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TRITIUM PRODUCTION BY NEUTRON IRRADIATION OF HELIUM-3

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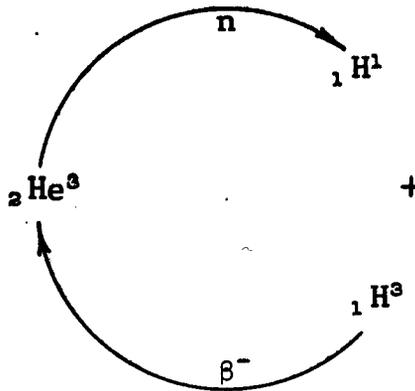
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TRITIUM PRODUCTION BY NEUTRON IRRADIATION OF HELIUM-3

The problem of producing tritium by the irradiation of helium-3 has several very interesting aspects. Among them are the following:

1. The reaction is cyclic, since helium-3 is the decay product of tritium, and the net result is the production of protium from neutrons.



2. The process is adaptable to a continuous loop through a reactor in a fairly straight forward manner. The separation of the isotopic mixture, tritium-protium, from helium-3, is not difficult. The gas mixture could be withdrawn from the reactor, separated, and the helium-3 returned. The protium-tritium mixture could be fed to a thermal diffusion, low-temperature distillation, or some other hydrogen isotope separation system.

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The reactor process has several interesting features, one being that the helium-3 can act (depending on the pressure) as a homogeneous reactor control which would vary as the helium-3 is burned up.

3. The fact that the only daughter of the tritium product is feed material is an unusual and fortuitous circumstance.

The stable gaseous isotopes sales program at Mound Laboratory predicts the sale of about 100 STP liters of helium-3 to France in FY 1965. If this material were irradiated for only 150 days at a flux of 10^{13} neutrons/cm²/sec (see Figure 4), it could result in the production of 25 liters, or over six (6) grams of tritium. If irradiated to the limit, it could result in twice this amount.

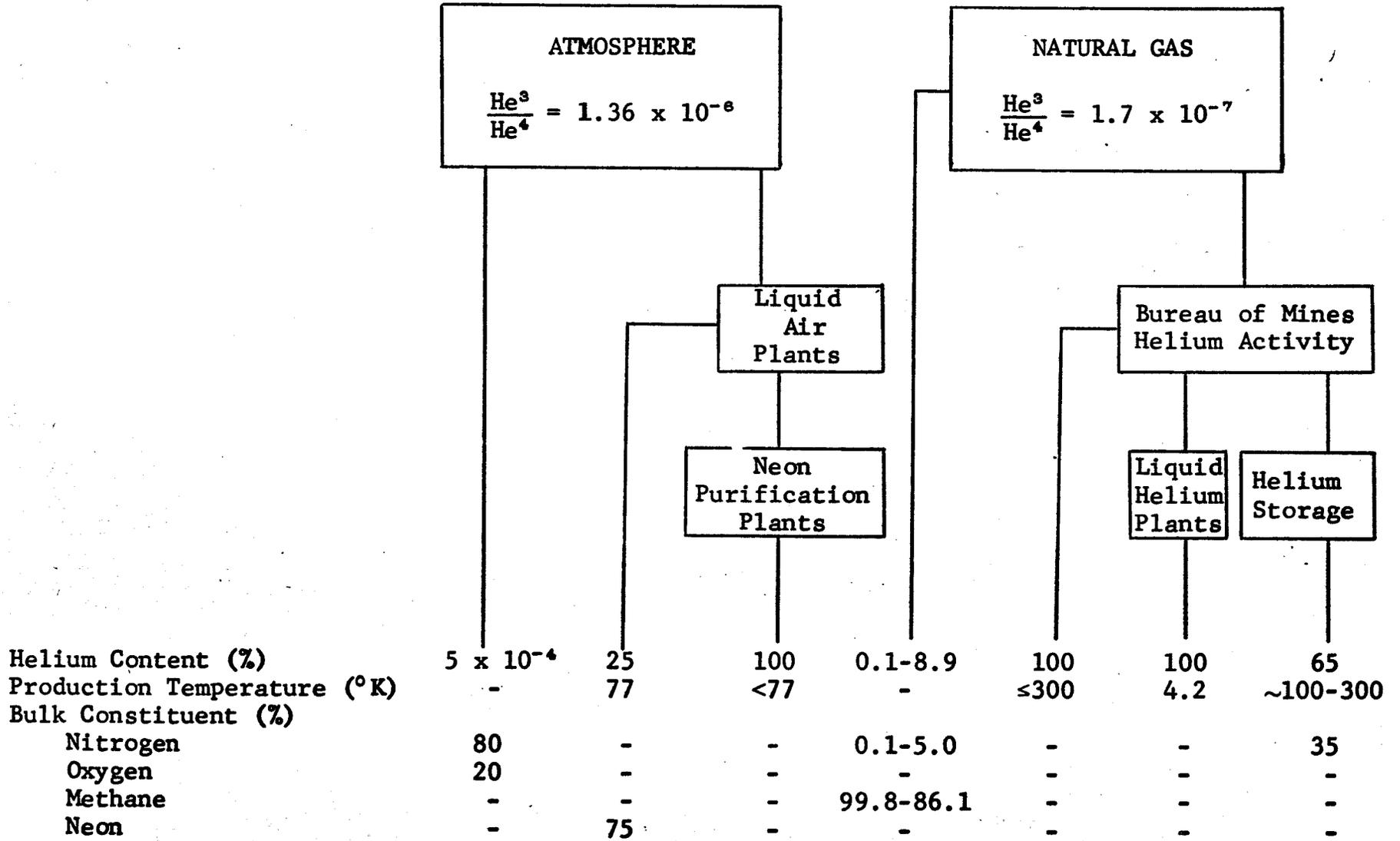
Since the cross section of helium-3 is nearly six times that of lithium-6 and the continuous process would seem more favorable than the batch-slug method, the cost of enriching helium-3 from nature could be considerably greater than that of enriching lithium-6 and still result in a competitive process.

One of the problems currently being studied at Mound Laboratory is the feasibility of the recovery of helium-3 from nature and other dilute sources.

Figure 1 gives a comparison of the natural sources of helium-3. While atmospheric helium is available to all, the United States has most of the well gas helium held captive. The most attractive atmospheric source in Figure 1 would seem to lie in the off gas from neon purification plants; however, it is estimated that the current United States production of neon is on the order of 10^6 STP liters per year which would result in less than one

Figure 1

COMPARISON OF NATURAL SOURCES OF HELIUM



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STP liter of helium-3. Based on the estimated annual United States production of nitrogen, the total helium available annually from the crude neon stream would contain less than 15 STP liters of helium-3. These crude neon streams are in many cases not even retained. The total helium-3 content of the atmosphere is about 2.4×10^{10} STP liters; however, it seems too dilute for consideration as a source.

Due to the large helium conservation program of the Bureau of Mines, there are approximately 14,000 STP liters of helium-3 made available annually mixed only with helium-4 and nitrogen. Since the process used by the helium conservation program is partly cryogenic, the addition of a heat flush or superleak device into the process may very well be economically feasible.

Furthermore, the capacity of major helium liquefaction plants throughout the United States was estimated to be over 1,000 liquid liters per hour by midyear 1965. The quantity of helium-3 available from this source would be approximately 1,000 STP liters per year. This source is especially important since the best known methods for the enrichment of very dilute helium-3 in helium-4 lie below the lambda point of helium-4, which is two degrees below the boiling point. It would seem possible to install a helium-3 separator in all helium liquefiers in such a manner that the helium-3 could be bled off either continually or by batches.

Another source of dilute helium-3, although not natural, is the thermal diffusion column raffinate stream at Mound Laboratory. This stream contains about 0.1 per cent helium-3 in total helium and is about 99 per cent helium. This material is produced at 5-10 STP liters per hour which results in an annual helium-3 loss of 50 to 100 STP liters.

The consideration of these sources for the production of

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tritium leads to Figure 2, which is a general schematic of a possible method of using three different feed materials for tritium production. Each stage in this schematic has several possible separation methods and Figure 2 is not meant to represent the best combination, e.g., the method indicated to recover helium from the atmosphere would not be practical. Partially enriched helium-3 - helium-4 streams may also be used directly in the reactor since helium-4 has essentially a zero cross section.

Figures 3, 4, and 5 were computed using reactor fluxes of 10^{12} , 10^{13} , and 10^{14} neutrons/cm²/sec, respectively, to show the relative concentrations of feed and product as a function of irradiation time. The equations used to construct the curves were,

$$\frac{N_{He}}{N_0} = \frac{\lambda_1}{\lambda_1 + \lambda_2} e^{-(\lambda_1 + \lambda_2)t} + \frac{\lambda_2}{\lambda_1 + \lambda_2}$$

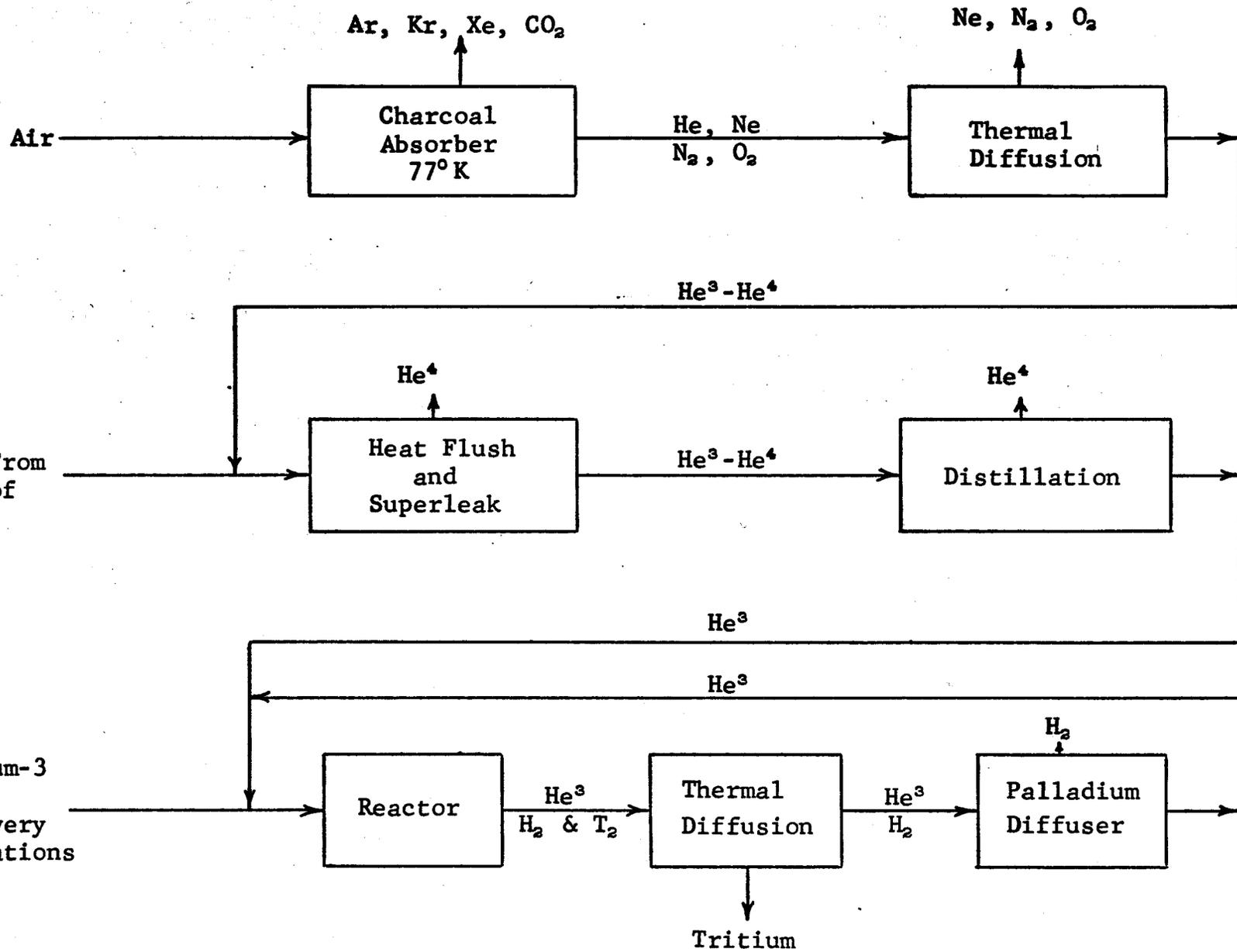
$$\frac{N_T}{N_0} = 1 - \frac{N_{He}}{N_0}$$

$$\frac{N_H}{N_0} = \left(\frac{\lambda_1}{\lambda_1 + \lambda_2} \right)^2 \left(1 - e^{-(\lambda_1 + \lambda_2)t} \right) + \frac{\lambda_1 \lambda_2}{\lambda_1 + \lambda_2} t$$

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Figure 2

PRODUCTION OF TRITIUM FROM HELIUM-3
Ar, Kr, Xe, CO₂

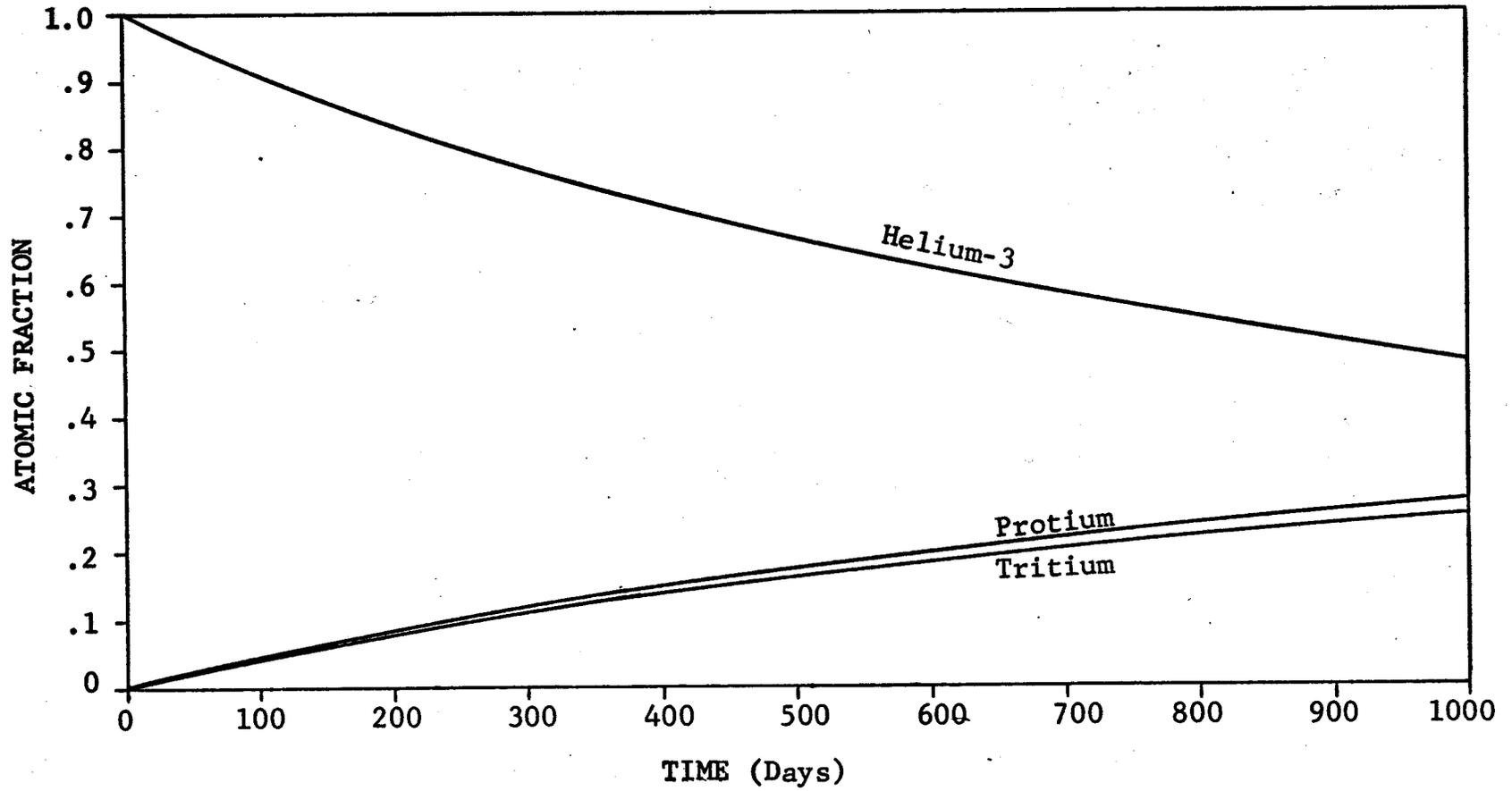


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Figure 3

RELATIVE ATOMIC FRACTIONS VS. IRRADIATION TIME
 $\phi = 10^{12}$ n/cm²/sec



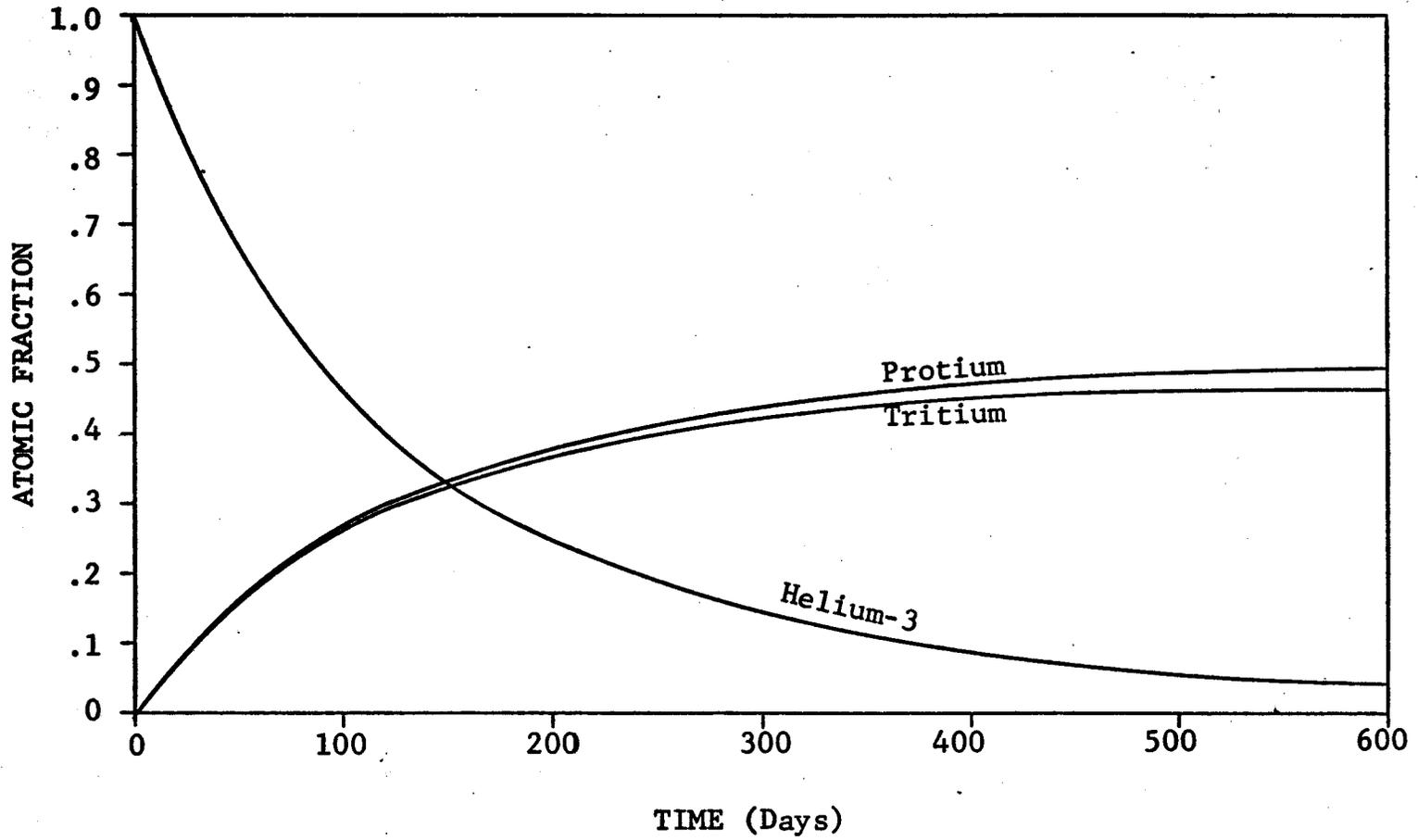
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Figure 4

RELATIVE ATOMIC FRACTIONS VS. IRRADIATION TIME
 $\phi = 10^{13}$ n/cm²/sec



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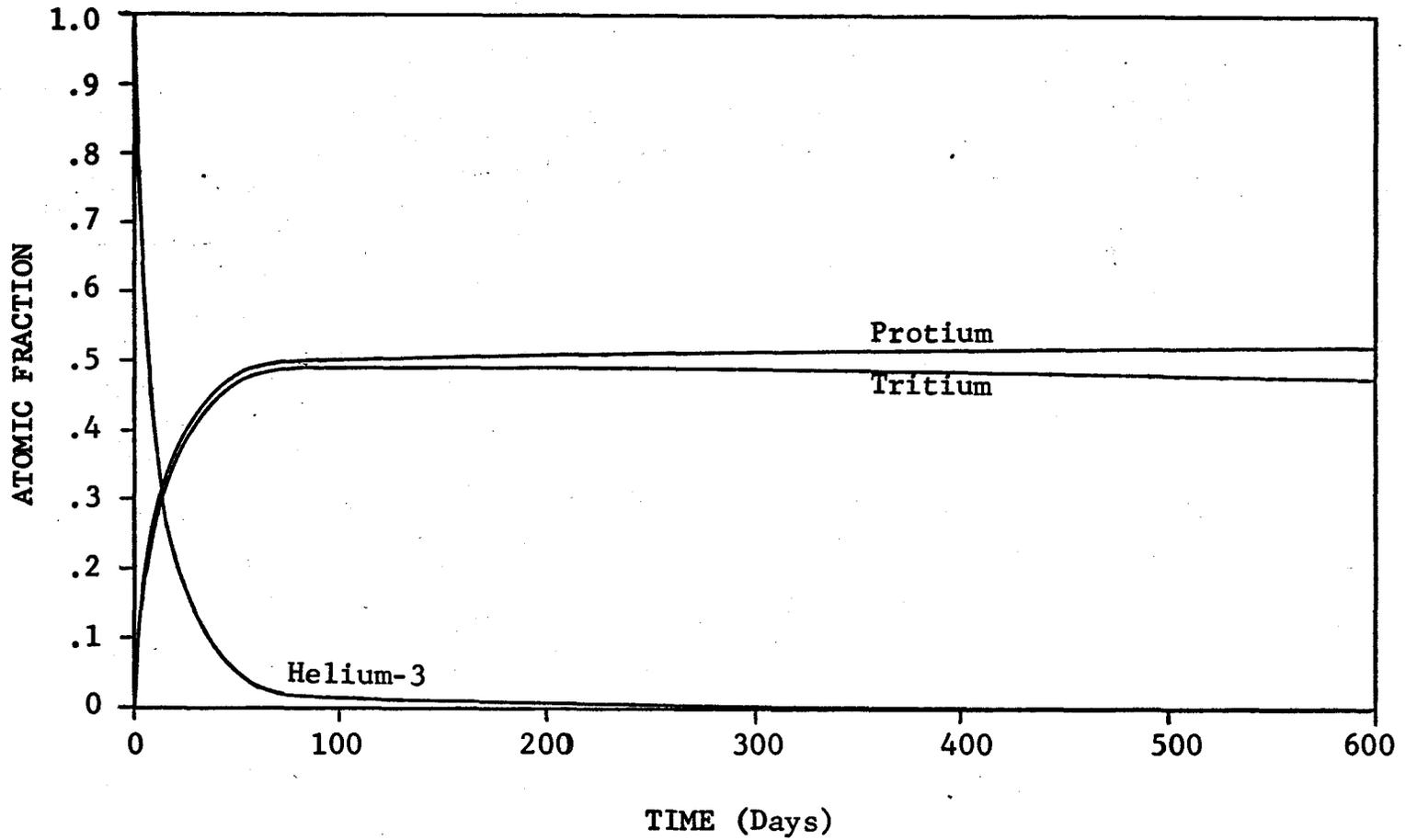
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Figure 5

RELATIVE ATOMIC FRACTIONS VS. IRRADIATION TIME

$$\phi = 10^{14} \text{ n/cm}^2/\text{sec}$$



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where

N_{He} = Number of helium atoms present at time t

N_T = Number of tritium atoms present at time t

N_H = Number of hydrogen atoms present at time t

N_0 = Number of helium atoms present at $t = 0$

$\lambda_1 = \phi\sigma$

ϕ = reactor flux

σ = cross section of helium-3 = 5400 barns

$\lambda_2 = 0.693/\tau_{1/2}$

$\tau_{1/2}$ = half-life of tritium = 12.26 years

From Figure 4, for example, it is seen that an essentially equi-molar mixture exists after 150 days of irradiation. The resulting mixture at any time is quite suitable for a thermal diffusion separation. The protium-tritium concentration as a function of time remains essentially 50-50 so one is faced with a separation of mass two from mass six in a system always resulting in a force of the equilibrium



to the right and the presence of an inert component of intermediate mass, helium of mass three, to act as a third component buffer to force the protium from the tritium. In addition, the inert third component is feed for the production plant.

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It would seem appropriate to evaluate methods, including cost analyses, of several possible separation methods for each step in Figure 2.

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