EA 451

The MITRE Corporation
Metrek Division

8005140017

April 9, 1980 W85-049

Richard H. Vollmer, Director
Division of Engineering
Nuclear Reactor Regulation
Nuclear Regulatory Commission
Bethesda Office, Room 542
Washington, D.C. 20555

Dear Mr. Vollmer:

Confirming my process disclosure to you in our meeting of March 21st and my prior meeting on March 10th with Messrs. Sydney Miner, Mark Greenberg and Jerrold Carter of your staff, I have developed a cryogenic separation process for the removal of radioactive Krypton 85 from the Three Mile Island nuclear reactor containment building. The process will reduce the estimated 57,000 curies radiation content to a total of 5 curies or 2.5 microcuries per SCF vent air.

The engineering and design details of my process disclosure are shown in:

- Figure 1: "Proposed Process Schematic Diagram for Krypton 85 Removal..."
- Table I: "Basic Process Scheme, Derivation of Kr^{85} Reduction Equation and Calculation of TMI Containment Building Reduction"
- Figure 2: "Reduction of Krypton 85 Curies Radiation..."

The basic process scheme is to link a cryogenic air separation plant to the containment building radioactive atmosphere in a closed recycle. The cryogenic plant will separate the atmosphere into its constituents: oxygen, nitrogen, argon, and enriched Kr + Kr^{85} fraction. All the constituents, except for this enriched fraction, will be recycled to the containment building. The Kr + Kr^{85} fraction will be filled into shielded metal gas cylinders, and the 57,000 curies removed for burial in a remote location. Depending on removal efficiency, the clean-up will require 3.6 to 16.2 days of continuous operation. The cryogenic separation technology has been known for more than 40 years (see M. Ruheman, "The Separation of Gases," 2nd Ed., Oxford Univ. Press, 1949, p. 228-236). The proprietary patentable features of my process are (1) use of a normal krypton fresh make-up feed to enable the delicate, complex vapor-liquid equilibria to function at low krypton solute concentrations in the distillation columns and to sweep out the Kr^{85} and (2) a process combination of air separation plant, krypton distillation column, and molecular sieve filter bed to remove the radioactive Krypton 85.

A 50 Tons/Day oxygen air separation plant will be required. The entire installation, including modifications to add a krypton distillation column, molecular sieve filter bed, supplementary cryogenic refrigeration, and a centrifugal compressor, will cost between $10 to $20 Million. A used air separation plant can be obtained on short notice from Union Carbide (Linde Div.) or Air Products. The fresh make-up gas feed (about 900 liters) can be readily obtained for about $0.70 per liter from three U.S. suppliers: Union Carbide (Linde Div.), Air Products, and AIRCO. Other equipment and installation materials can be purchased new or used from a variety of suppliers. The project schedule would require 11 months: procurement, fabrication modifications and installation - 9 months; start-up, debugging and optimization - 1 month; and removal of the Kr^{85} from the building - 1 month.

My credentials for the validity of the proposed process and project engineering details are as follows:

- Employed 1955-1969 with AIRCO, Inc., second largest cryogenic air separation plant gases manufacturer in U.S. Engaged in process engineering, design, project engineering, construction and capital investment planning for sixteen plants.

- Developed AIRCO engineering and design for proposed Krypton 85 removal from U.S.S. Savannah nuclear reactor, and installation of krypton recovery units in air separation plants.

The MITRE Corporation would be pleased to offer a proposal to NRC for consulting services to carry out the proposed process and project engineering/management details. We look forward to the opportunity to meet with Mr. Harold R. Denton, Director of NRC Nuclear Reactor Regulation, to discuss our proposal which you indicated that you will try to arrange. Should you want additional information or clarification, please telephone me at (703) 827-7198.

Sincerely,

Milton Lytton

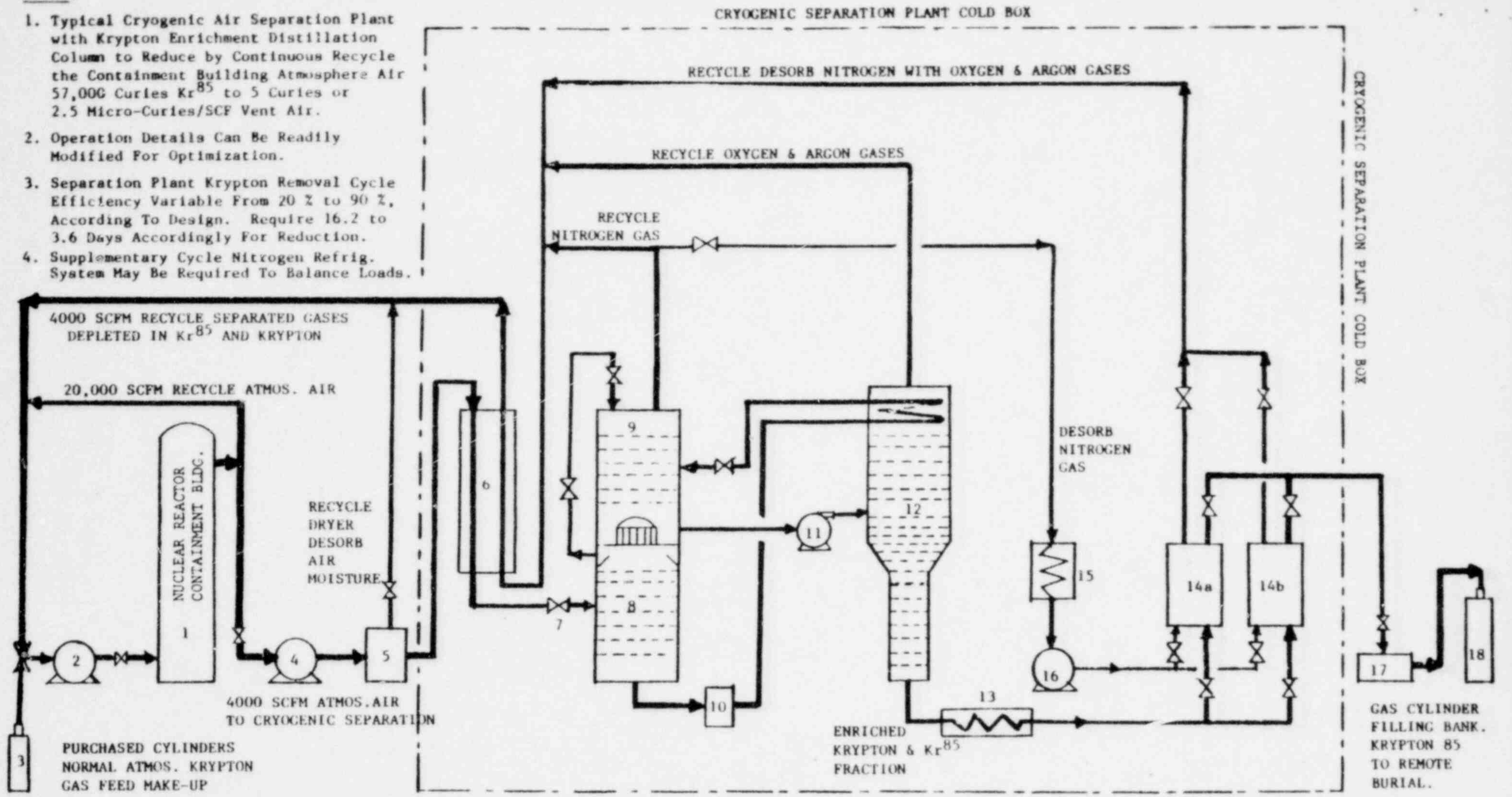
Milton Lytton

Enclosures

ML:njo

NOTES:

1. Typical Cryogenic Air Separation Plant with Krypton Enrichment Distillation Column to Reduce by Continuous Recycle the Containment Building Atmosphere Air 57,000 Curies Kr^{85} to 5 Curies or 2.5 Micro-Curies/SCF Vent Air.
2. Operation Details Can Be Readily Modified For Optimization.
3. Separation Plant Krypton Removal Cycle Efficiency Variable From 20 % to 90 %, According To Design. Require 16.2 to 3.6 Days Accordingly For Reduction.
4. Supplementary Cycle Nitrogen Refrig. System May Be Required To Balance Loads.



1. TMI Nuclear Reactor Containment Building; 2,000,000 Cu.Ft.; 57,000 Curies Krypton 85.
2. Existing Ventilating Air Blowers (2). Each 24,000 SCFM Air Flow.
3. Purchased Normal Atmospheric Krypton Feed Make-Up From Gas Cylinder. Total Required: 887 Liters.
4. Centrifugal Gas Compressor, 4000 SCFM at 85 PSIA Delivery.
5. Air Moisture Dryer, Silica or Alumina Bed, 40 °F, Dual Swing Vessels - Adsorb/Desorb.
6. Cryogenic Multi-Passage Reversing Exchangers; Remove CO_2 and H_2O by Cool-Down About -250 °F.
7. Joule-Thomson Expansion Valve to Cool-Down Air to about -280 °F to Form Liquid/Vapor Mix.

8,9. Double Distillation Column: 8. Nitrogen Section, 9. Oxygen Section

10. Molecular Sieve Filter for Hydrocarbon Impurities.
11. Liquid Oxygen Pump
12. Krypton Enrichment (Including Kr^{85}) Distillation Column - Bottoms: Kr^{85}, Kr, Xe, Ar, O_2.
13. Enriched Krypton Fraction (Including Kr^{85}) Vaporizer to Gasify.
14. Linde Molecular Sieve Filter, Dual Swing Vessels, Operate about -295 °F. Adsorb O_2 and Ar.
15. Nitrogen Warm Gas Desorb Heater for Molecular Sieve Bed Desorption of O_2 and Ar.
16. Desorb Gas Blower.
17. Enriched Krypton Mixture (Including Kr^{85}, Kr, Xe) Compressor For Filling Gas Cylinders.
18. Shielded Metal Krypton Mixture Gas Cylinder Filling Bank. Filled Cylinders Removed To Disposal.

FIGURE 1

PROPOSED

PROCESS SCHEMATIC DIAGRAM

FOR

KRYPTON 85 REMOVAL

FROM NUCLEAR REACTOR

CONTAINMENT BUILDING

THREE MILE ISLAND, PENNA.

CONFIDENTIAL
PROPRIETARY
DISCLOSURE

MILTON LYTTON
MITRE CORP.
MARCH 24, 1980

M. Lytton
The MITRE Corporation
March 24, 1980

TABLE I

BASIC PROCESS SCHEME
DERIVATION OF Kr^{85} REDUCTION EQUATION,
AND CALCULATION OF TMI CONTAINMENT BUILDING REDUCTION

GIVEN: Kr^{85} = 57,000 curies
Volume of TMI nuclear reactor containment building = 2,000,000 C.F.
Normal Kr in atmos. air = 1.0 ppm = 1.0SCF/1,000,000 SCF air
TMI containment bldg. air blowers = 24,000 SCFM (two each)

OBJECTIVE: Reduce total Kr^{85} within building to 5 curies, or 2.5 microcuries/SCF vent air

1. BASIC PROCESS SCHEME

The cryogenic plant and the containment building are in a closed recycle with only one stream being discharged, namely, an enriched concentrated krypton 85. It is necessary to maintain an inventory of normal krypton in the system of containment building atmosphere volume and the cryogenic plant in order to maintain a proper vapor-liquid equilibria in the cryogenic fractionation columns. Therefore, as normal krypton and krypton 85 are depleted in closed system recycle, purchased fresh normal krypton is fed into the recycle system to maintain the overall krypton inventory at 1.0 ppm or higher. The Kr^{85} is depleted continuously during the recycle and is replaced by krypton. This is the unique patent-sought feature of the process design. The closed recycle system returns the separated oxygen-nitrogen-argon streams from the cryogenic fractionation plant to the containment building atmosphere. The only change in the overall total system is the continuous depletion of krypton 85. Therefore, the derived equation below represents this significant change in the total system.

The enriched krypton 85 stream is further concentrated by gas passage through molecular sieve filter beds at cryogenic low temperature which effectively separate the krypton 85 from the oxygen and argon (which are in very much larger amounts). The separation is based on the sieve physical adsorption of the smaller molecules: oxygen - 3.46 Angstroms and argon - 3.40 Angstroms. The concentrated stream consists of about 93 percent Kr and Kr 85 and the remainder is xenon; nitrogen is extremely low. The Kr^{85} rich gas is filled into radiation shielded metal gas cylinders and removed for disposal.

See Process Diagram, Figure 1, for illustration.

2. REDUCTION EQUATION DERIVATION

A. Conversion of Curies Kr^{85} to Grams, Liters and SCF

curies/gram $= \frac{3.578 \times 10^5}{10.72 \times 85}$ — Kr^{85} Half Life = 10.72 yr. Kr^{85} Atomic Mass = 85

$= 392.7$ curies/g

$85 \frac{g}{g\text{-mol}} Kr^{85}$ $= 22.414$ liters/g-mol

$Kr^{85} = 0.2637$ liters/g

$= 1489$ curies/liter

Kr^{85} Bldg. Content $= \frac{57{,}000 \text{ curies}}{392.7 \text{ curies/g}} = 145.15$ g

$= \frac{57{,}000 \text{ curies}}{1489 \text{ curies/l}} = 38.28$ liters

$= \frac{38.28 \text{ liters}}{28.316 \text{ liters/CF}} = 1.352$ SCF

B. Equation for Reduction of Kr^{85}

Let:
- k_o = Kr^{85} bldg. content at time $t = 0$, = 1.352 SCF
- k_t = Kr^{85} bldg. content at any time t
- t = Time, minutes
- R = Cryogenic Plant Kr^{85} recovery efficiency
- A = Bldg. sweep air flow, SCFM to Cryogenic Plant
- V = Air volume of Containment Building
- Ψ = Kr^{85} removal coefficient

Assume:
- R = 0.9, 0.7, 0.6, 0.5, 0.4, 0.3, 0.2
- A = 4000 SCFM (1/6 of 24,000 SCFM Air Blower)
- V = 2,000,000 C.F. = 2,000,000 SCF

At time $t = o$, $k = k_o$

Kr^{85} Bldg. content at time $t = k_{t+1} = k_o - \Psi k_t . t$

where:

Kr^{85} Removal Coefficient, $\Psi = R \times \frac{A}{V}$

Differentiate:

$$\frac{dk}{dt} = - \Psi k$$

$$\frac{dk}{k} = - \Psi dt$$

Integrate:

$$\ln k_{t = 0} = - \Psi t + C$$

$$\therefore \quad C = \ln k_o \text{ at } t = 0$$

$$\ln k_t - \ln k_o = - \Psi t$$

$$\ln \frac{k_t}{k_o} = - \Psi t$$

$$\frac{k_t}{k_o} = e^{-\Psi t}$$

$$k_t = k_o e^{-\Psi t}$$

Now, given objective is residual Kr^{85} = 5 curies in Containment Building, or 5 microcuries/SCF

$$\text{Residual } Kr^{85} = k_t = \frac{5}{1489 \times 28.316} = 1.186 \times 10^{-4} \text{ SCF}$$

<u>Assume</u>: One 24,000 SCFM building blower in recycle operation.
4,000 SCFM side-stream is taken as feed to Cryogenic Plant.

$$\Psi = R \times \frac{4,000}{2,000,000} = 2.0 \times 10^{-3} R$$

$$\therefore \quad k_t = 1.352\, e^{-(2.0 \times 10^{-3} Rt)}$$

But

$$\text{Residual } Kr^{85} = k_t = 1.186 \times 10^{-4}$$

$$e^{-(2.0 \times 10^{-3} Rt)} = \frac{1.186 \times 10^{-4}}{1.352} = 0.8772 \times 10^{-4}$$

$$- 2.0 \times 10^{-3} Rt = \ln 0.8772 \times 10^{-4}$$

$$t = \frac{1}{R} \times \frac{\ln 0.8772 \times 10^{-4}}{- 2.0 \times 10^{-3}}$$

$$= \frac{1}{R} \times \frac{- 9.3416}{- 2.0 \times 10^{-3}}$$

Time to Complete Reduction of Kr^{85} to 5 Curies $= t = \frac{4671}{R}$

3. CALCULATION OF Kr^{85} REDUCTION

Assume: Recovery Efficiencies: 0.9 down to 0.2

R	TIME	
	MIN.	DAYS
0.9	5190	3.60
0.8	5839	4.05
0.7	6673	4.63
0.6	7785	5.41
0.5	9342	6.49
0.4	11677	8.11
0.3	15570	10.81
0.2	23355	16.22

4. CALCULATION OF PURCHASED NORMAL KRYPTON FEED MAKE-UP

Note: It is necessary to maintain an inventory of normal krypton in the system of containment building atmosphere volume and the cryogenic plant in order to maintain proper vapor-liquid equilibria in the cryogenic distillation columns. Therefore, as normal krypton and Krypton 85 are depleted in closed system recycle, purchased fresh normal krypton is fed into the recycle system to maintain the overall krypton inventory at 1.0 ppm or higher.

Normal Krypton in Atmosphere = 1.0 ppm = 1.0 SCF/1,000,000 SCF Air

In Containment Bldg.: Normal Kr = 2.000 SCF

Kr^{85} = 1.352

Total = 3.352

In one - 4000 SCFM sweep cycle:

$$\text{Removal of Kr} + Kr^{85} = R \times 3.352 \text{ SCF} \times \frac{4000 \text{ SCF/Min.}}{2{,}000{,}000 \text{ SCF}}$$

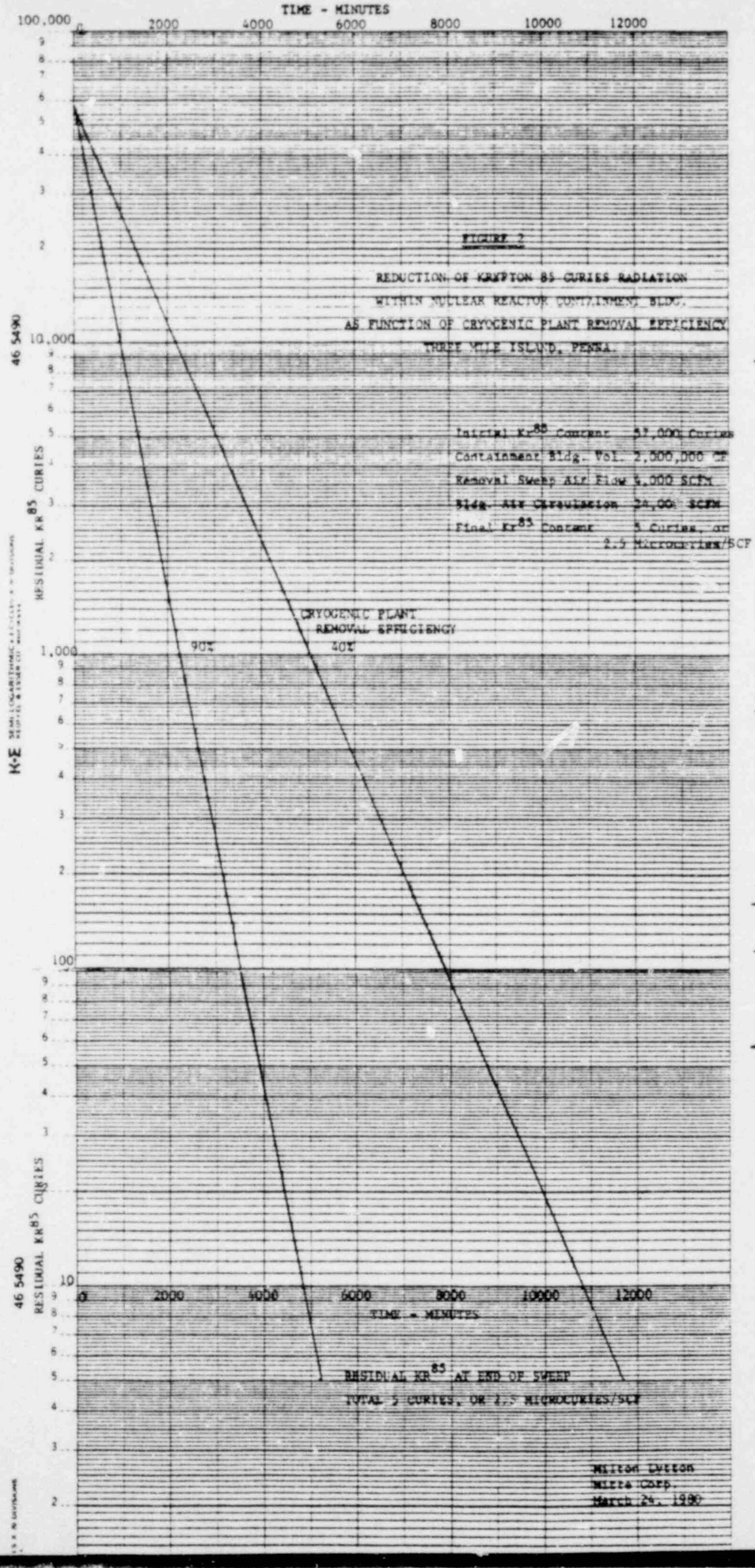
at R = 0.9:

$$\text{Krypton Make-Up} = 0.9 \times 3.352 \times \frac{4000}{2{,}000{,}000} \times 5190 \text{ Min.}$$
$$= 31.314 \text{ SCF} = 887 \text{ Liters}$$

at R = 0.2

$$\text{Krypton Make-Up} = 0.2 \times 3.352 \times \frac{4000}{2{,}000{,}000} \times 23{,}355 \text{ Min.}$$
$$= 31.314 \text{ SCF} = 887 \text{ Liters}$$

Therefore, Total Fresh Normal Krypton Make-Up is a Constant.



TIME - MINUTES

EA 426

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THEODORE LEE GAILLARD, JR.
9805 PARK LANE PLACE
DALLAS, TEXAS 75220

March 28, 1980

Chief Engineer,
Three Mile Island Nuclear Power Station,
Three Mile Island,

Pennsylvania

Dear Sirs:

Problem:
How to get slightly radioactive gas out of the powerplant's containment building, without venting directly into the atmosphere and causing undue concern by a worried populace.

Possible solution worth consideration:
Have you considered obtaining a high-altitude weather/atmospheric sounding balloon? The gas vent opening for the balloon could be sealed around the power plant's vent opening and the gas force-vented into the deflated balloon. When the gas has been exhausted from the building and/or the balloon filled, the balloon can be tied off or sealed at the throat, and the balloon containing the gas can then be transported by a variety of means to a desert or ocean area where the gas can be released (or buried). Such a balloon offers several advantages:

1) availability
2) designed to contain gas without leaking--and usually at some pressure since these balloons carry payloads of some weight to extremely high altitudes
3) immense volume--far more than would be needed, I suspect, to empty the building.

Perhaps this has already been suggested. If not, I hope it may be of some help.

Sincerely,

Theodore L. Gaillard, Jr.

Theodore L. Gaillard, Jr.
(214) 350-9152 (home)
(214) 363-6311 (work)

cc: Director, Nuclear Regulatory Commission