton control is not significantly different from that of the tritium control system.

The cost-effectiveness of a combined system for any group at risk can be estimated from the slope of a directed line segment which connects the origin and the appropriate arrow tip representing the combination. The steeper the slope, the greater the cost-effectiveness. It can be seen that the cost-effective excess of a combined system will be necessarily less than that of its most cost-effective component.

FIGURE D - 2 RISK REDUCTION (MORTALITY AND MORBIDITY) vs CUMULATIVE CONTROL SYSTEM COST

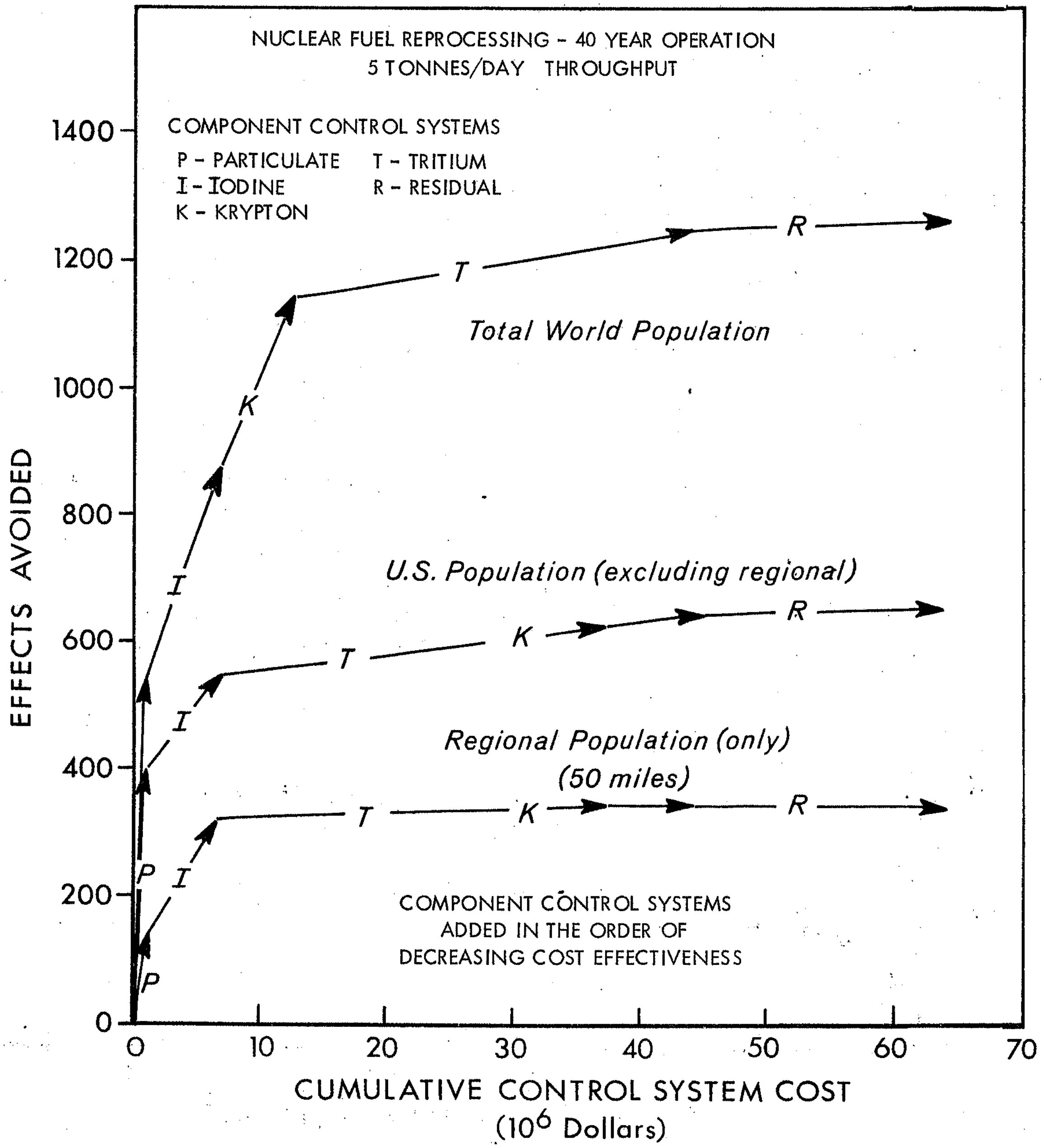
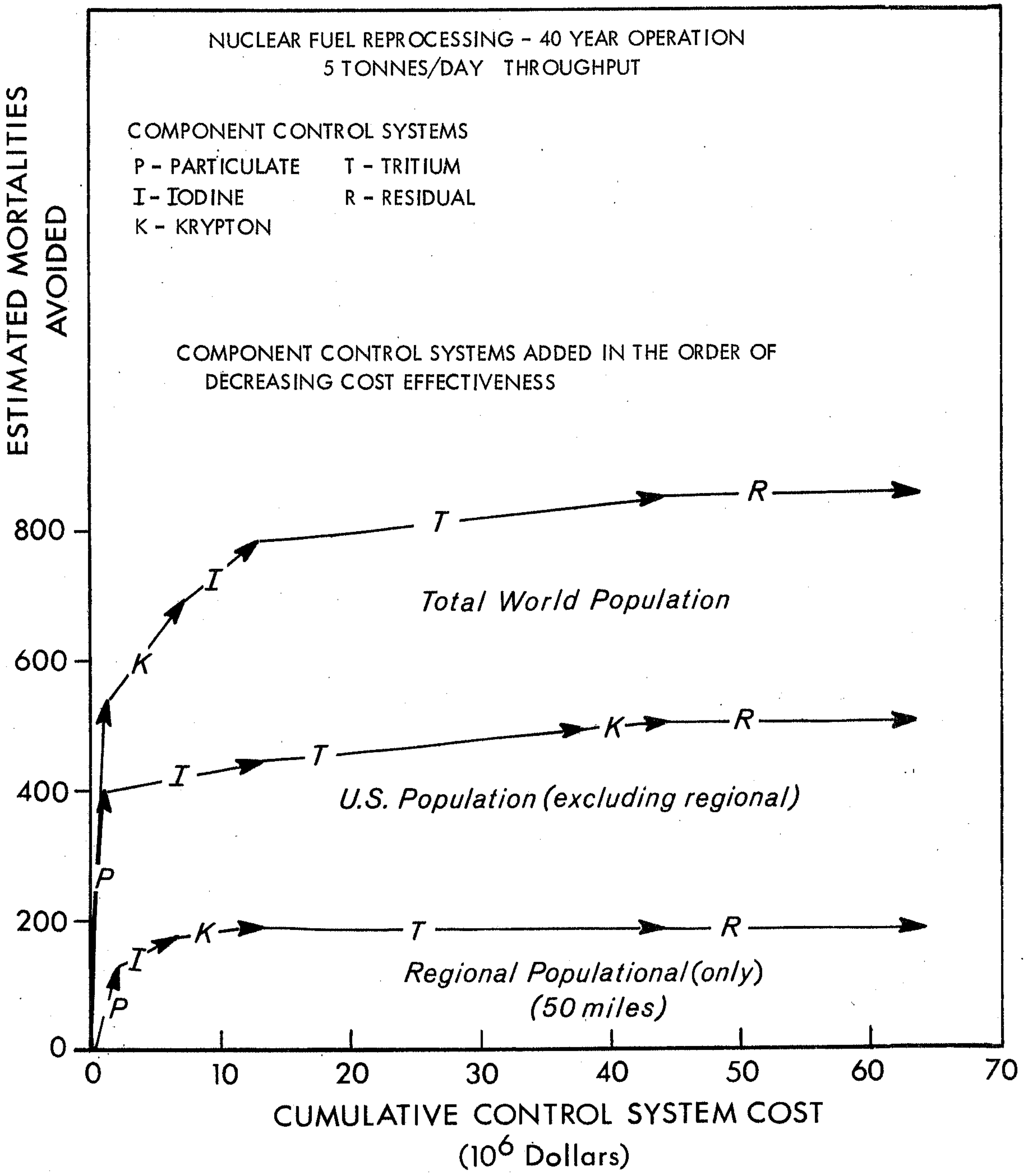


FIGURE D - 3 RISK REDUCTION (MORTALITY) vs CUMULATIVE CONTROL SYSTEM COST



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