

Stopping cross sections of oxygen for 0.3–2.0-MeV α particles in saturated alcohols and ethers*

E. K. L. Chau, R. B. Brown, A. S. Lodhi, D. Powers, S. Matteson, and S. R. Eisenbarth

Baylor University, Waco, Texas 76703

(Received 1 April 1977)

The stopping cross sections of three saturated alcohols, CH_3OH , $\text{C}_2\text{H}_5\text{OH}$, and $\text{C}_3\text{H}_7\text{OH}$, and two saturated ethers, $(\text{CH}_3)_2\text{O}$ and $(\text{C}_2\text{H}_5)_2\text{O}$, have been measured in a differentially pumped gas cell with probable random errors of 0.5–1.1%. The results confirm that $\epsilon(\text{CH}_2)$ quoted in a previous paper can be used to predict the stopping cross sections for single-bonded C–H–O compounds. The values for the stopping cross section of single-bonded oxygen are extracted and found to be 3–17% lower than $(1/2)\epsilon_{\text{expt}}(\text{O}_2)$, thereby demonstrating the effect of chemical bonding on the stopping cross section. Comparison is made between the stopping cross sections for oxygen and those calculated from metal-oxide yield ratios.

I. INTRODUCTION

Bragg's rule¹ states that the stopping cross section $\epsilon = dE/Ndx$ of a compound $X_m Y_n$ is given by the sum of the stopping cross sections of the elements

$$\epsilon(X_m Y_n) = m\epsilon(X) + n\epsilon(Y). \quad (1)$$

The validity of this rule has been tested by various groups,^{2–14} and departures have been observed that are caused by chemical bonding¹⁰ and possibly the physical state^{13,14} of the elemental constituents. On the other hand, Bragg's rule was observed to apply to 11 single-bonded gaseous or vapor compounds¹¹ for 0.3–2.0-MeV α particles, when due consideration was given to chemical bonding.

The stopping cross section $\epsilon(0)$ of oxygen and the applicability of Bragg's rule to its compounds are of particular interest because of the important role that oxygen plays in the semiconductor industry and in radiation dosimetry. Baglin and Ziegler¹² found that Bragg's rule held true to within 2% for Al_2O_3 and SiO_2 for 2-MeV α particles when $\epsilon(0)$ was given by $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$.¹⁵ Feng *et al.*,¹³ however, observed that $\epsilon(0)$ calculated from scattering-yield ratios and from assumed Bragg-rule validity was 6–22% lower than gaseous oxygen $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$. Their technique was to measure the scattering-yield ratios from two-layered metal-metal targets and metal to metal-oxide targets of MgO , Al_2O_3 , SiO_2 , $\alpha\text{-Fe}_2\text{O}_3$, and Fe_3O_4 . This deviation was attributed to a physical-state effect which has been further amplified by Ziegler *et al.*¹⁶ Langley and Blewer¹⁴ also observed a deviation from Bragg's rule for erbium oxide Er_2O_3 when use was made of the stopping cross sections of metallic erbium $\epsilon(\text{Er})$ and gaseous oxygen $\epsilon(0)$.¹⁷

Powers *et al.*¹⁰ found that the elemental stopping cross sections of carbon and hydrogen in simple gaseous and vapor hydrocarbons would change, de-

pending upon whether the compound consisted of purely single bonds, double bonds, triple bonds, or a mixture of single and double bonds. The set of elemental cross sections for carbon and hydrogen found¹¹ to be common to eleven gaseous (or vapor) single-bonded C–H, C–H–F, C–H–Br, and C–Br–F compounds could not be applied to gaseous (or vapor) compounds that contained double or triple bonds, thereby giving further evidence of the effect of chemical bonding on the stopping cross section.

In the present work, the approach of Lodhi and Powers¹¹ is extended to three saturated alcohols and two saturated ethers, which are in gas or vapor form and which consist of only single bonds. The stopping cross sections of oxygen common to these single-bonded compounds are extracted and are compared to $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$ of oxygen gas,¹⁵ and also to those of "solid oxygen" quoted in Feng *et al.*¹³ A set of values is also given for the stopping cross sections of the OH radical which is common to saturated alcohols.

II. EXPERIMENTAL PROCEDURE

The experimental technique and analysis were essentially the same as used in previous work^{9–11,18} and given in detail in Ref. 15. A He^+ ion beam from a 2-MeV Van de Graaff accelerator was deflected by a 10° magnetic analyzer and passed through a differentially pumped gas cell system with and without vapor. The He^+ ion energy after passing through the gas cell was measured by a 20° magnetic spectrometer. The pressure in the gas cell was measured by the following instruments: (i) a GM-100A, CVC Products, Inc. McLeod gauge, (ii) a U-tube manometer using Leybold-Heraeus HE-200 diffusion pump fluid, and (iii) more recently, an MKS type 221 capacitance manometer. All three instruments were consistent with each

other to within 1% error in the 0.5–3.0-Torr pressure region. Reference 19 discusses the use of the capacitance manometer for precision pressure measurements of <1% error. The capacitance manometer for this experiment was furnished with calibration data from the manufacturer which confirmed the actual error to be no greater than 0.25% when calibrated using an MKS Transfer Standard (<0.08% error) which in turn was calibrated with an absolute pressure standard traceable to the National Bureau of Standards using a CEC air dead weight tester (<0.015% error). The above absolute pressure measurements can also be obtained with less than 0.1% error using other methods,^{20,21} thereby showing that the absolute accuracy of calibration of the capacitance manometer used in this experiment is very reasonable.

The system for admitting vapor or gas into the gas cell was modified so that there were three supply lines: one for nitrogen, one for another gas, and one for the vapor from a liquid. Each supply line contained two needle valves to regulate the pressure and could be isolated from the gas cell by a high-vacuum valve. This arrangement enabled the stopping cross section of nitrogen (taken to be a reference standard) to be measured prior to measuring that of another compound. The U-tube manometer and capacitance manometer could then be checked or calibrated periodically against the McLeod gauge when nitrogen was in the gas cell. The stopping cross section of nitrogen was used as a reference standard to check the vapor measurements, since it had been previously measured and checked¹⁵ by two independent methods in this laboratory. The improvements in the ion-beam energy stability and position as well as improved pressure measurement accuracy in the gas cell since the publication of Ref. 15 have enhanced the accuracy of the stopping cross sections. The reproducibility of the stopping-cross-section measurements of nitrogen through numerous repetitions has demonstrated their reliability. An absolute error of $\pm 1\%$ was assigned, based on the comparison of our reference nitrogen curve to the independent measurements of the stopping cross section of nitrogen accumulated the past two years. During this time, the calibration of the incident ion beam energy was checked periodically, the capacitance manometer was checked against the McLeod gauge periodically, and different settings of the slits in front of the analyzing magnet have been used. All of these contributed to minimizing the systematic error in the reference nitrogen stopping-cross-section measurement. Systematic error in the vapor stopping-cross-section measurement was minimized in turn by measuring the stopping cross section of nitrogen at every in-

cident ion beam energy for which the vapor measurement was made. This procedure ensured that no systematic error greater than 1% would be introduced into the measurement.

The pressure of gas or vapor in the gas cell was typically between 0.5 and 3.0 Torr. The end correction¹⁵ to the effective length of the gas cell due to gas leaking out to the first differentially pumped section was less than 0.5%, when the pressure was kept within the above range. There were, however, a few data points that utilized higher pressures and required end corrections as great as 3%. Corrections to the mean energy¹⁵ of the ions in the gas cell were less than 1%. The methyl alcohol, propyl alcohol, and diethyl ether were all "analyzed reagent" grade from J. T. Baker Chem. Co., Philipsburg, N. J., and had purities of 99.9%, 99.8%, and 99.9%, respectively. The ethyl alcohol (purity greater than 99.9%) was from U. S. Industrial Chemicals Co., N. Y., N. Y. The dimethyl ether (purity 99.5%) was from Matheson Gas Products, La Porte, Tex.

The number of data points taken for each curve varied from 213 to 670. All stopping cross sections in this experiment were checked and remeasured at least on two different occasions. The methyl alcohol measurements were made over a span of three years and rechecked four different times. The Brice formula²² was not used to fit the experimental data points, because it could not give a faithful representation^{11,18} of all the data points. The functions employed in the least-squares fit were

$$\epsilon(E) = a_0 + a_1E + a_2E^2 + a_3E^3 + a_4E^4, \quad (2)$$

for the energy interval $0.3 \text{ MeV} \leq E \leq E_B$, and

$$\epsilon(E) = (C/E) \ln(DE) \quad (3)$$

for the energy interval $E_B < E \leq 2.0 \text{ MeV}$. The values of the parameters were chosen such that the two curves joined smoothly at $E = E_B$. The probable error of each curve was given by $0.6745(\sigma/\bar{\epsilon}_{\text{expt}}) \times 100\%$, where

$$\sigma = \left(\frac{\sum_{i=1}^n (\epsilon_{\text{expt}}^i - \epsilon_{\text{curve}}^i)^2}{n - m - 1} \right)^{1/2}, \quad (4)$$

$$\bar{\epsilon}_{\text{expt}} = \frac{1}{n} \sum_{i=1}^n \epsilon_{\text{expt}}^i,$$

n is the number of experimental data points within the appropriate energy range, and m is the degree of freedom for the curve-fit function. The curve-fit parameters are given in Table I.

TABLE I. Curve-fit parameters for the stopping cross sections of the five compounds. $\epsilon = a_0 + a_1E + a_2E^2 + a_3E^3 + a_4E^4$ for $0.3 \text{ MeV} \leq E \leq E_B$, $\epsilon = (C/E) \ln(DE)$ for $E_B < E \leq 2 \text{ MeV}$, where ϵ is in 10^{-15} eV cm^2 and E is in MeV. The two curves join smoothly at $E = E_B$.

	CH ₃ OH	C ₂ H ₅ OH	C ₃ H ₇ OH	(CH ₃) ₂ O	(C ₂ H ₅) ₂ O
a_0	47.46	76.56	117.01	80.73	136.31
a_1	399.81	563.35	660.15	542.93	858.58
a_2	-570.30	-786.16	-871.15	-745.73	-1109.06
a_3	329.07	440.12	445.03	406.17	536.72
a_4	-71.41	-93.48	-84.97	-83.95	-92.66
Probable error in the polynomial part (%)					
C	76.96	109.79	141.92	112.42	180.92
D	6.114	6.508	6.813	6.090	6.301
Probable error in the logarithmic part (%)					
E_B (MeV)	1.39	1.33	1.35	1.26	1.35
Number of data points	670	317	213	275	279

III. RESULTS

The values of the stopping cross sections of the five compounds measured in the present work are tabulated in Table II. The values for the two isomers, ethyl alcohol C₂H₅OH and dimethyl ether (CH₃)₂O, agree with each other to within 0.8%. When the stopping cross sections of the three alcohols are plotted as a function of the variable n according to the equation

$$\epsilon_{\text{expt}}(\text{C}_2\text{H}_{2n-1}\text{OH}) = n\epsilon(\text{CH}_2) + \epsilon(\text{HOH}) \quad (5)$$

at a fixed energy, the points form a straight line with standard deviation no greater than 0.13%.

Figure 1 shows the plot for energies 0.5, 1.0, 1.5, and 2.0 MeV. The slope of each straight line gives the stopping cross section $\epsilon(\text{CH}_2)$ which agrees to within 2% to the $\epsilon(\text{CH}_2)$ found previously¹¹ for alkanes (C-H compounds). The intercept of each straight line in Fig. 1 gives the stopping cross section $\epsilon(\text{HOH})$, which agrees at all energies to that of water vapor¹⁸ $\epsilon_{\text{expt}}(\text{H}_2\text{O})$ to within 1.7%. With the $\epsilon(\text{CH}_2)$ values quoted in Ref. 11 and the $\epsilon_{\text{expt}}(\text{H}_2\text{O})$

TABLE II. Tabulated values of the stopping cross sections of the five compounds measured. ϵ is in 10^{-15} eV cm^2 . Probable errors are given in Table I.

Energy (MeV)	CH ₃ OH	C ₂ H ₅ OH	C ₃ H ₇ OH	(CH ₃) ₂ O	(C ₂ H ₅) ₂ O
0.3	124.4	185.9	248.0	186.8	307.8
0.4	135.4	201.9	268.0	202.4	334.3
0.5	141.5	210.9	279.6	211.3	349.6
0.6	143.9	214.5	284.6	214.9	356.1
0.7	143.6	214.2	284.5	214.5	355.7
0.8	141.5	211.2	280.7	211.4	350.2
0.9	138.4	206.3	274.2	206.3	341.2
1.0	134.6	200.4	266.1	200.2	329.9
1.1	130.6	193.9	257.0	193.3	317.5
1.2	126.6	187.2	247.6	186.2	304.9
1.3	122.4	180.3	238.0	178.9	292.7
1.4	118.0	173.3	228.6	172.1	281.4
1.5	113.7	166.8	219.9	165.8	270.9
1.6	109.7	160.8	211.9	160.0	261.3
1.7	106.0	155.2	204.5	154.6	252.4
1.8	102.5	150.1	197.6	149.5	244.1
1.9	99.3	145.3	191.3	144.9	236.4
2.0	96.3	140.9	185.4	140.5	229.2

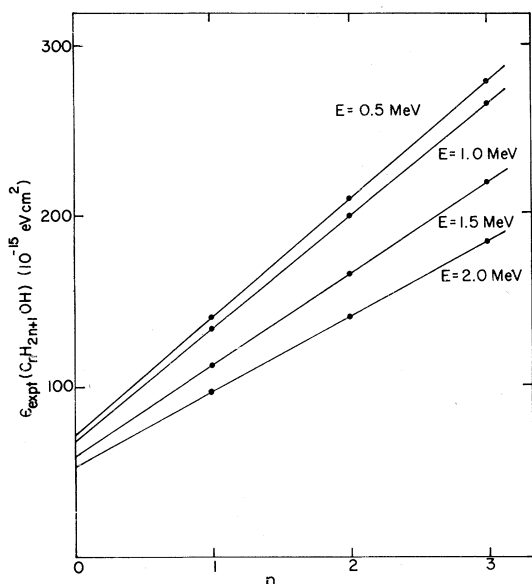


FIG. 1. Plot of molecular stopping cross section $\epsilon_{\text{expt}}(\text{C}_n\text{H}_{2n+1}\text{OH}) = n\epsilon(\text{CH}_2) + \epsilon(\text{HOH})$ for saturated alcohols versus n for energies 0.5, 1.0, 1.5, and 2.0 MeV. The experimental stopping cross sections for methyl alcohol, ethyl alcohol, and propyl alcohol correspond to $n = 1, 2,$ and $3,$ respectively. The slope of each straight line gives $\epsilon(\text{CH}_2)$, and the intercept gives $\epsilon(\text{HOH})$. The values of $\epsilon(\text{CH}_2)$ agree to those of Lodhi and Powers (Ref. 11) to within 2%, and $\epsilon(\text{HOH})$ agrees to $\epsilon_{\text{expt}}(\text{H}_2\text{O})$ (Ref. 18) to within 1.7%. The experimental stopping cross sections of dimethyl ether and diethyl ether (not shown on the figure) at the corresponding energies are essentially on the straight lines as well. The figure illustrates that $\epsilon(\text{CH}_2)$ can be used to predict the stopping cross sections of single-bonded C-H-O compounds.

values of Ref. 18, the stopping cross sections of the saturated alcohols and ethers of the present experiment can be calculated [for instance, $\epsilon_{\text{expt}}(\text{H}_2\text{O}) + \epsilon(\text{CH}_2)$ gives $\epsilon(\text{CH}_3\text{OH})$, $\epsilon_{\text{expt}}(\text{H}_2\text{O}) + 2\epsilon(\text{CH}_2)$ gives $\epsilon(\text{C}_2\text{H}_5\text{OH})$, etc.]. The predicted values and the experimental values for the stopping cross section of methyl alcohol CH_3OH agree to within 0.9%, those of ethyl alcohol $\text{C}_2\text{H}_5\text{OH}$ agree to within 1.3% (less than 1.0% over 90% of the entire energy interval of the curve), those of dimethyl ether $(\text{CH}_3)_2\text{O}$ agree to within 0.5%, those of propyl alcohol $\text{C}_3\text{H}_7\text{OH}$ agree to within 1.4% (less than 1.0% over 80% of the entire energy interval), and those of diethyl ether $(\text{C}_2\text{H}_5)_2\text{O}$ agree to within 0.5%.

When this scheme is repeated by use of the same value of $\epsilon(\text{CH}_2)$, but with the use of $\epsilon_{\text{expt}}(\text{CH}_3\text{OH})$ in place of $\epsilon_{\text{expt}}(\text{H}_2\text{O})$, the stopping cross sections of the other four compounds can be predicted to within 1.0% over the entire energy range of the curve. When the $\epsilon(\text{CH}_2)$ values obtained from the slopes of the plots shown in Fig. 1 were used, the agreement between the predicted values and the experimental val-

ues was also within 1.0%, except between $\epsilon_{\text{expt}}(\text{H}_2\text{O}) + 4\epsilon(\text{CH}_2)$ and $\epsilon_{\text{expt}}((\text{C}_2\text{H}_5)_2\text{O})$ which was within 1.3% (but less than 1.0% over 80% of the entire energy range of the curve), and also between $\epsilon_{\text{expt}}(\text{CH}_3\text{OH}) + 3\epsilon(\text{CH}_2)$ and $\epsilon_{\text{expt}}((\text{C}_2\text{H}_5)_2\text{O})$, which was within 1.1% (but less than 1.0% over 90% of the entire energy range of the curve). Thus, it is verified that the molecular stopping cross sections of saturated alcohols and ethers can be predicted to $\approx 1\%$ by use of integral multiples of $\epsilon(\text{CH}_2)$ from the alkanes of Ref. 11, along with $\epsilon_{\text{expt}}(\text{CH}_3\text{OH})$ of the present experiment or with $\epsilon_{\text{expt}}(\text{H}_2\text{O})$ of Ref. 18.

The chemical structure of the three alcohols and two ethers of the present experiment is given in Table III. For the alcohols, one H atom is bound to an O atom and the remaining H atoms to C atoms, whereas in the ethers, all H atoms are bound to C atoms. The fact that the molecular stopping cross sections of alcohols and ethers can be predicted to $\approx 1\%$ by use of $\epsilon(\text{CH}_2)$ from the alkanes means that for these five compounds of the present experiment a common value of $\epsilon_{\text{SB}}(\text{H})$ exists which is independent of whether the hydrogen is bound to an O atom or to a C atom. One can therefore obtain an $\epsilon_{\text{SB}}(\text{O})$ or $\epsilon_{\text{SB}}(\text{OH})$ by simply subtracting $2\epsilon_{\text{SB}}(\text{H})$ or $1\epsilon_{\text{SB}}(\text{H})$, respectively, of Ref. 11 from $\epsilon_{\text{expt}}(\text{H}_2\text{O})$ of Ref. 18. The values so obtained are given in Table IV, and the value of $\epsilon_{\text{SB}}(\text{O})$ is plotted as a function of energy in Fig. 2.

IV. DISCUSSION

The different values of $\epsilon(0)$ as a function of energy are plotted in Fig. 2, where the solid curve is $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$,¹⁵ the dotted curve is $\epsilon_{\text{expt}}(\text{H}_2\text{O})$ ¹⁸ - $\epsilon_{\text{expt}}(\text{H}_2)$,¹⁵ the short-dashed curve is $\epsilon_{\text{SB}}(0) = \epsilon_{\text{expt}}(\text{H}_2\text{O}) - 2\epsilon_{\text{SB}}(\text{H})$,¹¹ the long-dashed curve is " $\epsilon_{\text{solid}}(\text{O})$ " proposed by Ziegler and Chu,¹⁶ the dash-dotted curve is $\frac{1}{3}[\epsilon(\text{Er}_2\text{O}_3) - 2\epsilon(\text{Er})]$,¹⁴ and the values of $\epsilon_{\text{solid}}(\text{O})$ at 2 MeV obtained from $\alpha\text{-Fe}_2\text{O}_3$, Fe_3O_4 , SiO_2 , MgO , and Al_2O_3 by Feng *et al.*¹³ are also given. The error bars of the dotted curve and the short-dashed curve were computed in quadrature for the probable random errors of the individual components in each curve. The error bar of the dash-dotted curve was computed in the same way, using the absolute error (2%) of the stopping cross section of erbium and erbium oxide quoted in Ref. 14. No error was quoted for the semiempirical curve of Ref. 16.

The dashed curve $\epsilon_{\text{SB}}(0)$ is lower than the solid curve $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$ by 17% at low energy. This difference decreases to 3.3% at 2.0 MeV, where the error bars overlap. The fact that $\epsilon_{\text{SB}}(\text{O})$ is lower than $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$ agrees qualitatively with the concept found previously^{10,11} that double-bonded compounds give a higher stopping cross section, since gase-

TABLE III. Chemical structure of compounds used in present experiment.

Compound	Chemical formula	Chemical structure
Methyl alcohol	CH ₃ OH	<pre> H H — C — O — H H </pre>
Ethyl alcohol	C ₂ H ₅ OH	<pre> H H H — C — C — O — H H H </pre>
Propyl alcohol	C ₃ H ₇ OH	<pre> H H H H — C — C — C — O — H H H H </pre>
Dimethyl ether	(CH ₃) ₂ O	<pre> H H H — C — O — C — H H H </pre>
Diethyl ether	(C ₂ H ₅) ₂ O	<pre> H H H H H — C — C — O — C — C — H H H H H </pre>

ous oxygen O₂ is double bonded. Also, the closer agreement at 2.0 MeV is consistent with the concept that a chemical-bonding effect should be less as the energy increases (in the region to the right of the stopping cross section peak) due to less in-

TABLE IV. Tabulated values and probable errors of stopping cross section of O and OH, calculated from $\epsilon_{\text{expt}}(\text{H}_2\text{O})$ and $\epsilon_{\text{SB}}(\text{H})$. These values are applicable to single-bonded C-H-O compounds. ϵ is in 10^{-15} eV cm².

Energy (MeV)	$\epsilon_{\text{SB}}(\text{O})$	$\epsilon_{\text{SB}}(\text{OH})$
0.4	36.8 ± 0.9	52.8 ± 0.7
0.5	38.7 ± 1.0	55.3 ± 0.8
0.6	40.0 ± 1.0	56.7 ± 0.8
0.7	41.1 ± 1.0	57.3 ± 0.8
0.8	41.9 ± 1.0	57.4 ± 0.8
0.9	42.3 ± 1.0	57.0 ± 0.8
1.0	42.1 ± 0.9	56.1 ± 0.7
1.1	42.0 ± 0.9	55.2 ± 0.7
1.2	41.8 ± 0.9	54.2 ± 0.7
1.3	41.2 ± 0.9	52.95 ± 0.7
1.4	40.5 ± 0.9	51.6 ± 0.7
1.5	39.5 ± 0.9	50.05 ± 0.7
1.6	38.5 ± 0.9	48.6 ± 0.7
1.7	37.7 ± 0.8	47.3 ± 0.7
1.8	36.7 ± 0.8	45.95 ± 0.6
1.9	35.8 ± 0.8	44.7 ± 0.6
2.0	34.8 ± 0.7	43.4 ± 0.6

teraction between the incident particles and the valence electrons of the target atoms. The fact that the dotted curve $\epsilon_{\text{expt}}(\text{H}_2\text{O}) - \epsilon_{\text{expt}}(\text{H}_2)$ is lower than $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$, but greater by 5–10% than $\epsilon_{\text{SB}}(\text{O})$, reveals the effect of chemical bonding on the energy-loss process, particularly at lower energies.

The bond between the two H atoms in a hydrogen molecule is formed by the overlapping of 1s orbitals, so it is stronger than the bonds between H and O, or H and C, formed by the overlapping of a 1s and an sp^3 hybridized orbital which possesses some p character. This relative bond strength can be shown from the following numerical values: at 298°K the bond dissociation energy²³ for the hydrogen molecule (104.2 kcal/mole) is slightly higher than that for the OH radical (102.3 kcal/mole) and for methane (103.2 kcal/mole). The ionization potential²³ of the hydrogen molecule (15.43 eV) is also higher than that of the OH radical (13.18 eV) and methane (12.99 eV). A higher value of the mean excitation potential I in the Bethe-Bloch formula²⁴ gives a lower stopping cross section than a lower I value, so that $\epsilon_{\text{SB}}(\text{H})$ might be expected to be slightly higher than $\frac{1}{2}\epsilon_{\text{expt}}(\text{H}_2)$, a result that was seen in Ref. 11. Since the bonds involving H atoms in the single-bonded C-H-O compounds are of the $s-sp^3$ type, $\epsilon_{\text{SB}}(\text{H})$ should be used rather than $\frac{1}{2}\epsilon_{\text{expt}}(\text{H}_2)$. Hence, the short-dashed curve $\epsilon_{\text{SB}}(\text{O})$ in Fig. 2 should give a more meaningful representation of the

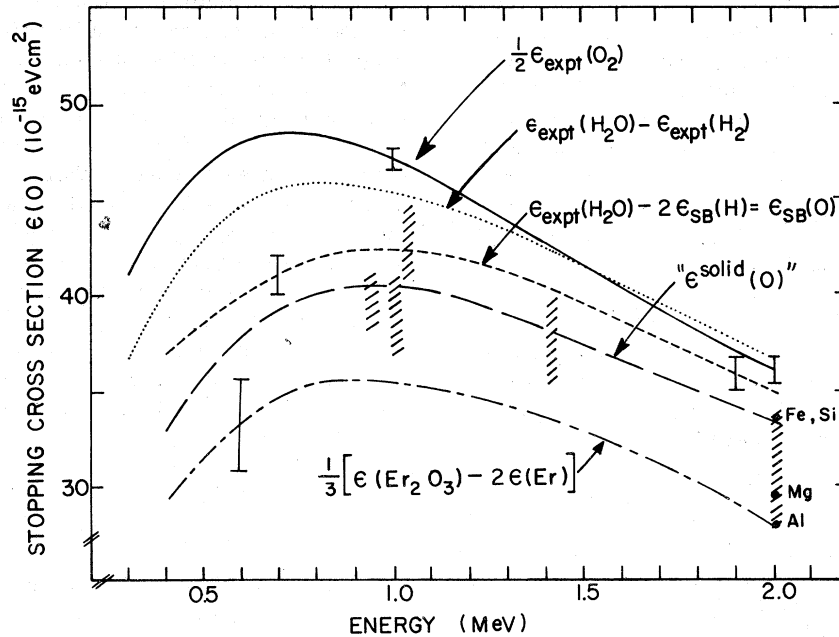


FIG. 2. Stopping cross section $\epsilon(O)$ of oxygen as a function of energy. The solid curve is one half the experimental value for molecular oxygen $\epsilon_{\text{expt}}(\text{O}_2)$ given in Ref. 15. Dotted curve, $\epsilon_{\text{expt}}(\text{H}_2\text{O})$ of vapor (Ref. 18) less $\epsilon_{\text{expt}}(\text{H}_2)$ from Ref. 15. Short-dashed curve, $\epsilon_{\text{expt}}(\text{H}_2\text{O}) - 2\epsilon_{\text{SB}}(\text{H}) = \epsilon_{\text{SB}}(\text{O})$, where $\epsilon_{\text{SB}}(\text{H})$ is given in Ref. 11; this curve is 3–17% lower than $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$ and clearly demonstrates the effect of chemical bonding on the stopping cross section. The bottom curve is found by subtracting $\frac{2}{3}\epsilon(\text{Er})$ from $\frac{1}{3}\epsilon(\text{Er}_2\text{O}_3)$ in Ref. 14. Long-dashed curve, a semiempirical curve (Ref. 16) which estimates the value of $\epsilon(\text{O})$ in solid compounds containing oxygen. Closed circles, “ $\epsilon^{\text{solid}}(\text{O})$ ” obtained from metal oxides by Feng (Ref. 13). Cross-hatched areas, the range of “ $\epsilon^{\text{solid}}(\text{O})$ ” values calculated from the yield ratios of Feng (Ref. 13) using various groups of experimental and adjusted stopping cross sections of metals (see text).

stopping cross sections of single-bonded oxygen compounds than $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$ or $\epsilon_{\text{expt}}(\text{H}_2\text{O}) - \epsilon_{\text{expt}}(\text{H}_2)$.

In Fig. 2, it is seen that $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2) > \epsilon_{\text{SB}}(\text{O}) > \epsilon_{\text{Er}_2\text{O}_3}(\text{O})$. Gaseous oxygen is purely covalent; $\epsilon_{\text{SB}}(\text{O})$ was calculated from $\epsilon_{\text{expt}}(\text{H}_2\text{O})$, and water possesses 39% ionic character, according to Pauling's electronegativity scale²⁵; erbium oxide possesses 74–76% ionic character. Thus, one might possibly suspect that the larger the percentage of ionic character of the compound, the lower the stopping cross section of oxygen in that compound. The values of the stopping cross section for “solid oxygen” of Feng *et al.*,¹³ however, did not agree to this approach, because Al_2O_3 possesses less ionic character (63%) than MgO (74%), yet the stopping cross section of their “solid oxygen” from Al_2O_3 was lower than that from MgO .

At this point, it would seem appropriate to comment on the values of the stopping cross section for “solid oxygen” by Feng *et al.* In their analysis, the ratios of $[\epsilon]_{\text{metal}}^{\text{O}}$ to $[\epsilon]_{\text{metal}}^{\text{metal}}$ {where $[\epsilon]_A^B = k_A \epsilon_B(E_{1n}) + |1/\cos\theta| \epsilon_B(E_{\text{out}})$ } were obtained from the metal to metallic oxide scattering-yield ratios, and the $[\epsilon]$ ratios were converted to the ratios of $\epsilon^{\text{solid}}(\text{O})$ to $\epsilon(\text{metal})$, using the semiempirical values for the metallic stopping cross sections of Ziegler and

Chu,²⁶ and finally the value of $\epsilon^{\text{solid}}(\text{O})$ was obtained by multiplying that ratio by the experimental metallic stopping cross section corresponding to each metallic oxide. The experimental metallic stopping cross sections they used were $\epsilon(\text{Fe})$, $\epsilon(\text{Mg})$, and $\epsilon(\text{Al})$ from Chu and Powers,²⁷ and $\epsilon(\text{Si})$ from Eisen *et al.*²⁸ However, neither the semiempirical values from Ziegler and Chu nor the published metallic stopping cross sections could satisfy their metal-to-metal yield ratios, and deviations up to 15% are found. In light of this finding, other reported metallic stopping cross sections have been used to calculate “ $\epsilon^{\text{solid}}(\text{O})$,” such as $\epsilon(\text{Al})$ by Porat and Ramavataram,²⁹ or $\epsilon(\text{Al})$ by Feng,³⁰ and $\epsilon(\text{Si})$ by Lin *et al.*,³¹ but none of the combinations will satisfy their metal-to-metal yield ratios. The reported metallic stopping cross sections have been adjusted arbitrarily so as to obtain agreement to their metal-to-metal yield ratios within 2.5%; for instance, $\epsilon(\text{Si})$ by Lin *et al.* was unchanged, $\epsilon(\text{Al})$ by Porat and Ramavataram was increased by 3%, $\epsilon(\text{Fe})$ by Chu and Powers was decreased by 7%, and $\epsilon(\text{Mg})$ by Chu and Powers was increased by 2%. Of course, there could be more than one way to adjust the reported metallic stopping cross sections to obtain agree-

ment to their metal-to-metal yield ratios, so the adjustment has been made in accordance with the theoretical Z_2 dependence of stopping cross sections.³² Their calculation, made only at 2.0 MeV, has also been extended to find the stopping cross sections of " $\epsilon^{\text{solid}}(\text{O})$ " at lower energies, viz., 0.94 MeV for MgO, 1.0 MeV for Al_2O_3 , 1.03 MeV for SiO_2 , and 1.41 MeV for $\alpha\text{-Fe}_2\text{O}_3$ and Fe_3O_4 . It is found that the values of " $\epsilon^{\text{solid}}(\text{O})$ " are a sensitive function of the choice of metallic stopping cross sections, and a variation as great as 17% is found, as is seen by the cross-hatched areas in Fig. 2. However, it is interesting that all of these values calculated from their yield ratios are below the curve $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$, and the majority are below the $\epsilon_{\text{SB}}(\text{O})$ curve. Hence, if there is indeed a common set of stopping cross sections for oxygen among the metallic oxides, it would be expected to be below the $\epsilon_{\text{SB}}(\text{O})$ curve between 1 to 2 MeV.

The long-dashed curve in Fig. 2 shows the semi-empirical value for $\epsilon^{\text{solid}}(\text{O})$ proposed by Ziegler and Chu,¹⁸ and agrees with the separate calculations made from the yield ratios of Feng *et al.* On the other hand, selfconsistency of the results from the yield ratios was the evidence that Feng *et al.* took for the existence of a common set of $\epsilon^{\text{solid}}(\text{O})$ among the metallic oxides, but we found that the relative contributions of stopping cross sections of oxygen varied by as much as 6% among the five oxides, rather than 2.7% as given in their paper. Therefore, although the approach by Feng *et al.* did demonstrate clearly a lower stopping cross section of oxygen for metal oxides than $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$, it is evi-

dent that more information on the stopping cross sections of metals and metallic oxides is needed to support the assumption (used by Langley¹⁴ as well as by Feng *et al.*) that $\epsilon(\text{metal})$ does not change from pure metal to oxide, and that there is indeed a common set of $\epsilon^{\text{solid}}(\text{O})$ values among the metallic oxides as stated by these authors. The careful measurement of ϵ for several metal oxides (same metal) would be very helpful since a plot of $\epsilon(\text{metal})$ vs $\epsilon(\text{oxygen})$ for a fixed He^+ ion energy (similar to the plot used¹⁰ by Powers *et al.* for hydrocarbons, and preferably at the energy of the maximum of the stopping-cross-section curve) would explicitly reveal whether or not $\epsilon(\text{metal})$, $\epsilon(\text{O})$, or both $\epsilon(\text{metal})$ and $\epsilon(\text{O})$ change in a metallic oxide.

V. CONCLUSION

The present work confirms that the values of the stopping cross sections $\epsilon(\text{CH}_2)$ and $\epsilon_{\text{SB}}(\text{H})$ of Ref. 11 can be used to predict the stopping cross sections of single-bonded C-H-O compounds for saturated ethers and alcohols. The values of $\epsilon_{\text{SB}}(\text{O})$ and $\epsilon_{\text{SB}}(\text{OH})$ were calculated; the fact that $\epsilon_{\text{SB}}(\text{O})$ is 3-17% lower than $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$ demonstrates the effect of chemical bonding on the stopping cross section. The $\epsilon_{\text{SB}}(\text{O})$ values found for the gas and vapor compounds of this experiment, although lower than $\frac{1}{2}\epsilon_{\text{expt}}(\text{O}_2)$, are still higher than the $\epsilon(\text{O})$ values predicted from the metal oxide yield ratios of Feng *et al.*

*Research supported in part by the Robert A. Welch Foundation, Houston, Texas, 77002.

¹W. H. Bragg and R. Kleeman, *Philos. Mag.* **10**, S 318 (1905).

²D. Thompson, Univ. of Calif. Radiation Laboratory Report No. UCRL-1910, 1952 (unpublished).

³H. K. Reynolds, D. N. F. Dunbar, W. A. Wenzel, and W. Whaling, *Phys. Rev.* **92**, 742 (1953).

⁴J. T. Park and E. J. Zimmerman, *Phys. Rev.* **131**, 1611 (1963).

⁵R. J. B. Palmer, *Proc. Phys. Soc. London* **87**, 681 (1966).

⁶E. Rotondi, National Research Council of Canada Report No. NRC-9076, 1966 (unpublished).

⁷D. A. Thompson and W. D. Mackintosh, *J. Appl. Phys.* **42**, 3969 (1971).

⁸P. D. Bourland and D. Powers, *Phys. Rev. B* **3**, 3635 (1971).

⁹D. Powers, W. K. Chu, R. J. Robinson, and A. S. Lodhi, *Phys. Rev. A* **6**, 1425 (1972).

¹⁰D. Powers, A. S. Lodhi, W. K. Lin, and H. L. Cox, Jr., *Thin Solid Films* **19**, 205 (1973).

¹¹A. S. Lodhi and D. Powers, *Phys. Rev. A* **10**, 2131

(1974).

¹²J. E. E. Baglin and J. F. Ziegler, *J. Appl. Phys.* **45**, 1413 (1974).

¹³J. S.-Y. Feng, W. K. Chu, and M.-A. Nicolet, *Phys. Rev. B* **10**, 3781 (1974).

¹⁴R. A. Langley and R. S. Blewer, *Nucl. Instrum. Methods* **132**, 109 (1976).

¹⁵P. D. Bourland, W. K. Chu, and D. Powers, *Phys. Rev. B* **3**, 3625 (1971).

¹⁶J. F. Ziegler and W. K. Chu, *J. Appl. Phys.* **47**, 2239 (1976); J. F. Ziegler, W. K. Chu, and J. S.-Y. Feng, *Appl. Phys. Lett.* **27**, 387 (1975).

¹⁷R. A. Langley, *Phys. Rev. B* **12**, 3575 (1975).

¹⁸S. Matteson, D. Powers, and E. K. L. Chau, *Phys. Rev. A* **15**, 856 (1977).

¹⁹G. Lorient and T. Moran, *Rev. Sci. Instrum.* **46**, 140 (1975).

²⁰S. Ruthberg, *J. Vac. Sci. Technol.* **6**, 401 (1969).

²¹K. F. Poulter, *J. Phys. E* **10**, 101 (1977).

²²D. K. Brice, *Phys. Rev. A* **6**, 1791 (1972).

²³V. I. Vedeneyev, L. V. Gurvich, V. N. Kondrat'yev, V. A. Medvedev, and Ye. L. Frankevich, *Bond Energies, Ionization Potentials, and Electron Affinities*

- (St. Martin's, New York, 1966).
- ²⁴F. Bloch, *Ann. Phys. (Leipz.)* 16, 285 (1933); *Z. Phys.* 81, 363 (1933).
- ²⁵L. Pauling, *The Nature of the Chemical Bond* (Cornell University, Ithaca, N. Y., 1960).
- ²⁶J. F. Ziegler and W. K. Chu, *At. Data Nucl. Data Tables* 13, 463 (1974).
- ²⁷W. K. Chu and D. Powers, *Phys. Rev.* 187, 478 (1969).
- ²⁸F. H. Eisen, C. J. Clark, J. Böttiger, and J. M. Poate, *Radiat. Eff.* 13, 93 (1972).
- ²⁹D. I. Porat and K. Ramavataram, *Proc. Phys. Soc. London* 78, 1135 (1961).
- ³⁰J. S.-Y. Feng, *J. Appl. Phys.* 46, 444 (1975).
- ³¹W. K. Lin, H. G. Olson, and D. Powers, *J. Appl. Phys.* 44, 3631 (1973).
- ³²W. K. Chu and D. Powers, *Phys. Lett.* 38A, 267 (1972).