Proceedings of the 6th International Symposium
on
Advanced Nuclear Energy Research
—INNOVATIVE LASER TECHNOLOGIES IN NUCLEAR ENERGY—

SEPARATION OF BORON ISOTOPES BY INFRARED LASER

Kazuya.SUZUKI
Department of Chemistry and Fuel Research
Tokai Reseach Establishment, JAERI
Tokaimura, Ibarakiken, 319-11, Japan

Vibrationally excited chemical reaction of boron tribromide(BBr_3) with oxygen(O_2) is utilized to separate ^{10}B and ^{11}B. Infrared absorption of $^{10}BBr_3$ is at 11.68μ and that of $^{11}BBr_3$ is at 12.18μ. The wavelengths of ammonia laser made in the laboratory were mainly 11.71μ,12.08μ and 12.26μ. Irradiation was done by focussing the laser with ZnSe lens on the sample gas (mixture of 1.5 torr of natural BBr_3 and 4.5 torr of O_2) in the reaction cell. Depletions of $^{10}BBr_3$ and $^{11}BBr_3$ due to chemical reaction of BBr_3 with O_2 was measured with infrared spectrometer. The maximum separation factor $\beta(^{10}B/^{11}B)$ obtained was about 4.5

Keywords: Boron Isotopes, Ammonia laser

1. Introduction

Since R.V.Ambartsumyan, et.al have proved boron isotope separation by CO2 laser irradiation of boron trichloride (BCl_3) [1], many investigations on laser-induced reaction of BCl_3 with various acceptors have been reported[2,3,4,5]. It has become clear that strong laser field is necessary for dissociation of BCl_3 due to large B-Cl bond energy [10] and low density of vibrational energy levels of BCl_3. Increase of laser field brings about break down of BCl_3 gas and leads to decrease of selectivity of ^{10}B and ^{11}B. To overcome this difficulty, R.V.Ambartsumyan, et al proposed two frequency laser irradiation method and have shown increase of osmium isotope separation.[6,7] Developing this method, we tried multifrequency laser irradiation of BBr_3 molecule. By using NH_3 laser with multifrequency oscillation, the optimum combination of irradiation wavelengths of the laser for selective excitation of $^{10}BBr_3$ was researched.

2. Experimental

The arrangement of NH_3 laser and the irradiation system is shown in Fig.1. The pumping source of NH_3 laser was TEA CO_2 laser(Lumonics Type 202). The NH_3 laser tube was 40 mm i.d., 3.6 m long, made of stainless steal and set up collinear to CO2 laser[8,9]. The entrance window of the tube was NaCl and the exit of it was uncoated ZnSe. Resonator of NH_3 laser consisted of ZnSe output window of the CO_2 laser and that of NH_3 laser. The mixtures of NH_3 gas with nitrogen(N_2) gas and various inert gases were used as operating gas. The gas used in the experiment was mainly 1) 6%-0.2% NH_3 in N_2 , 2)6% NH_3 in He and 3)6% NH_3 in Ar. In this layout of the NH_3 laser without a grating, simultaneous oscillation of multi-wavelengths occurred. Distribution of energy of NH_3 laser lines depends on the pumping CO_2 lines. In case of pumping by 9R(30) CO_2 laser, the output NH_3 laser lines were sP(7,0) 12.08μ , aP(6,0) 12.26μ, aP(5,K) K=1,2,3,... 12.00μ , sP(5,0) 11.53μ , and aP(4,0) 11.71μ. Distribution of output of the laser lines controlled by changing the ratio of NH3 to buffer gas, which causes rotational relaxation of NH_3 molecule.
Freon gases (CH_3F, CCl_2F_2 and CCl_4) in a cell(40 mm i.d.,15 cm length) with NaCl or KCl windows were used as an optical filter, to select the suitable NH_3 laser lines and also to absorb residual CO_2 laser radiation.

Relative outputs of NH_3 laser lines were measured through a monochrometer(JASCO CT-10) with a joulemeter(Gentec ED-200, ED-500) and pulse forms of them were observed through the monochrometer with a HgCdTe detector(LABIMEX) at room temperature.

Preparation and irradiation of the sample BBr_3 was done as follows. The BBr_3 reagent from E.Merk in natural isotope composition($^{10}BBr_3$ 20%, $^{11}BBr_3$ 80%) was distilled in vacuum and the mixture of 1.5 torr BBr_3 and 4.5 torr of O_2 was enclosed in a pyrex cell with NaCl windows. The size of the cell was 1.5 cm i.d. and 15 cm long. Grease-free stopcocks were used in the vacuum line for treating BBr_3 gas.

Irradiations on the sample were done by focussing the laser beam through ZnSE lens on the center of the cell. The focal length of the lens was 50 cm. After irradiation depletions of concentration of $^{10}BBr_3$ and $^{11}BBr_3$ due to reaction with O_2 were measured by infrared spectrometer(JASCO A-202).

3. Results and Discussion

The fundamental vibration ν_3 of $^{10}BBr_3$ is at 856.8 cm^{-1} and that of $^{11}BBr3$ is at 820.4 cm^{-1} in our measurement. The laser line 11.71μ(854 cm^{-1}) is nearest to ν_3 of $^{10}BBr_3$ and the line 12.26μ(816 cm^{-1}) is nearest to that of $^{11}BBr_3$.

In Fig.2 to in Fig.4, depletions of $^{10}BBr_3$ and $^{11}BBr3$ of BBr_3 in natural abundance irradiated with NH_3 laser pulse, generated by 9R(30) CO_2 laser pumping are shown. Also, infrared spectra of BBr_3 and the position of the NH_3 laser lines, which mainly excite $^{10}BBr_3$ are shown in small window.

In Fig.2, the result of irradiation with NH3 11.71μ line, which is coincident with ν_3 peak of $^{10}BBr3$, is shown. Although the pulse energy of the laser is comparatively large(460mJ), irradiation with this line seems to be not effective for excitation of $^{10}BBr_3$.

In Fig.3, result of irradiation with the 12.00μ and 12.08μ lines is shown. Infrared absorption of $^{10}BBr_3$ in ground state does not exist in this region and these lines are not effective.

In Fig.4, the result of irradiation with the 11.71μ line, together with the 12.00μ line and 12.08μ line is shown. The total pulse energy is equal to the case of Fig.5. It is clearly seem that addition of the 12.00μ line and 12.08μ line promotes excitation of $^{10}BBr_3$. This results suggests that infrared absorption of excited $^{10}BBr_3$ seems to exist in 12.00μ to 12.08μ region.

Summarizing the results of Fig.2 to Fig.4, fractional depletion per pulse α of $^{10}BBr_3$ and that of $^{11}BBr_3$ is shown in Table.1 and also the values of single step $^{10}B/^{11}B$ separation factor β , which is defined as the ratio of α of $^{10}BBr_3$ to that of $^{11}BBr_3$ is shown.

From the experimental results of Fig.2 to Fig.4, one can fully recognize effectiveness of multifrequency irradiation, which promotes sequential excitation along with vibrational levels of BBr_3 molecule.

References

1. R.V.Ambartsumyan, V.S.Letkhov, E.A.Ryabov, N.V.Chekalin: ZhETF Pis.Red. 20, 597(1974) [JETP Lett., 20, 273(1974)]
2. S.M.Freud, J.J.Ritter: Chem.Phys.Lett., 32, 255(1975)
3. R.V.Ambartumyan, Yu.A.Gorokhov, V.S.Letkhov, G.N.Makarov, N.V.Chechalin: Kvant.Elektron. (Mos) 2, 2197(1975)

[Sov.J.Quant.Electron. 5, 1196(1976)]

4. S.D.Rockwood, J.W.Hudson: Chem.Phys.Lett. 34, 542(1975)

5. H.Kojima, T.Fukumi, K.Fukui, K.Naito: J.Phys.Chem. 84,2528(1980)

6. R.V.Ambartsumyan, Yu.A.Gorokhov, V.S.Letokhov, G.N.Makarov, A.A.Puretskii, N.P.Furzikhov: Pis'ma Zh.Eksp.Teor.Fiz. 23, No4, 217(1976) [JETP Lett.,23, No4, 194(1976)]

7. R.V.Ambartsumyan, Yu.A.Gorokhov, G.N.Makarov, A.A.Puretskii, N.P.Furzikhov: Kvantovaya Elektron(Moscow) 4, 1590-1591(1977) [Soviet J.Quantum Electron. Vol 7, No 7, 904-905 (1977)]

8. K.Midorikawa, I.Matsuda, M.Obara, T.Fujioka: Optics Comm., 32, 447(1980)

9. K.Midorikawa, I.Matsuda, M.Obara, T.Fujioka: Opt.Lett., 6, 177(1981)

10. R.T.Sanderson: Chemical Bond and Bond Energy, p180, Academic Press(1971)

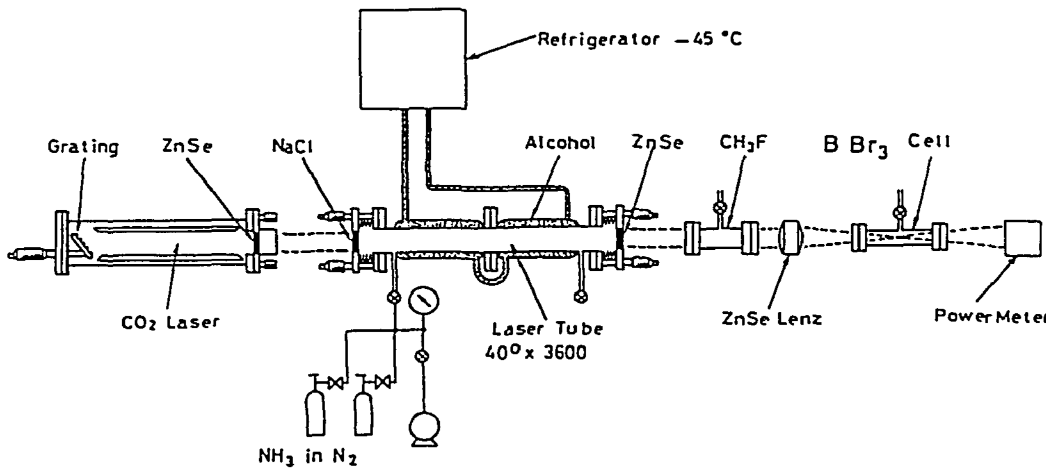

Fig.1. Experimental arrangement of the NH_3 laser pumped with the CO_2 laser, the freon filter, ZnSe lens (f=50 cm) and the reaction cell

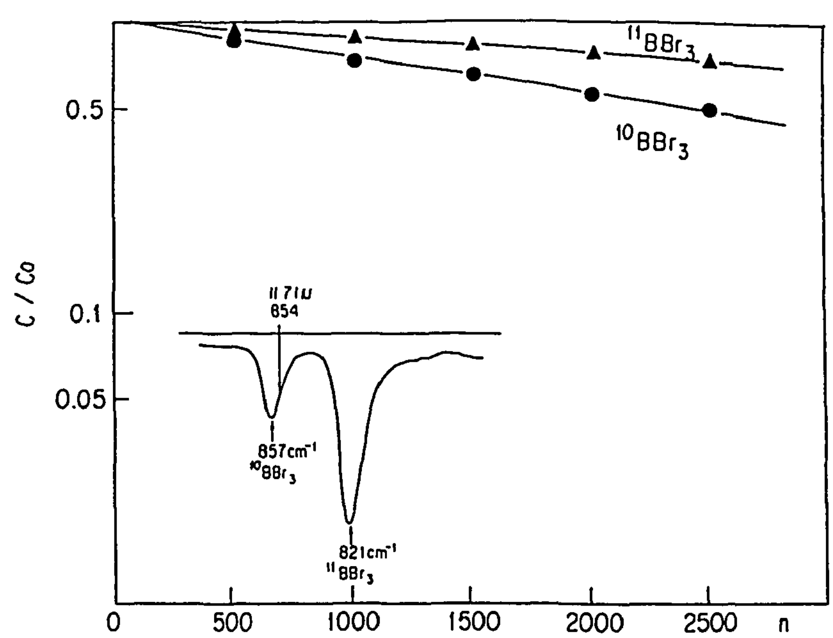

Fig.2. Relative concentration(C/C_0, C is concentration of BBr3 in the cell after n pulse irradiation, C_0 is initial concentration) of $^{10}BBr_3$ and $^{11}BBr_3$ vs number of laser pulse(n). The NH_3 laser pulse was generated by pumping of NH_3 gas(0.2% NH_3 in N_2, 400 torr) with CO_2 laser line 9R(30). Filter is CH_3F(300 torr). The pulse energy E is 460 mJ(11.71μ 98%, 12.26μ 2%)

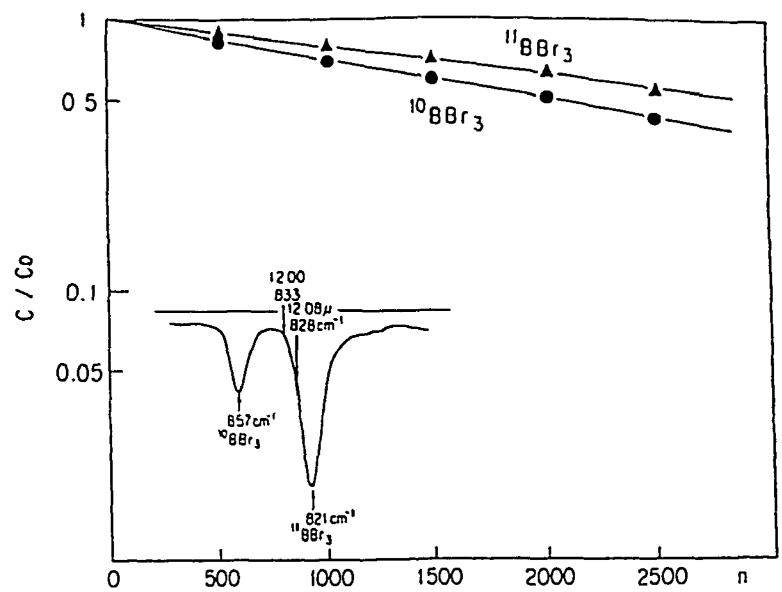

Fig.3. Relative concentration (C/C_0)of BBr_3 vs number of laser pulse(n). Pulse energy of NH_3 laser E=251 mJ(12.00μ 36%, 12.08μ 60%, 12.26μ 4%), Operating NH_3 gas (6% NH_3 in He, 30 torr), pumped with CO_2 laser line 9R(30). Filter, CH_3F 300 torr.

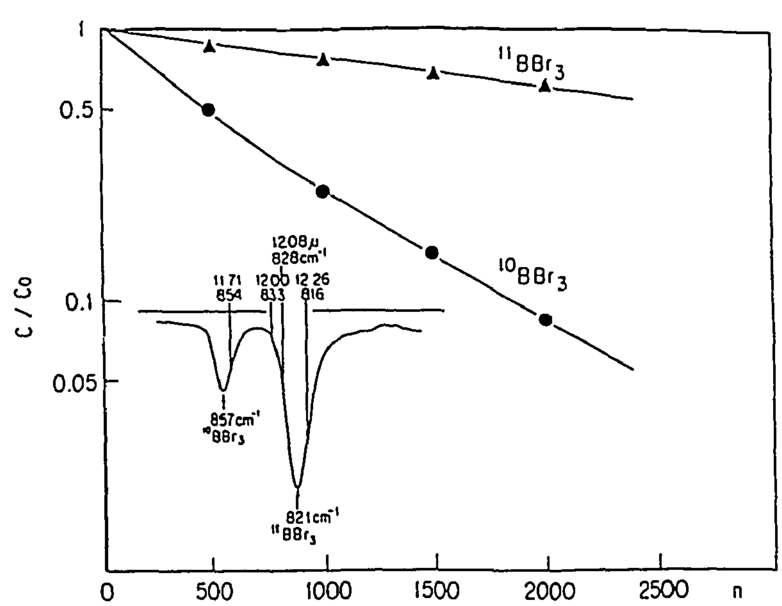

Fig.4. Relative concentration(C/C_0) of BBr_3 vs number of laser pulse(n). Pulse energy of NH_3 laser E=460 mJ(11.71μ 34%, 12.00μ 34%, 12.08μ 22%, 12.26μ 10%), Operating NH_3 gas (0.5% NH_3 in N_2, 180 torr), pumped with CO_2 laser line 9R(30). Filter, CH_3F 300 torr and CCl_4 80 torr.

Table.1. Results of the experiments (Fig.4 to Fig.6).

Fractional depletion per laser pulse α and separation factor β

Laser energy (mJ)	Wavelength (μ)	α of $^{10}BBr_3$ ($\times10^{-4}$)	α of $^{11}BBr_3$ ($\times10^{-4}$)	β($^{10}B/^{11}B$)
460	11.71(98%) 12.26(2%)	2.27	1.20	2.31
251	12.00(36%) 12.08(60%) 12.26(4%)	3.33	2.25	1.48
460	11.71(34%) 12.00(34%) 12.08(22%) 12.26(10%)	14.68	3.25	4.51