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Multiple Reflection Reactor for Infrared Laser Isotope Separation

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Abstract

A new photoreactor for laser isotope separation is demonstrated in which the laser beam is reflected and focused several times between two concave mirrors in a compartment while the reacting gas is continuously supplied and extracted. This reactor utilizes the entire nonuniform non-Gaussian beam in the transverse direction. Its process capability was tested experimentally and found to agree with the performance predicted from the proposed mathematical model.

INTRODUCTION

Laser isotope separation is based on isotopically selective laser-induced reactions. For example, when the gas mixture C_2TClF_4/C_2HClF_4 is exposed to a CO_2 laser beam at an appropriate wavelength, C_2TClF_4 is selectively dissociated to form tritium-containing products (*1*). This reaction, called infrared multiphoton dissociation, is known to be described well by the relations $q = (\Phi/\Phi_c)^n$ for $\Phi \leq \Phi_c$ and $q = 1$ for $\Phi > \Phi_c$, (*2*), where q, Φ, Φ_c, and n are the reaction probability per pulse, fluence, the critical fluence, and a parameter for power dependency, respectively. Φ_c is the minimum fluence to yield $q = 1$. The value of Φ_c depends on the conditions of the reactant (pressure, temperature) and of the laser beam (wavenumber, pulse shape), but not on the fluence or the geometry of beam focusing.

When the value of Φ_c exceeds the acceptable fluence for the window materials, Φ_w, we have to focus the laser beam to have reactions occur at the center of the compartment while avoiding damage to the windows.

We have developed a new photoreactor (*3*) (shown schematically in Fig. 1b) of a compartment-in-series type that improves the residence time distribution of the reactant gas. In this reactor the laser beam is focused only once in each compartment. According to our multiphoton absorption data for CTF_3/CHF_3 and C_2TClF_4/C_2HClF_4, the optical path lengths to use up most of the laser energy may exceed several tens of meters. The chemical reactor theory (*4*) of the compartment-in-series type suggests that the number of compartments does not significantly affect the performance of the ractor if it exceeds a certain number, such as 10. Therefore, it may be wise to have multiple reflections in one compartment to shorten the total length of the photoreactor and reduce the number of compartments. Such a reactor is illustrated in Fig. 1(c). In this report an attempt is made to establish the design procedure of a single multiple reflection reactor (shown in Fig. 1a) which will become one compartment of the practical reactor shown in Fig. 1(c).

EXPERIMENTAL

A multiple reflection reactor is schematically shown in Fig. 2. Chlorotetrafluoroethane (C_2TClF_4/C_2HClF_4) containing 0.1 ppm tritium was supplied from a reservoir (1) (5 L) through a precalibrated mass-flow controller (2) and a mass-flow valve (3) to the reactor (4). The pressure therein was detected by a capacitance manometer (5) and regulated by a servo-valve (6) to 5.0 torr. The reactor was operated at 25°C.

The entire multimode beam from a TEA CO_2 laser (7) was used without passing through an aperture. Thus the beam was neither Gaussian nor uniform. First, it was attenuated and focused by a BaF_2 lens (focal length 52.5 cm) into the cell. Then the beam was focused three more times by the concave mirrors (r = 46.7 cm) placed (face-to-face, 84.6 cm apart) in the cell. No attempt was made to stir the gas in the reactor since we know from previous work that the reactor could be treated as a well-mixed reactor at this pressure (*5*). The outlet gas was carefully sampled by using a bypass between the servo-valve and the vacuum pump so that the gas flow was not disturbed. The sample gas was then introduced into a radio-gas chromatograph and the depletions of C_2TClF_4 and C_2HClF_4 were measured in the same manner as reported in an earlier paper (*1*).

Prior to the continuous-flow experiment, the cell was used as a batch reactor and the depletions of both isotopic compounds were measured in the same way.

RESULTS AND DISCUSSION

The concentration of C_2HClF_4 was found to be unchanged at the outlet of the reactor for all the experimental runs. This corresponds to the high selectivity of this working molecule. Thus, no further discussion will be

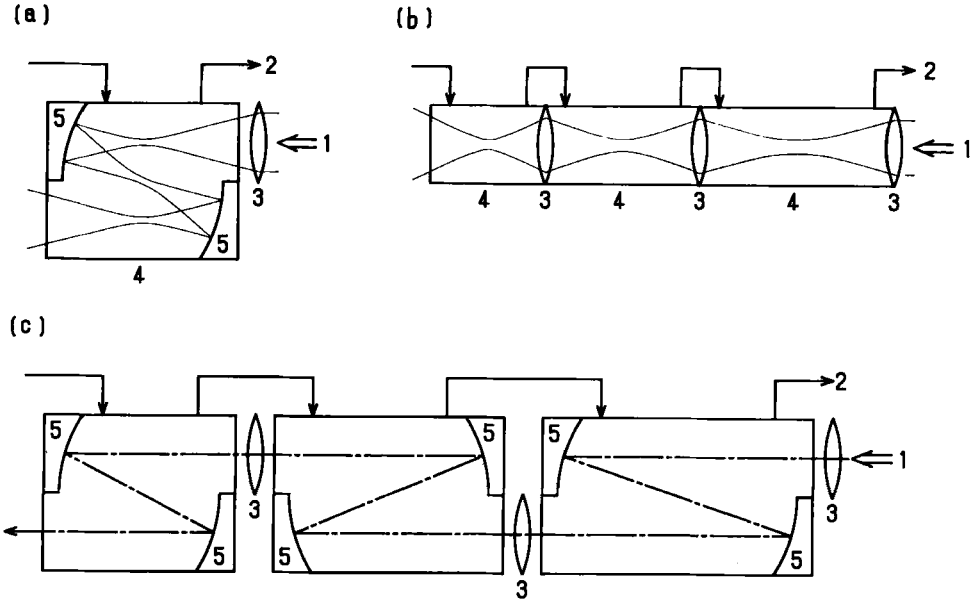

FIG. 1. Schematic illustration of photoreactors of the single compartment type with multiple reflection (a), of compartment-in-series type with single focusing in each compartment (b), and of compartment-in-series type with multiple reflection (c): 1) laser beam, 2) reacting gas, 3) lens, 4) reactor compartment, 5) mirror.

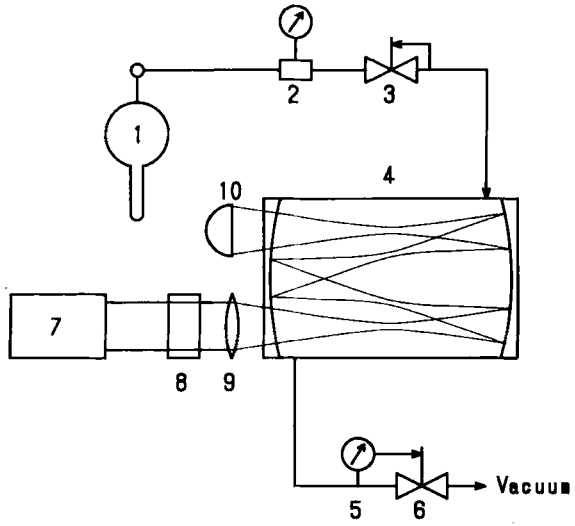

FIG. 2. Experimental set-up: 1) gas reservoir, 2) mass-flow controller, 3) mass-flow valve, 4) multiple reflection reactor, 5) capacitance manometer, 6) servo-valve for auto-pressure control, 7) TEA CO_2 laser, 8) attenuator, 9) BaF_2 lens, 10) pyroelectric detector.

made on C_2HClF_4. The total reaction volume ΣV_R per pulse for C_2TClF_4 was obtained from the batch irradiation experiment by using the relation

$$\Sigma V_R = -(V_{cell}/t) \ln (1 - X_T) \tag{1}$$

where X_T is the fraction of C_2TClF_5 dissociated after t-pulse irradiation and V_{cell} is the cell volume (8460 cm^3). Figure 3 shows the dependence of ΣV_R on the incident pulse energy, where ΣV_R is the sum of the four reaction volumes in the cell. From the fourth-power dependence observed in Fig. 3, we conclude that the reaction was unsaturated in the focal zones. The mass-balance equation of C_2TClF_4 in the continuous reactor is given by

$$V_{cell}\frac{dC}{d\theta} = (C_0 - C)Q - Ch\Sigma V_R \tag{2}$$

where C_0, C, Q, h, and θ denote the inlet concentration, the outlet concentration, the volumetric flow rate, the laser pulse repetition rate, and time, respectively. Using the initial condition $C = C_0$ at $\theta = 0$, we obtain:

$$\frac{C}{C_0} = 1 - X_T = \frac{1 + \dfrac{h\Sigma V_R}{Q} \exp\left\{-\left(\dfrac{h\Sigma V_R + Q}{V_{cell}}\right)\theta\right\}}{1 + \dfrac{h\Sigma V_R}{Q}} \tag{3}$$

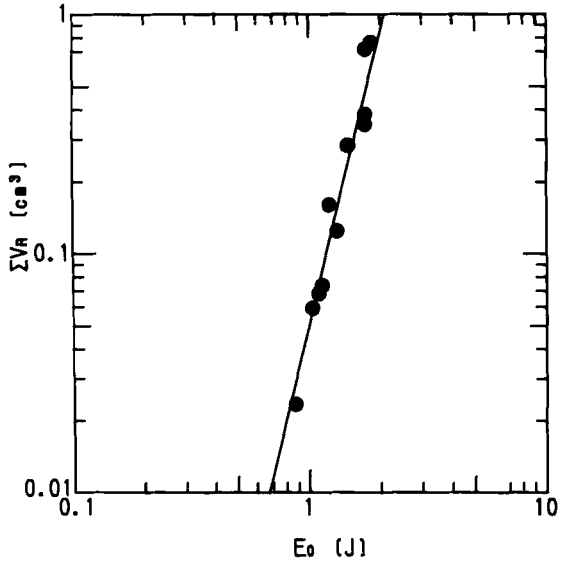

FIG. 3. Results of the batch irradiation experiment: V_R as a function of E_0.

From the exponent in Eq. (3), the characteristic transient time θ_{trans} is $V_{cell}(h\Sigma V_R + Q)^{-1}$. Figure 4 shows the transient behavior of the reactor: $1 - X_T$ as a function of time and the incident pulse energy E_0.The transient performances of the reactor are shown in Figs. 5 and 6 for the variables Q and h, respectively. In Figs. 4, 5, and 6, only one of the three parameters (E_0, Q, h) was varied from the standard operating condition (E_0 = 1.73 J, Q = 1.83 cm^3/s, and h = 5 s^{-1}). It was confirmed experimentally that $3\theta_{trans}$ was sufficient for the system to reach the steady-state.

The steady-state behavior of the reactor is given by

$$1 - X_T = (1 + h\Sigma V_R/Q)^{-1} \tag{4}$$

The steady-state experimental results shown in Figs. 7, 8, and 9 are reproduced well by Eq. (4).

CONCLUSION

A multiple reflection photoreactor in which the laser beam was focused four times was designed and tested. The transient and steady-state behaviors of the reactor are described well by our proposed model. Such multiple reflection will be necessary for the laser isotope separation of tritium using such working molecules as CTF_3/CHF_3 and C_2TClF_4/C_2HClF_4 where an optical path length of several tens of meters will be required. The reactor

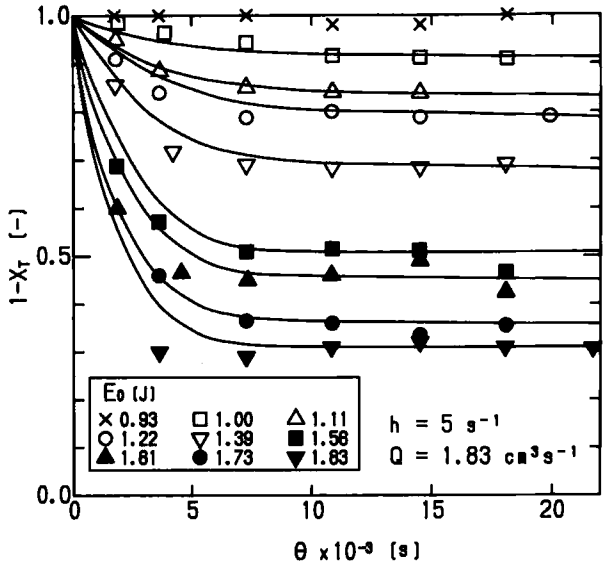

FIG. 4. Transient behavior of the reactor: $1 - X_T$ as a function of time θ and E_0.

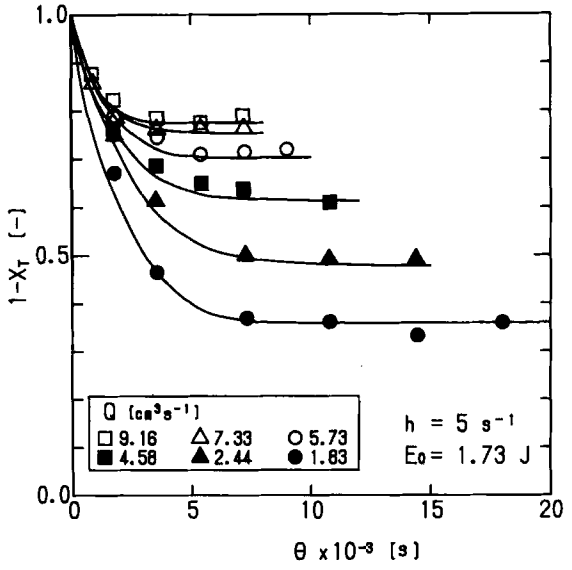

FIG. 5. Transient behavior of the reactor: $1 - X_T$ as a function of time θ and volumetric flow rate Q.

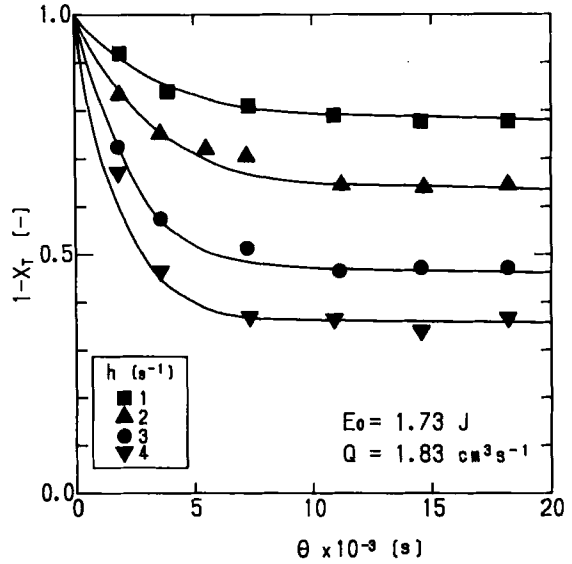

FIG. 6. Transient behavior of the reactor: $1 - X_T$ as a function of time θ and laser repetition rate h.

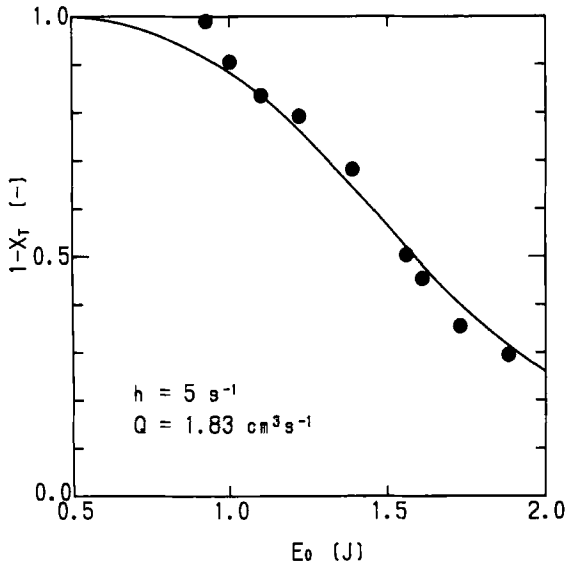

FIG. 7. Steady-state behavior of the reactor: $1 - X_T$ vs E_0. The solid line is from Eq. (4).

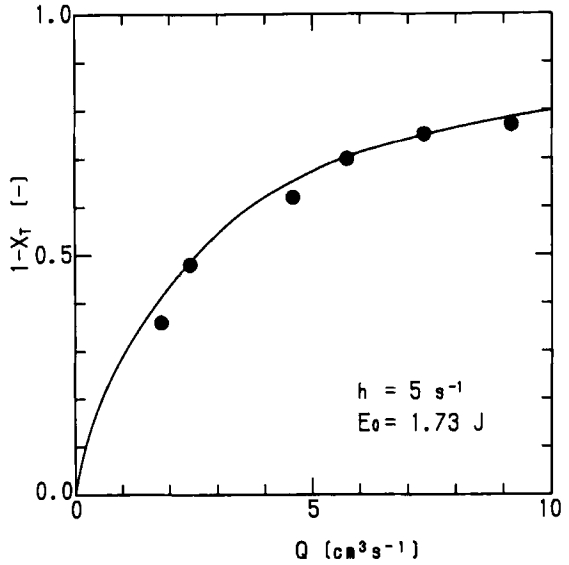

FIG. 8. Steady-state behavior of the reactor: $1 - X_T$ vs Q. The solid line is from Eq. (4).

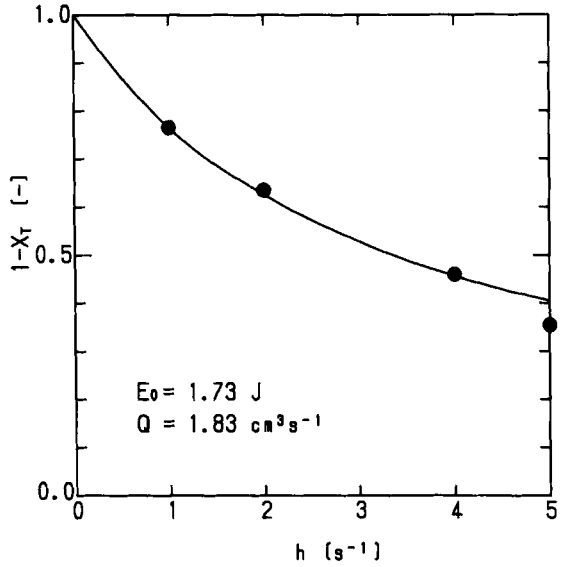

FIG. 9. Steady-state behavior of the reactor: $1 - X_T$ vs h. The solid line is from Eq. (4).

length may be shortened to the order of 10 m by using a reactor of the compartment-in-series type with multiple reflection in each compartment.

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