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CROSS-SECTION FOR THE REACTION $O(-)+O_2$ YIELDS $O_2(-)+O$ AT RELATI--ETC(U)

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CROSS-SECTION FOR THE REACTION $O^- + O_2 \rightarrow O_2^- + O$ AT
RELATIVE KINETIC ENERGIES FROM THRESHOLD TO 3 eV

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ABSTRACT

The cross-section for the charge-transfer reaction of O^- with O_2 is determined in the relative kinetic energy range from 1 eV (threshold) to about 3 eV, using a combination of experimental and theoretical ion-swarm techniques. Reaction rate constants are measured in a helium-buffered flow-drift tube in the range $50 \leq E/N \leq 110$ Td, where E is the DC electric field strength, N is the helium gas number density and 1 Townsend (Td) $\equiv 10^{-17}$ V cm². Velocity distributions are computed using Monte Carlo techniques for O^- ions drifting in helium at E/N values in this range. These distributions and the drift-tube rate constants define the reaction cross-section at low collision energies, a region in which it has been difficult to make reliable ion-beam measurements.

INTRODUCTION

The endothermic [1,2] charge-transfer reaction



provides an excellent illustration of the difficulty of measuring reaction cross-sections at low relative kinetic energies. For example, this reaction has been the subject of several beam studies [3–8] during the past decade, but there are factor-of-five differences in the magnitudes of the cross-sections obtained in the energy range 0.5–10 eV. In addition to these beam investigations, there have been numerous studies of reaction (1) in oxygen-buffered

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drift tubes [9–16], which have provided rate-constant data over a 0.09–3.5-eV relative kinetic energy range. In addition to there being factor-of-ten differences between some of these results, the major obstacle to obtaining the reaction cross-section from these rate constant data is that the required velocity distributions of O^- ions in the molecular oxygen buffer gas are apparently beyond the current state of the art for both theory and experiment.

However, the motion of atomic ions in drift tubes containing an atomic (i.e. rare-gas) buffer gas is now very well understood [17–22]. This makes it possible to obtain the cross-section of reaction (1) at low energies. In the present study, a helium-buffered flow-drift tube is used to measure the rate constant over the relative kinetic energy range 0.5–1.7 eV. Speed distributions are computed for O^- ions in helium and are used to obtain the reaction cross-section at these energies.

EXPERIMENTAL METHOD

The apparatus used here has been described in detail elsewhere [23–25]. Its basic features and operation are summarized here, along with a few details that are unique to the type of measurement made in the present study.

The O^- ions are formed upstream from the reaction region by dissociative electron attachment in a helium buffer gas containing a trace ($\leq 0.1\%$ compared to He) of oxygen. The buffer-gas flow carries the O^- ions into the drift-reaction region of the apparatus, where they rapidly equilibrate to a mean energy corresponding to the selected ratio of the static electric field strength E to the helium number density N . The O^- signal is monitored while O_2 neutral reactant molecules are added in small quantities ($\leq 1.5\%$ compared to He) to the reaction region. The decline of the O^- signal as a function of added O_2 depends on the number of O_2 molecules added, the time that the reacting species O^- and O_2 are exposed to each other, and the reaction rate constant. The first two of these are known; thus, the measured decline of the O^- signal as a function of added O_2 yields the rate constant (when other O_2 -dependent O^- losses can be ignored, as described below). By varying E/N , the reaction rate constant can be obtained over a range of energies.

Since reaction (1) has a threshold at 1 eV, its rate constant will be small over the mean relative energy range accessible to the present apparatus, 0.04–1.7 eV, and will be small indeed at mean relative energies ≤ 1 eV. Consequently, there will be only a small decline in the O^- signal as a function of added O_2 . For example, at 0.6 eV, the addition of 1.3% O_2 produces a decline in the O^- signal of 9.4%. In such cases, any small non-reactive loss of O^- that is also a function of added O_2 can constitute a large fraction of this observed decline in the O^- signal and, if unaccounted for, can cause the rate constant that is deduced from this decline to be erroneously large. There-

fore, special techniques are required to reliably measure the rate constant of reaction (1) near threshold.

Such a major non-reactive loss arises because, as the reactant O_2 is added, the O^- ions are no longer drifting and diffusing in pure helium, but rather a mixture of O_2 and He [19]. The problem is exacerbated for slow reactions in which one must add a relatively large amount of neutral reactant gas in order to obtain a significant primary-ion decline. Tests made with non-reacting ion-neutral combinations show that flow, drift and diffusive alterations arising from a few percent mixture can cause apparent "rate constants" of approximately $10^{-12} \text{ cm}^3 \text{ s}^{-1}$, thereby setting the lower limit on rate constants measured with this apparatus using the customary methods [23,24]. The alteration of the low-field drift properties by the mixture could be accounted for using Blanc's law [26], but here an empirical approach is used to correct for all types of non-reactive loss, thereby decreasing the lower rate-constant limit by a factor of about five. Apparent "rate constants" are determined separately for non-reactive ion-neutral species selected to simulate closely O^- and O_2 . These values are subtracted from the rate constants obtained from the observed decline (containing both reactive and non-reactive contributions) of the O^- signal as a function of O_2 addition. The difference should closely approximate the reactive loss alone.

MEASURED RATE CONSTANTS

The contributions arising from non-reactive loss are estimated by observing the magnitude of the decline of F^- ion signals as a few percent of O_2 or N_2 are added through the neutral reactant inlet [23]. The electron affinity of F is sufficiently high, 3.399 eV [2], to prevent collisional detachment of F^- at the mean relative kinetic energies, $\leq 2 \text{ eV}$, encountered here. Furthermore, any ion-molecule reactions are about this endothermic [27,28]. Thus, the decline observed in the F^- signal as a function of O_2 or N_2 addition could arise only from non-reactive loss. The F^- and N_2 or O_2 reduced masses are quite similar to those of O^- and O_2 . Furthermore, the mobilities of O^- with O_2 , determined in the customary way [24] from the decline of the O^- and F^- ions in helium are also quite similar [23,29]. Thus, there are reasons to believe that the apparent "rate constants" observed for F^- with N_2 or O_2 are representative of the O^- non-reactive loss caused by the addition of a few percent of O_2 into helium.

These apparent "rate constants" are given by the open symbols in Fig. 1. The F^- , N_2 and F^- , O_2 data are the same, within the scatter of the measurements. The solid symbols in Fig. 1 are the rate constants obtained for O^- signal as a function of O_2 addition; thus they represent varying contributions of both reactive and non-reactive loss. These data and the F^- data merge at mean relative kinetic energies of about 0.4 eV. Below this energy, we interpret the O^- , O_2 data only as apparent "rate constants" alone, representing only non-reactive loss. Above this energy, we interpret the differences be-

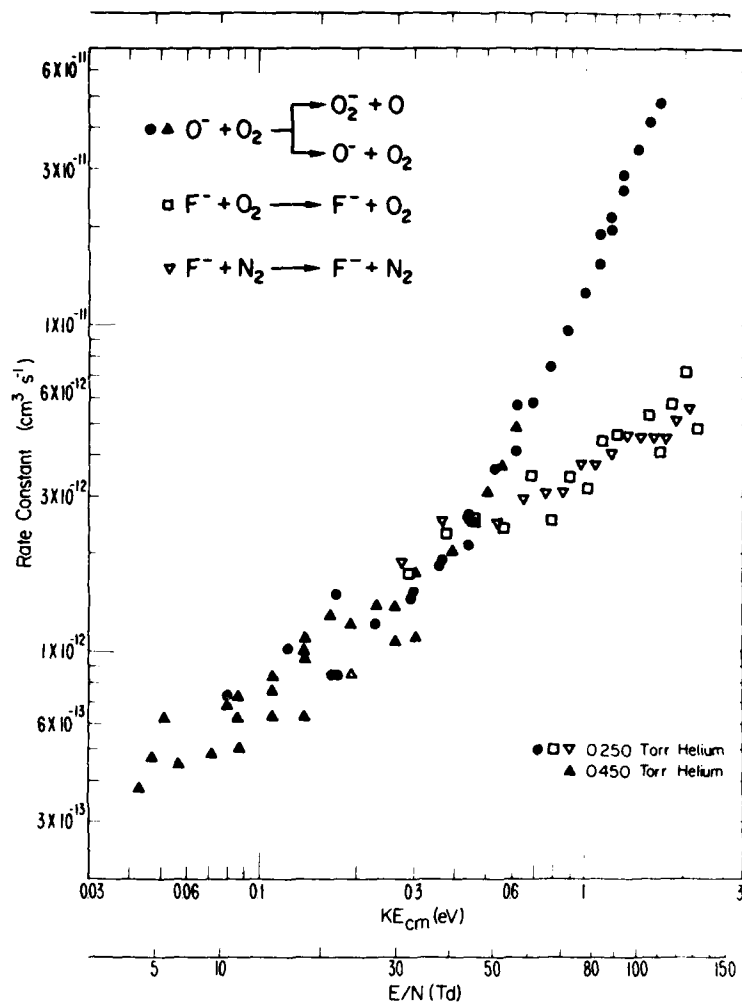


Fig. 1. "Rate constants" determined from the reactive and non-reactive losses of the O^- signal as a function of O_2 number density and determined from the non-reactive losses of the F^- signal as a function of O_2 and N_2 number densities (separately). The abscissae are (a) the mean relative O^- , O_2 and F^- , O_2 , N_2 kinetic energy and (b) the ratio of the electric field strength E and He buffer gas number density N , the latter expressed in Townsends, where $1 \text{ Td} \equiv 10^{-17} \text{ V cm}^2$. The He, O_2 , and N_2 temperature is 300 K.

tween the O^- , O_2 and F^- , O_2 , N_2 data as the rate constant for the reactive loss of O^- ions with O_2 , i.e. reaction (1). The values corresponding to these differences are given in Fig. 2.

The error bars given in Fig. 2 reflect the uncertainty in precision that arises from taking the difference of similar numbers. They are based on two standard errors in the predicted values of rate constants from a linear least-squares fit to the F^- data in Fig. 1. As the difference grows larger, these errors become smaller. The precision of the data with regard to the hori-

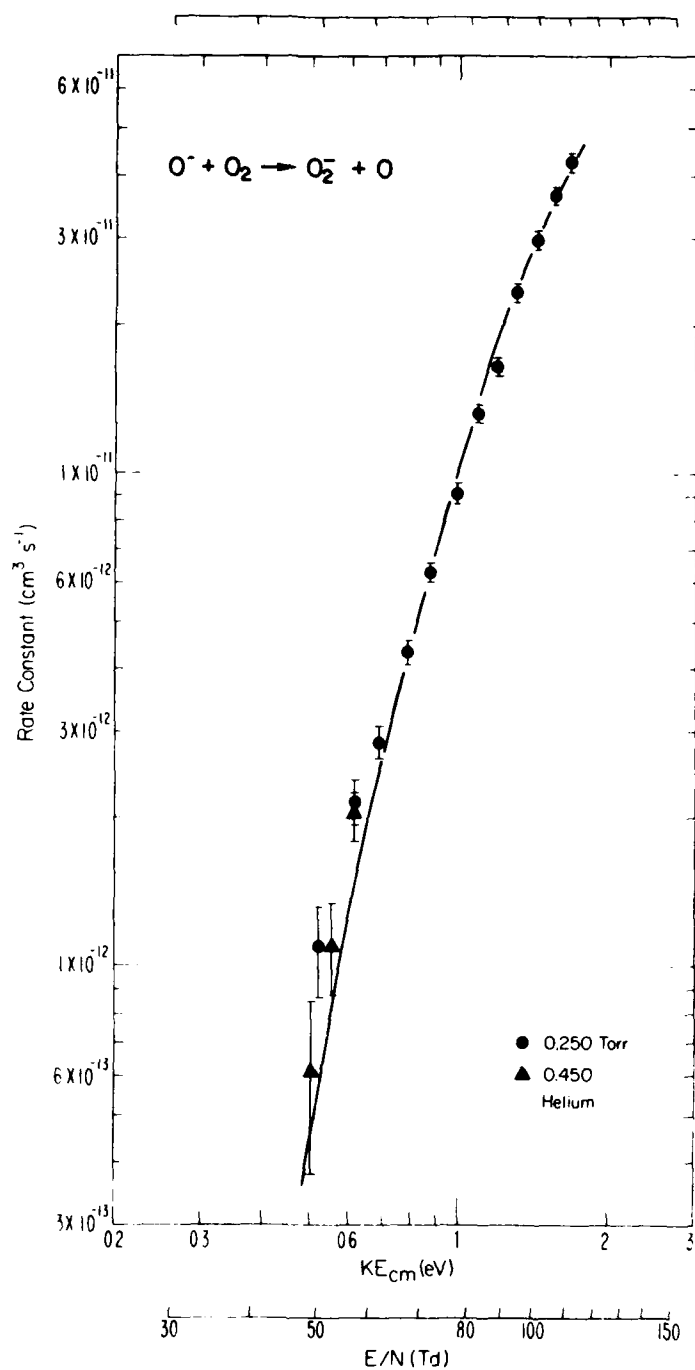


Fig. 2. Rate constants for the charge-transfer reaction of O^- with O_2 as a function of (a) the mean relative O^- , O_2 kinetic energy and (b) the ratio of electric field strength E and the He buffer gas number density N , the latter expressed in Townsends, where $1 \text{ Td} \equiv 10^{-17} \text{ V cm}^2$. The He and O_2 temperatures are 300 K. The vertical bars on the data are estimates of the precision. The accuracy is approximately $\pm 60\%$ at low energies and $\pm 30\%$ at high energies.

zontal scales, E/N and KE_{cm} , is small for all measurements, being only about the width of the symbols. The effects of systematic error are not shown. These could be as large as 30–60% [24] in the rate constant, the larger values being associated with the lower-energy points, which rest most strongly on the assumption that F^- simulates O^- in these measurements. The accuracy of the data with regard to E/N is probably better than 3%, the error arising largely from the pressure calibration.

Although reaction (1) is only one of several O^- reactive loss processes that can occur at these energies, the data in Fig. 2 are very likely to correspond to this reaction alone, for the following reasons. Firstly, beginning at a mean relative kinetic energy of roughly 0.4 eV, the decline of the O^- signal is accompanied by an increase in the O_2^- signal. Although unknown ion sampling discrimination between O^- and O_2^- prevents a quantitative statement, an estimate of this discrimination shows that reaction (1) accounts for most of the O^- loss, indicating that associative detachment



is probably only a minor part of the loss observed here. Reaction (2) is known to have a thermal rate less than $10^{-12} \text{ cm}^3 \text{ s}^{-1}$ [30] and such reactions often decrease with increasing relative kinetic energy [25]. Furthermore, the relatively high threshold of the collisional detachment process



makes it unlikely to contribute substantially to the present data.

The mean relative kinetic energy scales in Figs. 1 and 2 were established as described earlier [21], using the "Wannier expression" [31] based on the point-charge, induced-dipole model, which has been shown experimentally [19,22] and theoretically [17,18,20, 21,32] to yield a very good estimate. However, in spite of its accuracy, the mean relative kinetic energy of the O^- and O_2 reactants is not sufficient characterization for such a strongly energy-dependent reaction [17,20] and is given in Figs. 1 and 2 only as a general indication of the energy behavior. The observed drift-tube rate constant is a phenomenological quantity that can depend sensitively on the properties of the high-energy tail of the O^- -in-He ion speed distribution through an integral of the speed distribution and the reaction cross-section. The latter is the only unique and fundamental quantity in such a case and its extraction from the data of Fig. 2 will now be described.

THEORETICAL ANALYSIS

The E/N -dependent rate constants obtained in a drift-tube experiment are related to the fundamental kinetic-energy-dependent reaction cross-section, $\sigma(\epsilon)$, by

$$k(E/N) = \iint |v - u| \sigma(\epsilon) f_1(v, E/N) f_n(u) dv du \quad (4)$$

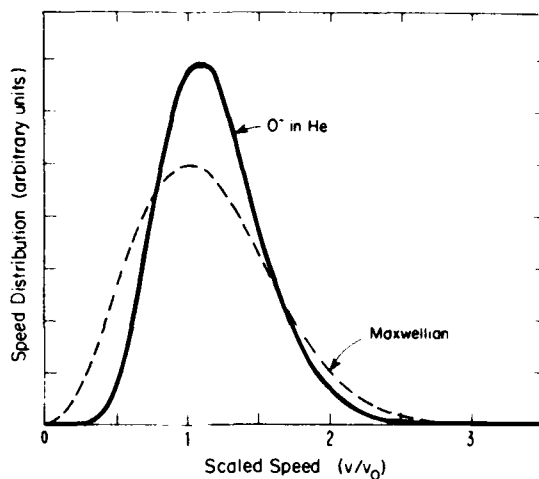


Fig. 3. Speed distributions of O^- ions in He at 115 Td (1 Townsend $\equiv 10^{-17}$ V cm²). The solid curve is calculated using Monte Carlo techniques. The line thickness corresponds approximately to the precision of the calculation. The dashed curve is a Maxwellian distribution corresponding to the same mean energy: 2.7 eV lab frame. The abscissa is a scaled speed, where v_0 is the speed corresponding to the peak of the Maxwellian distribution.

where $f_n(u)$ is the velocity distribution of the O_2 neutral reactant at the ambient He buffer temperature, $f_i(v, E/N)$ is the velocity distribution of the O^- ions at the established E/N , and $\epsilon = 0.5 \mu |v - u|^2$, where μ is the O^- , O_2 reduced mass. (The dependence of k , f_i , and f_n on the buffer gas temperature is not stated explicitly here, since the present study was confined to 300 K throughout.) Clearly, the key to relating the observed drift-tube rate constants $k(E/N)$ to the fundamental reaction cross-section is the ion velocity distribution, $f_i(v, E/N)$.

Early approaches to this problem employed trial analytical functions for $f_i(v, E/N)$ containing estimated parameters [33–35]. However, a rigorous approach has now become feasible following the development of analytic [20] and Monte Carlo [21] solutions of the relevant Boltzmann transport equation. In the present study we make use of the velocity distributions calculated in the Monte Carlo simulations of the motion of O^- ions through helium gas at 300 K [21,36]. We assume that the small amounts of the reactant O_2 molecules used in the experiment do not significantly alter the ion velocity distributions. The estimated errors discussed above have been deliberately made larger to account for any possible weakness in this assumption.

Monte Carlo simulations were carried out for $E/N = 35, 55, 70, 85, 100$, and 115 Td (1 Townsend $\equiv 10^{-17}$ V cm²). Figure 3 shows the speed distribution obtained at $E/N = 115$ Td. The abscissa is the ratio of the ion speed, v , to the speed v_0 that corresponds to the peak of a Maxwellian distribution of the same average energy: 2.7 eV lab frame or 1.8 eV O^- , O_2 center-of-mass frame.

Various forms were assumed for $\sigma(\epsilon)$ with parameters that were adjusted in order to fit the observed reaction rates in Fig. 2. The constraints placed on the cross-section were that it vanishes at collision energies near 1.022 eV, that it does not become negative within the energy range of interest and that it does not oscillate rapidly. Attempts to remove the first constraint, i.e. to use the present data to determine the threshold precisely, failed. As has been pointed out by Russ et al. [37], drift-tube measurements must extend down to very small values ($k \lesssim 10^{-13}$ or 10^{-14} cm³ s⁻¹) to establish a meaningful threshold.

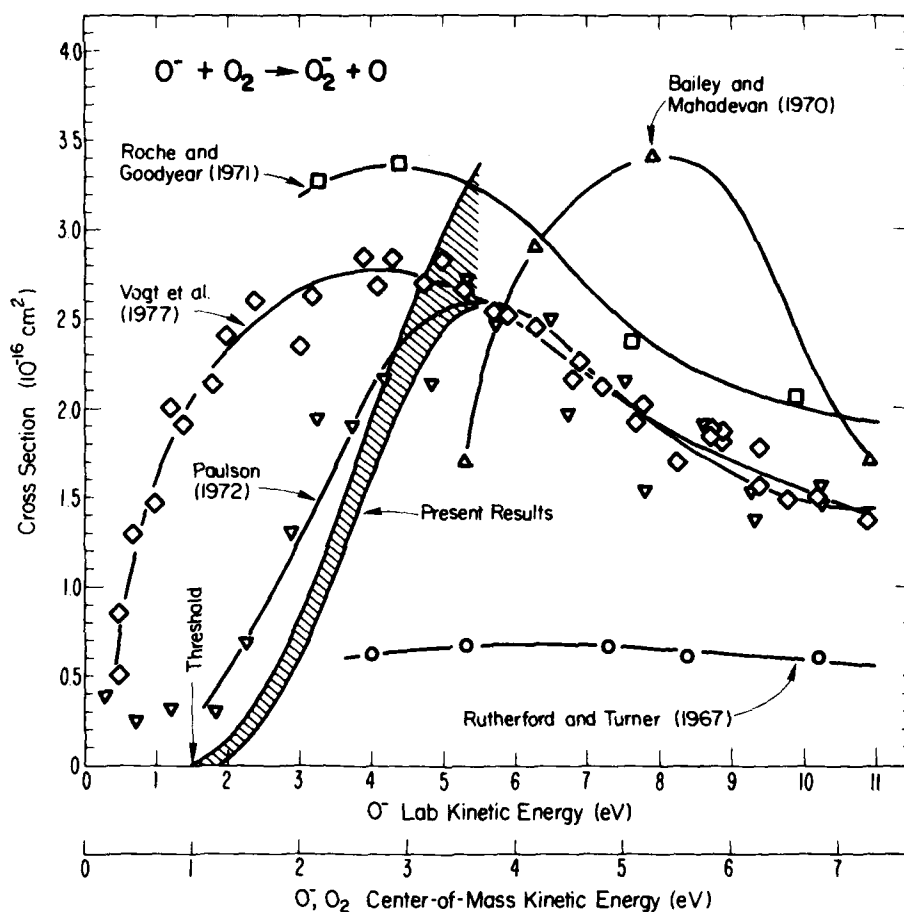


Fig. 4. Cross-section for the charge-transfer reaction of O^- with O_2 as a function of relative kinetic energy. The dashed area represents the present results and their precision, determined from the rate constants in Fig. 2, Monte Carlo speed distributions as given in Fig. 3, and eqn. (4). These data are considered accurate to $\pm 60\%$ at low energies and $\pm 30\%$ at high energies. The open symbols are the results of earlier ion-beam studies: $\odot$ ref. 3 (stated $\pm$ factor-of-two uncertainty); $\triangle$ ref. 4; $\square$ ref. 6 (stated $\pm 20\%$ uncertainty, at least); ∇ ref. 7 (factor-of-two uncertainty, J.F. Paulson, private communication, 1979); and $\diamond$ ref. 8.

The form used here, and which appears to provide reasonable flexibility, is

$$\sigma(v) = \sum_{n=1}^4 c_n [(v - v')/v']^{\beta+n\gamma} \exp\{\alpha [(v - v')/v']^2\} \quad (5)$$

where c_n , α , β , γ are adjustable parameters and v' is the threshold speed corresponding to an energy of 1.022 eV. In each fit, the parameters α , β and γ are fixed at one set of values and the c_n adjusted to obtain the minimum-variance fit to the observed rate constants. Acceptable fits were obtained with the values of the parameters $0 \leq \alpha \leq 1.5$, $1 \leq \beta \leq 3$, and $\gamma = 1$ or 2. One such fit is given in Fig. 2 as the solid line. The most difficult data points to fit are those of lowest energy, which is understandable in view of the possible larger inaccuracies in these data.

Thus, the ranges of these parameters over which the fits are acceptable establish an uncertainty in the cross-section deduced in this manner. The uncertainty is reflected by the width of the dashed area in Fig. 4; this represents the cross-section of reaction (1) obtained in this study. As indicated, this uncertainty is much smaller for relative energies less than 2 eV than it is for higher energies, since the latter represent extensions beyond the range of the drift-tube rate constants. Although these uncertainties do grow with increasing energy, it is interesting to note that the drift-tube rate constants place useful constraints on the cross-section at energies lying at about a factor of two beyond the range of the data. In addition to this indicated "precision", one must add about $\pm 60\%$ (low energy) to $\pm 30\%$ (high energy) absolute accuracy associated with the measurement of $k(E/N)$.

DISCUSSION

Figure 4 gives the cross-section data determined for reaction (1) in previous ion-beam experiments [3,4,6–8]. Although all these studies agree that the cross-section has a maximum at lab energies 4–8 eV and a minimum at 10–35 eV (not shown in Fig. 4), there is considerable uncertainty about the magnitude of the cross-section, particularly at energies near threshold. It is in this low-energy region that the present measurements make their contribution. The agreement between the drift-tube and beam measurements is best with the data of Paulson [7], the difference being typically within $0.3 \times 10^{-16} \text{ cm}^2$, or less than about 50%, which is certainly within the combined experimental uncertainties.

There have been many measurements of the rate constants, $k(E/N)$, of reaction (1) in oxygen-buffered drift tubes [10–16,38]. These data are given by the open symbols in Fig. 5. Because of the large O^- , O_2 collision frequency in this situation, these measurements can be made with relatively high sensitivity, thereby providing nearly five decades of dynamic range of $k(E/N)$ measurements. The abscissa of Fig. 5 is the O^- , O_2 mean center-of-mass kinetic energy, calculated [23] using the Wannier expression [31] and

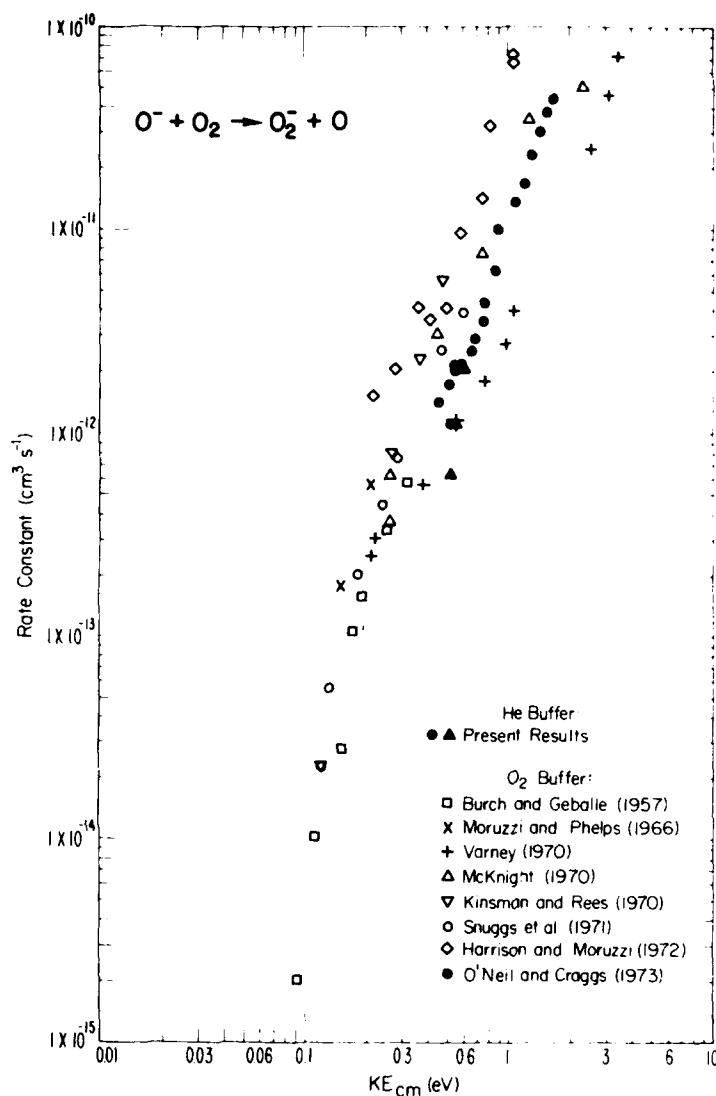


Fig. 5. Rate constants for the charge-transfer reaction of O^- with O_2 as a function of mean O^- , O_2 relative kinetic energy. The solid symbols are the present results, measured in a helium-buffered flow-drift tube. The open and line symbols are values determined in oxygen-buffered static drift tubes: □ ref. 9; × ref. 10; + ref. 11; △ ref. 12; ▽ ref. 13; ○ ref. 14; ◇ ref. 15; and ⊕ ref. 16.

the mobilities of O^- in O_2 [39,40]. Figure 5 also contains the rate constants deduced in the present study, represented by the filled symbols. In principle, the differences between the two types of data — $k(E/N)$ measured in an O_2 buffer gas and measured in a He buffer gas — reflect the differences between the O^- -in- O_2 and O^- -in-He velocity distributions. Such differences have been explored for O^+ -in-Ar and O^+ -in-He distributions using the strongly

energy-dependent reactions of O^+ with O_2 and N_2 [19–21].

However, in the present situation, the O_2 -buffered data do not form a sufficiently homogeneous set to warrant a detailed treatment of the differences. The "scatter" in the O_2 buffered data no doubt reflects varying systematic differences between the different experiments. In spite of this scatter, the trends of the two types of data in Fig. 5 tend to suggest some general features of the O^- -in- O_2 and O^- -in-He velocity distributions. The O^- -in- O_2 $k(E/N)$ values appear to be larger than the O^- -in-He values for $KE_{cm} \leq 1$ eV. Since the high-velocity tail of the speed distribution dominates the integrals in eqn. (2), this suggests that the tail is more pronounced for the O^- -in- O_2 distributions than it is for the O^- -in-He distributions. This would be expected from theoretical considerations [41], which find that the speed distributions for light ions in heavy gases are generally broader than those for heavy ions in light gases. There has been an attempt to directly measure O^- -in- O_2 speed distributions [42]; however, the mean energies deduced from these measurements differ so substantially (by a factor of two) from those given by the Wannier expression [31], which is generally believed to be accurate within several percent [17,18,32], that these data appear to be quite uncertain. The best test for differences between O^- -in- O_2 and O^- -in-He velocity distributions, either experimental or theoretical, would be to measure the rate constants for reaction (1) separately in O_2 and He buffer gases in the same apparatus. It is hoped that the comparisons given in Fig. 5 will stimulate such studies, just as the comparisons in Fig. 4 suggest the utility of further ion-beam measurements at low collision energies.

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