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MACROSCOPIC ISOTOPE SEPARATION OF URANIUM BY SELECTIVE PHOTOIONIZATION

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MACROSCOPIC ISOTOPE SEPARATION OF URANIUM BY SELECTIVE PHOTOIONIZATION

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Washington D. C., May 28 - 30, 1975

In this presentation, I will give the results of an experiment in which milligram quantities of uranium were enriched using selective photoionization. Basically, the technique involved using two, colinear laser beams to ionize ^{235}U atoms emanating from a high temperature oven. The ions were then electrostatically separated from the neutral atoms and collected on a beryllium plate.

Figure 1 is an energy level diagram depicting the photoionization process used. The uranium temperature in this experiment was about 2600°K. At this temperature, the first metastable level in uranium which is at 620 wavenumbers, is thermally populated to about 27%. In the ground state, the population is about 45%. A xenon ion laser operating at 3781 Å, was used to selectively excite ^{235}U atoms from the 620 level to a higher lying level. To reach the ionization continuum, a second photon with a wavelength shorter than 4400 Å is required. For this second photon, the output of a krypton ion laser was used, which has two strong lines at 3507 Å and 3564 Å.

In Figure 2, the isotopic absorption lines in uranium for the 3781 Å transition are compared with the xenon laser emission characteristics. For the uranium lines, Doppler broadening at 2600°K results in a linewidth of about 2 GHz, FWHM. The isotope shift for this transition is 3 GHz, ^{235}U occurring at the higher frequency. The emission line from the xenon laser has a mode envelope bandwidth of about 4 GHz and overlaps both the ^{235}U and ^{238}U absorption lines. Consequently, it is necessary to etalon tune the laser to avoid excitation of ^{238}U. To this end, a temperature stabilized, intra-cavity etalon was used, which had a F.S.R. of 10 GHz and 10% reflective surfaces. Figure 3 shows the laser characteristics obtained with the etalon. The output bandwidth was narrowed to about 0.3 GHz, with three longitudinal modes oscillating. The relative tuning range obtained, shown by the dotted curve, is about a factor of two broader than the natural bandwidth shown in Figure 2. This is attributed to a significant amount of homogeneous broadening in the laser With the laser tuned to the peak of ^{235}U absorption line, the output power was about 70 mW.

To maintain precise tuning of the xenon laser, a real-time monitoring system was set up as shown in Figure 4. The small amount of laser radiation transmitted through the rear mirror was split into two beams. One was fed to a spectrum analyzer to monitor the laser bandwidth. The other beam was split again, 50% to a reference photo-diode, B, and 50% focused through a UI_4 microwave discharge lamp containing 70% ^{235}U. The number of free ^{235}U atoms generated in the lamp were such that 40% of the xenon laser beam was absorbed. The transmitted radiation was fed to another photo-diode, B.

The signals from diodes A and B were then fed to a ratiometer and the ratio of A/B was monitored on an oscilloscope or chart recorder. The etalon in the xenon laser was then temperature tuned to obtain a minimum A/B ratio, thereby indicating maximum absorption at the peak of the ^{235}U line. The etalon temperature was maintained to $\pm$ .01°C which provided sufficient frequency stability for a period of hours.

The optical arrangement of the experiment is shown in Figure 5. A vacuum chamber, which housed the uranium vapor source, was placed between the xenon and krypton lasers. The laser beams were colinear and the optics were arranged such that the vacuum chamber was internal to the krypton laser cavity. Mirror M_2 acted simultaneously as an output coupler for the xenon laser and a high reflector for the krypton laser. The normal output of the krypton laser was 1-watt. With this arrangement, the circulating krypton radiation through the vacuum chamber was about 30 watts. This technique was possible because of the small photoionization cross-section in uranium. The estimated round trip absorption is only about 10^{-5}. The xenon laser beam made essentially one pass through the system, with a corresponding absorption of about 0.5%. Lenses L_1 and L_2 focused the two laser beams to 0.2 mm within the vacuum chamber.

With the vacuum chamber intra-cavity to the krypton laser, it was extremely important to keep the chamber windows free from uranium vapor deposition and other vacuum system contaminants. Initially, significant contamination would occur within a few minutes of operation time. A gas scattering cell, shown in Figure 6, was developed to circumvent this problem. The

chamber windows, which were actually the end sections from commercial gas laser tubes, were mounted onto hollow flanges on the chamber housing. Xenon gas was continually fed into the side of the flange and into the chamber through a 15-cm aluminum tube with a 3-mm bore. The xenon flow was such that a quasi-static pressure of about 0.5 Torr was maintained in the vicinity near the Brewster window. The vacuum pumping system was adequate to provide an overall chamber pressure of about 2×10^{-5} Torr. The collision of extraheous uranium atoms with the xenon gas prevented them from reaching the Brewster windows.

An additional procedure was required to prevent long-term migration of other vacuum system contaminants, such as organic molucules from the pump system, which would eventually deposit on the windows and interact with the high intensity krypton laser beam. (The intensity at the windows was about 30 kW/cm^2.) A d.c. discharge wasmaintained in the xenon gas in a short region near the window by applying 600 volts to a stinger electrode, as shown in the Figure. With these techniques, no apparent contamination of the windows was incurred with several hours of running time.

Figure 7 is a schematic of the uranium source and collection system. A 10-cm long uranium oven with a 1-mm by 8-cm exit slit provided an atom density of about 3×10^{13} cm^{-3} in the path of the laser beams. Photoionized ^{235}U atoms generated by the lasers were separated from the uranium vapor stream by the electric field produced by a beryllium collector plate, biased at -1500 volts, located next to the oven. The ions were deflected into roughly parabolic trajectories and made a deposition on the collector plate

in an area of approximately 5-mm by 8-cm. Thermally ionized ^{238}U atoms emanating from the oven were also attracted to the plate, thereby diluting the collected ^{235}U sample. The neutral atoms were allowed to condense onto a water-cooled, U-shaped copper block located above the oven.

Figure 8 is a drawing showing the spatial distribution of uranium on the collector plate. The contours shown were obtained from autoradiographs of the plate. Calculated trajectories indicated two, more-or-less distinct bands, which were clearly visible on the plate.

A continuous run of the system was made for about two hours. The collected sample was dissolved off the collector plate with nitric acid and then evaporated to dryness. The compound, uranyl nitrate, thus formed was then analyzed for its isotopic content. The resulting sample, shown in a test tube in Figure 9, contained 4 mg of reactor grade uranium, which is about 3% ^{235}U. This represents an enrichment of a factor of 4 over the natural isotopic content of 0.7%.

The amount of enrichment obtainable is obviously dependent on the available laser power, since the dilution by ^{238}U is from a fixed amount of thermal ions. Figure 10 shows what one would obtain from this system with higher laser powers. The rate of collection of ^{235}U is shown as a function of the intra-cavity krypton laser power for three fixed amounts of xenon laser power. The experimental separation run which was just described, with a krypton power of 30 watts and a xenon power of 70 mW, approximately 0.1 mg of ^{235}U were produced in two hours, which is a rate of 50 µg/hour. By increasing the power of each laser by 10-fold, one would obtain a ^{235}U production rate of about 0.5 mg/hour. I might add at this point, if the uranium oven were increased in length from 10 cm to 1-meter, the ^{235}U production rate would

essentially increase by a factor 10 for the same amount of laser power. This is because of the small absorptions discussed earlier.

Figure 11 shows the corresponding enrichment obtainable in the presence of thermal ions. Again, with a 10-fold increase in laser power, an enrichment of about 20% is obtainable.

In summary, the macroscopic separation of uranium isotopes has been obtained using selective photoionization. The 3% enrichment obtained, which was influenced by thermal ions, is not an inherent limitation, but depends directly on the laser power available. This experiment was performed as part of a program to investigate the viability of laser separation processes based upon atomic vapors. Further experimental studies will be necessary to ascertain the economics of such systems.

PHOTOIONIZATION, ENERGY LEVELS

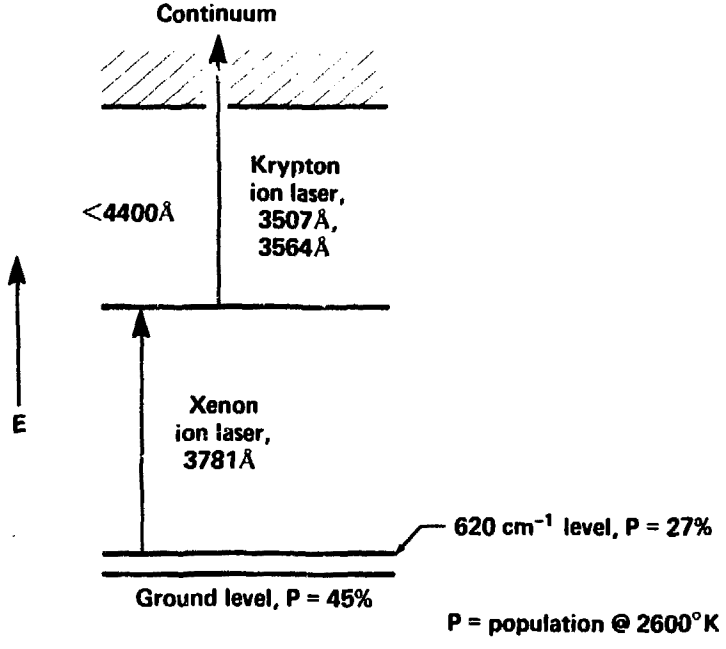

FIGURE 1

Xe EMISSION VS URANIUM ABSORPTION–A

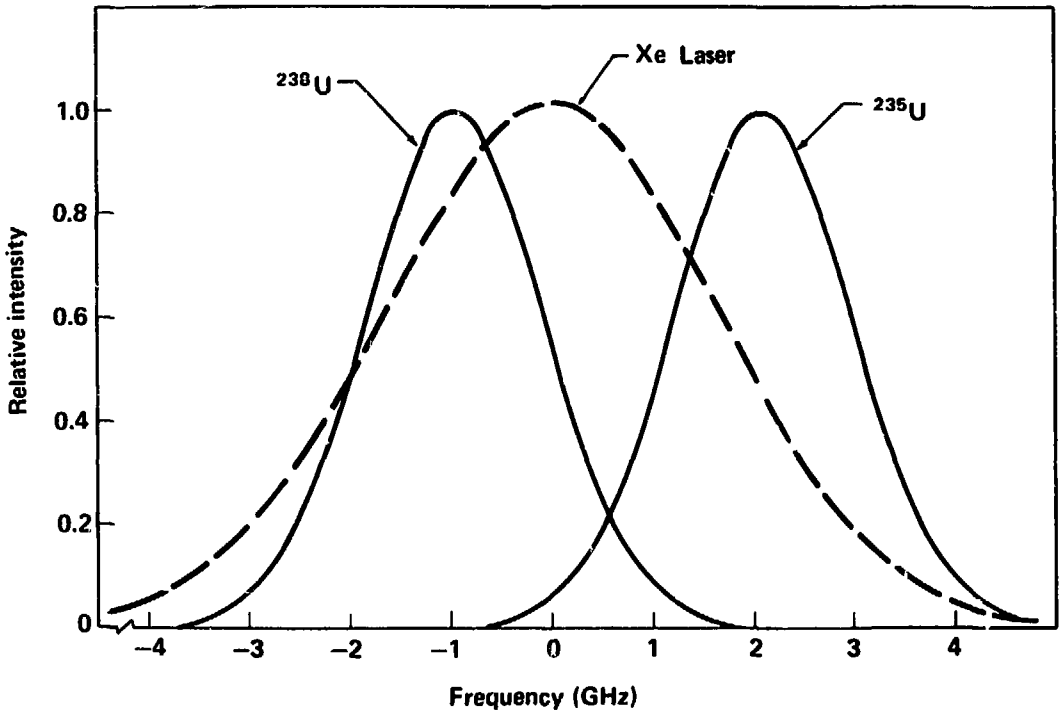

FIGURE 2

Xe EMISSION VS URANIUM ABSORPTION

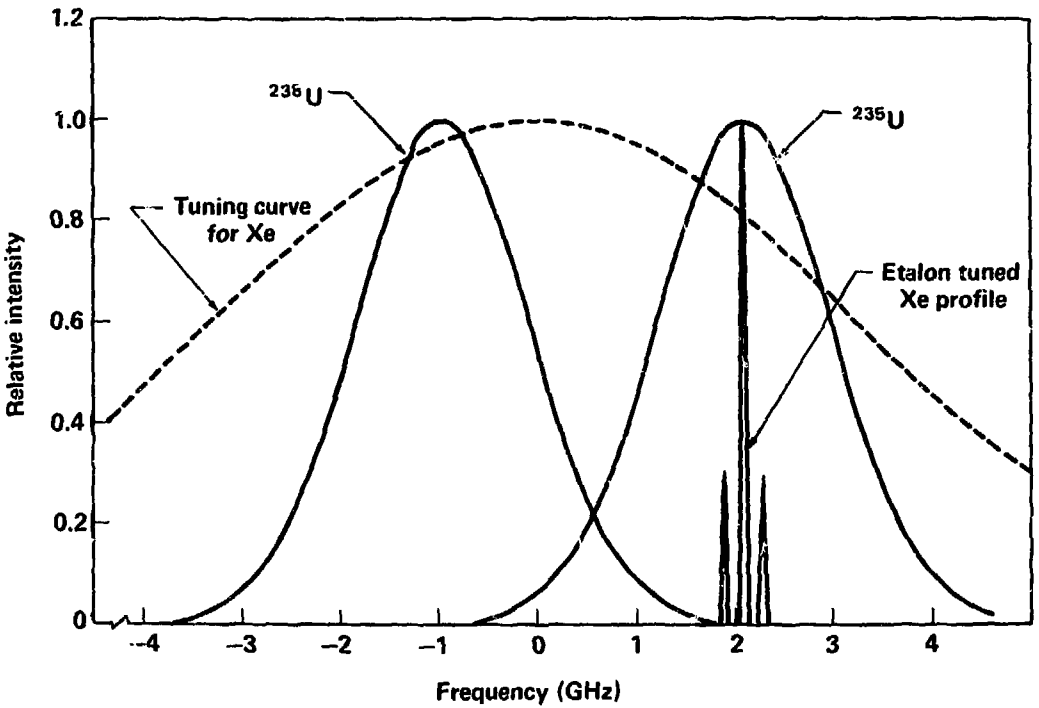

FIGURE 3

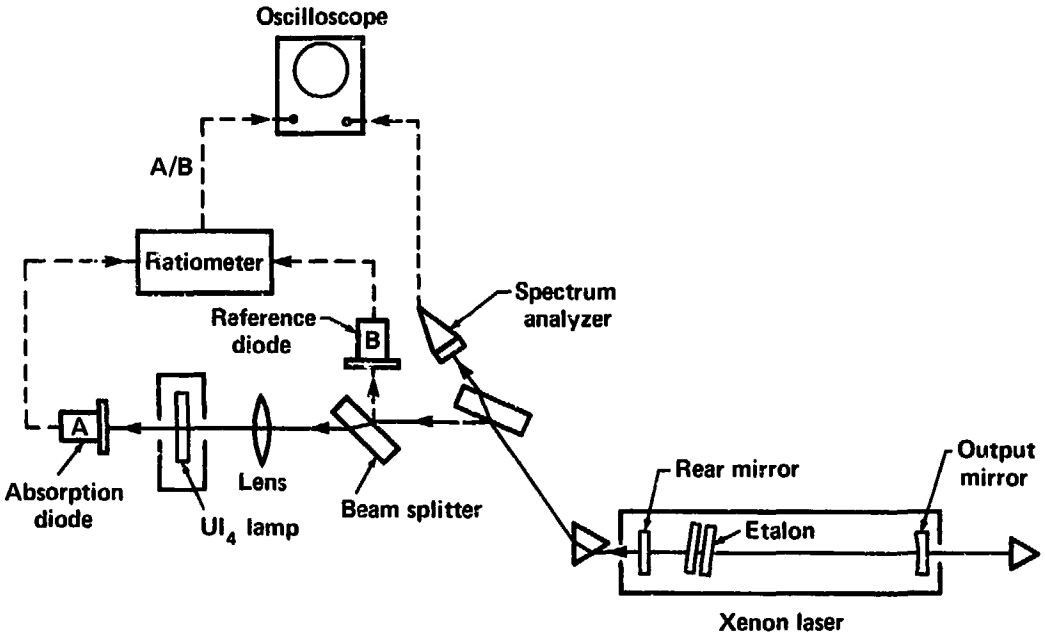
Oscilloscope
A/B
Ratiometer
Reference diode
B
Spectrum analyzer
A
Absorption diode
Lens
UI_4 lamp
Beam splitter
Rear mirror
Output mirror
Etalon
Xenon laser

FIGURE 4

SYSTEM OPTICAL GEOMETRY

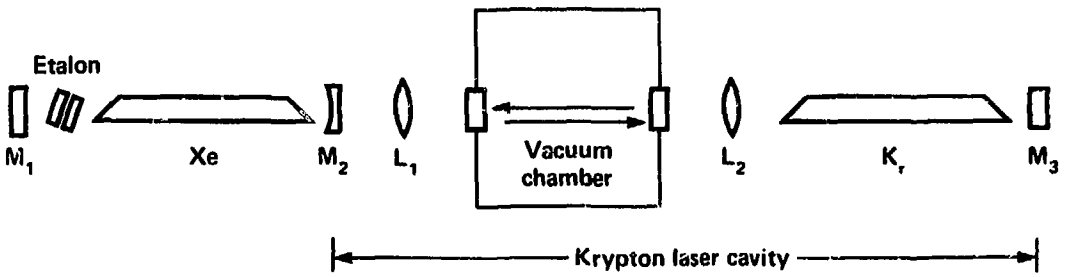

FIGURE 5

GAS SCATTERING CELL

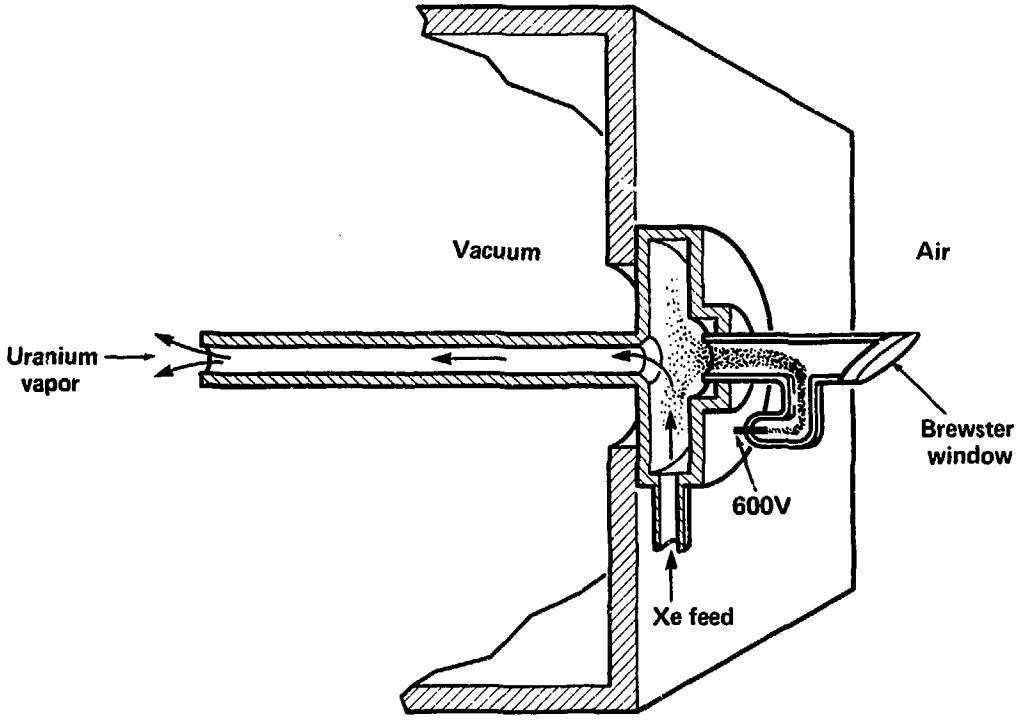

FIGURE 6

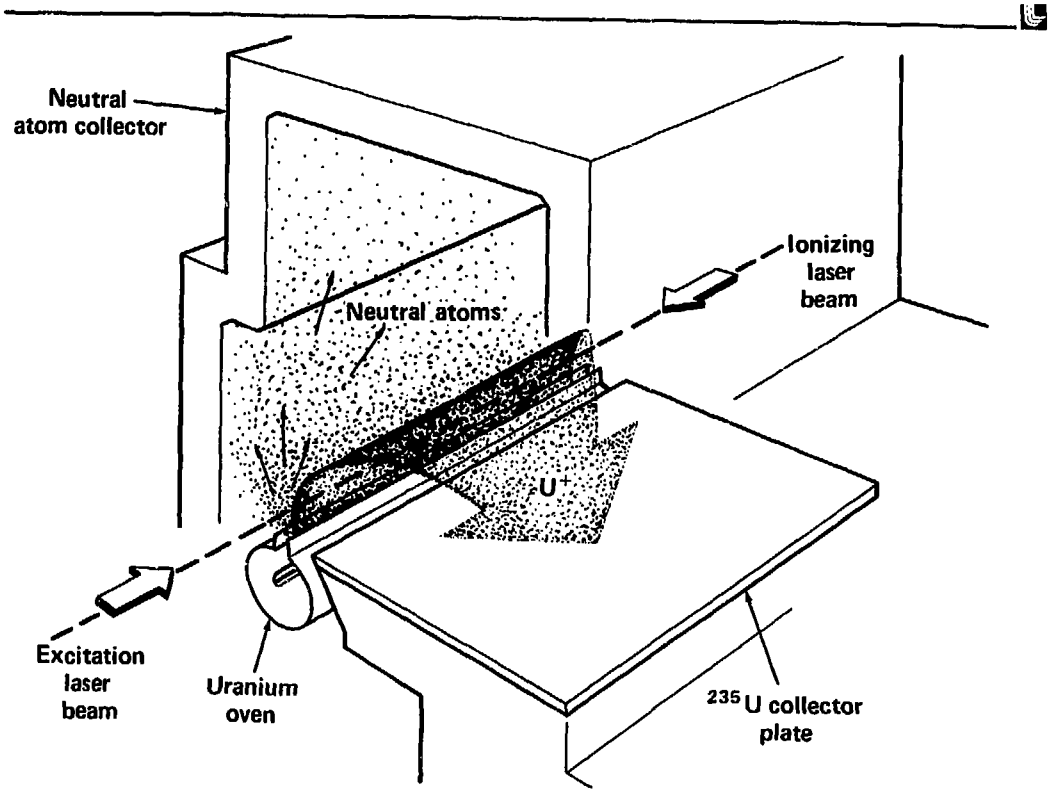
Neutral
atom collector
Ionizing
laser
beam
Neutral atoms
U+
Excitation
laser
beam
Uranium
oven
235U collector
plate

FIGURE 7

URANIUM ION SPATIAL DISTRIBUTION ON ^{235}U COLLECTOR PLATE

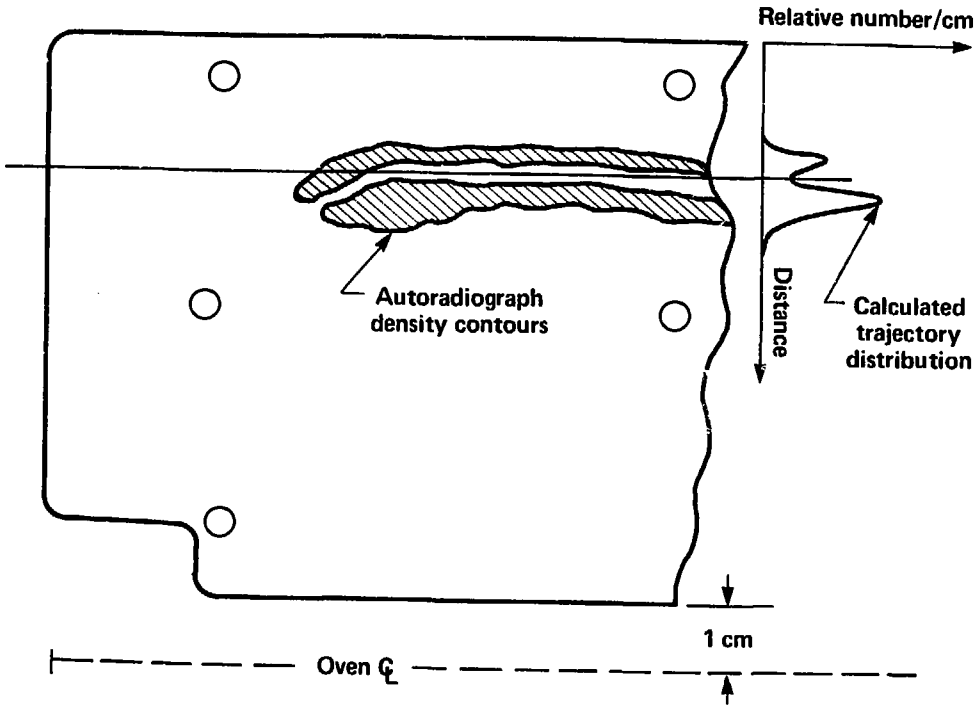

FIGURE 8

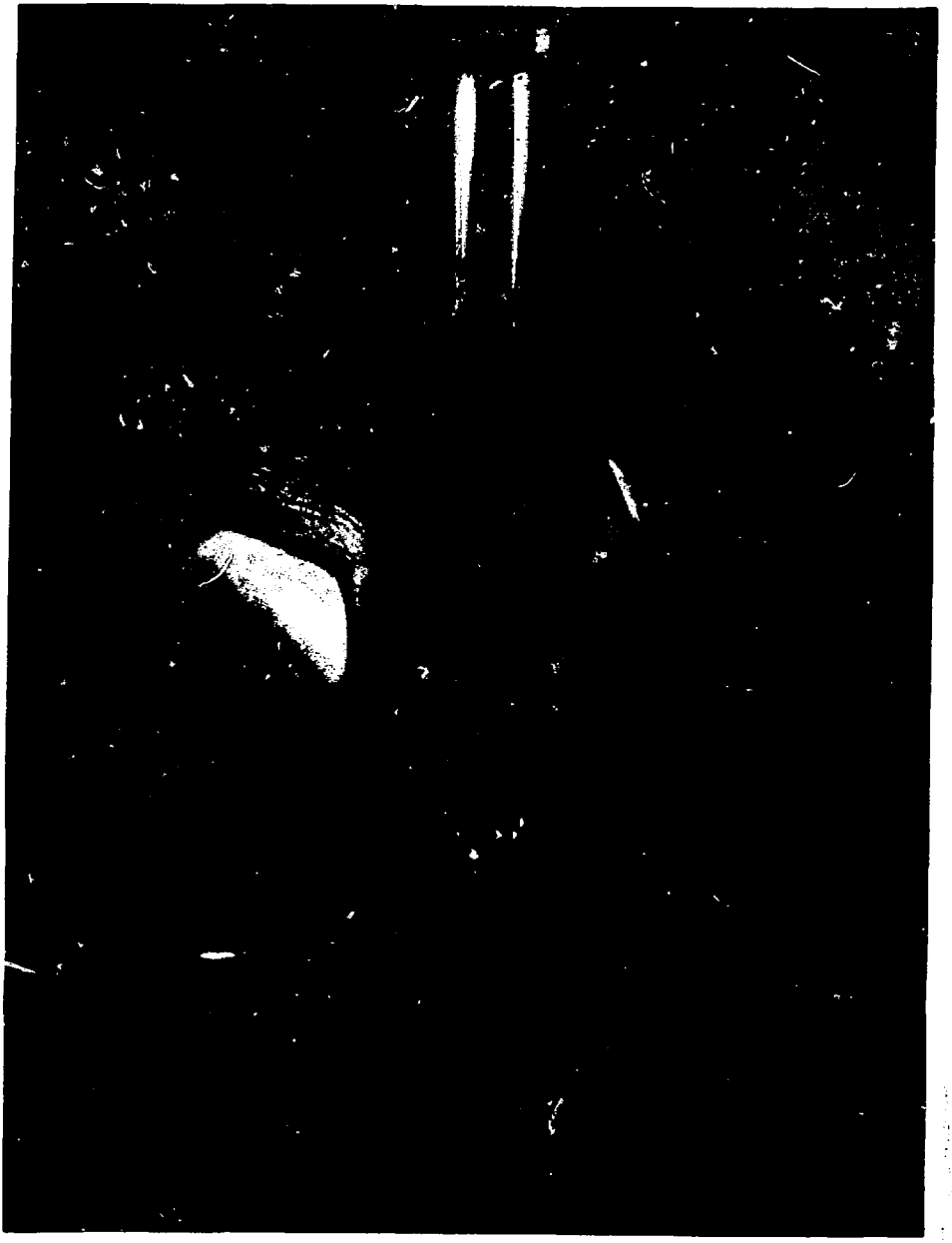

FIGURE 9

^{235}U PRODUCTION RATE VS LASER POWER

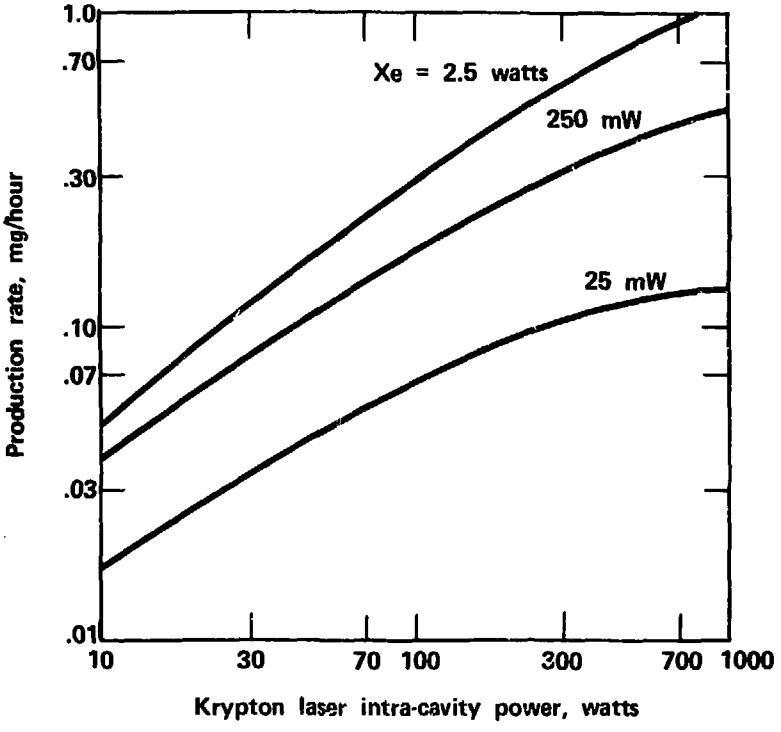

FIGURE 10

ENRICHMENT VS LASER POWER

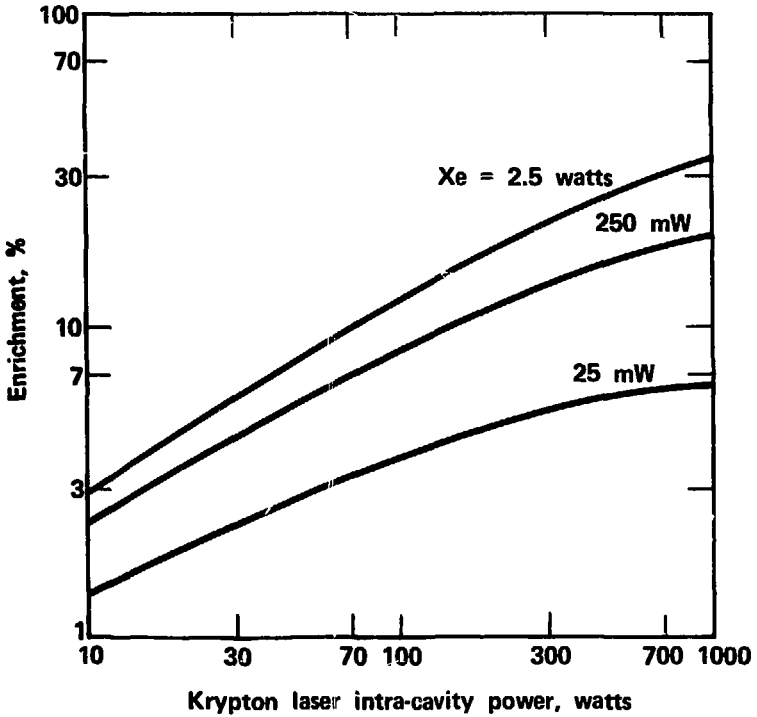

FIGURE 11