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DETERMINATION OF THE GLOBAL RECOMBINATION RATE COEFFICIENT FOR THE ISX-B TOKAMAK *

R.A. LANGLEY and H.C. HOWE

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, USA

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The global recombination rate coefficient for hydrogen has been measured for the ISX-B tokamak vacuum vessel for various surface conditions. The measurements were performed by observing the rate of decrease of gas pressure in the vessel during a glow discharge. The parameters of the glow discharge and the complete experimental method are described. Previously published analytic and numerical models are used for data analysis. The effects of surface contamination on the results are described. For "unclean" wall conditions $\sigma k_r = 1.8 \times 10^{-28}$ cm^4/atom·s at 296 K and increases to $\sigma k_r = 4.4 \times 10^{-28}$ cm^4/atoms·s for "clean" conditions and remains constant until subsequent exposure to air.

1. Introduction

The hydrogen isotopes deuterium and tritium are used as fuel for magnetically confined reactors. Important areas of concern in dealing with the interaction of hydrogen with the reactor wall and in-vessel components (e.g., limiters) in fueling, recycling, and hydrogen inventory in the solid materials. The retention of hydrogen in the walls is mainly effected by its surface recombination rate at the wall and diffusion in the solid bulk of the wall. As has been shown previously [1,2] the effective parameter is $\sigma k_r/D$ where σ is the surface roughness factor, k_r is the surface recombination rate and D is the bulk diffusion coefficient.

Laboratory measurements have been made of σk_r and D. The results for σk_r for stainless steel are found to vary by up to four orders of magnitude at any particular temperature [3] depending on, with decreasing uncertainty, surface condition, specific type of material, and technique of measurement used. Reported values of k_r are in actuality σk_r. Experimental measurements of σ have shown increases above the geometrical area of up to a factor of 20 depending on surface conditioning, e.g., mechanical polishing and electropolishing. The results for D for stainless steel have been measured but have an uncertainty of perhaps a factor of 2 [4]. Uncertainties quoted by individual experimenters are much less, usually of the order ± 20%. Since many of the measurements are sensitive to surface effects it is not too surprising that significant differences are observed in the measured diffusion coefficient.

A previous paper [2] details both a simple analytic approach and a numerical model or obtaining results of $\sigma k_r/D$ from the experimental measurements. In addition it lists values for all pertinent physical parameters of the experimental system. The present experimental method allows determination of the global recombination coefficient for the vacuum vessel actually used for plasma studies and does not rely on laboratory simulators [1,5]. The remainder of this paper describes the experimental method, presents the experimental results, details the numerical modelling results for one set of experimental data and finally discusses the implications of the results.

2. Experimental method

The object is to measure the time evolution of the H_2 number density in the vacuum vessel during a glow discharge and correlate this with $\sigma k_r/D$, and is accomplished by measuring the time evolution of the pressure. The experimental method is to pump the vacuum chamber to its lowest ultimate pressure, usually 2×10^{-7} Torr, then close off all diagnostic ports and pumping ports so

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that there is *no* pumping. The wall outgassing rate is then determined by measuring the pressure rise as a function of time; typical outgassing rates are 5×10^{-10} Torr/cm^2 s. During some of these studies an RGA was used to determine the time evolution of the desorbed gas composition; the major components were: mass 2, $\approx 80\%$; mass 18, $\approx 18\%$; mass 28, $\approx 1\%$; and mass 15 and 44, $\approx 0.2\%$. Following this measurement H_2 gas is admitted to a pressure of about 25 mTorr and a glow discharge struck by applying a DC voltage between the torus wall and a rectangular stainless steel electrode with surface area of 0.055 m^2 inserted vertically into the central volume of the torus, shown schematically in fig. 1. Current versus voltage scans were made to determine the type of glow discharge, see fig. 2. As suspected the glow obtained was an "abnormal" glow discharge [6]. The gas is only ionized to a few percent and the discharge current is sustained by ions accelerated across the dark space hitting the wall with the concomidant emission of secondary electrons which are in turn accelerated from the wall across the dark space and into the glow region.

Usually the glow discharge was started with approximately 25 mTorr of H_2 in the vessel. At this pressure and down to about 15 mTorr the glow was visually observed to be uniform and steady; below 15 mTorr the glow became somewhat unsteady and usually the glow discharge was purposely terminated above this pressure. At an applied voltage of 550 V the current was steady and constant down to about 2 mTorr; below 2 mTorr the glow discharge became tenuous and extinguished at about 0.5 mTorr. At 550 V a pressure of at least 15 mTorr was required to initiate a glow discharge.

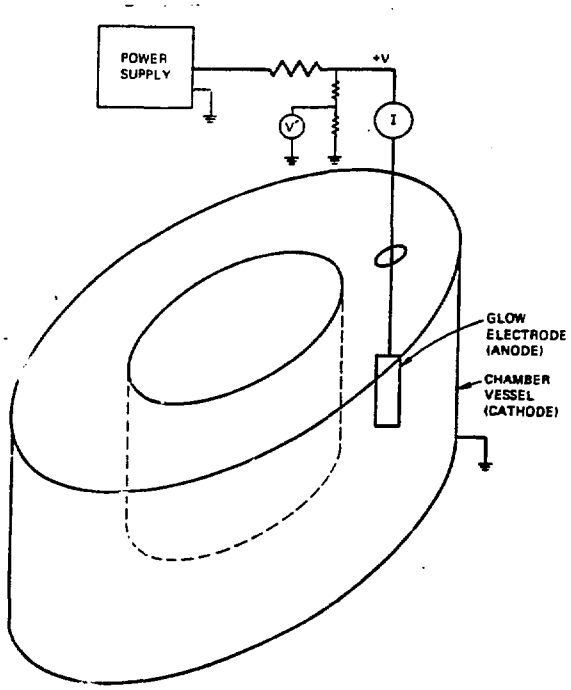

Fig. 1. Schematic of experimental set-up.

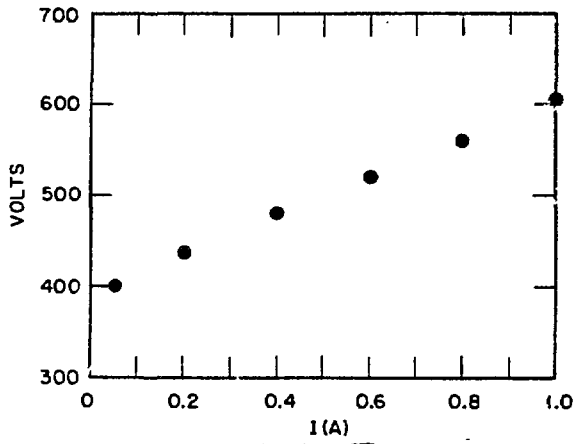

Fig. 2. Plot of voltage versus current for glow discharge in the ISX-B tokamak vacuum vessel.

During many of the discharges the background gas composition was monitored using a residual gas analyzer (RGA) with a highly attenuated flow. For "clean" conditions only small changes were observed in the major impurities (CH_4, H_2O and CO), while for "unclean" conditions large fractions of CH_4 were observed to be formed. Since the formation of CH_4 depletes the number of H_2 molecules in the vacuum vessel the pressure would be expected to decrease. This is discussed in detail in the next section.

3. Results

Fig. 3 presents the data for a typical "clean" vacuum vessel. The definition of clean used here is two fold, i.e., (1) the tokamak had been low power discharged cleaned [7] for at least five days at eight hours per day following a previous opening to air and (2) the impurity levels, as measured by the attenuated flow RGA, remained insignificant during the initiation and steady state portions of the glow discharge.

The initial rate of pressure decrease, $\Delta P/\Delta t$, can be

correlated directly with the incident flux since at initiation of the glow discharge and for a short time afterward, depending on the implantation depth and diffusion coefficient, there is no outgoing flux due to recombination, only that due to reflection. The relation between $\Delta P/\Delta t$ and the flux is given by

$$F_p = N_L \frac{V}{A} \frac{\Delta P}{\Delta t} \frac{1}{1-(R_{H^\circ} + R_{H^+})},$$

where N_L is Loschmidt's number, V the chamber volume and A the wall area exposed to the glow discharge. R_{H° and RD^+ are the effective reflection coefficients for H_2^+ incident on the surface with the emission of H° and H^+ respectively. Since the energy of the reflected particles is always lower than the energy of the incident H_2^+ and H^+ particles the reflected H^+ particles return to the wall whereas the H° particles can move freely across the cathode drop. For energies less than 1 keV, which is the case here, $R_{H^+}/R_{H^\circ} \ll 1$ and we need only be concerned with the fate of the reflfected H° particles. It appears probable that the reflected H° particles collide mainly with the background gas particles, H_2, and slow down until they adsorb on the wall [8]. If this is true then the effective R_{H° is much smaller than the measured and calculated reflections coefficients. For the energy range of incident particles used in this experiment, i.e. up to about 500 eV, only calculated reflection coefficients are available. Data from Oen and Robinson [9] indicate that R is slowly decreasing with increasing energy and a value between 0.2 and 0.5 is appropriate for the actual reflection coefficient, while a value of $\leqslant 0.1$ is more probably appropriate for the effective reflection coefficient.

As a consistency check the flux determined by the initial pressure drop was compared with that determined by the discharge current. In order to directly compare the two, assumptions must be made concerning the average charge per particle, f, of the particles hitting the wall, and the secondary electron coefficient, γ, for incident H_2^+ and H^+ on hydrogen covered stainless steel surfaces. The flux as determined from the discharge current is given by

$$F_I = \frac{I}{eA} \cdot \frac{f}{1+\gamma}, \tag{2}$$

where I is the current. Previous measurements of the ion energy distributions for hydrogen glow discharges have shown that H_2^+ is the dominant ion at $\approx 85\%$ while H^+ was found at a level of $\approx 15\%$ therefore an appropriate value for f is 1.85 [10]. Adsorbed layers have been found to significantly influence γ: monolayers of gas atoms have been found to sometimes increase and sometimes decrease the yield at low ion energy. Most reproducible measurements of γ have been made for clean (flash heated) surfaces or "gas-covered" (probably oxygen and nitrogen) surfaces. Previous results for incident hydrogen found that γ was independent of the substrate for "gas-covered" surfaces of Cu [11], Ag [12], Au [11] and stainless steel [13]. In addition Ray and Barnett found that $\gamma(H^+, E) \sim \gamma(H^\circ; E)$ for $E > 100$ eV [11]. Values of γ for H_2^+ and H^+ on a H_2 gas-covered stainless steel surface are difficult to predict, but indications are the γ may be fairly low, i.e. less than 0.05, because of the low electron density in the adsorbed hydrogen layer [14].

Eqs. (1) and (2) contain both measured and estimated quantities, i.e., f, $R(R_{H^\circ} + R_{H^+})$ and γ. In order to quantize these estimated quantities we note that $F_p = F_I$ and using eqs. (1) and (2) defined a quantity

$$\beta = N_L V \frac{\Delta P}{\Delta t} \frac{e}{I}, \tag{3a}$$

which contains only constants and directly measured quantities. Also

$$\beta = \frac{1-R}{1+\gamma} \cdot f \tag{3b}$$

where both R and γ can only be estimated with rather large uncertainties and would be expected to be dependent on surface conditions. As noted previously f can be estimated with a rather small uncertainty and would be expected to be independent of surface conditions. Experimentally it is observed that β, as determined by eq. (3a), varies from 0.5 for uncleaned surfaces to about 1.8 for cleaned surfaces. This implies that for clean conditions $(1-R)/(1+\gamma)$ is almost unity and both R and are small and the incident flux can be determined uniquely using either eq. (1) or (2), while for unclean conditions $(1-R)/(1+\gamma)$ is about 0.3 and either R or γ or both increase significantly and no unique flux can be obtained directly, only estimated.

As the glow discharge progresses in time the pressure decreases because some of the implanted particles diffuse away from the surface and into the bulk. The remainder of the particles diffuse to the surface and recombine to form H_2 and desorb to the gas phase. This decrease is easily seen in fig. 3 until the discharge is terminated at 40 min at which time the pressure rises because the incident flux has been reduced to zero while the previously implanted hydrogen continues to diffuse both into the bulk and to the surface where it recombines and desorbs. An analytic model [2] has been presented which describes the discharge-on portion of the pressure vs. time curve to within about 5% when compared with a complete numerical treatment which

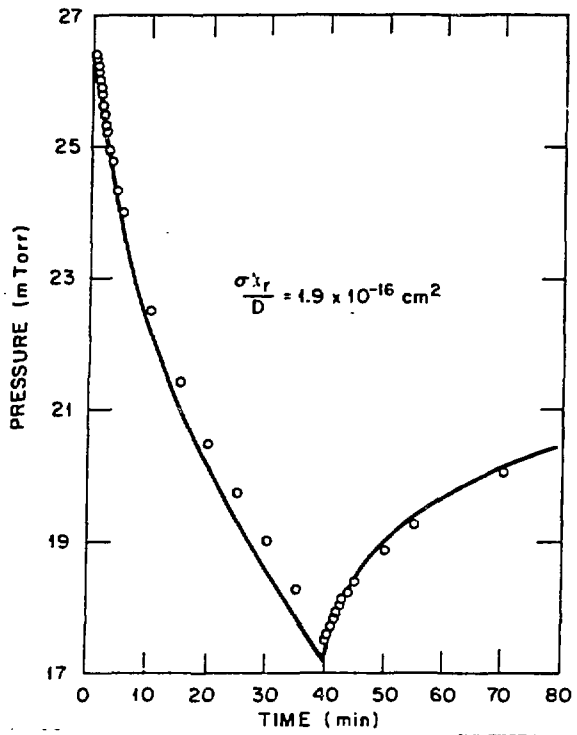

Fig. 3. Typical plot of pressure versus time response curve for a clean vacuum vessel. Open circles are experimental points; solid curve is obtainedby using numerical model using $\sigma k_r/D = 1.9\times10^{-16}$ cm^2 as determined from analytic approach.

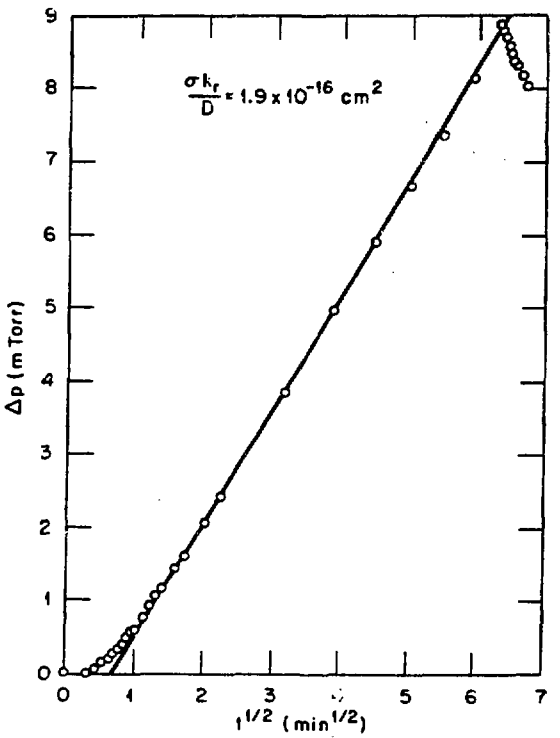

Fig. 4. Plot of the change in pressure versus square root of time. Open circles are experimental points of fig. 3 and solid line is best fit to linear portion. The value of $\sigma k_r/D$ is determined from the analytic approach.

includes implantation, diffusion, and recombination [2]. The results of the analytic model predict that

$$\frac{\sigma k_r}{D} \alpha (T_w A/V\, 1/K)^2 \phi_0 \text{ cm}^2, \quad (4)$$

where k is the measured slope of the pressures. the square root of time, e.g. see fig. 4. We estimate the uncertainty in the wall area exposed to the glow discharge to be less than $\pm 10\%$, in the chamber volume to be $\pm 5\%$, in the slope (K) to be $+5\%$ and in the deposited flux to be $\pm 10\%$ for "clean" conditions. These combined uncertainties result in an uncertainty in $\sigma k_r/D$ of $\pm 25\%$ for "clean" conditions and $\pm 40\%$ for "unclean" conditions. An analytic formulation has not been found to adequately model the recovery (discharge-off) phenomena but the numerical treatment does provide reasonable agreement, $\pm 10\%$, with the experimental results. Further analysis of this recovery phenomena is unders study.

For an "unclean" vacuum vessel, the initial phase of the pressure versus time response does not behave the same as for a "clean" vacuum vessel. An example of results for such a condition is shown in fig. 5. An RGA with an attenuated flow rate was used to measure the impurities as a function of time. On initiation of the glow discharge masses 16 and 28 were observed to increase significantly while other masses increased only slightly Almost all of the mass 16 signal is due to CH_4, as determined by comparing the magnitudes of the mass 12, 14, 15 signals to the mass 16 signal and the mass 28 signal is almost all due to CO as determined by comparing the mass 28 to the mass 14 and 12 signals. This rise in CH_4 partial pressure corresponds both in time and magnitude to the initial drop in pressure, see fig. 5. The plateau level in the pressure versus time curve following the increase in both the CH_4 and CO concentration is probably indicative of the complete consumption of all exposed physisorbed carbon and oxygen followed by migration of these impurities to parts of the vacuum vessel inaccessible to the glow discharge and their subsequent adsorption.

Fig. 3 shows one set of experimental results for a

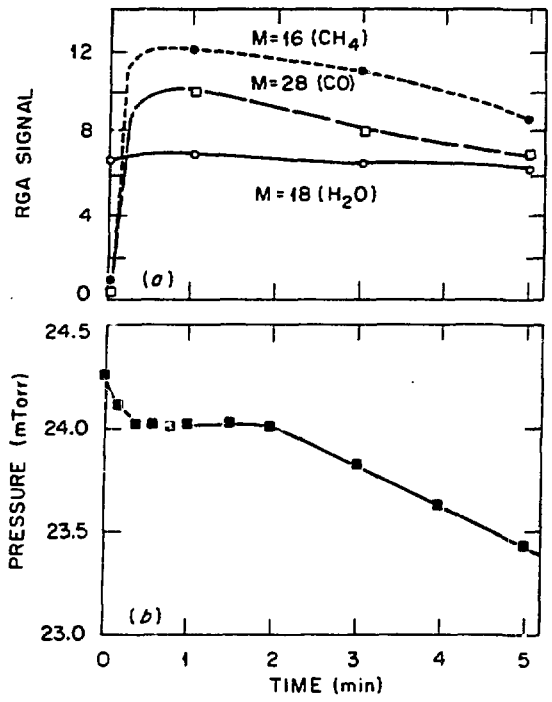

Fig. 5. Plot of initial pressure for an "unclean" vacuum system and of residual gas analyzer signal for masses 16, 18, and 28 versus time.

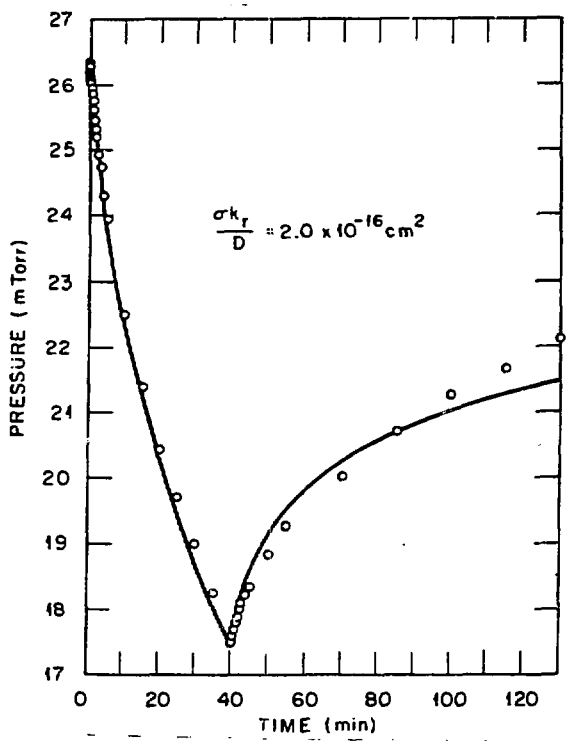

Fig. 6. Plot of pressure versus time for data of fig. 3. Solid line is the results of the numerical model using $\sigma k_r/D = 2.0 \times 10^{-16}$ cm^2.

"clean" vessel where the starting pressure was 26 mTorr. The discharge was on for 40 min and then turned off. The change in pressure versus the square root of time is shown in fig. 4. The analytic approach [2], which applies only to the "discharge-on" condition, yields a value of $\sigma k_r/D$ of 1.9×10^{-16} cm^2 which when used in the numerical model provides the solid line in fig. 3. The calculated pressure response is lower than the experimentally measured values indicating that a slightly higher $\sigma k_r/D$ is appropriate. The results using $\sigma k_r/D = 2.0 \times 10^{-16}$ cm^2 are shown in fig. 6 where the agreement is quite good for the "discharge-on" condition. The analytic approach is found to consistently underestimate $\sigma k_r/D$ by about 5% when compared with the numerical approach. This difference is almost insignificant considering the uncertainties in σ and D are relatively large.

The pressure-time response when the glow discharge is terminated is dependent on the amount and distribution of hydrogen in the wall, D, and σk_r. Experimentally it is observed that the pressure change is linear with $t^{1/2}$ after a short, < 1 min., delay.

Experiments were performed over a time period exceeding12 months and resulted in measurements of $\sigma k_r/D$ for surface conditions which varied from air exposed to extensive combined discharge cleaning and tokamak discharges. Titanium gettering is occasionally performed in ISX-B with the application of a few monolayers of titanium to approximately 70% of the wall area [15]. For all measurements reported here the titanium film was fully saturated and is thought to influence the results in only a minor way. After a prolonged exposure to air $\sigma k_r/D = 0.9 \times 10^{-16}$ cm^2 and increased to 2.2×10^{-16} cm^2 as the walls were cleaned. These results are shown on an Arrhenius plot in fig. 7 assuming $d = 2.0 \times 10^{-12}$ cm^2/s. The size of the symbol represents the scatter in the data and the uncertainties are shown by the bars. Also shown are results of laboratory experiments and theoretical calculations. For laboratory measurements higher values of σk_r were obtained with sputter-cleaned surfaces while oxide-covered surfaces are generally represented by the lower values of σk_r.

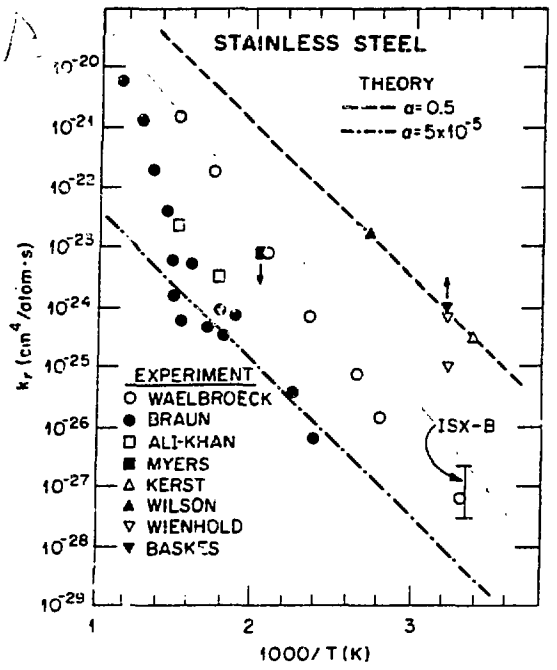

Fig. 7. Arrhenius plot of σk_r for both laboratory experiments and theory. Here D is taken as 2.0×10^{-12} cm^2/s for the present experimental data. See text for explanation of data points and uncertainty limits. References for data are: Waelbroeck [16], Braun [17], Ali-Khan [81], Myers [19], Kerst [20], Wilson [21, Wienhold [22], and Baskes [23]. Baskes theoretical approach uses the molecular sticking coefficient, α, as an input variable; results are shown for two values.

These results indicate that the ISX vacuum chamber has, for "clean" conditions, a stable surface oxide layer as opposed to an oxide-free sputter-cleaned surface. Although D_2 was not used extensively as the working gas, initial results for $\sigma k_r/D$ were the same for both gases even though the measured discharge current using D_2 was approximately 20% less at the same applied voltage

4. Conclusions

The major conclusions are:

(1) Surface chemistry effects can significantly affect the results for "unclean" walls.

(2) The factor β varies significantly with surface cleanliness; from 0.5 for an air exposed surface to about 1.8 for a clean surface.

(3) For the clean ISX-B vacuum vessel at room temperature $\sigma k_r/D = 2.2\times10^{-16}$ cm^2.

(4) After an air exposure $\sigma k_r/D$ increases with increasing cleanliness to 2.2×10^{-16} cm^2, thereafter it remains constant with discharge cleaning and increasing number of tokamak discharges.

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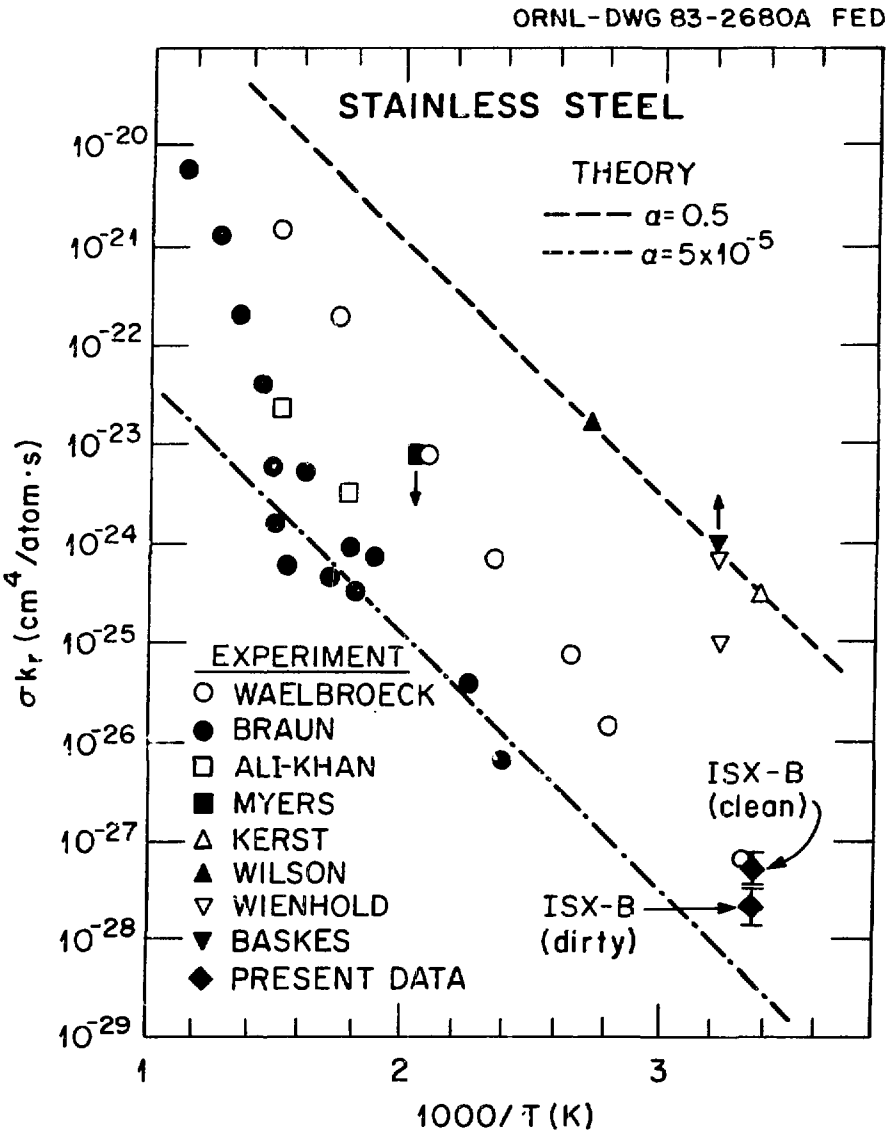

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