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Bromine and Oxygen Quenching of Positronium in Silica Gel

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Bromine and Oxygen Quenching of Positronium in Silica Gel*

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Abstract

Positron lifetime spectra for various concentrations of bromine and oxygen acsorbed on silica gel are investigated. Two-photon angular correlation experiments are also performed on some of these samples to investigate the quenching process of bromine and oxygen. The cross section for bromine quenching of orthopositronium in silica gel is about two orders of magnitude larger than that for oxygen quenching. The process of bromine quenching of positronium is found to be chemical quenching. Oxygen quenching of positronium in silica gel is found to be conversion quenching. The interaction between positronium and bromine or oxygen in silica gel is found to be similar to that found in the gaseous state. The energy of Ps annihilating in the pores of silica gels is found to be 0.25 ± 0.10eV.

Key Words: Positronium Quenching, Silica Gel, Bromine and Oxygen

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I. Introduction

Recently, porous oxides have been used to produce long life and high intensity orthopositronium for investigating physical and chemical properties of positronium and molecule-positronium interaction [1; 2; 3]. In our previous paper we demonstrated that silica gel is a good substrate for studying inhibition and quenching of positronium [1]. Silica gel is composed of closely bonded primary particles of silica [4]. The voids between these particles are called pores. When a positron enters such a medium, it may annihilate directly with an electron or it may capture an electron to form positronium in either a singlet state or triplet state. Singlet positronium (p-Ps) will annihilate quickly while triplet positronium (o-Ps) may have a chance to diffuse into the pores and survive much longer [5].

Experimentally, three positron lifetimes were observed from measurements of positron annihilation in silica gels [1; 2; 6]. The shortest meanlife, $\tau_1 \sim 10^{-10}$ sec, can be attributed to the direct annihilation of free positrons and the annihilation of p-Ps. The second meanlife, $\tau_2 \sim 10^{-9}$ sec, is attributed to the pickoff annihilation of o-Ps inside silica particles. The longest meanlife, $\tau_3 \sim 10^{-8}$ sec, has been shown to be due to the annihilation of o-Ps in the pores or voids between particles [1; 5]. The value of τ_3 not only depends on the pore size but also is highly dependent on the interaction between o-Ps and the molecules on the pore surface.

Previously, we have studied positronium lifetimes in various

molecules, such as iodine, carbon tetrachloride, water, n-hexane, nitromethane, etc., adsorbed on silica gels [1; 2]. Among these molecules, iodine was found to be an extremely effective quencher of positronium. Even one micro-mole of iodine adsorbed on one gram of silica gel was sufficient to quench the τ_3-component. In this paper, we investigated the positronium quenching effect due to another halogen, bromine, as well as oxygen adsorbed in silica gel. We also studied quenching of positronium using the method of two-gamma angular correlation.

II. Experimental

The positron lifetime spectra were measured by using a delayed coincidence system with two time-to-amplitude converters (TAC). The timing pulses produced by the photomultipliers are split into two pairs and fed into two different ranges of TAC. The short-range TAC has a linear region of 30 nsec, and the other has a linear region of 300 nsec. This apparatus had an operational time resolution of f.w.h.m. = 0.5 nsec. The details of this apparatus were described previously [7].

Davison grade 05 silica gel was used. Davison technical literature gives the following information about grade 05 gel: surface area = 800 m^2/g, pore volume = 0.43 cc/g, and bulk density = 0.72 g/cm^3. The procedure for preparing the samples for lifetime measurements is as follows. A McBain type of adsorption system with a precision cathetometer was used for preparing oxygen adsorbed samples. A schematic layout of the adsorption apparatus is shown in Figure 1. A glass bucket, containing 2.5 g of dried silica gel and a positron

source, was suspended on a quartz spiral spring. The spring was hung in a long glass tube which connected to the vacuum line. A large glass flask was also connected onto the same system. The whole system was first outgassed for 24 hours at a pressure of about 10^{-5} Torr. Pure oxygen gas was then transferred and stored in the flask. The positron lifetime measurement was performed on dried silica gel first. Then a small amount of pure oxygen gas was transferred from the flask to the glass tube containing the silica gel sample. A cathetometer was used to determine the concentration of oxygen adsorbed on silica gel by measuring the extension of the calibrated quartz spring. The positron lifetime spectrum was measured again. The procedure for adsorption of bromine on silica gel was similar to that of iodine and other liquids reported previously [1; 2].

A standard long slit type of angular correlation apparatus was used for investigating the conversion quenching of positronium. A detailed description of this system was reported previously [8]. A typical counting rate in the center of a distribution was about 20 counts per minute using plastic scintillators 1.5" diameter and 6" long with a 1 mCi Na^{22} source and 0.8 mrad slits. More than 20,000 counts were accumulated for each point in the center region of the distribution.

III. Results

Positron lifetime spectra for bromine-silica gel and oxygen-silica gel systems were all decomposed into three lifetime components,

τ_1, τ_2, and τ_3, as in the procedure described previously [1; 6]. The values of the longest meanlife τ_3 and the relative intensities of the two long lifetime components, I_2 and I_3, of positron annihilating in various concentrations of bromine adsorbed silica gel are shown in Figure 2. The concentration of bromine is in a unit of 1 micro-mole of bromine per gram of silica gel. Similarly, the values of τ_3, I_2, and I_3 at various concentrations of oxygen adsorbed on silica gel are shown in Figure 3. The concentration of oxygen is in a unit of 10^{-4} mole per gram of gel.

Two-photon angular correlation curves of annihilating positron-electron pairs in degassed silica gel, air-saturated silica gel, oxygen adsorbed silica gel (0.9×10^{-4} mole of oxygen per gram of silica gel), and two different concentrations of bromine adsorbed silica gels, 1 micro-mole of bromine per gram of gel and 100 micro-moles of bromine per gram of gel, are shown in Figure 4. All angular distributions in Figure 4 are corrected for a finite angular resolution and normalized to the same area. For comparison, we also plot momentum distributions of annihilating positron-electron pairs in these samples in Figure 5, in which all momentum distributions are normalized to the same height of the most probable momentum of the high momentum component. The intensities of the low momentum components I_N for all samples are listed in Table 1. Their corresponding values of τ_3, I_2 and I_3 are also listed in Table 1.

IV. Discussion and Conclusion

A. Bromine quenching of positronium in silica gel

As shown in Figure 2, the longest meanlife τ_3 is drastically quenched by the presence of a small concentration of bromine adsorbed on silica gel. This is similar to the quenching effect of iodine in silica gel as reported previously [1]. The annihilation rate λ_3, or $1/\tau_3$, increases linearly with the concentration of bromine as shown in Figure 6. The quenching rate constant K_q can be obtained from the slope of λ_3. It was found that the quenching rate constant is

$$K_q = \frac{d\lambda_3}{dM} = 1.50 \times 10^{14} \text{ g-mole}^{-1}\text{sec}^{-1}$$

which is the same order of magnitude as that for iodine quenching in silica gel [1], or equivalent to $3.43 \times 10^{-10} \text{cm}^3\text{sec}^{-1}$.

To understand the origin of bromine quenching of positronium, we performed positron angular correlation measurements for two different concentrations of bromine adsorbed silica gel. The intensity of the low momentum component I_N for the sample with a concentration of 1×10^{-6} mole of bromine per gram of gel is reduced from 16% for the pure silica gel to about 11%, and the momentum distribution of the low momentum component becomes broader than that of pure silica gel. This clearly indicates that bromine quenching of positronium in silica gel is not conversion quenching. Therefore, chemical quenching is a likely explanation.

For a concentration of bromine in silica gel of 1×10^{-4} mole/g, no long positron lifetime was observed, and the narrow component of the angular distribution practically disappeared. This shows that bromine

adsorbed on silica gel has a very high value of Z_{eff} which has also been observed in a bromine-gas system [9].

B. Oxygen quenching of positronium in silica gel

As shown in Figure 3, the values of τ_3 and I_3 both decrease and the value of I_2 increases at the expense of I_3 as the concentration of oxygen is increased. This indicates that oxygen in silica gel is a good positronium quenching agent and is not an inhibitor to the formation of positronium. The annihilation rate λ_3 plotted against the concentration of oxygen is shown in Figure 7. As in the bromine-silica gel sample, a linear relationship between λ_3 and the concentration (M) of oxygen in silica gel is observed. The quenching rate constant for oxygen quenching in silica gel is found to be

$$K_q = 7.66 \times 10^{11} \text{ g-mole}^{-1}\text{sec}^{-1},$$

or equivalent to $1.75 \times 10^{-12} \text{cm}^3\text{sec}^{-1}$, which is similar to the results obtained from oxygen quenching of o-Ps in gases, $(0.8 \text{ to } 2.4) \times 10^{-12} \text{cm}^3 \text{sec}^{-1}$ [10].

From Figure 5 the intensity of the low momentum component I_N for air saturated silica gel is found to be 25%, about 9% more than the I_N of degassed silica gel. The momentum distribution of the low momentum component for air saturated silica gel is sharper than that of degassed silica gel. The most probable momentum of p-Ps, i.e., the maximum of the low momentum component, in air saturated silica gel was determined to be 1.1×10^{-3} mc or equivalent to a kinetic energy of 0.3 eV, while the most probable momentum of p-Ps observed in degassed silica gel was

1.5×10^{-3} mc, corresponding to 0.56 eV. This is attributed to the conversion quenching of o-Ps in the pores of silica gel by oxygen molecules. The concentration of oxygen in air saturated silica gel was determined to be 0.34×10^{-4} mole per gram of silica gel. The sharpness of the momentum distribution of the low momentum component and its intensity I_N were further intensified when pure oxygen was introduced into silica gel at a pressure of 700 mm Hg. The oxygen concentration was measured to be 0.9×10^{-4} mole per gram of gel. The value of I_N at this oxygen concentration was 28%. This shows that most of the o-Ps inside the pores was converted into p-Ps by oxygen molecules. The momentum distribution of the low momentum component was very sharp with a maximum at 1.0×10^{-3} mc. By comparing this distribution with that of degassed silica gel, we found that all converted Ps has a momentum in the range of $(1.0 \pm 0.2) \times 10^{-3}$ mc corresponding to a kinetic energy of 0.25 ± 0.10 eV. This is close to the calculated zero energy of Ps inside the pores of silica gel, $\sim$0.2 eV, using a potential model which assumes Ps atoms being "trapped" or "adsorbed" near the pore surface [6]. It is interesting to note that this kinetic energy of Ps in the pores of silica gel is similar to the observed kinetic energy of o-Ps escaping from MgO powder, 0.28 ± 0.10 eV [11].

From the above results, we can conclude that the process of bromine quenching of positronium in silica gel is chemical quenching and oxygen quenching of positronium in silica gel is via the conversion process, and that the interaction between positronium and bromine or

oxygen in silica gel is similar to that in the gaseous state. Our finding agrees very well with the results obtained by Goldanskii and his colleagues [12].

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References

1. S. Y. Chuang, S. J. Tao: J. Chem. Phys. 52, 749 (1970).

2. S. Y. Chuang, S. J. Tao: J. Chem. Phys. 54, 4902 (1971).

3. D. J. Judd, Y.K. Lee, L. Madansky, E. R. Carlson, V. W. Hughes, B. Zundell: Bull. Am. Phys. Soc. 18, 83 (1973).

4. E. G. Acher: J. Colloid Interface Sci. 32, 41 (1970).

5. W. Brandt, R. Paulin: Phys. Rev. Letters 21, 193 (1968).

6. S. Y. Chuang, S. J. Tao: Can. J. Phys. 51, 820 (1973).

7. S. J. Tao: Phys. Rev. A2, 1669 (1970).

8. J. B. Wang, S. Y. Chuang, S. J. Tao: Nucl. Inst. Meth. 108, 253 (1973).

9. S. Y. Chuang, S. J. Tao: to be published.

10. G. J. Celitans, S. J. Tao, J. H. Green: Proc. Phys. Soc. 83, 833 (1964); A. D. Mokrushin, V. I. Goldanskii: Soviet Phys. Jetp. 26, 314 (1968).

11. S. M. Curry, A. L. Schawlow: Phys. Lett. A 37, 5 (1971).

12. V. I. Goldanskii, A. D. Mokrushin, A. O. Tatur, V. P. Shantarovich: Kinet. Katal. 13, 961 (1972).

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Table 1

Comparison Between the Results from Lifetime and Angular Correlation Methods for Positronium Annihilation in Silica Gel 05[†]

Silica Gel	τ_3(nsec)	I_3(%)	I_2(%)	I_2+I_3(%)	Low Momentum component		
					I_N(%)	Peak Energy (eV)	Width
Degassed	32.0±0.5	29.0±1.5	4.0±0.5	33±2	16±1	0.56	Medium
Air	17.0±0.5	28.0±1.5	5.0±0.5	33±2	25±1	0.30	Sharp
Oxygen (1×10^{-4} mole/g)	9.6±0.4	22.0±1.5	8.0±0.5	30±2	28±1	0.25	Sharp
Bromine (1×10^{-6} mole/g)	8.0±0.3	16.0±1.5	9.0±0.5	25±2	11±1	0.65	Broad
Bromine (1×10^{-4} mole/g)	-	0	-	-	0	-	-

[†]Silica Gel 05: Pore volume - 0.43 cc/g
Average pore diameter - 22 Å
Surface area - 800 m^2/g

Figure Captions

Figure 1 Diagram of sorption apparatus for lifetime measurements.

Figure 2 τ_3, I_3, and I_2 as functions of the concentration of bromine in silica gel.

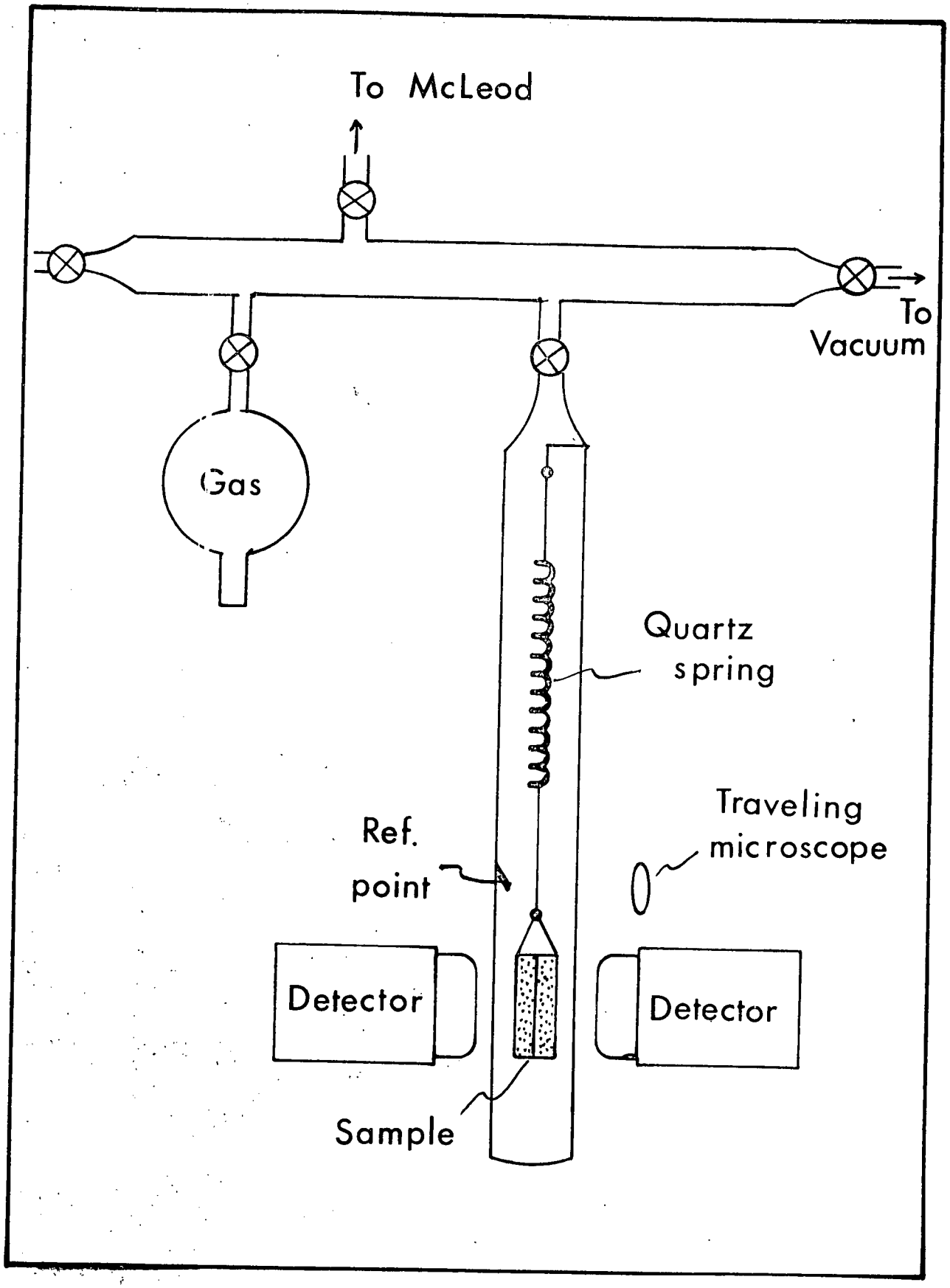
Figure 3 τ_3, I_3 and I_2 as functions of the concentration of oxygen in silica gel.

Figure 4 Angular distributions of annihilating positron-electron pairs in (A) oxygen adsorbed silica gel (0.9×10^{-4} mole of oxygen per gram of gel), (B) air saturated silica gel, (C) pure silica gel (degassed), (D) bromine adsorbed silica gel (1×10^{-6} mole/g), and (E) bromine adsorbed silica gel (1×10^{-4} mole/g). All curves are corrected for random background and for a finite angular resolution and normalized to the same area.

Figure 5 Momentum distributions of annihilating positron-electron pairs in (A) oxygen adsorbed silica gel (0.9×10^{-4} mole of oxygen per gram of gel), (B) air saturated silica gel, (C) pure silica gel (degassed), (D) bromine adsorbed silica gel (1×10^{-6} mole/g), and (E) bromine adsorbed silica gel (1×10^{-4} mole/g). All curves are normalized to the same height of the most probable momentum of the high momentum component.

Figure 6 Annihilation rate λ_3, or $1/\tau_3$, as a function of the concentration of bromine in silica gel.

Figure 7 Annihilation rate λ_3, or $1/\tau_3$, as a function of the concentration of oxygen in silica gel.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To
Vacuum
Gas
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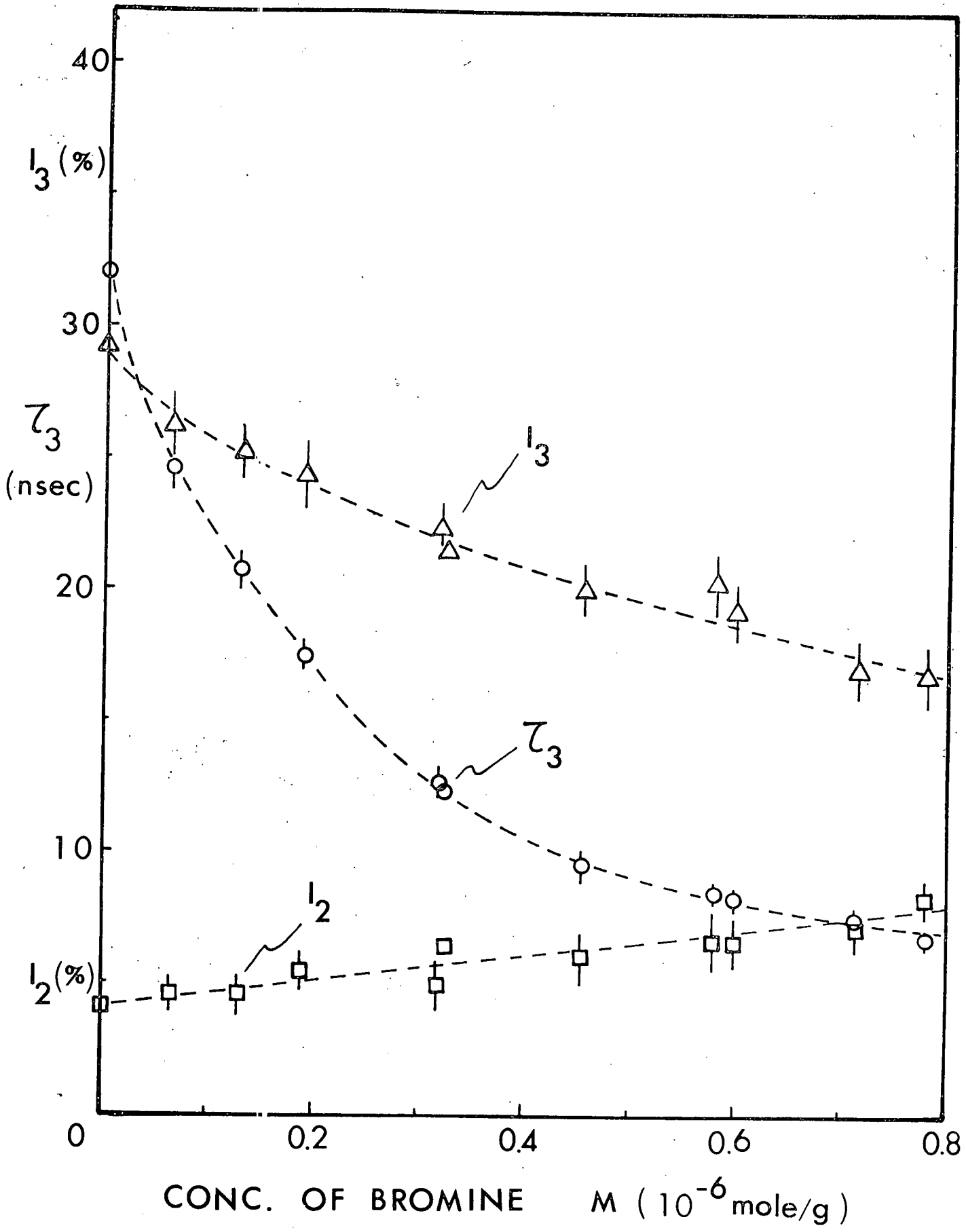
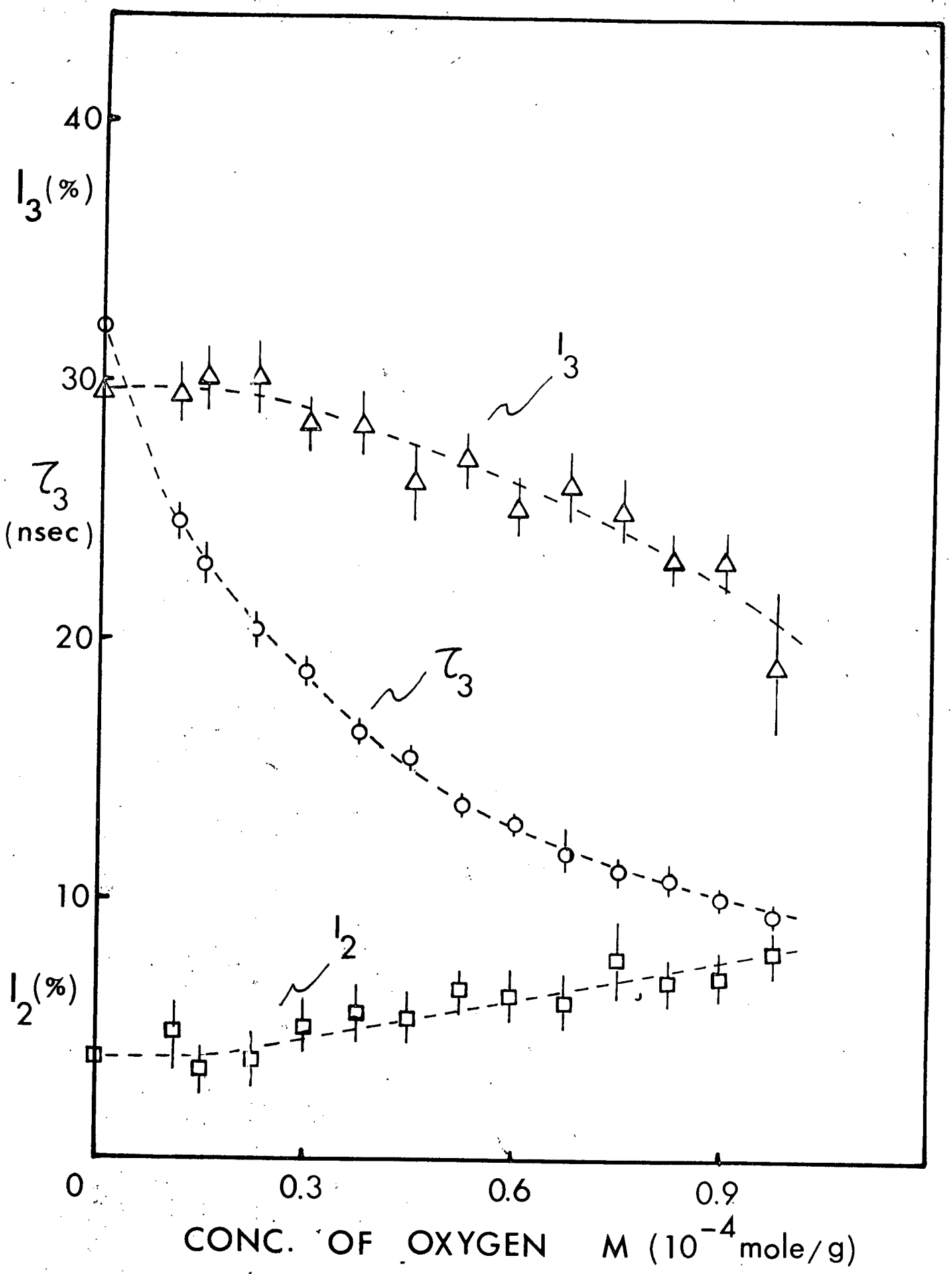
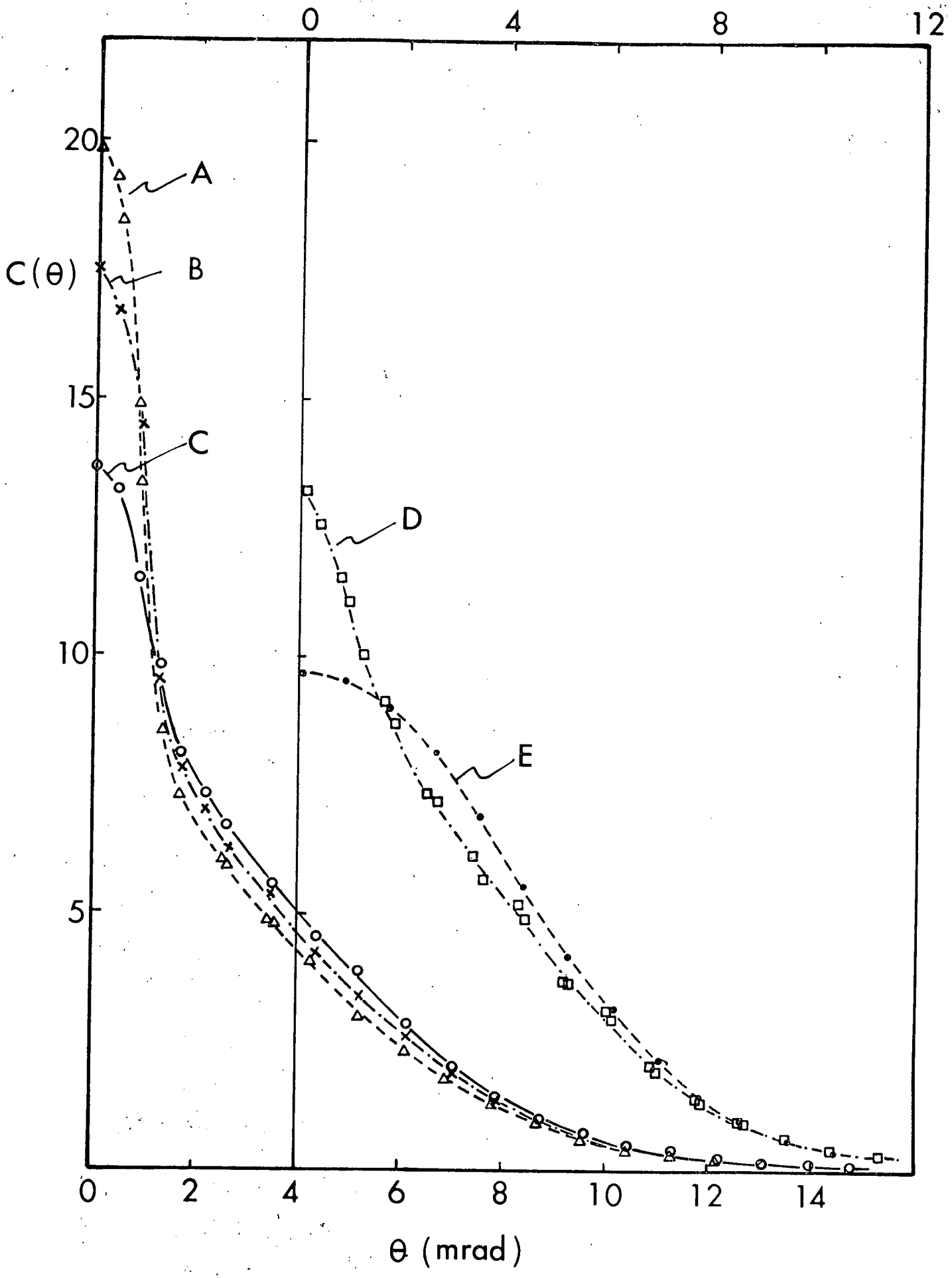
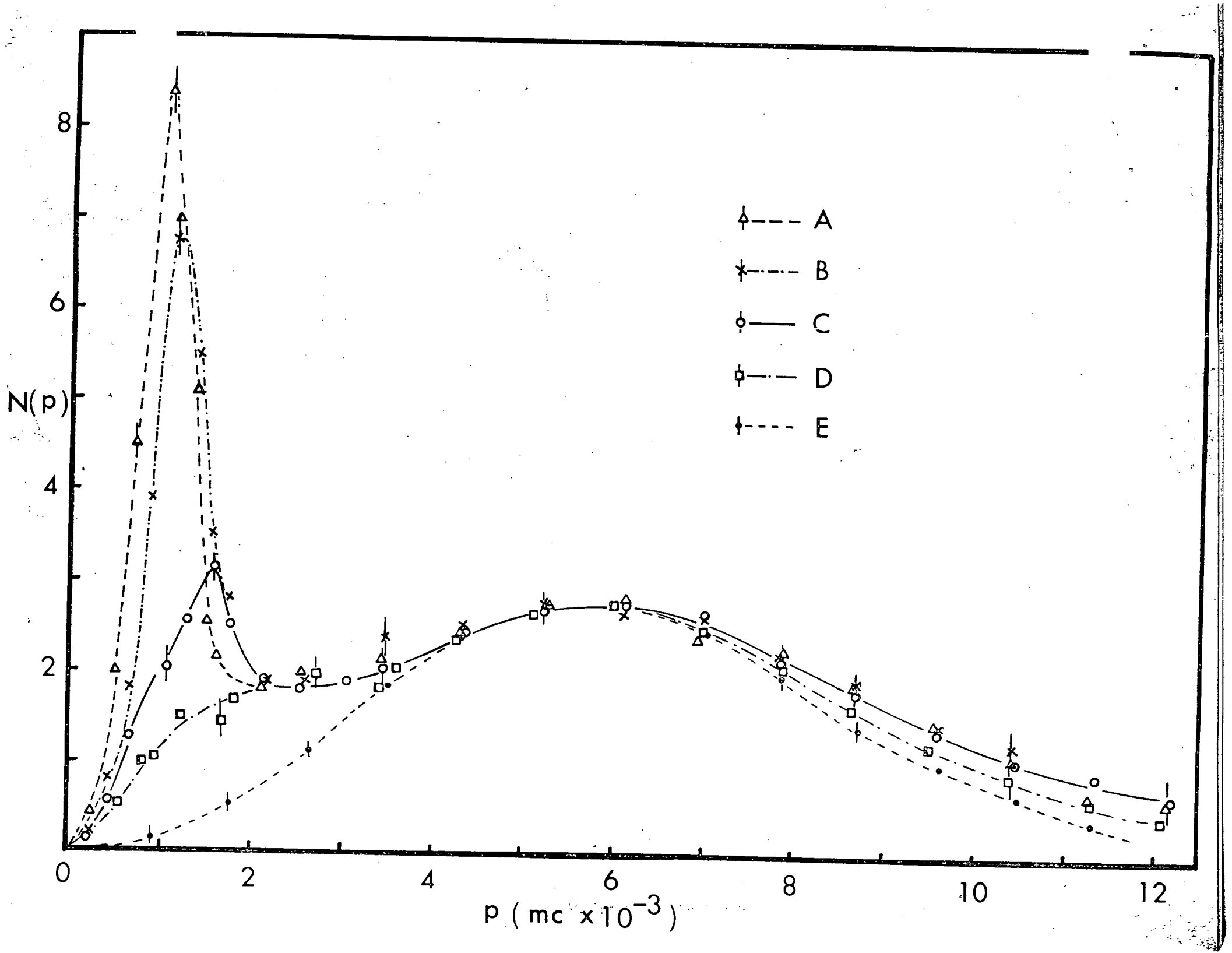
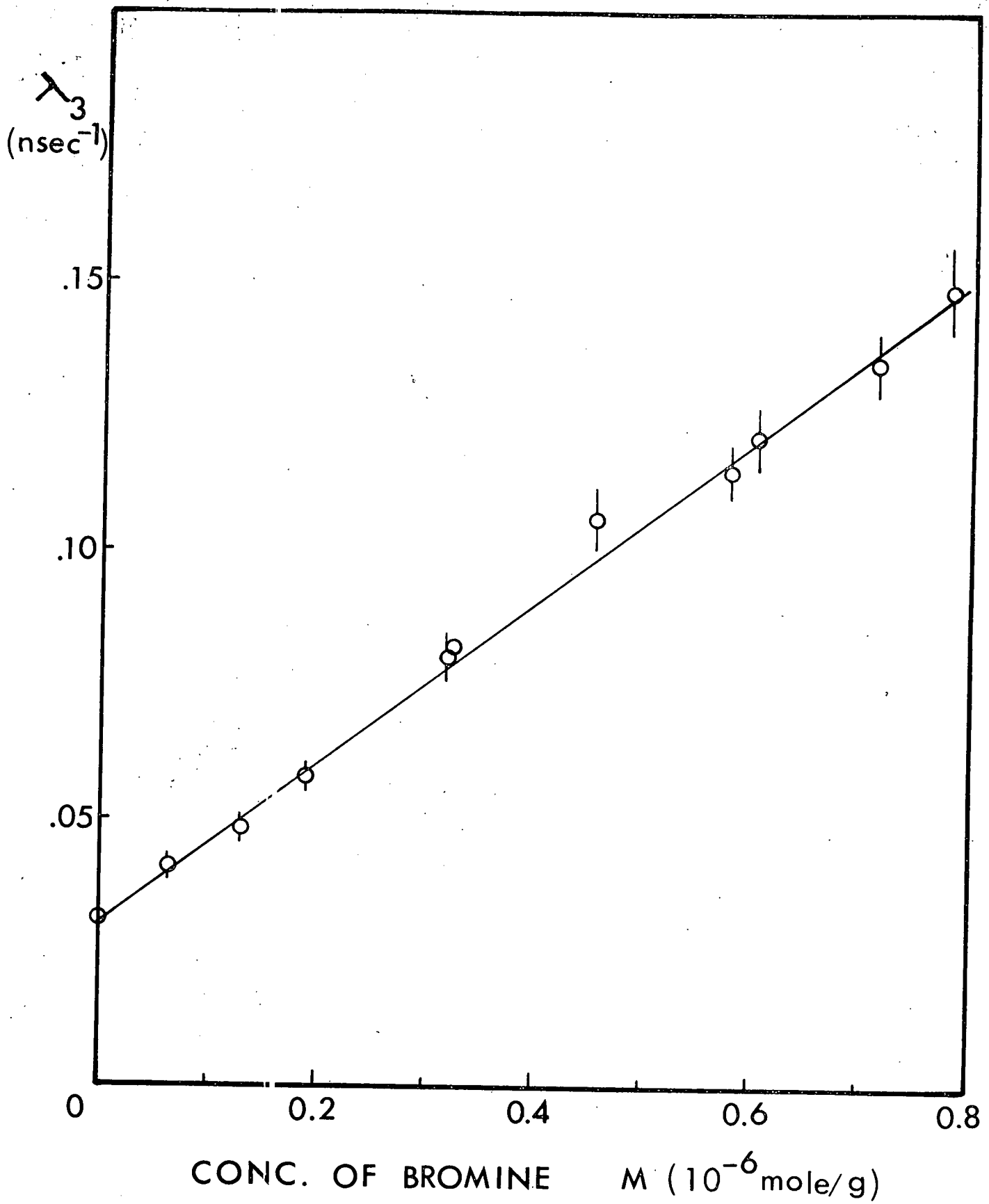
spring
Traveling
microscope
Ref.
point
Detector
Detector
Sample

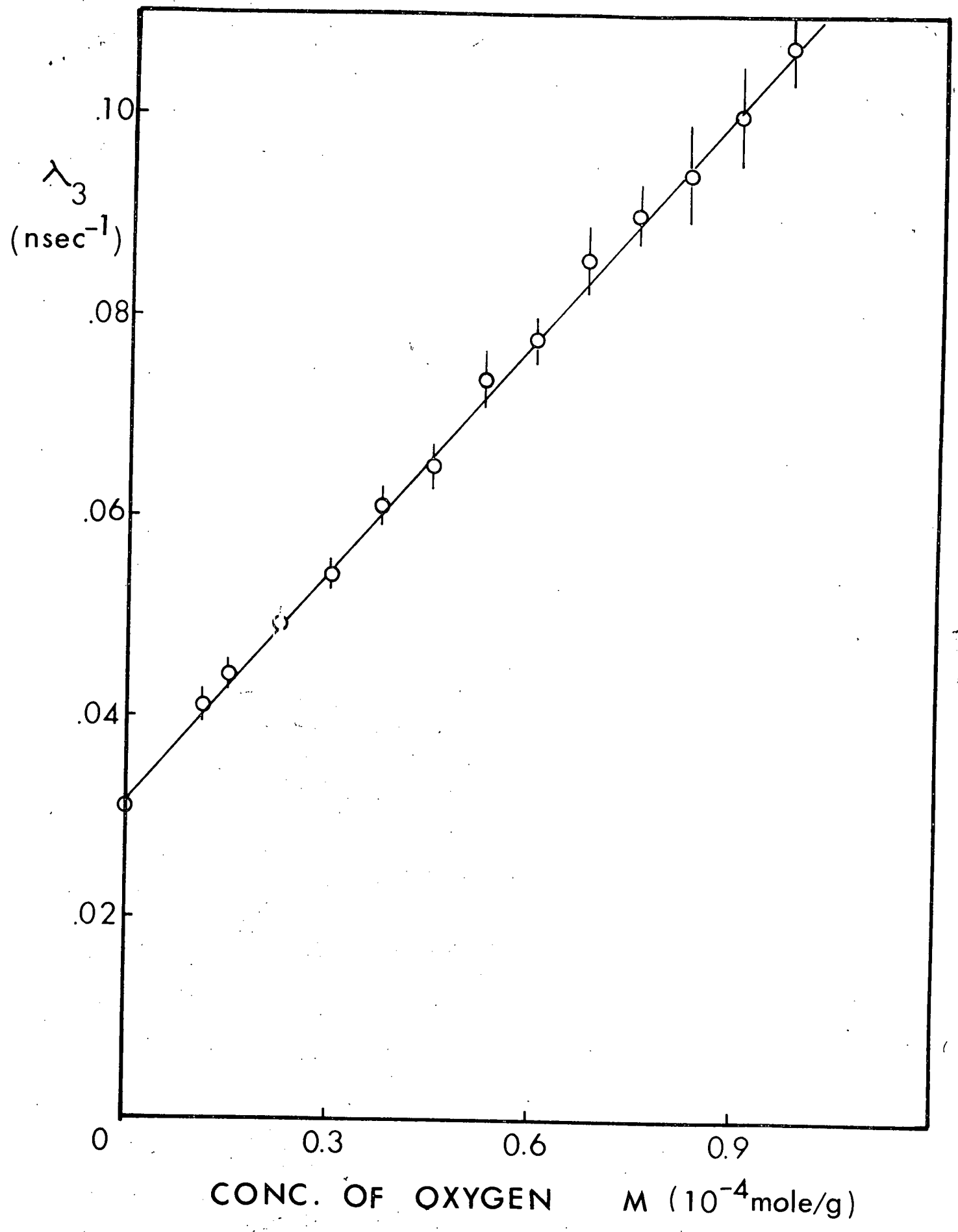

40
I_3 (%)
30
τ_3
(nsec)
20
I_3
τ_3
10
I_2
I_2(%)
0
0.2
0.4
0.6
0.8
CONC. OF BROMINE M (10^{-6} mole/g)


40
I_3(%)
30
τ_3
(nsec)
20
10
I_2(%)
0
0.3
0.6
0.9
I_3
τ_3
I_2
CONC. OF OXYGEN M (10^{-4} mole/g)


0
4
8
12
20
15
10
5
A
B
C
D
E
C(θ)
0
2
4
6
8
10
12
14
θ (mrad)


A
B
C
D
E
N(p)
p (mc x 10^-3)
0
2
4
6
8
10
12


λ_3 (nsec^{-1})
.15
.10
.05
0
0.2
0.4
0.6
0.8
CONC. OF BROMINE M (10^{-6} mole/g)


.10
λ_3
(nsec^{-1})
.08
.06
.04
.02
0
0.3
0.6
0.9
CONC. OF OXYGEN M (10^{-4}mole/g)