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Three-Compartment Photoreactor for Laser Isotope Separation of Tritium

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Abstract

A new photoreactor has been designed and tested for laser isotope separation of tritium using trifluoromethane as a working substance. The reactor consists of three compartments in series which are stirred efficiently with recirculation pumps independently. The CO_2 laser beam is sequentially focused with BaF_2 lenses serving as the beam entrance windows of the compartments. In this reactor the reaction volume is maximized while the window crystals are kept undamaged. The experimental results showed that the performance of such a reactor can be suitably predicted from the dissociation characteristics obtained from batch irradiation experiments.

INTRODUCTION

In order to remove tritium from the effluent water of a nuclear fuel reprocessing plant, a few conventional hydrogen isotope separation techniques have been considered for practical application. Recently, the authors showed that an isotopically selective dissociation reaction induced by an infrared laser such as the CO_2 laser can be utilized to separate tritium (*1, 2*). Currently, the separation factor of this laser

method exceeds 10^4 at one stage at an operational pressure as high as 200 torr (*3*). The authors also showed that continuous operation is possible when a single continuous stirred tank reactor (CSTR) is used and the laser beam is focused with a lens serving as the window of the compartment (*4*). This single CSTR is the first continuous reactor for laser tritium isotope separation, but it is not designed to utilize fully the laser energy in the direction of beam propagation; only a small fraction of the laser energy is absorbed by the working substance.

In the new photoreactor we propose in this report, the laser beam is consecutively focused as it passes through continuously stirred compartments in series. Furthermore, the focal length of the lens is sequentially reduced so that the focal fluence is kept at the optimum value (*5*) while the pulse energy is gradually decreased due to photon absorption by the resonant and off-resonant substances as well as by the loss due to the optical devices.

In this report the performance of this new integral reactor is investigated experimentally.

EXPERIMENT

The schematic diagram of the experimental apparatus is shown in Fig. 1. The reactor compartments are made of stainless steel (2 cm in diameter and 49, 47, and 45 cm in length) connected in series in the direction of beam propagation. The focal lengths of the KCl lenses are 24.5, 12.0, and 11.5 cm in the same direction, so that the focal fluence in each compartment is approximately the same although the pulse energy decreases by ~10% for each compartment. The central portion of the cell (40 cm) was immersed in a thermally insulated box filled with crushed Dry Ice, and the sample gas was cooled to −78°C. Since absorption is negligible for such a small cell, the energy loss is mostly due to the loss by lenses. The gaseous working substance, trifluoromethane (CTF_3/CHF_3, T/H = 0.01 ppm), was prepared in a similar manner as reported elsewhere (*2*). The sample gas thus prepared was supplied from a reservoir (5 L) to the 3-compartment continuous stirred tank reactor at a constant mass flow rate. The pressure level of the reactor compartment was maintained at 85 torr. The fluctuations of the mass flow rate and the pressure were 2 and 5%, respectively, and no long-term drifts were observed. The gas in each compartment was mixed sufficiently with a recirculation pump so that the recirculation rate was at least 30 times larger than the sample feed rate.

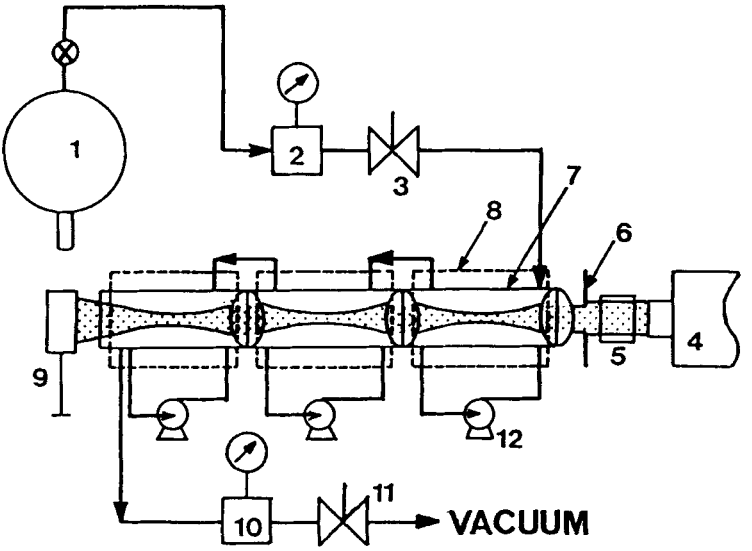

FIG. 1. Schematic diagram of the experimental apparatus: (1) gas reservoir, (2) mass-flow meter, (3) mass-flow valve, (4) TEA CO_2 laser, (5) attenuator, (6) aperture, (7) reactor, (8) thermal insulation vessel filled with crushed Dry Ice, (9) pyroelectric energy meter, (10) capacitance manometer, (11) needle valve, (12) recirculation pump.

The multimode laser beam from a TEA CO_2 laser (Lumonics 821) has a relatively uniform transverse profile of the fluence. The central uniform portion is used after the beam has passed through an aperture (17 mm in diameter) and BaF_2/KCl/KBr/CaF_2 attenuators. The laser line is tuned to the 9P(8) line of the CO_2 laser at 1057 cm^{-1}. The laser pulse consists of a spike (~100 ns FWHM) and a 1-μs tail. The pulse energy was measured with a pyroelectric energy meter (Lumonics 20D). After the elapse of the time required to reach the steady-state concentration at the outlet of the reactor (usually 3 to 5 times the mean residence time of the system), the outlet gas was sampled and injected into a home-made radio-gas chromatograph with a Porapak Q column (4 mm i.d., 4 m in length). The depletion of CTF_3 was measured with a proportional counter (internal gas-flow type) by comparing the radioactivities of the sample at the reactor outlet and of the reference. The thermal conductivity detector measurement indicated that no detectable depletion of CHF_3 was observed under the experimental conditions chosen.

MODELING

The fractional conversion per pulse in infrared-laser-induced multiphoton dissociation (IRMPD), q, increases very rapidly with the increase of the fluence Φ until q saturates to unity. The dissociation characteristics of the IRMPD are usually described by the cumulative log-normal distribution function written as (*6*)

$$q = \frac{1}{\sqrt{2\pi}\sigma} \int_{-\infty}^{\Phi} \exp\left\{-\frac{(\ln \Phi - \ln \Phi_s)^2}{2\sigma^2}\right\} d \ln \Phi \tag{1}$$

where the parameters Φ_s and σ are the saturation fluence and the standard deviation, respectively. When the fluence is equal to the saturation fluence, q is equal to 0.5. For given experimental conditions (gas pressure, temperature, laser wavenumber, pulse duration, etc.), the values of Φ_s and σ are determined from the batch irradiation experiment.

As the next step of the modeling of the reactor performance, the quantity called the reaction volume, V_R, is defined in relation to q as follows:

$$V_R = \int_V q \, dV \tag{2}$$

The transverse profile of the fluence for the multimode CO_2 laser beam can be treated as uniform. The envelope of the axial profile of the fluence is given by

$$r^2 = r_f^2\{1 + (Z/a)^2\} \tag{3}$$

where r, r_f, Z, and a are the beam radius, the beam radius at the focal point, the axial distance from the focal point, and the characteristic axial distance, respectively. The schematic diagram of the irradiation experiment with a focused beam is shown in Fig. 2. Since the transverse and axial fluence profiles are determined when the focal length of the lens, the beam radius, and the effective beam divergence are given, only numerical integration of Eq. (2) over the inner volume of the reaction chamber is needed to predict V_R. Based on the mass balance equation for each reactor compartment, the fraction of the resonant substance CTF_3 undissociated at the outlet of the last compartment is written as

$$1 - X = \prod^{i} \left(\frac{1}{1 + V_{Ri}h/Q}\right) \tag{4}$$

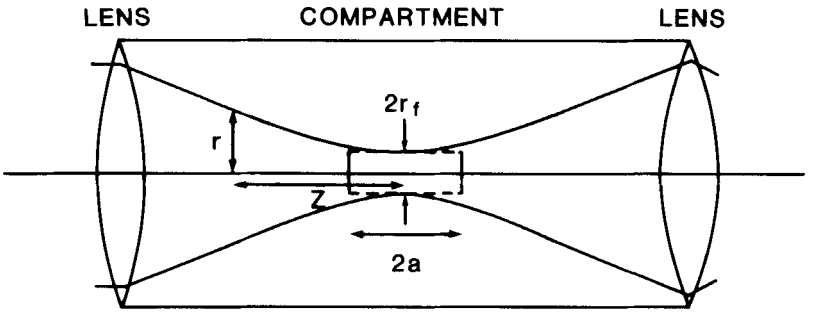

FIG. 2. Schematic diagram of irradiation geometry.

where V_{Ri}, Q, and h denote the reaction volume in the ith compartment, the volumetric flow rate of the working substance, and the laser pulse repetition rate, respectively. Since the focal lengths of the lenses decrease in the direction of beam propagation, the values of the reaction volume also decrease accordingly.

RESULTS AND DISCUSSION

Figure 3 shows the steady-state result of the 3-compartment photoreactor: the fraction undissociated at the outlet of the reactor $(1 - X)$ is plotted as a function of the volumetric flow rate Q with the pulse energy and the repetition rate fixed at 0.74 J and 5.0 Hz, respectively. The subscripts T and H denote CTF_3 and CHF_3, respectively. Figure 4 shows the relation between $(1 - X)$ and the pulse energy E_0 when h is 5.0 Hz and Q is 0.218 cm^3/s. Figure 5 illustrates the relation between $(1 - X)$ and h when E and Q are 0.74 J and 0.218 cm^3/s, respectively. The reproducibility of the measurement of $(1 - X)$ is within 2%. The depletion of the off-resonant CHF_3 was negligible. Since it was confirmed elsewhere (*3*) that the T/H-selectivity under this experimental condition exceeds 10^4 at one stage, it is unnecessary in our case to take the dissociation of CHF_3 into consideration for the design of the reactor. Therefore, only the dissociation of CTF_3 will be considered in the following discussion.

The solid curves in Figs. 3 to 5 were obtained from theoretical calculations based on the procedure discussed in the preceding section: The dissociation characteristic curve $q(\Phi)$ was calculated from Eqs. (2)

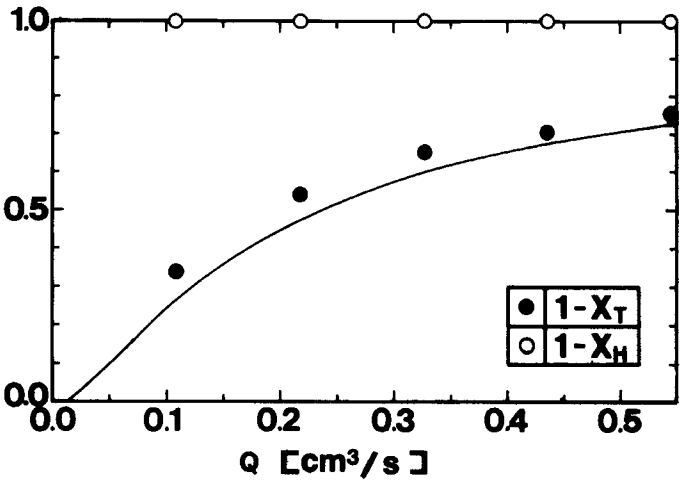

FIG. 3. Fraction of undissociated CTF_3 at the outlet of the reactor, $(1 - X)$, as a function of the volumetric flow rate Q. ($E_0 = 0.71$ J, $h = 5.0\ s^{-1}$.)

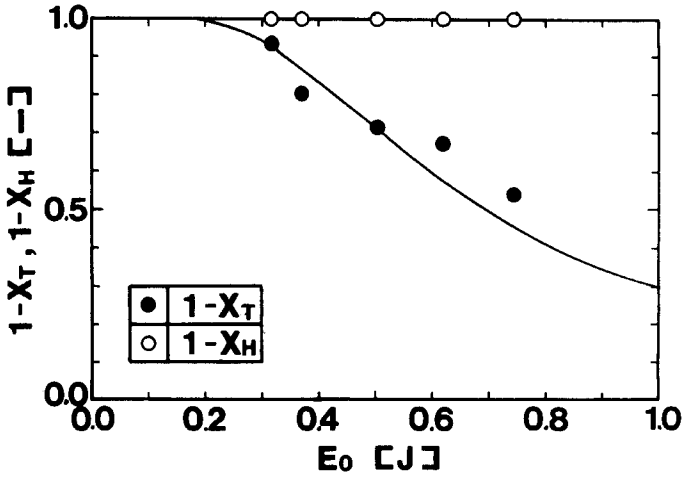

FIG. 4. Fraction of undissociated CTF_3 at the outlet of the reactor, $(1 - X)$, as a function of the laser pulse energy E_0 at the beam inlet window of the reactor. ($Q = 0.218\ cm^3/s$, $h = 5.0\ s^{-1}$.)

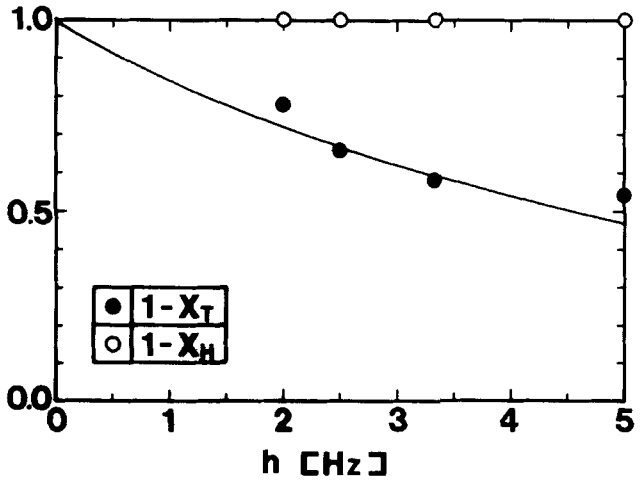

FIG. 5. Fraction of undissociated CTF_3 at the outlet of the reactor, $(1 - X)$, as a function of the laser repetition rate h. (E_0 = 0.74 J, Q = 0.218 cm^3/s.)

and (3) using the values $\sigma = 0.38$ and $\Phi_s = 70$ J/cm^2 calculated from the data reported in the preceding paper (*4*). The agreement of theory with experiment was found to be satisfactory. Consequently, the design procedure of such a photoreactor was experimentally validated.

The most efficient reactor for laser isotope separation of tritium may be a plug-flow reactor with constant fluence profile in the direction of the beam propagation as well as in the transverse direction. Since the laser energy is inevitably consumed as it is absorbed by the resonant and off-resonant substances (CTF_3 and CHF_3, respectively), this ideal reactor may resemble a conical tube with a total reflection mirror on the internal surface where the cross section of the reactor is so designed that the fluence is uniform axially, irrespective of the decrease in pulse energy. Such a reactor is not practical since the damage thresholds of available mirrors and lenses are usually considerably lower than the value of Φ_s. Consequently, a conical reactor with a total reflection mirror on the interior surface may not be easy to manufacture. Our new reactor gives a very efficient alternative. Chemical reactor theory (*7*) indicates that the performance of such CSTRs in series approaches that of the corresponding plug-flow reactor as the number of compartments increases. A simple calculation shows that 10 compartments are sufficient from such a viewpoint.

CONCLUSION

In laser isotope separation of tritium, a new reactor of continuously stirred tanks in series was found satisfactory in utilizing the laser energy in the direction of beam propagation. In this reactor the focal length of the lenses is reduced sequentially in the beam direction so that the focal fluence is similar in spite of the decrease of pulse energy. The design procedure and the performance of such a reactor were experimentally validated.

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