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DESORPTION AND RELATED PHENOMENA RELEVANT TO FUSION DEVICES

EDITED BY A.KOMA

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Edited by
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Abstract

Desorption and related phenomena involved in the plasma-wall interactions in fusion devices are briefly reviewed. Discussed are: ion-induced desorption(Section II), electron-stimulated desorption (III), photodesorption (IV), ion-induced re-emission of H, D and He atoms (V), chemisorption on carbide surfaces (VI), and theory of desorption (VII). A rather comprehensive bibliography is attached and typical data on the relevant quantities are shown when available.

CONTENTS

Desorption on the First Walls of Nuclear Fusion Devices

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The surface of a solid is generally covered with foreign atoms and molecules. Those foreign atoms leave the surface and go into the vacuum due to the heat or the impact of incident atoms, ions, electrons and photons. This is so-called desorption and plays significant rolls in the plasma-wall interactions in two ways, which occur at the surface of the first walls of the nuclear fusion devices. One is the impurity introduction process. Such impurity atoms as C or O on the first wall go into the plasma due to the desorption and cause the serious radiation loss. The other is the fuel gas recycling process. The hydrogen isotopes are adsorbed on the surface of the first wall and go back to the plasma due to the desorption. This process makes important contribution to the energy and particle balances of the plasma. Thus it is urgently needed to make clear the desorption on the surface of the first wall. This is the purpose of the present compilation of the data related to desoptions.

As for the impurity introduction process from the first wall into the plasma, sputtering has been mostly notified. Actually the desorption occurs much more easily than the sputtering. One

of the reason is the low binding energy (usually less than 1 eV) of the impurity atoms adsorbed to the surface of the first wall. This binding energy is considerably lower than the one involved in the sputtering. Even if such impurity atoms as C and O are chemically and strongly bound to the first wall, they react with impinging hydrogen isotopes and changes their forms into CD_4 or D_2O, which are bound to the surface physically and loosely. Thus the detailed understanding of desorption process is needed to reduce the impurity introduction to the plasma. The knowledge is also useful to the effective discharge cleaning of the surface of the first wall.

Various desorption processes occur on the surface of the first wall. Themal desorption is the most popular one, and many data are available about them. Ion-induced, electron-induced and photo-induced desorptions are the ones charcteristic to the plasma wall interactions. Section II, III and IV of the present report are devoted to those desorption processes, respectively. Comparison of the yield for each desorption mechanism is shown in Table 1 and Table 2. Table 1 is the results of Bauer's estimation for C and O on stainless steel[1]. It indicates that ion-induced desorption will make the largest contribution to the impurity introduction to the plasma due to the desorption]. Table 2 shows the result of Wilson's estimation on the numbers of desorbed deuterons, which contribute to the fuel gas recycling. Again ion-induced desorption seems to be the leading desorption process in the fuel gas recycling.

Incident Particles	Desorption Yield (atoms/particle)	Incident Flux ($cm^{-2}s^{-1}$)	Released Impurity ($cm^{-2}s^{-1}$)
D	2	1×10^{16}	2×10^{16}
electron	5×10^{-3}	4×10^{16}	2×10^{14}
photon	4×10^{-4}	10^{17}	4×10^{13}

Table 1. Evaluation of desorption of C and O on stainless steel[1]

Incident Particle	Cross Section (cm^2)	Incident Flux ($cm^{-2}s^{-1}$)	Desorbed D /Adsorbed D
D	10^{-16}	10^{16}	1
electron	10^{-17}	5×10^{16}	0.5
photon	$10^{-20} - 10^{-18}$	10^{18}	0.01 - 1

Table 2 Evaluation of fuel gas recycling due to desorption[2]

The followings are remaining problems to be solved for further understanding of the desorption process at the first walls of the nuclear fusion devices.

Even if a clean surface is once obtained by the dischrge cleaning, there still remains a problem. There are a large amount of impurities inside the first wall materials, and large gradient of impurity concentration is formed by the removal of impurity atoms on the surface of the first wall. That concentration gradient acts as a motive force for the diffusion of the impurities from the inside of the wall to the surface. Thus the desorption process on the first wall must be made clear in connection with

the diffusion process in the first wall materials. In this connection Section V of the present report is devoted to the ion-induced detrapping, replacement and release of deuterium and helium atoms.

Such low z materials as graphite and carbides are promising candidate materials as limitter, armour and coating materials. But there are few data on the desorption on low z materials. It is needed to produce and compile desorption data for low z materials. In this connection chemisorption on the carbide surface is reviewed in Section VI of the present report.

Data for photo-desorption are also lacking. In addition to the data for cross-section, informations about intensity and energy distributions of photons impinging on the first wall are needed as well.

The knowledge about the coverage of hydrogen isotopes on the first wall material is needed in the estimation of desorption yield of hydrogen isotopes. But those data are lacking mainly because Auger electron spectroscopy or ion scattering spectroscopy, which are successfully used to check the surface coverage of atoms other than hydrogen isotopes, are essentially insensitive to hydro gen isotopes. Development of new technique is needed to detect hydrogen isotopes on the surface with high sensitivity.

Desorption and adsorption is much more sensitive to the surface conditions than sputtering. The surface of the first wall chages their forms by sputtering and blistering etc.. In some compound materials, the surface composition changes as well. Thus the data for those "technical" surfaces are needed for the reli-

able estimations of desorption yields at the actual first wall, which are lacking as well.

Fianlly theoretical analysis is also needed for systematic understanding of the process and to make reasonable estimation when few experimental data are available. Present status and problems in the theory of desorption are reviewed in Section VII of the present report.

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ION INDUCED DESORPTION

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1. Introduction

Historically ion induced desorption of adsorbed species has been well known for many years as a phenomenon in glow discharge cleaning to obtain atomically clean surfaces [1-13]. The systematic studies, however, have been made only for this decade, giving the measured desorption yields (atoms/ion or molecules/ion) or the desorption cross sections(cm^2) under well controlled surface conditions as to surface preparation[10-45].

As for the theoretical approach and computer simulations to ion induced desorption, there have been the theoretical estimates due to Winters and Sigmund [22] and computer simulations tried by Taglauer et al. [24, 26, 29, 39], both of which use the binary elastic collision model essentially. Agreement between experimental and theoretical values is not good about both of numerical values and their dependences on the primary ion energy and incident angle of ions.

2. Desorption Yields [10-20]

The measurements of desorption yields (atoms/ion or molecules/ion) have been performed mainly for the technological surfaces of vacuum devices in order to research the optimum conditions of vacuum bakeout and glow discharge cleaning [10-13, 18, 19]. And also, the yields for the condensed gas

layers on cryogenic surfaces have been measured [14-16].

(a) Measured systems (Table 1)

(b) Effect of vacuum bakeout temperature (Fig.1-Fig.8)

(c) Effect of glow discharge cleaning [9, 46, 47] (Fig.9- Fig.11)

(d) Condensed gas layers (Fig.12-Fig.14)

(e) Chemical aspects [9] (Fig.15)

3. Desorption Cross Cections [21-45]

The desorption cross sections in cm^2 can be measured, without knowing the surface atomic density of adsorbates, from monitoring the relative change of the remaining surface coverage as a function of irradiating ion dose, by using surface analytical methods such as ion scattering spectroscopy (ISS) [25], Auger electron spectroscopy (AES) [28], secondary ion mass spectrometry (SIMS) [34], and the analysis using ion induced photon (IPP) [34]. There are other complicated methods which use residual gas analysis (RGA) [22,42]. As to the species of projectile, inert gas ions have been mainly used. Therefore the knowledge about the chemical effect on desorption due to chemically active gas ions are very lacked at present [9, 28, 41, 42].

(a) Measured systems (Table 2)

(b) Dependence on the ion energy (Fig.16-Fig.25)

(c) Theoretical estimates and computer simulations [22, 24, 26, 29, 39, 45] (Fig. 25, 30, 32 and 33)

(d) Dependence on the heat of adsorption (Fig.29 and 30)

(e) Dependence on the incident angle of ions (Fig.31-Fig.33)

(f) Effect of surface damages [40]

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Table Captions

Table 1 Substrate-adsorbate-ion combinations for the data compilation of ion induced desorption yields.

Table 2 Substrate-adsorbate-ion combinations for the data compilation of ion induced desorption cross sections.

Figure Captions

Fig. 1 Desorption yields as a function of vacuum bakeout temperature for 24 h [19].

Fig. 2 "

Fig. 3 "

Fig. 4 "

Fig. 5 "

Fig. 6 "

Fig. 7 Desorption yields as a function of K^+ ion energy for 316 L + N stainless steel after vacuum bakeout at 300°C for 24 h [18].

Fig. 8 Desorption yields as a function of the ion energy for the ISR stainless steel vacuum chamber (316 L + N) after vacuum bakeouts at 300°C and 340°C for 24 h (ISR proton beam current = 3A) [10].

Fig. 9 The dependence of the desorption yields on the ion energy (1) before and (2) after an Ar glow discharge cleaning (measured at 6.0 and 9.5 A ISR proton beam currents respectively). Negative yields indicate the phenomenom of beam pumping [10].

Fig. 10 The electron and K^+ ion induced desorption yields for 316 L + N stainless steel before and after an in situ pure Ar glow discharge [11].

Fig. 11 The electron and K^+ ion induced desorption yields for pure Al before and after an in situ pure Ar glow discharge [13].

Fig. 12 Initial yields of desorbed hydrogen as a function of condesed gas thickness when bombarded by 5 and 20 keV protons. The exposure of the surface to hydrogen to form the condensed layer is given. The surface temperature of Cu substrate is 3.2°K [14].

Fig. 13 Initial yields of desorbed CO as a fucntion of CO exposure when bombarded by 5 keV protons. The surface temperature of Cu substrate is 4.2°K [15].

Fig. 14 Desorption yields of H_2O molecules by D^+ ions as a function of incident ion energy. The surface temperature of the stainless steel substrate is 77°K [16].

Fig. 15 Desorption yields when bombarded by Ar^+ ions (< 520 eV). The substrate 304 L stainless steel was baked at 270°C for more than 8 hr (marked B) and then exposed to CO_2, H_2, CO, air O_2, CH_4, Ar, H_2O or N_2 at 1×10^{-5} Torr for 2 h [20].

Fig. 16 Ion energy dependence of desorption cross sections measured by Sagara et al. (43).

Fig. 17 Ion energy dependence of desorption cross sections measured by Taglauer et al. (23, 29, 33), McDonald et al. (34) and Sagara et al. (45).

Fig. 18 Ion energy dependence of desorption cross sections measured by Taglauer et al. (23, 26, 33), Van den Berg et al. (27).

Fig. 19 Ion energy dependence of desorption cross sections measured by Windawi and Katzer (28), Taglauer et al. (24) and Sagara et al. (45).

Fig. 20 The desorption cross section measured by Taglauer and Heiland (40).

Fig. 21 Ion energy dependence of desorption cross sections measured by Sagara et al. (37, 43, 45).

Fig. 22 Ion energy dependence of desorption cross sections measured by Tsuchidate et al. (44) and Sagara et al. (37, 45).

Fig. 23 Ion energy dependence of desorption cross sections measured by Sagara et al. (45).

Fig. 24 Ion energy dependence of desorption cross sections measured by Winters and Sigmund (22).

Fig. 25 Ion energy dependence of desorption cross sections measured by Taglauer et al. (29, 31).

Fig. 26 Desorption cross sections as a function of ion energy for S on 316 stainless steel by hydrogen ion impact at 45° incidence from normal to surface [41].

Fig. 27 Desorption cross sections as a function of ion energy for CO on 304 stainless steel by various ions at the normal incidence to surface [42].

Fig. 28 Calculated cross sections for the desorption of S from Ni (110) with prjectiles of the given atomic species, MORLAY: numerical code; W + S: theory of Winters and Sigmund [39].

Fig. 29 Dependence of the desorption cross sections for O_2 and CO on various metals as a function of the inverse heat of adsorption [35, 36, 39].

Fig. 30 Calculated desorption cross sections for oxygen on various metals. Here E_B corresponds to Q in Fig. 29. The dashed line is a linear fit to the data points including the origin [35, 36, 39].

Fig. 31 Dependence of the desorption cross section on the angle of the incoming ion beam relative to the surface for oxygen on Si (111) [39].

Fig. 32 Calculated and experimental values for the desorption cross section of CO on Ni as a function of the incident angle of Ne^+ ions relative to the surface [29].

Fig. 33 Measured and calculated dependence of the desorption cross section on the incident angle relative to the surface [24].

Table I

Substrate	Adsorbate	Ion(keV)	Fig.no.	Method and references	Condition
Al	residual gases	$^{15}N_2^+$,K^+(2)	11	RGA*(12,13)	bake and Ar discharge
Cu(poly.)	H_2 , D_2	H^+,D^+(5-20)	12	RGA(14)	condensed at 2.5-4.2 K
	He , N_2	H^+ (5)		RGA(15)	〃
	CO , Ar	H^+ (5)	13	RGA(15)	〃
Stainless steel	H_2O	D^+(5-35)	14	RGA(16)	〃 at 77 K
304	CO	Ar^+ (1)		RGA(17)	.66 atoms/ion
	CO	Ar^+(<0.52)	15	RGA(20)	
	CH_4,N_2	〃	〃	〃	
	H_2O,O_2	〃	〃	〃	
	CO_2,Ar	〃	〃	〃	
316L+N	residual gases	H^+(0-3)	8,9	RGA(10)	bake and Ar discharge
	〃	K^+(2)	10	RGA(11)	Ar discharge
	〃	$^{15}N_2^+$,K^+ (1.4,0-2)	1,7	RGA(12,13 18,19)	bake
Inconel 600	〃	K^+(1.4)	3	RGA(13,19)	bake
718	〃	K^+(1.4)	4	RGA(18,19)	bake
Al alloy 5086	〃	K^+(1.4)	6	RGA(19)	bake
6061	CO	Ar^+(1)		RGA(17)	.76 atoms/ion
Ti alloy 13V11Cr3Al	〃	$^{15}N_2^+$ (2)		RGA(13)	bake
14V6Al	〃	K^+ (1.4)	2	RGA(19)	bake
Cu-OFHC	〃	K^+ (1.4)	5	RGA(13,19)	bake

* RGA : Residual Gas Analysis

Table 2 (No. 1)

Substrate	Adsorbate	Ion(keV)	Angle (from normal)	Fig.No.	Method and references
Si(111)	O_2	He (1.0)	20-75	31	ISS*1 (39)
Ti	H_2	He (1.5)	45	0.003atoms/ion	RGA*2 (43)
		Ar (1.5)	45	0.01 atoms/ion	RGA (43)
	CO	He (1.5)	45	16	ISS (43)
		He (0.5)	30	29	ISS (35,36)
		Ar (1.5)	45	16	ISS (43)
	O_2	He (0.5)	30	29	ISS (35,36)
Ni(poly.)	CO	He,Ne (2.0)	45	17	ISS IIP*3 (34,38)
		Ar (2.0)	45	17	IIP SIMS*4 (34,38)
		He,Ne(0.3-2.6)	45	17	ISS (45)
	S	H (0.25-1.0)	30	19	AES*5 (28)
		He,Ne(0.2-2.6)	45	19	ISS (45)
		Ar (0.5)	30	19	AES (28)
Ni(110)	CO	He (0.4-1.2)	60	17	ISS (23,35)
		Ne (1.0)	60	17	ISS (33)
	O_2	^{3}He,Ne(0.6-1.6)	60	18	ISS (23,26,33)
		H (0.5)	30	29	ISS (35)
		Ne (6.0)	75	18	ISS (27)
		Ar (0.3-1.8)	60	18	ISS (23)
	S	He,Ne,Ar (0.2-1.8)	60	19	ISS (24)
Ni(111)	CO	Ne (0.7)	20-75	17,32	ISS (29)
	S	Ne (0.2-1.4)	60	19	ISS (24)
		Ne (1.0)	20-70	33	ISS (24)
Cu(poly.)	O_2	He (0.5)	30	20,29	ISS (35,40)
Mo(poly.)	CO	He (0.5-2.5)	45	21	ISS (37)
		Ar (1.5)	45	21	ISS (43)
	O_2	He (0.5-2.5)	45	22	ISS (32,37)
		He,Ne,Ar (0.5-2.5)	0	22	AES (44)
Mo(110)	CO	He,Ne,Ar (0.4-2.6)	45	21	ISS (45)
	O_2	He,Ne,Ar (0.4-2.5)	45	22	ISS (45)
	S	He,Ne,Ar (0.3-2.8)	45	23	ISS (45)
Pd	O_2	He (0.5)	30	29	ISS (35)
Ta	O_2	He (0.5)	30	29	ISS (35)
W(poly.)	$^{15}N_2$	He,Ne,Ar,Kr,Xe (0.02-0.5)	0	24	flash filament (22)
	O_2	He (0.5)	30	29	ISS (35)
W(100)	H_2	^{3}He (0.8-1.8)	60	25	ISS (29,31)

Table 2 (No. 2)

Substrate	Adsorbate	Ion(keV)	Angle (from normal)	Fig.No.	Method and references
Pt	O_2	He (0.5)	30	29	ISS (35)
Stainless steel 316	S	H (0.3-1.0)	45	26	SIMS (41)
304	CO	H,D(0.1-0.5)	0	27	RGA (42)
		He,Ne,Ar (0.2-0.9)	0	27	RGA (42)

*1 ISS : Ion Scattering Spectroscopy
*2 RGA : Residual Gass Analysis
*3 IIP : Ion induced Photon
*4 SIMS: Secondary Ion Mass Spectrometry
*5 AES : Auger Electron Spectroscopy

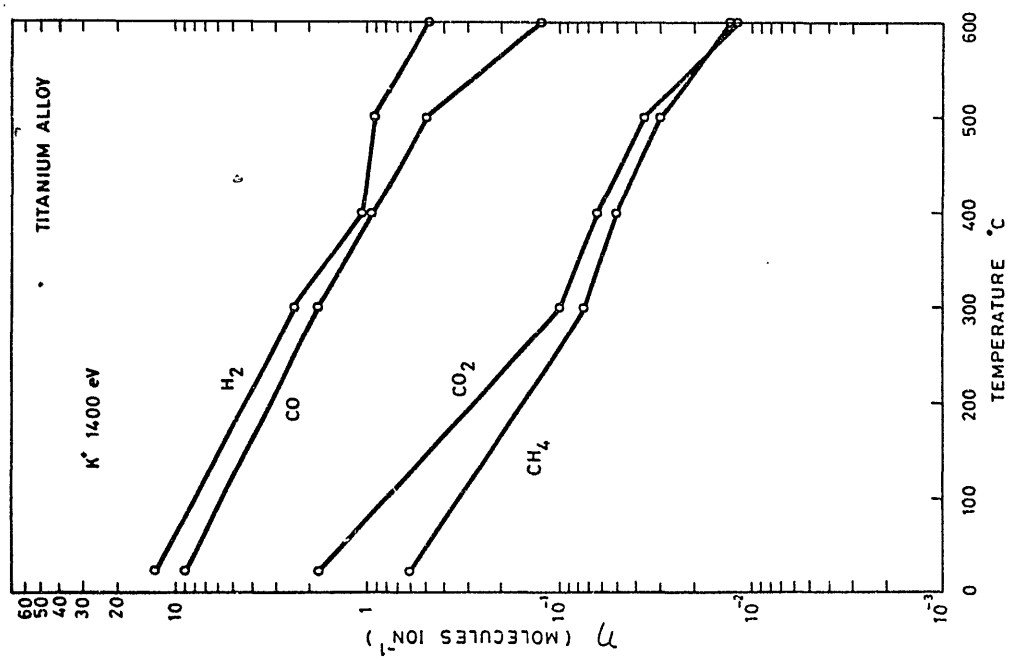
TITANIUM ALLOY
K° 1400 eV
H2
CO
CO2
CH4
η (MOLECULES ION-1)
TEMPERATURE °C
60
50
40
30
20
10
1
10-1
10-2
10-3
0
100
200
300
400
500
600

Fig. 2

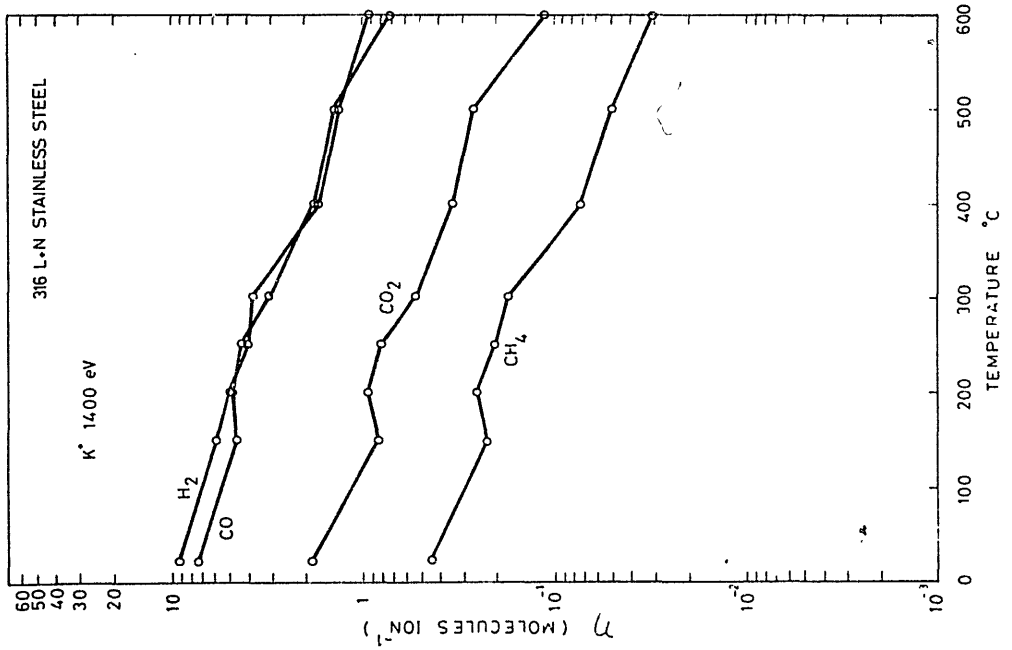
316 L-N STAINLESS STEEL
K° 1400 eV
H2
CO
CO2
CH4
η (MOLECULES ION-1)
TEMPERATURE °C
60
50
40
30
20
10
1
10-1
10-2
10-3
0
100
200
300
400
500
600

Fig. 1

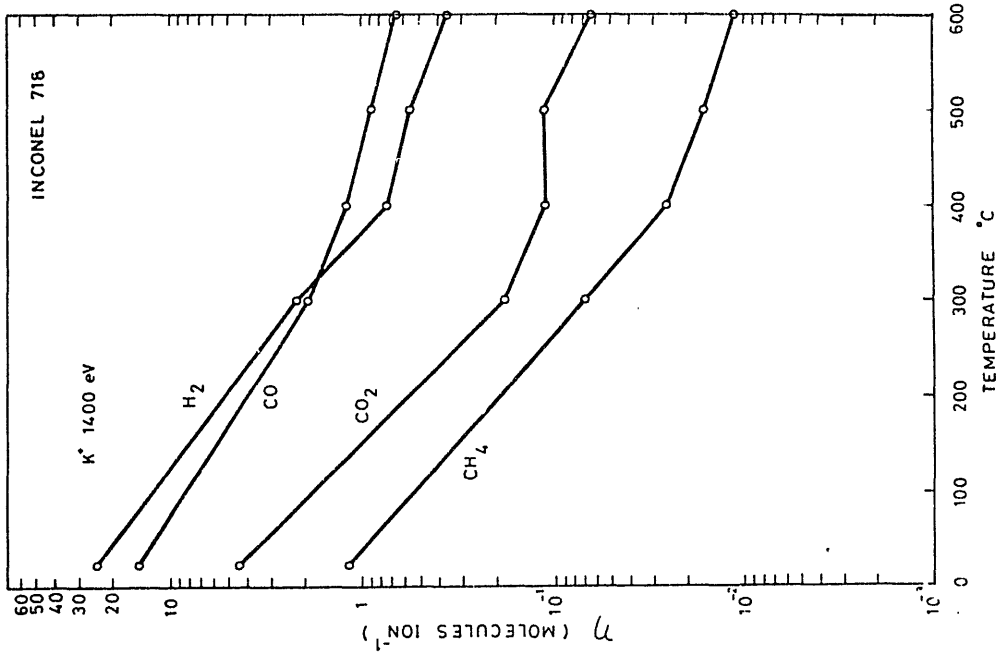
INCONEL 716
K' 1400 eV
H2
CO
CO2
CH4
η (MOLECULES ION-1)
TEMPERATURE °C

Fig. 4

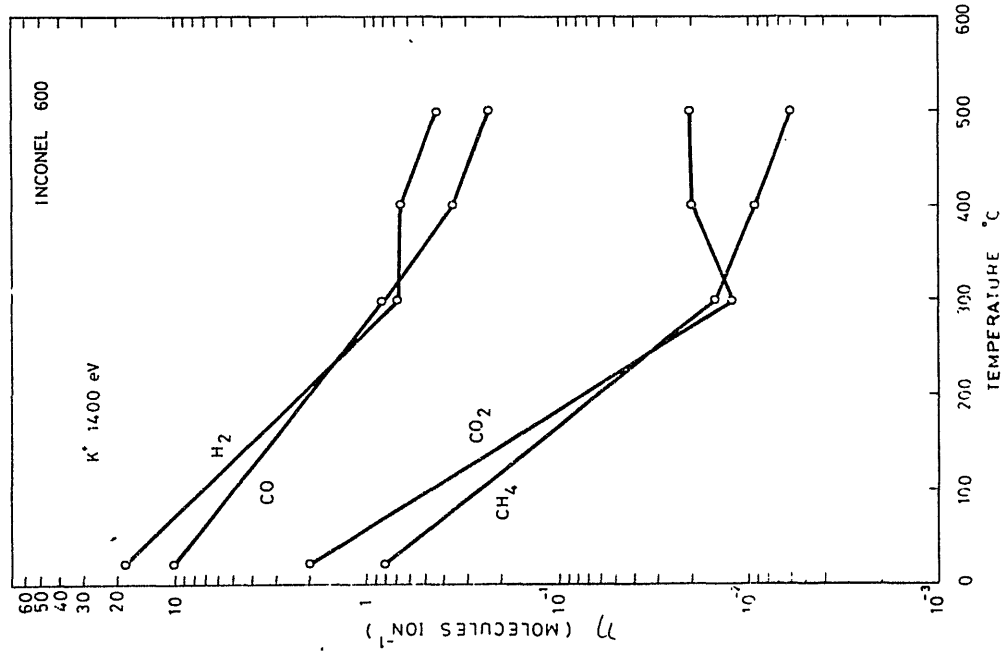
INCONEL 600
K' 1400 eV
H2
CO
CO2
CH4
η (MOLECULES ION-1)
TEMPERATURE °C

Fig. 3

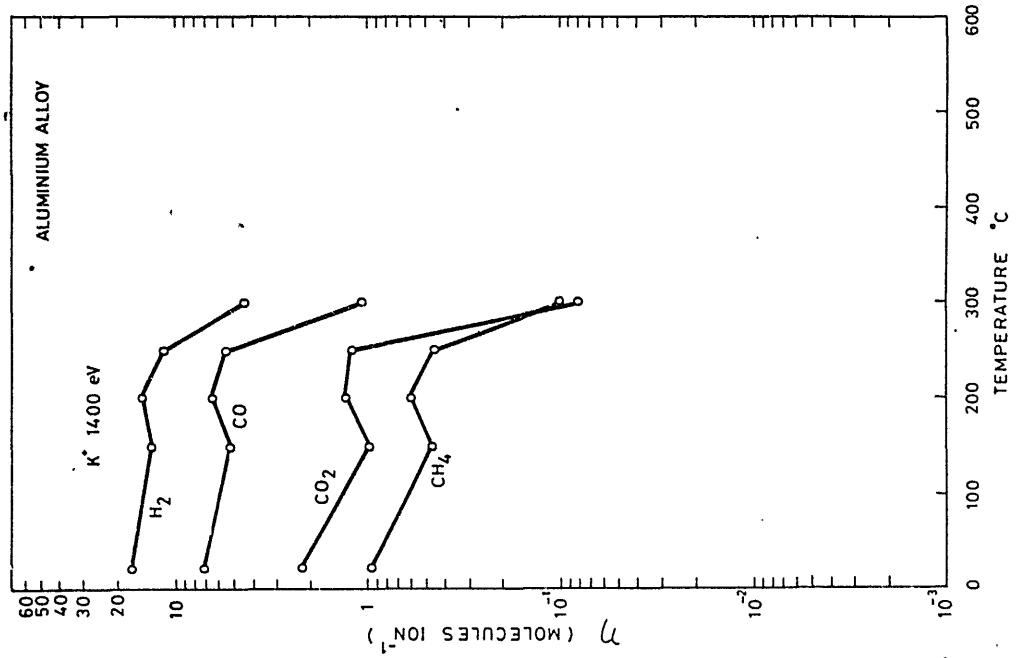
ALUMINIUM ALLOY
K* 1400 eV
H2
CO
CO2
CH4
η (MOLECULES ION-1)
TEMPERATURE °C

Fig. 6

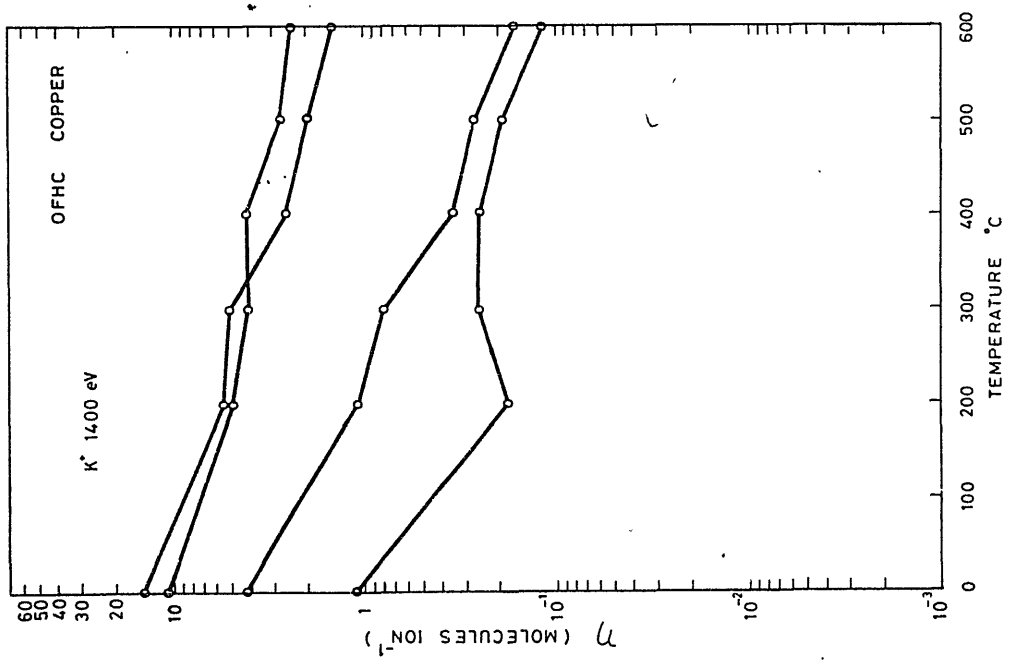
OFHC COPPER
K* 1400 eV
η (MOLECULES ION-1)
TEMPERATURE °C

Fig. 5

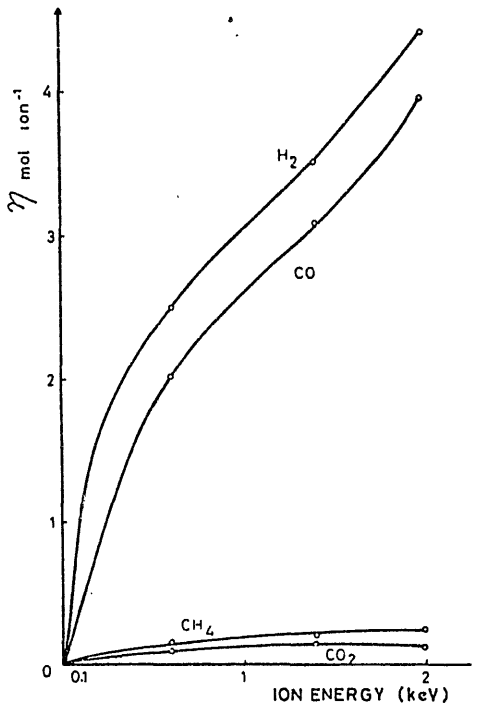
η mol ion⁻¹
4
3
2
1
0
H2
CO
CH4
CO2
0.1
1
2
ION ENERGY (keV)

Fig. 7

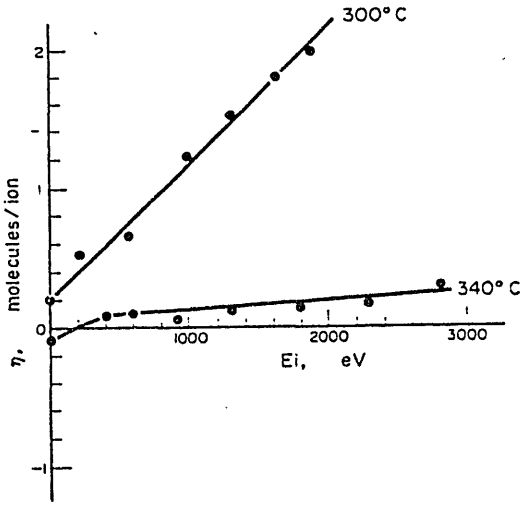
300° C
340° C
molecules/ion
η,
2
1
0
-1
1000
2000
3000
Ei, eV

Fig. 8

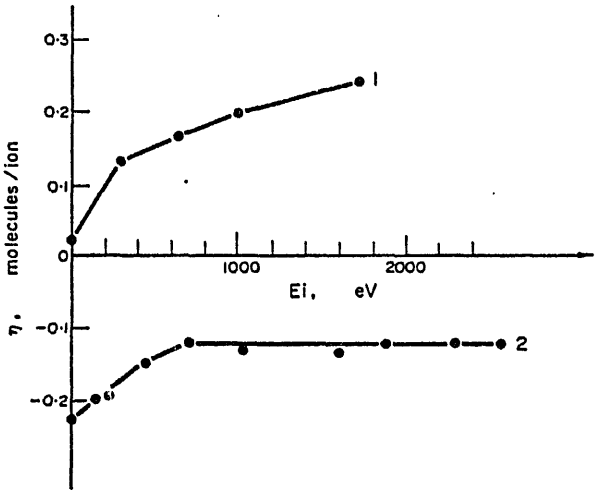
0·3
0·2
0·1
0
−0·1
−0·2
η, molecules / ion
1000
2000
Ei, eV
1
2

Fig. 9

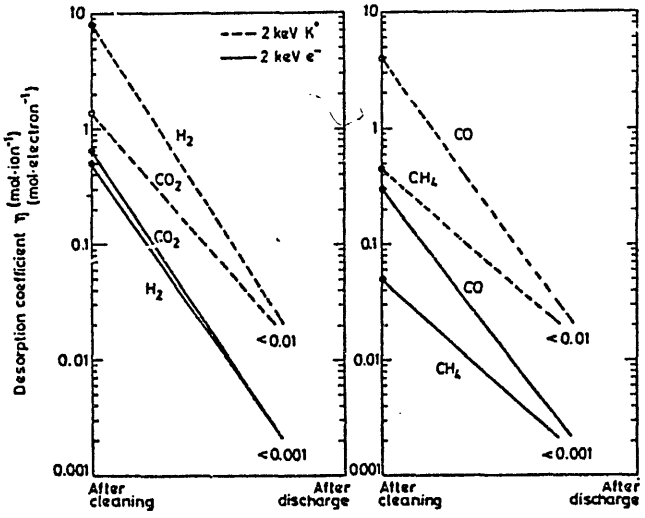
10
1
0.1
0.01
0.001
Desorption coefficient η (mol·ion⁻¹) (mol·electron⁻¹)
2 keV K⁺
2 keV e⁻
H₂
CO₂
CO₂
H₂
<0.01
<0.001
CO
CH₄
CO
CH₄
<0.01
<0.001
After cleaning
After discharge
After cleaning
After discharge

Fig. 10

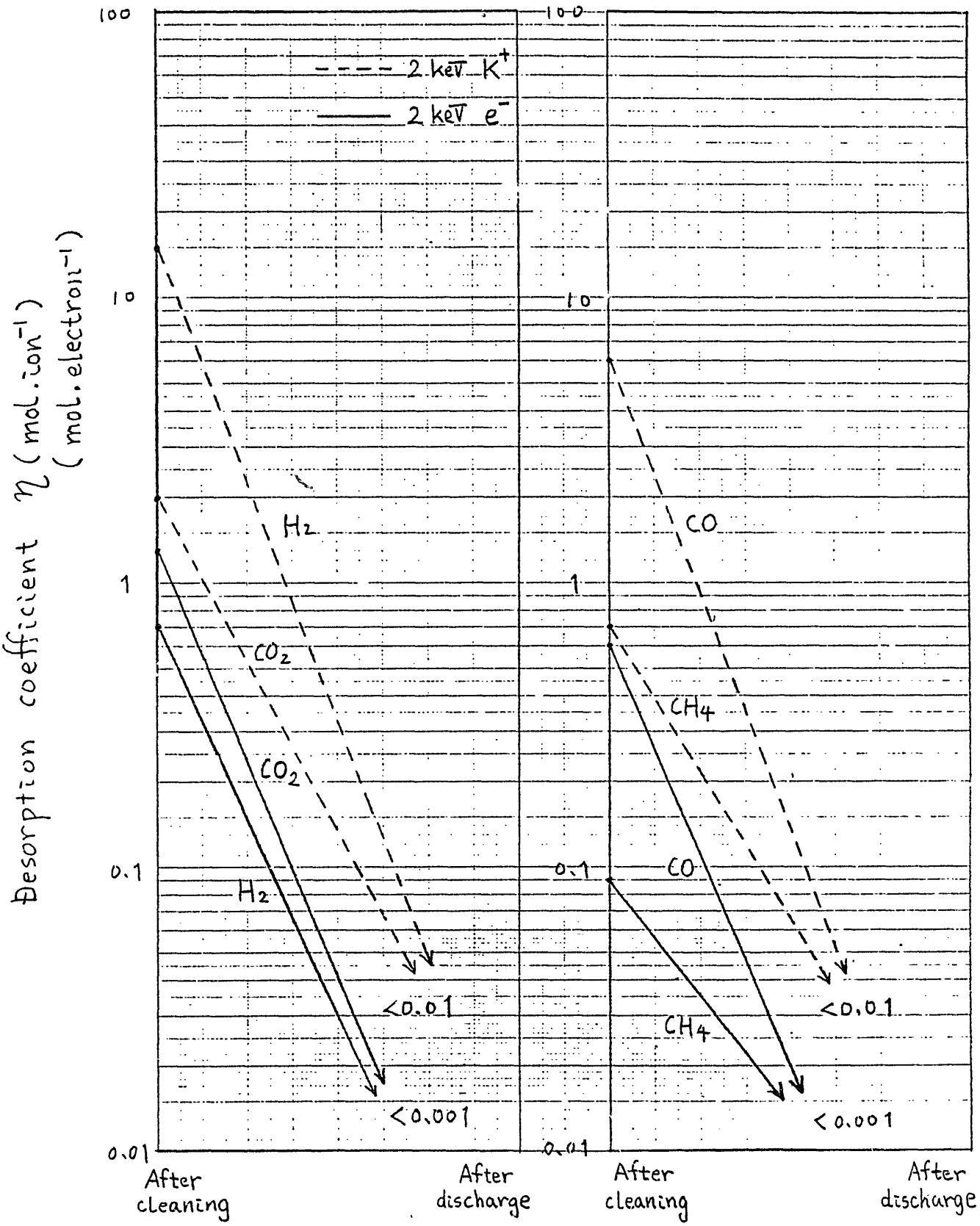
Desorption coefficient η (mol.ion⁻¹) (mol.electron⁻¹)
100
10
1
0.1
0.01
2 keV K⁺
2 keV e⁻
H₂
CO₂
CO₂
H₂
<0.01
<0.001
CO
CH₄
CO
CH₄
<0.01
<0.001
After cleaning
After discharge
After cleaning
After discharge

Fig. 11

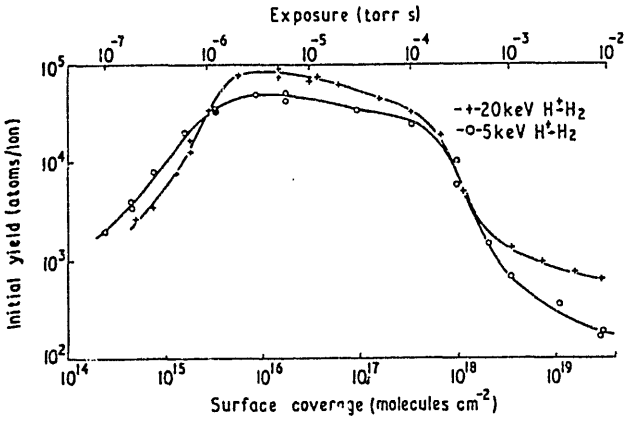

Fig. 12

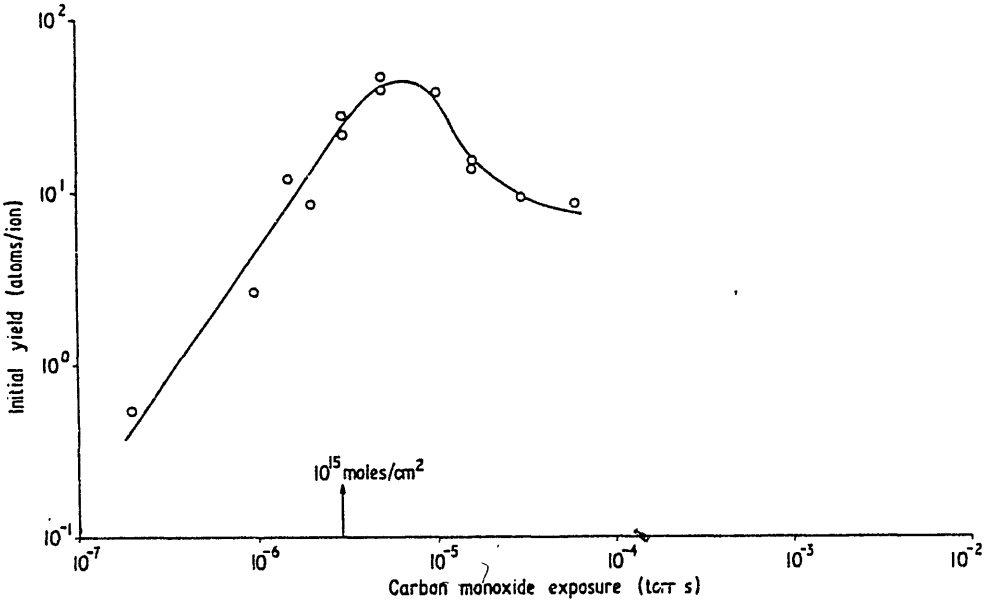

Fig. 13

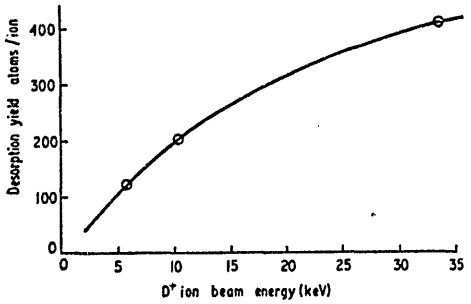

Fig. 14

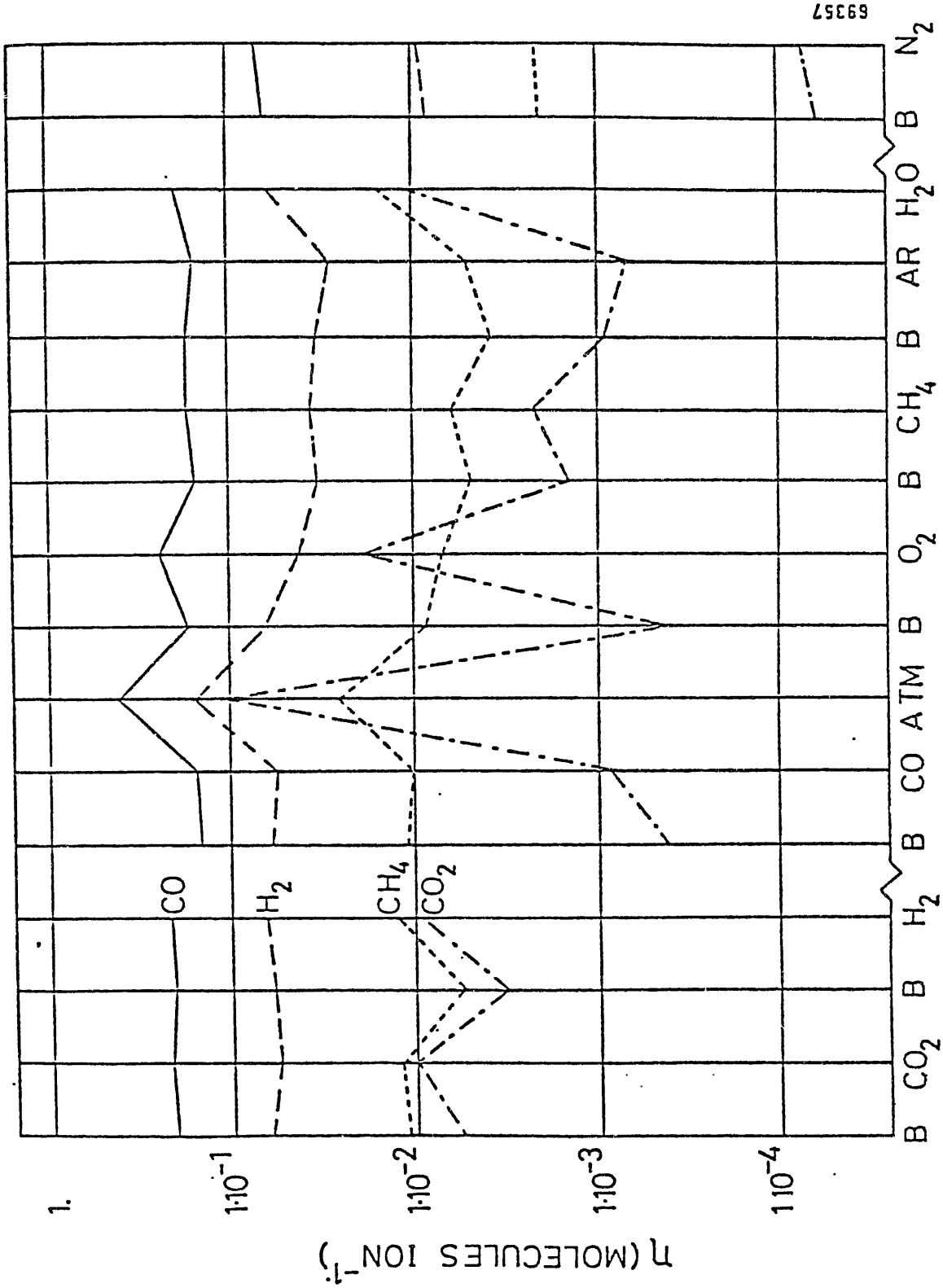
η (MOLECULES ION^{-1})
1.
$1 \cdot 10^{-1}$
$1 \cdot 10^{-2}$
$1 \cdot 10^{-3}$
$1 \cdot 10^{-4}$
CO
H_2
CH_4
CO_2
B CO_2 B H_2 B CO ATM B O_2 B CH_4 B AR H_2O B N_2

Fig. 15

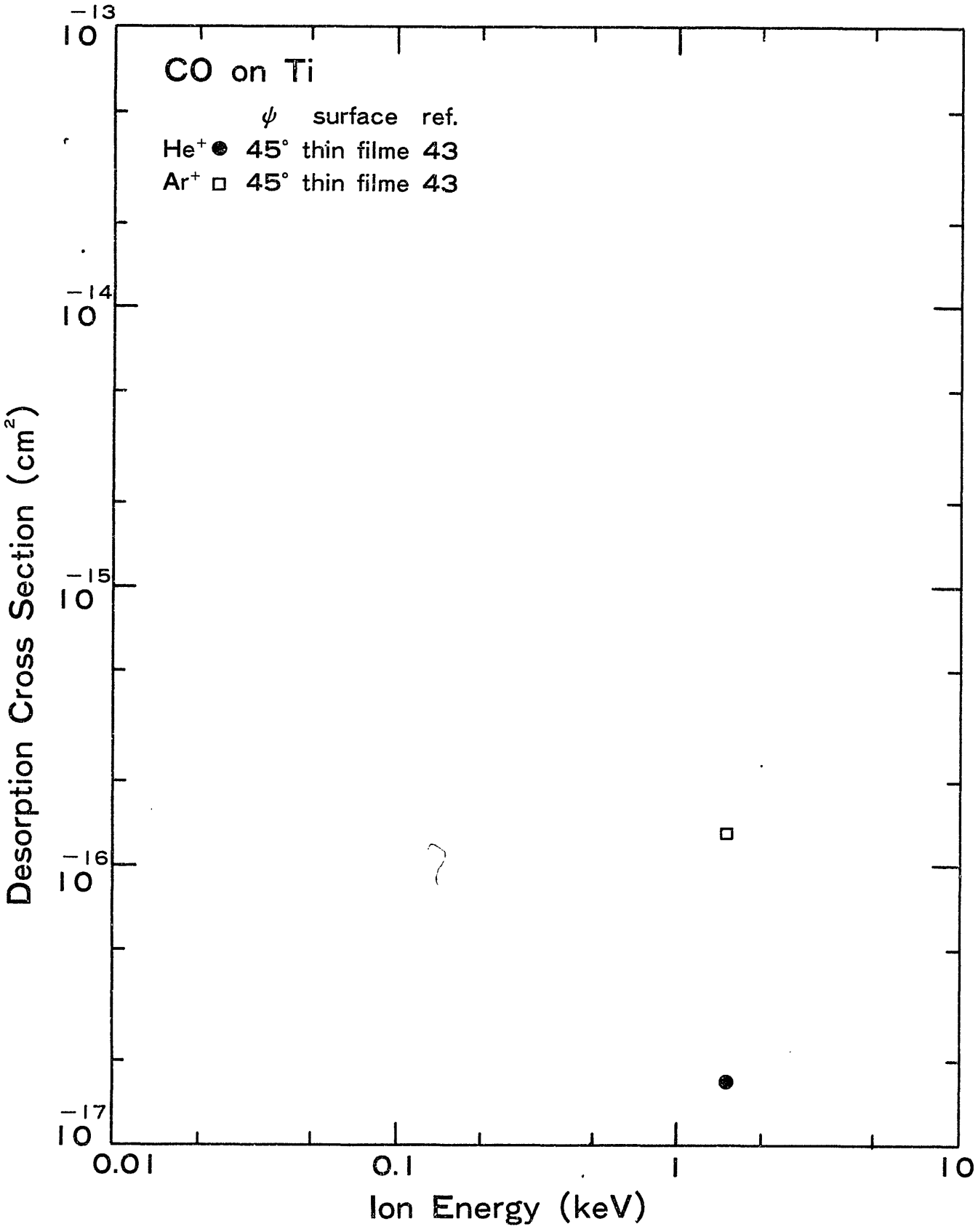
CO on Ti
ψ surface ref.
He+ ● 45° thin filme 43
Ar+ □ 45° thin filme 43
Desorption Cross Section (cm²)
10⁻¹³
10⁻¹⁴
10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
0.01
0.1
1
10
Ion Energy (keV)

Fig. 16

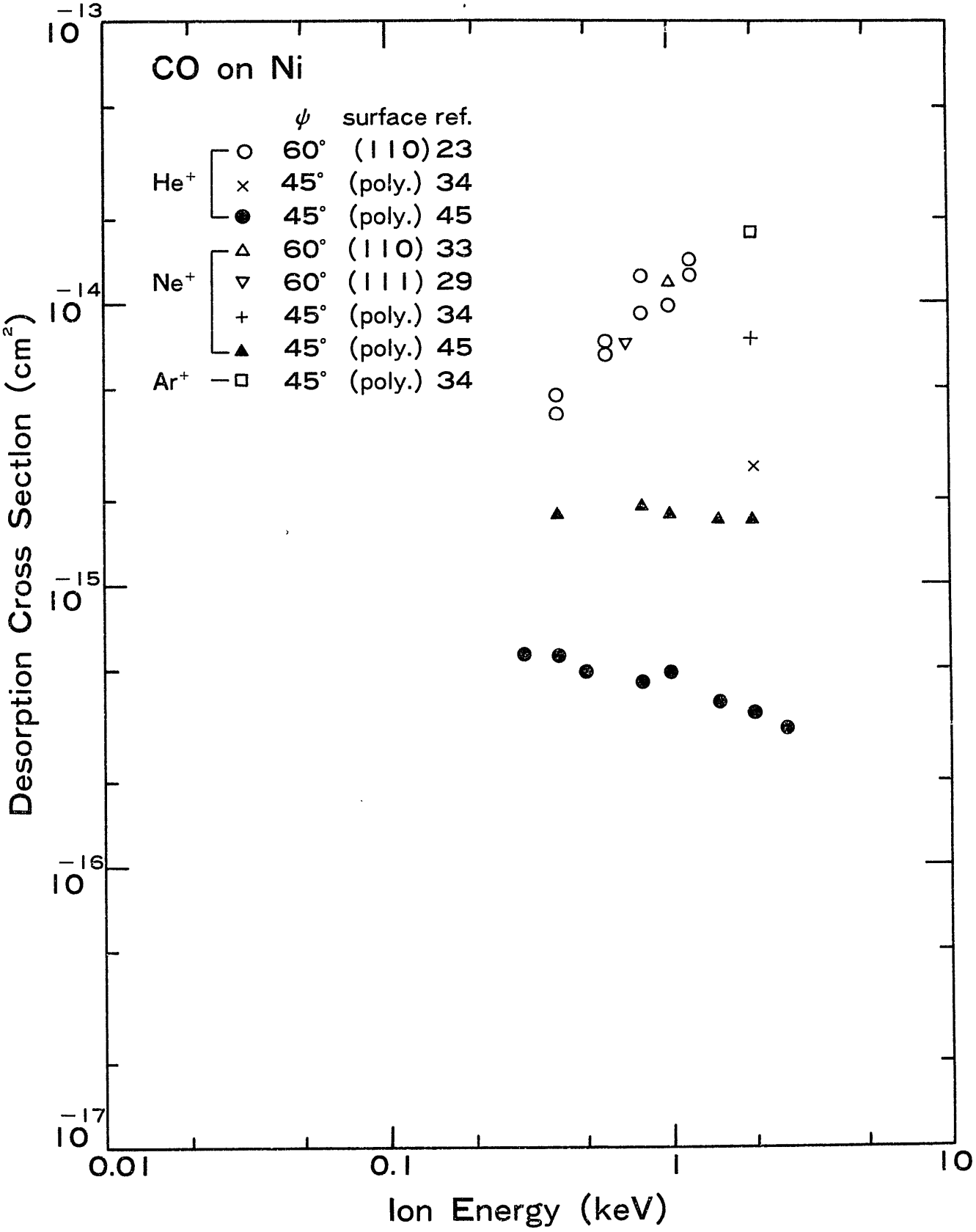
CO on Ni
ψ surface ref.
He+ 60° (110) 23
45° (poly.) 34
45° (poly.) 45
Ne+ 60° (110) 33
60° (111) 29
45° (poly.) 34
45° (poly.) 45
Ar+ 45° (poly.) 34
Desorption Cross Section (cm²)
10⁻¹³
10⁻¹⁴
10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
0.01
0.1
1
10
Ion Energy (keV)

Fig. 17

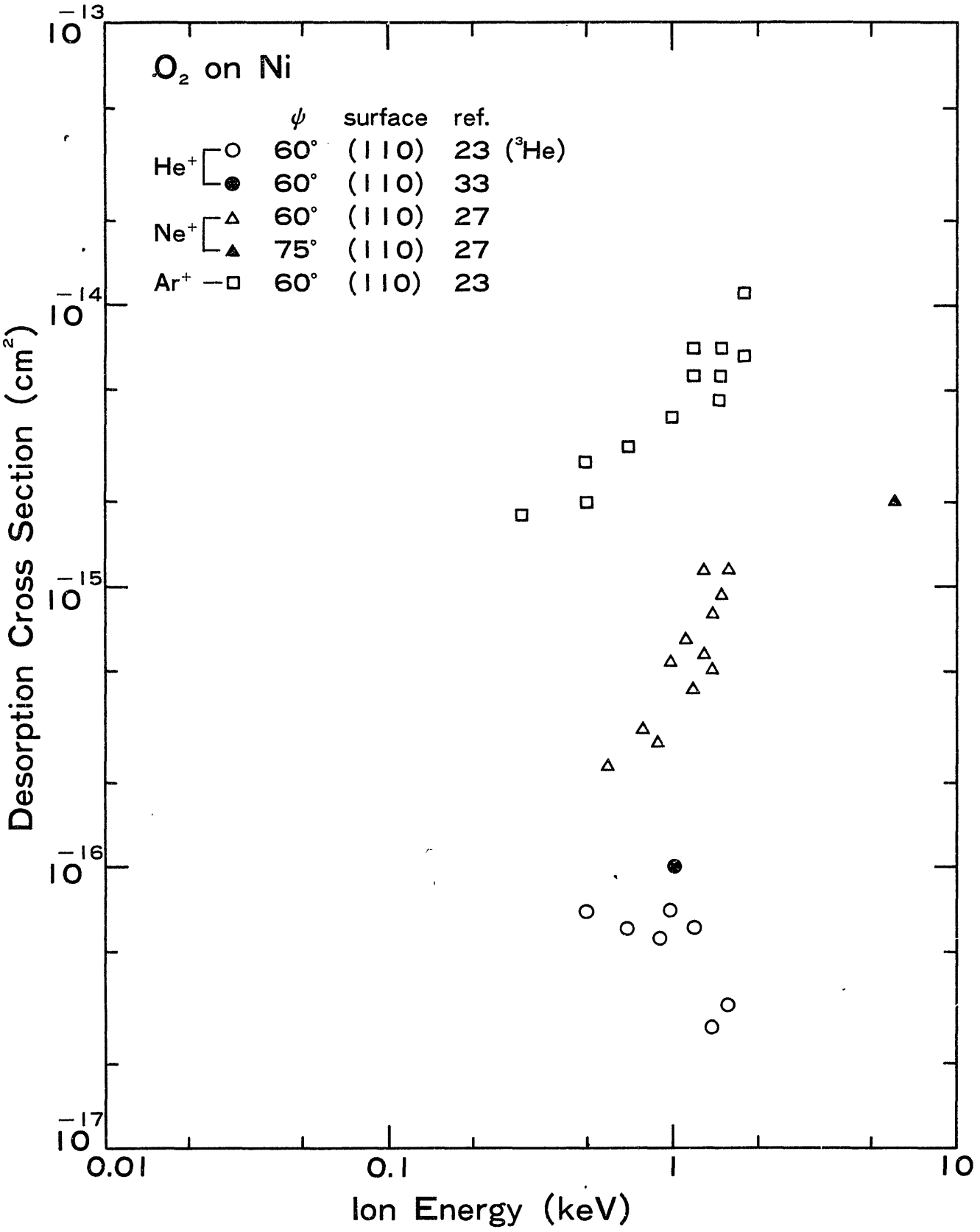
O_2 on Ni
ψ surface ref.
He+ 60° (110) 23 (3He)
60° (110) 33
Ne+ 60° (110) 27
75° (110) 27
Ar+ 60° (110) 23
Desorption Cross Section (cm2)
10-13
10-14
10-15
10-16
10-17
0.01
0.1
1
10
Ion Energy (keV)

Fig. 18

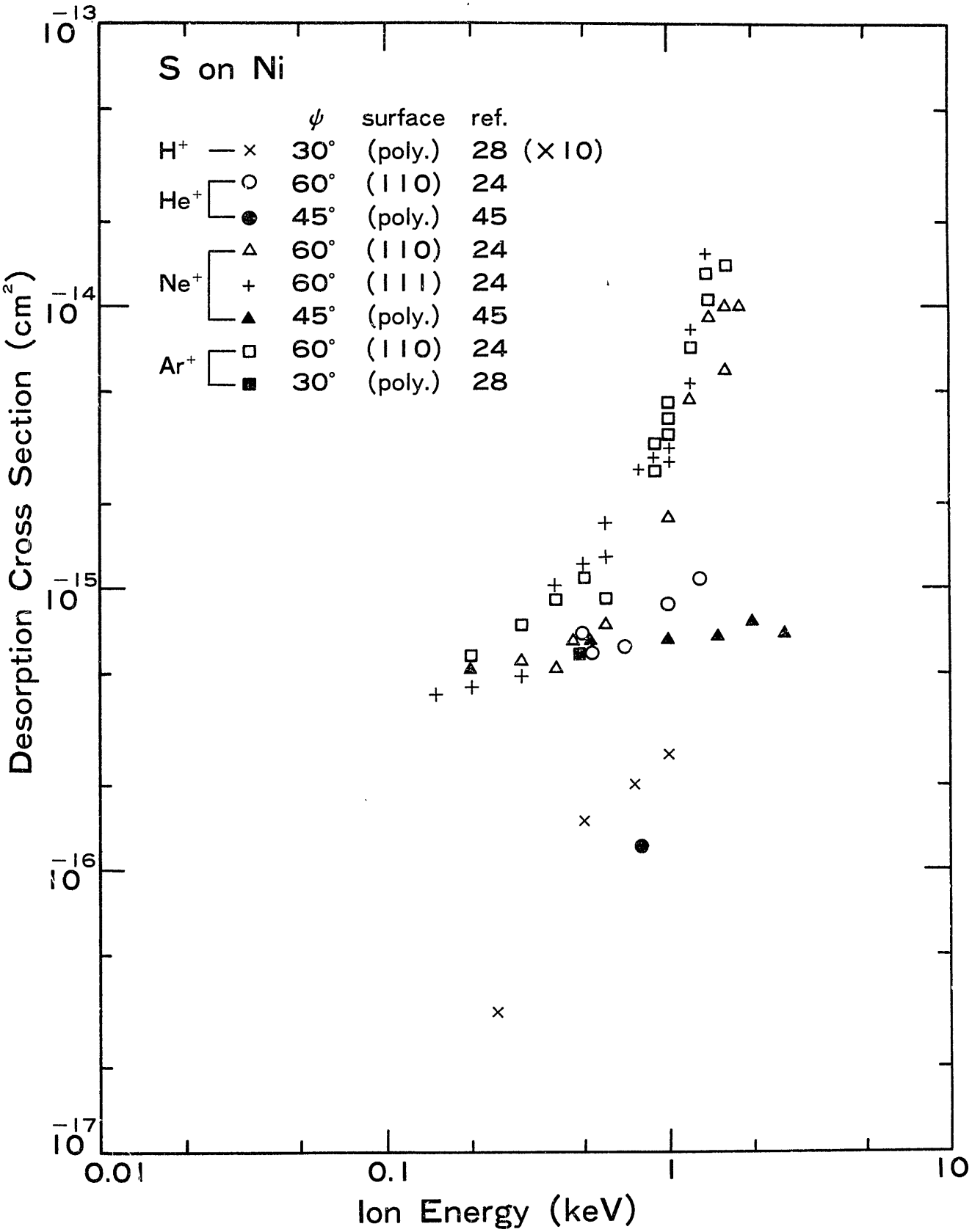
S on Ni
ψ surface ref.
H+ 30° (poly.) 28 (×10)
He+ 60° (110) 24
45° (poly.) 45
Ne+ 60° (110) 24
60° (111) 24
45° (poly.) 45
Ar+ 60° (110) 24
30° (poly.) 28
Desorption Cross Section (cm²)
Ion Energy (keV)
10⁻¹³
10⁻¹⁴
10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
0.01
0.1
1
10

Fig. 19

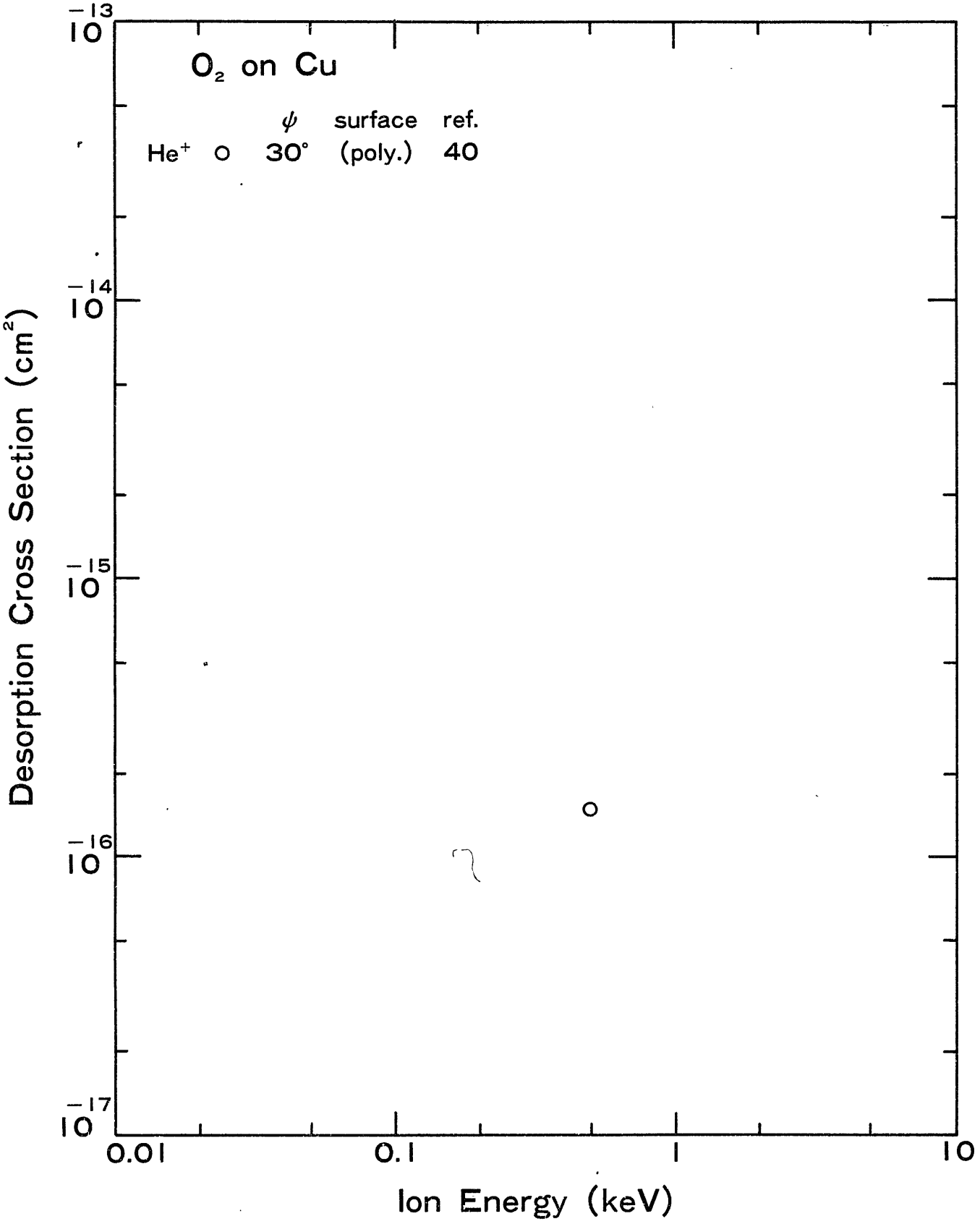
O_2 on Cu
ψ surface ref.
He^+ o 30° (poly.) 40
Desorption Cross Section (cm^2)
10^{-13}
10^{-14}
10^{-15}
10^{-16}
10^{-17}
0.01
0.1
1
10
Ion Energy (keV)

Fig. 20

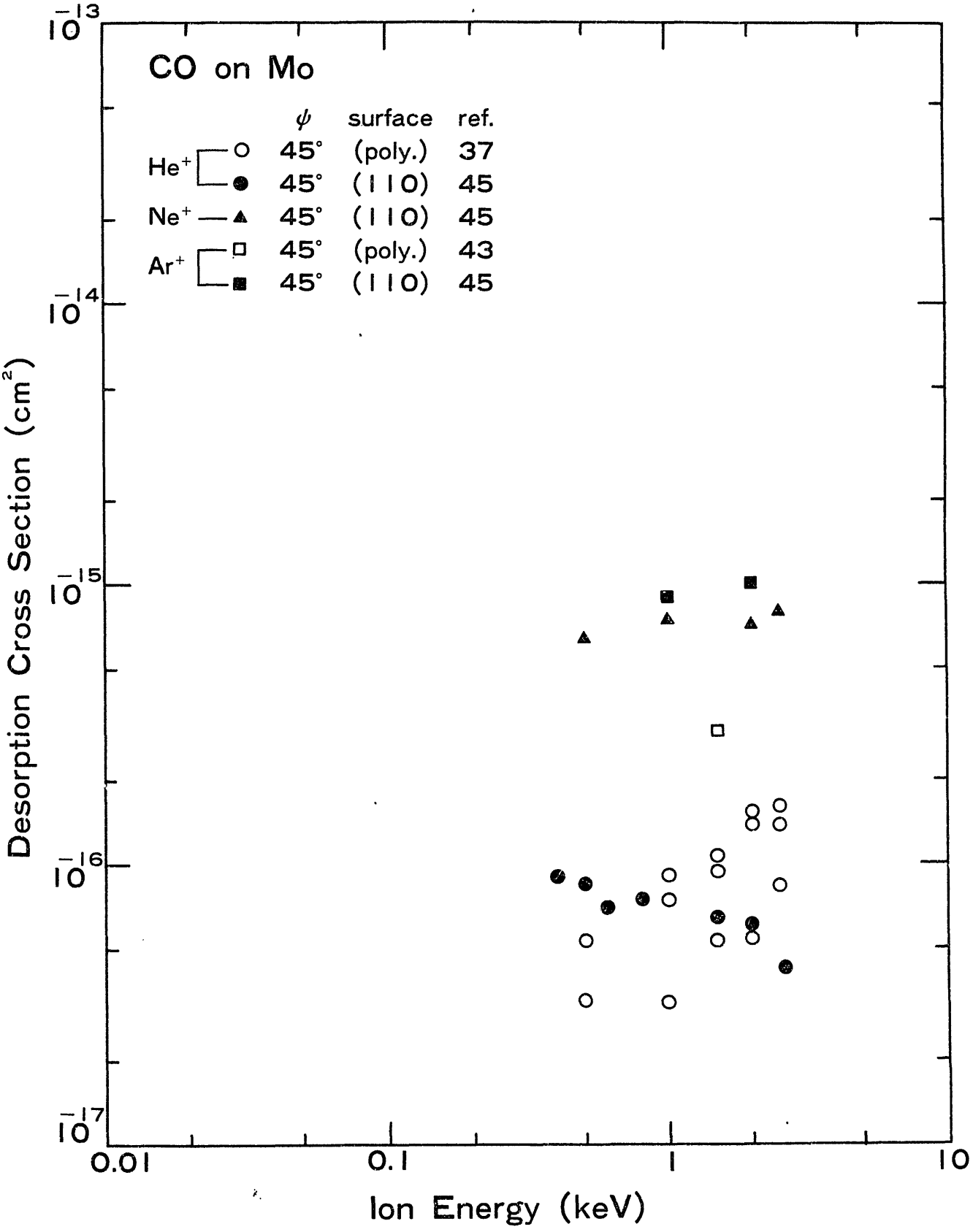
CO on Mo
ψ surface ref.
He+ 45° (poly.) 37
45° (110) 45
Ne+ 45° (110) 45
Ar+ 45° (poly.) 43
45° (110) 45
Desorption Cross Section (cm²)
10⁻¹³
10⁻¹⁴
10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
0.01
0.1
1
10
Ion Energy (keV)

Fig. 21

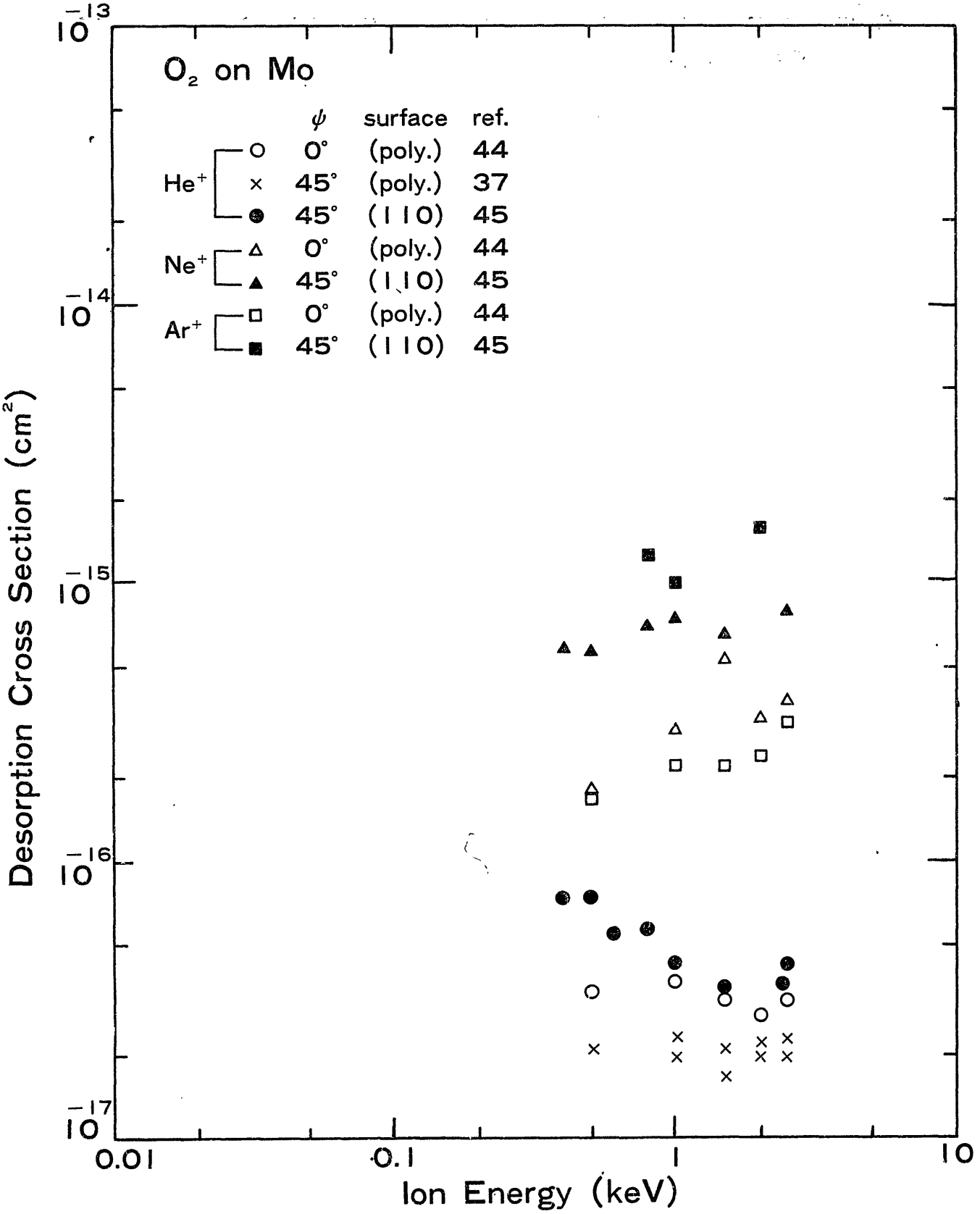
O_2 on Mo
ψ surface ref.
He+ ○ 0° (poly.) 44
× 45° (poly.) 37
● 45° (110) 45
Ne+ △ 0° (poly.) 44
▲ 45° (110) 45
Ar+ □ 0° (poly.) 44
■ 45° (110) 45
Desorption Cross Section (cm²)
10⁻¹³
10⁻¹⁴
10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
0.01
0.1
1
10
Ion Energy (keV)

Fig. 22

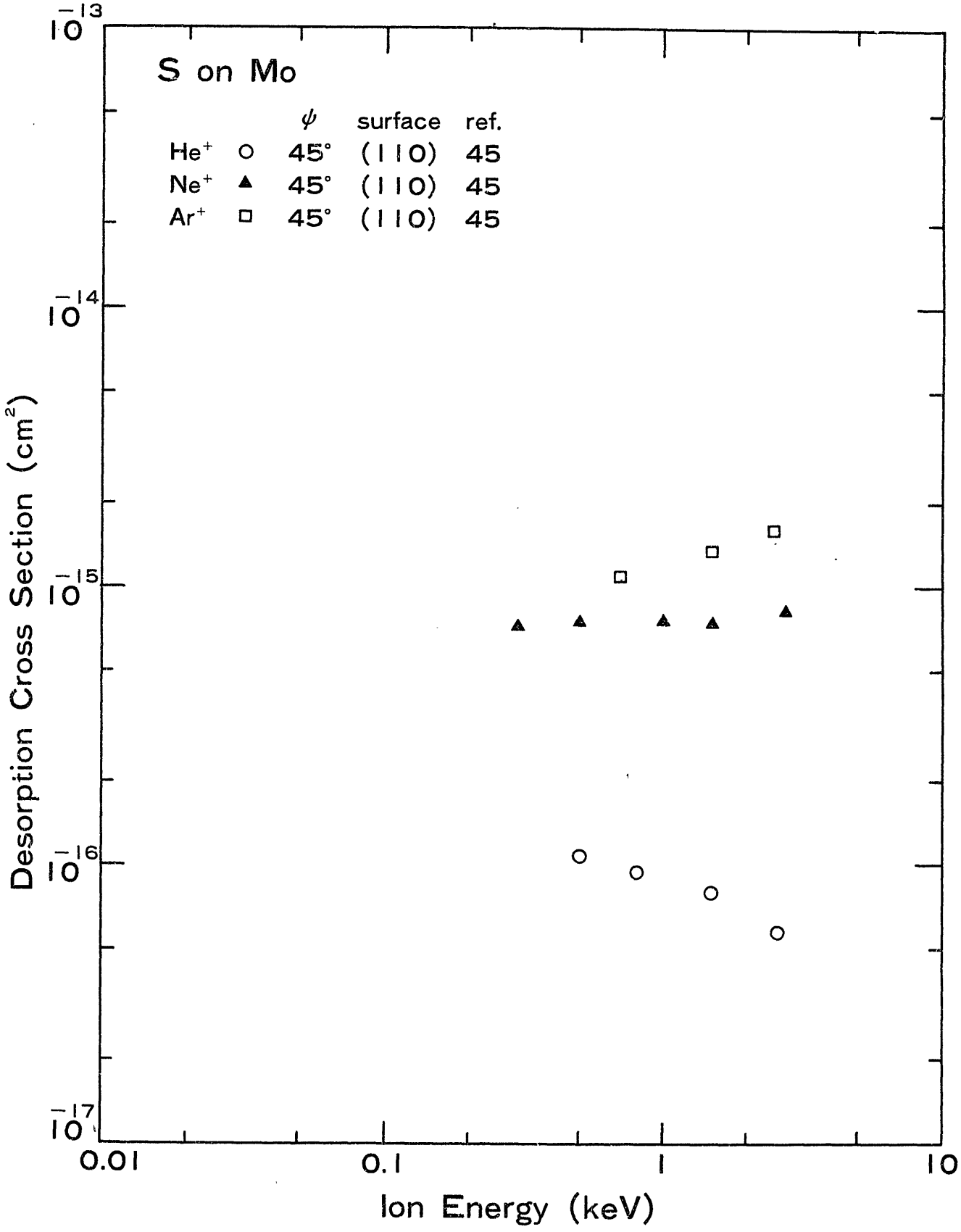
S on Mo
ψ surface ref.
He+ 45° (110) 45
Ne+ 45° (110) 45
Ar+ 45° (110) 45
10^-13
10^-14
10^-15
10^-16
10^-17
Desorption Cross Section (cm^2)
0.01
0.1
1
10
Ion Energy (keV)

Fig. 23

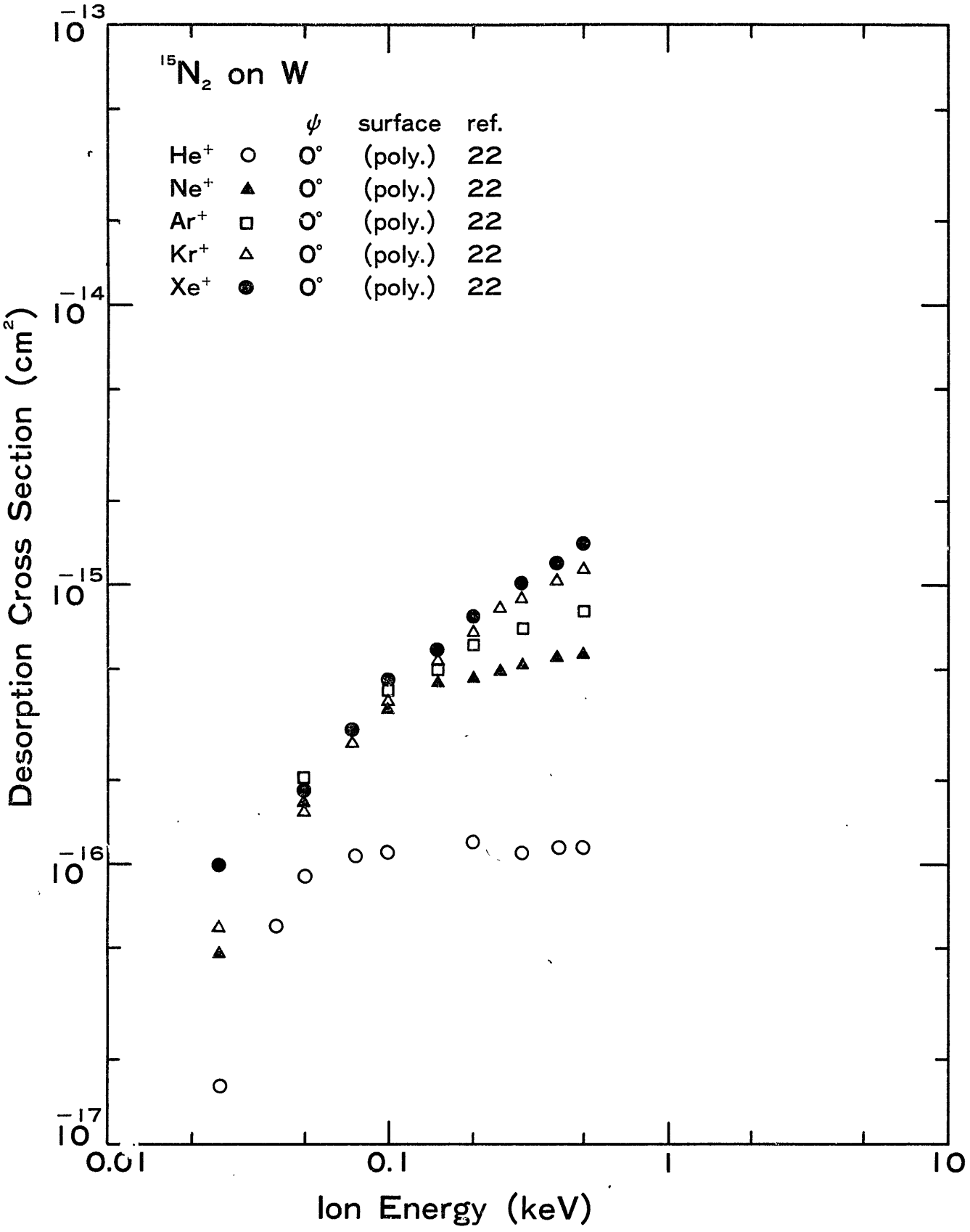
$^{15}N_2$ on W
ψ surface ref.
He+ ○ 0° (poly.) 22
Ne+ ▲ 0° (poly.) 22
Ar+ □ 0° (poly.) 22
Kr+ △ 0° (poly.) 22
Xe+ ● 0° (poly.) 22
Desorption Cross Section (cm²)
10⁻¹³
10⁻¹⁴
10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
0.01
0.1
1
10
Ion Energy (keV)

Fig. 24

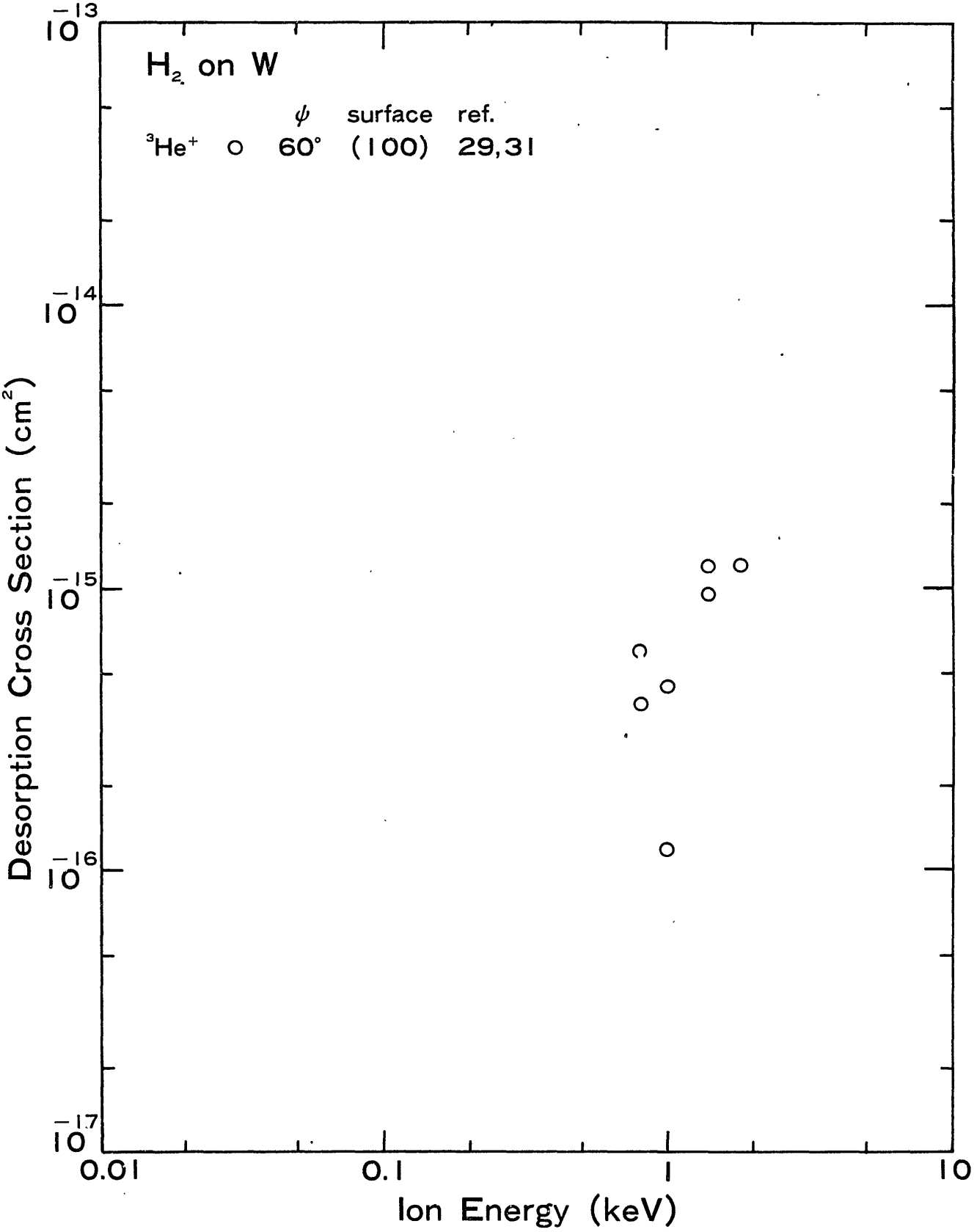
H2 on W
ψ surface ref.
3He+ ○ 60° (100) 29,31
10^-13
10^-14
10^-15
10^-16
10^-17
Desorption Cross Section (cm^2)
0.01
0.1
1
10
Ion Energy (keV)

Fig. 25

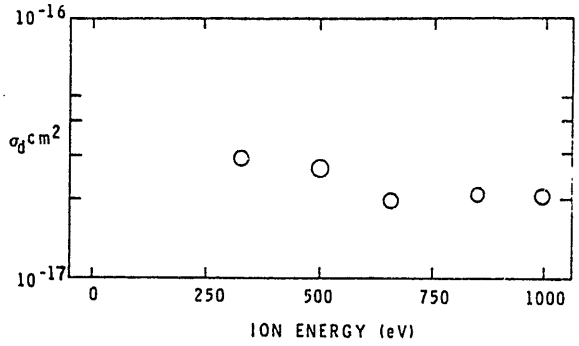
10^{-16}
$\sigma_d cm^2$
10^{-17}
0
250
500
750
1000
ION ENERGY (eV)

Fig. 26

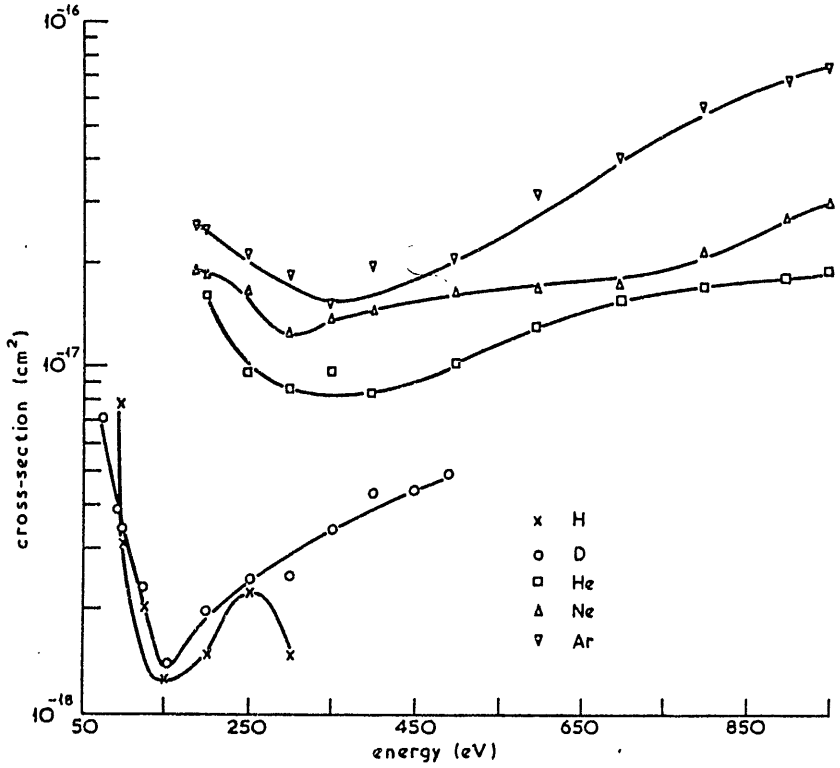
10^{-16}
10^{-17}
10^{-18}
cross-section (cm^2)
50
250
450
650
850
energy (eV)
x H
o D
□ He
Δ Ne
▽ Ar

Fig. 27

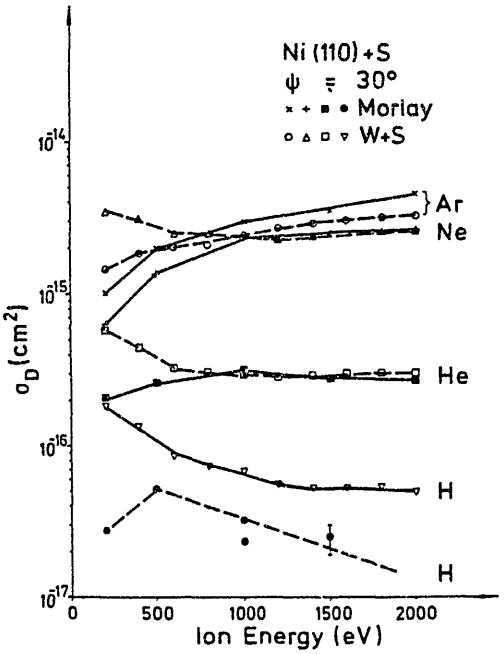
Ni (110)+S
ψ = 30°
Morlay
W+S
Ar
Ne
He
H
H
σ_D (cm^2)
10^{14}
10^{15}
10^{16}
10^{17}
0
500
1000
1500
2000
Ion Energy (eV)

Fig. 28

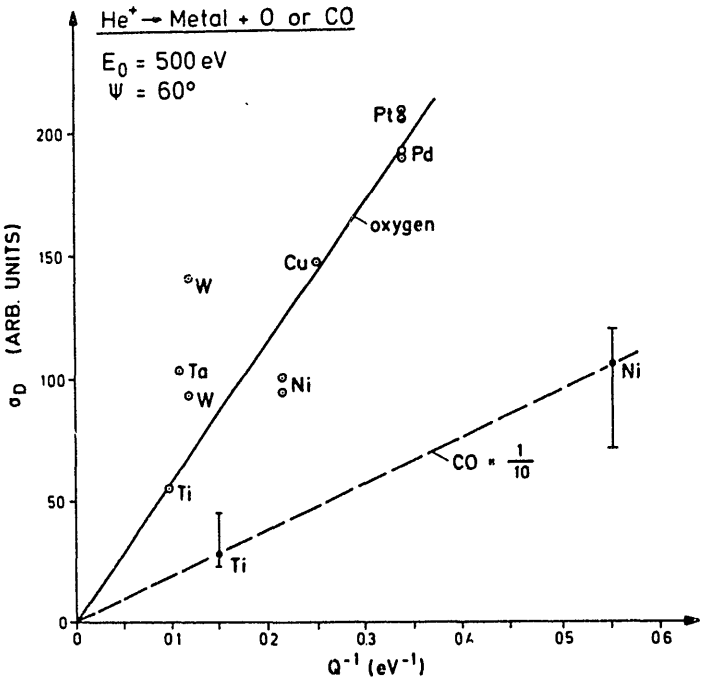
He$^+$ → Metal + O or CO
E_0 = 500 eV
Ψ = 60°
Pt8
Pd
oxygen
Cu
W
Ta
W
Ni
Ti
Ti
Ni
CO = 1/10
σ_D (ARB. UNITS)
200
150
100
50
0
0
01
02
03
04
05
06
Q^{-1} (eV^{-1})

Fig. 29

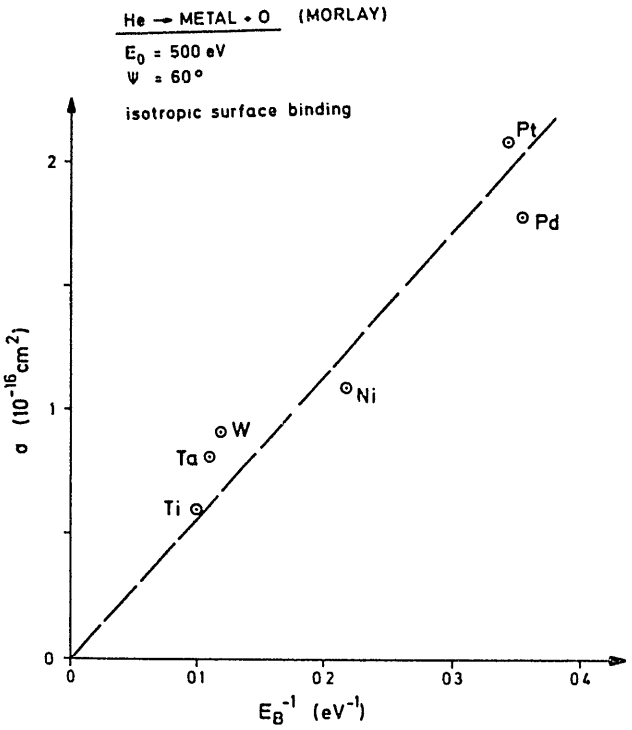
He → METAL + O (MORLAY)
E0 = 500 eV
Ψ = 60°
isotropic surface binding
Pt
Pd
Ni
W
Ta
Ti
σ (10^-16 cm^2)
E_B^-1 (eV^-1)
0
1
2
0
01
02
03
04

Fig. 30

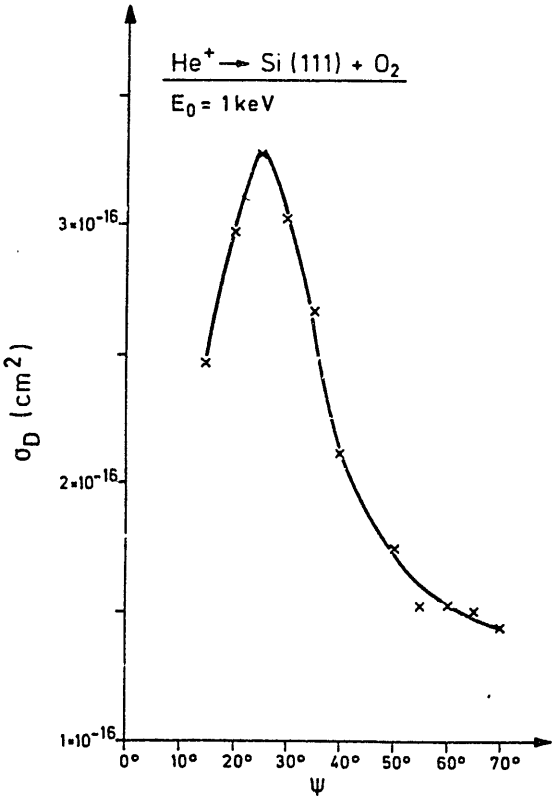
He+ → Si (111) + O2
E0 = 1 keV
σD (cm^2)
3×10^-16
2×10^-16
1×10^-16
0°
10°
20°
30°
40°
50°
60°
70°
Ψ

Fig. 31

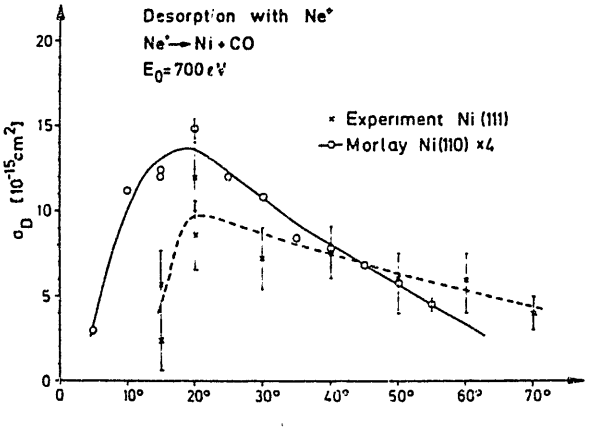
Desorption with Ne+
Ne+ → Ni + CO
E0 = 700 eV
× Experiment Ni (111)
o Morlay Ni(110) ×4
σD (10-15cm2)
0
5
10
15
20
0
10°
20°
30°
40°
50°
60°
70°

Fig. 32

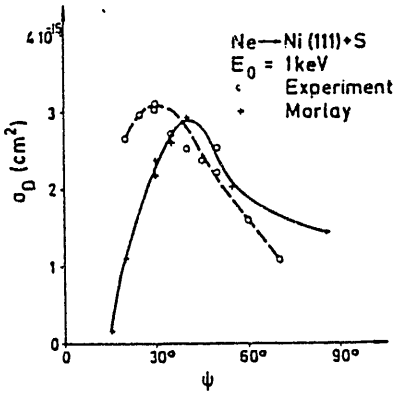
Ne—Ni (111)+S
E0 = 1keV
Experiment
Morlay
σD (cm2)
4 10-15
3
2
1
0
0
30°
60°
90°
ψ

Fig. 33

Electron Stimulated Desorptions

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Many investigations of electron stimulated desorptions(ESD) have been carried out with various experimental techniques in the past ten years [1-357]. In this report results of ESD experiments are described shortly. Detail descriptions of ESD studies are given in the cited reviews [288, 306, 345].

ESD cross sections and mechanisms

ESD cross sections are mostly measured by decays of adsorbate concentrations. When electron current densities are high enough to cause the decay by ESD, the total ESD cross sections δ are determined from the time dependence of the concentrations:

$$dN/dt = -\delta N dn/dt,$$

where N is the density of adsorbed atoms, and dn/dt the number of incoming electrons per unit time and area. Several energies of incoming electrons, between 10 eV and 3keV, were used in ESD experiments. Most of ESD cross sections were measured indirectly by the decay of signal intensities of the Auger electrons from adsorbates with low electron current densities. The values of ESD cross sections are shown in Table I. For ESD of neutral atoms the cross sections lie between 10^{-16} and $10^{-18}cm^2$, while the cross sections for the desorption of ions are between 10^{-20} and $10^{-25}cm^2$.

In the ESD mechanisms electronic excitations are mainly involved. Two models are presented to explain the ESD experimental results[288, 345]. One of the models is described as follows: TheFranck-Condon transition from the ground state of the adsorbate system to a repulsive neutral or ionic state of the complex of adsorbed and substrate atoms is induced by electron impact. The neutral or ionic particles move away from the surface. In the case of ionic desorption ions excited by the primary transition are reneutralized or recaptured by an electron tunneling from the metal substrate into the hole in the surface level. The ionic desorption probability P_+ is given by

$$P_+ = q_+/q_{ex} = \exp(-\int_{x_0}^{\infty} R(x)/v(x)dx), \qquad (1)$$

where q_+ is ionic cross section, q_{ex} primary excitation cross section, R(x) recapture probability at the distance x from the surface, and v(x) classical velocity of ions at x. This model of the recapture process is in good agreement with the experimental results of the isotope effects of ESD with Hydrogen and Deuterium adsorptions. Therefore

$$\ln(I(H^+)/I(D^+)) = -0.41 \ln(P_+(H^+)).$$

The experimental results for the isotope effects are shown in Table II. For β_2 state on W(100) and (110) surfaces values of calculated q_{ex} from the experimental value of $I(H^+)/I(D^+)$ are in very good agreement with each other.

Recently another ESD mechanism was presented by Knotek and Feibelman[211] to explain threshold for ESD of positive ions (O^+, OH^+ and F^+) from metal oxides. From the experimental positive ion yields and the electron energy loss spectrum, they suggested that the desorptions induced by the interatomic Auger

type process are dominant compared with the direct ionization process of adsorbates.

Angular distributions of ESD ions

In many investigations of angular resolved ESD the expriment by Madey et. al. [296] is a new development of the exprimental technique to show as visual patterns of the angular distributions by using a phosphorescent screen with a multi-channel-electron= multiplier-plate. The ESD-ion-angular-distribution (ESDIAD) patterns strongly depend on the adsorbate states. By assuming that the direction of the desorption agree with the direction of the potential gradient at the surface, the sites of adsorbed atoms are estimated from the symmetry and the orientation of the ESDIAD patterns.

For H_2 on W(100) surface single spot is observed in the ESDIAD patterns of H^+ ions in the direction normal to the surface. The peak width is about 21 degrees. Such single peak is observed in ESDIAD patterns of F^+ ions with about 10 degrees peak width from SF_6 on W(111) and (011) surfaces. For SF_6 on O_2 covered W-surfaces, the ESDIAD patterns, however, show the symmetries of the tungsten surfaces.

ESDIAD patterns of O^+ for O_2 on W(100) surface depend strongly on the adsorbate states. For the β_2 state of low coverage O_2 on W(100), adsorbed with 2×10^{-6} Torr. Sec at 400K, the ESDIAD pattern at 300K has four hold symmetry as shown schematically in Fig. 1a. The pattern goes away for heating

the substrate up to 700K. For the β_1 state, which is prepared by heating the substrate after the adsorption of O_2 with $40x10^{-6}$ Torr. Sec at 100K, the ESDIAD pattern shows very different behaviors from the pattern for the low coverage state(β_2 state). After the heating less than 600K the ESDIAD patterns taken at 100K show 45 degrees rotation from the orientation of the low coverage patterns as shown in Fig. 1b. For the heating more than 600K each spot in the pattern is splitted into two spots as shown in Fig. 1c. Different ESDIAD patterns for high coverage O_2 on W(100) adsorbed at high temperature above 700K are observed. The patterns taken at 400K after adsorptions of O_2 with $20x10^{-6}$Torr.sec. at 700K and $40x10^{-6}$Torr.sec. at 795K show the same symmetry and the orientation of the pattern from low coverage surface. For adsorptions with $40x10^{-6}$Torr.sec. at 705K and $120x10^{-6}$Torr.sec. at 700K, the pattern observed above is added by the splitted 40° rotating four hold pattern. For adsorption with $40x10^{-6}$Torr.sec. at 630K the pattern is nearly the same as that shown in Fig. 1c.

For O_2 on W(111) a structureless circular spot is observed in ESDIAD pattern at low oxygen exposures(about $0.25x10^{-6}$Torr. sec.). The direction of the ions is normal to the surface. At high exposurs more than $5x10^{-6}$Torr.sec. the ESDIAD pattern becomes the triangular shape. For heating after exposure of $40x10^{-6}$Torr. sec. at 100K the pattern depends on the heating temperatures above 600K. Above the temperature 600K the ESDIAD pattern becomes sharp , and no ion desorption can be observed in the direction normal to the surface. The angle of ion emission between the center of the pattern and the most intense regions of the spots is 34 ± 4 degrees. For the pattern without heating, the angle is 27 degrees.

For the adsorption of 40×10^{-6} Torr.sec. exposures at about 400K, the ESDIAD pattern disappeares after heating to about 600K, while the ESDIAD pattern for adsorption at 100K becomes sharp and intense for heating.

For adsorption of CO on W(111) ESDIAD single peak is observed in the direction normal to the surface. However, most of the observations show off-normal peaks in 5 - 20 degrees from normal direction for O^+ ions desorbed CO on metal surfaces (W, Mo, Pd, and Ru).

Energy distributions

Most of the energies of desorbed ions stimulated by electrons distribute below 15 eV. For one monolayer coverage of oxygen atoms on W(100), there is one symmetric peak at 300K and after annealing with above 950K with peak maximum at 7.5 eV and 8.0 eV respectively. The halfwidth of the respective peaks are 2.7 and 2.1 eVs. After the intermediate annealing temperature range an additional peak at 5.5 eV is measured. The peak energy depends on O_2 coverage. At 300K, the peak energy is 7.8 eV up to about 3/4 monolayer(ML) coverage and shifts to 7.5 eV for more than 3/4ML. The peak at 5.5 eV is observed between 1/2ML and 3/4ML. The maximun intensity is obtained at 0.6ML coverage. After 1100K annealing peak at 7.8 eV is shifted to 8.3 eV. These energy peak shifts are explained by adsorbate structural changes. For CO and O_2 on W(111) kinetic energy of the desorbing O^+ ions has a value of 6.3 eV after O_2 exposure and the value is shifted to 5.5 eV after CO exposure.

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Table I

Adsorbates	Substrates	Cross sections (cm^2)	E_p (eV)	Ref.
H_2 (β_2)	W(100)	6×10^{-23} for H^+	100	
(β_2)	W(110)	5×10^{-22} for H^+	100	
(κ)	CO/W(100)	8×10^{-20} for H^+	100	
	W(100)	1.5×10^{-19}	120	
CO	Mo	$3.7\text{-}16\times10^{-17}$	50-300	
		$7\text{-}14\times10^{-20}$ for C	50-300	
	W	3×10^{-18}, 8.6×10^{-20}	120	
		2×10^{-21} for β	2.5keV	
		1.55×10^{-18} for α_2	2.5keV	
	Rh(110)	10^{-22}		
	Ir	$0.8\text{-}1.7\times10^{-17}$	86	
		1×10^{-17}	2.5keV	
O_2	W	6×10^{-20} for high cover.	120	
		2×10^{-19} for low cover.	120	
		7×10^{-15} for O_2	150	
		10^{-15} for O^+	150	
	Te	3×10^{-18}	2keV	
F	W	3×10^{-19} (total)	120	
		3×10^{-20} for F^+	120	
Na	MgO	2×10^{-18}	300	
S	C(diamond)	5×10^{-20}		
Xe	Ni	1×10^{-17}		
Ba	W	6×10^{-20} (total)	200	
		4.4×10^{-22} for Ba^+	200	

Table II

State	$q_+(H^+)$ cm^2	$I(H^+)/I(D^+)$	calc. q_{ex} cm^2
β_2/W(100)	6×10^{-23}	154	1.4×10^{-17}
β_2/W(110)	5×10^{-22}	68	1.3×10^{-17}
K/CO on W(100)	8×10^{-20}	5.7	6×10^{-18}

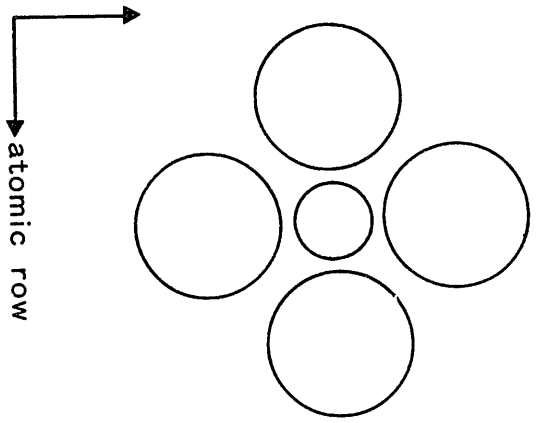
Figure captions

Fig. 1 Schematic ESDIAD patterns for $O_2/W(100)$.

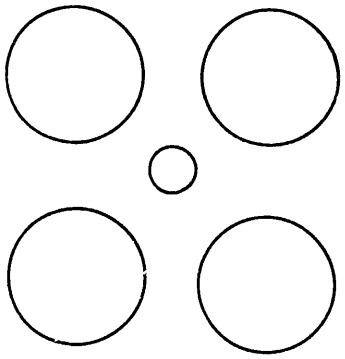
(a) low coverage β_2 state.

(b) high coverage β_1 state annealed at below 600K.

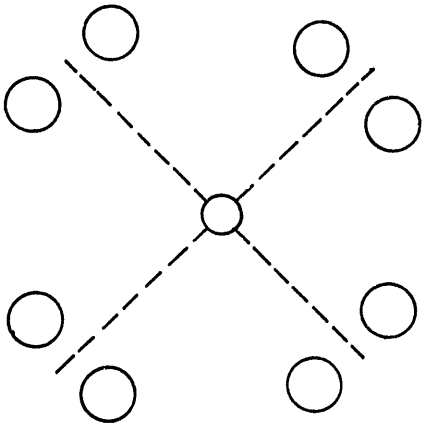
(c) high coverage β_1 state annealed at above 600K.



(a)



(b)



(c)

Fig. 1

Photodesorption

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Photodesorption is the desorption caused by incident photon flux. Thermal and quantum mechanical processes are considered in photodesorption. In the thermal mechanism photodesorption is essentially the same as themal desorption. In the quantum mechanical mechanism the adsorbed atoms are knocked off due to the direct interactions with incident photons or by photoelectrons produced by incident photons. Although only limitted data are available on photodesorption, the cross-section of photodesorption is estimated as about $10^{-(18\sim 21)}$ cm^2.

In the followings published papers related to photodesrption are listed. Papers on photodesorption on the metal surfaces are listed in the first group, and those on the semiconductor surfaces and theoretcal ones are listed in the second and the third groups, respectively.

Photo Disorption from Metal Surfaces

γ-ray region

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"Radiation induced outgassing in an aluminum system."

H. Galron: Vacuum 22 (1972) 229,
"Desorption of water vapour by gamma irradiation."

H. Dobrozembsky: J. Vac. Sci. Technol. 13 (1976) 467,
"Operating experience with an UHV system in the vicinity of a fission-reactor core."

X-ray region

S. Brumbach & M. Kaminsky: J. Appl. Phys. 40 (7)(1976) 2844,
"Gas release from Al_2O_3 and stainless steel surfaces under X-ray irradiation".

Soft X-ray region

G.E. Fisher & R.A. Mack: J. Vac. Sci. Technol.2 (1965) 123,
"Vacuum design problems of high current electron storage rings"

M. Bernardini & L. Malter: J. Vac. Sci. Technol.2 (1965)130,
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R. Franchy & D. Menzel: Proc. Intern. Vac. Congr. 7th(2) (1977 Vienna) 1209,
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R. Franchy & D. Menzel: Phys. Rev. Letters 43(12) (1979) 865
"Adsorbate core ionization as primary process in electron- and photon-stimulated desorption from metal surfaces."

R. Jaeger, J. Feldhaus, J. Haase, J. Stohr, Z. Hussain, D. Menzel & D. Norman: Phys. Rev. Letters 45(23)(1980) 1870,
"Surface extended X-ray absorption fine structure by means of Photon-stimulated ion desorption; 0 on Mo (100)."

VUV,Visible region

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Ion Induced Detrapping, Replacement and Release of Hydrogen, Deuterium and Helium Atoms.

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Particle fluxes from wall to plasma in magnetically confined fusion devices are classified by three groups: wall atoms released by erosion processes such as sputtering and blistering; surface impurity atoms, which are adsorbed on the wall, desorbed by energetic ions and neutrals; and working gas from the wall released by energetic ions and neutrals. Understanding the behavior of working gas, that is recycling, is important for estimation of tritium inventory and control of plasma density in the devices [1 - 3].

Recycling of working gas is controlled by several pocesses, i.e., bachscattering of incident ions, trapping, energetic particle-induced release of trapped atoms, thermal diffusion of trapped atoms and desorption of working gas adsorbed on the surfaces. It has recently been recognized that energetic particle-indeced release plays an important role in recycling [3] . The physical mechanism of the release process is still not fully understood, but in practice the pobability of release can be easily described by release cross section, since recent results of isotopic replacement experiments show that there is a region where the release rate of trapped atoms decreases exponentially with time, i.e., the release cross section σ is given by

$$-\frac{dn}{dt} \propto \exp\ (-J_0 \sigma t)$$

where n is areal density of retained gas and J_0 is incident flux of isotopic ions used to release the trapped gas.

The release cross sections of deuterium from stainless steels by proton bombardment reported [4-7] are shown in Fig.1. The result that the release

cross section at 300 K (curve (a)) has larger value than that at 77 K (curve (b)) indicates that the ion induced release depends on the temperature of the target [4]. Thomas [5] has measured similar temperature dependence, which was obrained by using the relation that σ is inverse saturated areal density of deuterium and not using subsequent proton bombardment for releasing denterium atoms. It has been reported by Blewer et al.[6] that the decrease of retained deuterium atoms by subsequent bombardment eith protons, which was measured by D (^{3}He, H)^{4}He nuclear reaction analysis, was fitted by a two-term inverse exponential function which is given as follows:

$$n_D = n_1 \exp(-\sigma_1 n_H) + n_2 \exp(-\sigma_2 n_H)$$

where σ_1 and σ_2 are two release cross sections, and n_1 and n_2 are the corresponding areal densities of trapped deuterium atoms at saturation. The values of σ_1 and σ_2 are shown as the curves of (c) and (d) in Fig. 1.

The release cross sections of deuterium from graphites by proton bombardment have been investigated by a few groups[8-10] as shown in Fig. 2. Underwood et al.[9] have reported the results of replacement experiments using not only the same energy for deuterons and protons but also the 10 KeV deuterons and the protons whose energies ranged from 10 to 30 KeV in order to investigate mechanisms of the ion induced release. Erents [10] has pointed out that HD production must be considered for calculation of the magnitude of release cross section, suggestion that inclusion of HD production makes the cross section be lower than the value of higher energy measurement which neglected HD production [8].

The release cross section of deuterium from molybdenum by subsequent proton bombardment has been investigated [11], using not only same energy for deuterons and protons from 0.5 to 6 keV but also the 1 keV deuterons and the protons whose energy ranged from 0.5 to 6 keV. In Fig. 3 are shown

the release cross sections apparently obtained by the initial decreae of the measured release rates, although the release curves were fitted by two release cross sections in ref.[11]. The cross section strongly depends on the pre-implanted deuterium dose, i.e., the cross section decreases with increasing deuteron dose which can be ascribed to radiation damage effect.

The experiments described above are concerned with the replacement of deuterium by hydrogen at low incident energy. There is a series of experiment of detrapping by incident ions whose energy is more than 100 KeV. The importance of investigating high energy ion induced detrapping is increasing, since high energy He ions produced by D-T burning in fusion devices impact absorbed D and T in the wall. Scherzer et al. [12] have reported the radiation-induced detrapping of deuterium in BeO by 790 KeV He^+ and by 2.2 MeV H^+. Their results show that the decrease of trapped deuterium atoms can be fitted by two-term inverse exponential function as similarly as the results of 316 SS [6] and that the value of σ_1, which dominates the initial detrapping of deuterium atoms in BeO by 790 KeV He^+ ions is almost 2 orders of magnitude larger than the value of σ_1 obtained by low energy H^+ ions in 316 SS. Roth et al.[13] have reported the release cross sections of deuterium in graphites, whose materials are pyrolytic graphite and high purity flexible graphite known as Papyex, due to bombardments with H^+, He^+ and Ne^+ ions at high incident energy. In Fig. 4 are shown the above results, indicating that the release cross section increases strongly with increasing the atomic number of the incident ions for subsequent bombardment.

The above detrapping process due to high energy bombardments is different from the replacement process of implanted gas by subsequent im;oantation of another gas whose projected range is not far-off range of the previous implantation [4 - 11], because the ranges of the H^+ and He^+ ions are far beyond the implanted deuteron Layer. Scherzer et al.[12] have suggested that the detrapping may be induced by electronic excitation rather than by nuclear

collison cascades because of the high value of detrapping coefficient and the ratio of initial detrapping yields for 790 KeV He^+ to 2.2 MeV H^+ bombardment which is similar to the ratio of electronic stopping powers.

The mechanism of the replacement of implanted gas by subsequent bombardment with isotopic ions at low energy is not fully understood. Three possible mechanism mechanisms canbe considered at least to describe experimental isotope exchange data. The first model assumes that the trapped gas atoms are activated by collision cascades caused by incoming particles and energetically ejected from the surface as a resulr of collision cascades like sputtering. The second model assumes that n atoms can be bound at each trapping site and the binding energy of traps decreases drastically when the saturation value is exceeded and weakly bound atoms escape. This model assumes ideal mixing between the newly implanted and the previously trapped gas, which means that if saturation is reached, the ratio of isotope which are released is proportional to the ratio of lacal concentration of atoms in a trapping site. The third model proposes that trapped gas atoms are expelled from sites by the action of the ion beam and migrate be bombardment-induced diffusion until they find unsaturated traps, eventually reaching the surface by a repetition of the trapping and detrapping processes.

The exponential decrease of release rate with time can be expressed by the first model as follows:

$$\frac{dn}{dt} = -J_0 \sigma n = -J_0 \sigma n_0 \exp(-\sigma J_0 t)$$

where the notations are the same as the previous formula. Braganza et al. [4] have pointed out that this expression can not explain the threshold dose required before any release process in 304 SS. They have proposed that there are two types of sites, i.e., shallow site with low binding energy and deep site with high binding energy, and that release process occurs, after deep sites are

fully saturated, assuming in the above expression n is proportional to the fraction of shallow sites which are filled. The second model can predict the threshold dose for release assumimg only one type of trapping site. Hydrogen replacement of deuterium can be expressed in this model as follows:

$$n_D = n_{sat}\exp(-\sigma n_H) \qquad \text{and } \sigma = 1/n_{sat}$$

where n_{sat} is the maximum areal density at saturation and n_D is the number of D remaining in the lattice after a replacement dose of n_H has been incident. This σ corresponds to the release cross section in the first model. Since the replacement of D in 316 SS [6] was described by a two-term inverse exponential function, Blewer et al. have proposed two kinds of trapped atoms such as am easily replaced component of trapped deuterium and a replacement resistant component, even though their midel is based on the ideal mixing. Doyle et al. [14] have reported that the replacement behavior of D in 316 SS measured by Blewer et al. is in good agreement with a ideal mixing model providing the depth dependence of its model is taken into account, assuming only two parameters, i.e., the range distribution and saturation concentration without knowledge of the number of the types of trapping sites. Underwood et al. [9] have supported the mixing model on the base of their experimental results that the release rate of deuterium atoms pre-implanted at 10 KeV is effectively ziro for subsequent bombardment with the 30 KeV protons. They have ascribed the results to very small range overlap between the 10 KeV deuterons and the 30 KeV protons, whereas the damage profile of 30 KeV protons still overlaps the range of 10 KeV deuterons to a large extent. Hotston [15] have introduced a model which assumes two different trapping sites, namely, shallow ones and deep ones, and also assunes radiation -induced diffusion due to the incoming flux. This model also describes the Blewer's results well by assuming reasonable values for the concentration of deep trapping sites, the cross sections for collisions between beam ion and trapped atoms in both shallow

sites and deep sites, respectively, and scattering function describing the scattering of the protons as they penetrate the target. Schulz et al.[16] have reported the release of implanted ^{3}He by subsequent bombardment with deuterons and vice versa at low energy. From the experimental result that the initial release is independent of whether the range of the He is larger, equal or smaller than the range of the D ions, they have concluded that the release process is not simple exchange in saturated traps, but it may be dominated by bombardment-induced diffusion in a near-surface layer. The results of Yamada et al.[11] also show that the release cross sections of the 1 keV deuterons by the proton bombardment depends weakly on proton energy compared with the cross sections of deuterons whose energies have the same as proton energies, whereas the energy of proton bombardment has influence on the amount of release of deuterium atoms preimplanted at 1 keV.

These models described above can give fairly good explanation for the experimental results of the trapping and the release of deuterium with a suitable choice of parameters, but several questions about the mechanism of release are still unresolved and it is difficult to choose one mechanism for the release process. For example, the number of types of trapping sites assumed in these models is not necessarily the same as the number of types of trapping sites obtained by the results desorption. It seems difficult to relate the number of the release cross sections directly to the number of types of trapping sites. Since the release curve is obtained as a result that the gas atoms are released after they go through a lot of traps with several activation energies, the concept of release cross section is too simple to explain the release data, even though it is useful to evaluate quantitative amount of release. A model to explain the release mechanism must take the number and the properties of trapping sites into account more exactly and then it should include the damage effect on the number of trapping sites and properties of sites. In order to understand the release mechamism, the energy and temperature effect on the release process

should be investigated intensively, where the wide range of incident energy for replacement must be employed to study not only the effect of incident energy transfer to both gas atoms and lattice atoms but also the effect of range overlap between the pre-implanted ions and the subsequent implanted ions.

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Figure Captions

Fig.1 Cross section for the release of deuterium from stainless steels by proton bombardment as a function of incident energy. The proton beams have the same energy as the deuteron beams do. (a) 304 SS target at 300 K [4]; (b) 304 SS target at 77 K [4]; (c) σ_1 of 316 SS target at -120 C [6]; (d) σ_2 of 316 SS target at -120 C [6]; (e) 304 SS target at room temperature [7]; (f) 304 SS target at 300 K [5]; (g) 304 SS target at 90 K [5].

Fig.2 Cross section for the release of deuterium from graphites by proton bombardment as a function of incident energy. Target temperature is 300 K. The proton beams have the same energy as the deuteron beams do, for the cases of (a) [8], (c) [9] and (d) [10]. In the case of (b) [9], the 10 keV deuterons implanted are released by the protons whose energy ranges from 10 to 30 keV.

Fig.3 Cross section for the release of deuterium from molybdenum by proton bombardment as a function of incident energy at room temperature [11]. The same energy is used for the deuterons and the protons for the cases (a) and (b), and the deuteron energy is fixed at 1 keV in the cases of (c) and (d). The pre-implanted deuteron dose are 1.1×10^{18} D^+/cm^2 for (a) and (c), and 2.3×10^{17} D^+/cm^2 for (b) and (d).

Fig.4 Cross section for the release of deuterium from BeO and Graphites by high energy H^+, $^3He^+$, N^+ ion bombardments [12,13]. Energy of deuteron bombardment, species of ions for subsequent bombardment, target and target temperature are written in the figure. The value of σ_1 and σ_2 are shown in the figure if the detrapping cross sections were determined by assuming a two-term inverse exponential function.

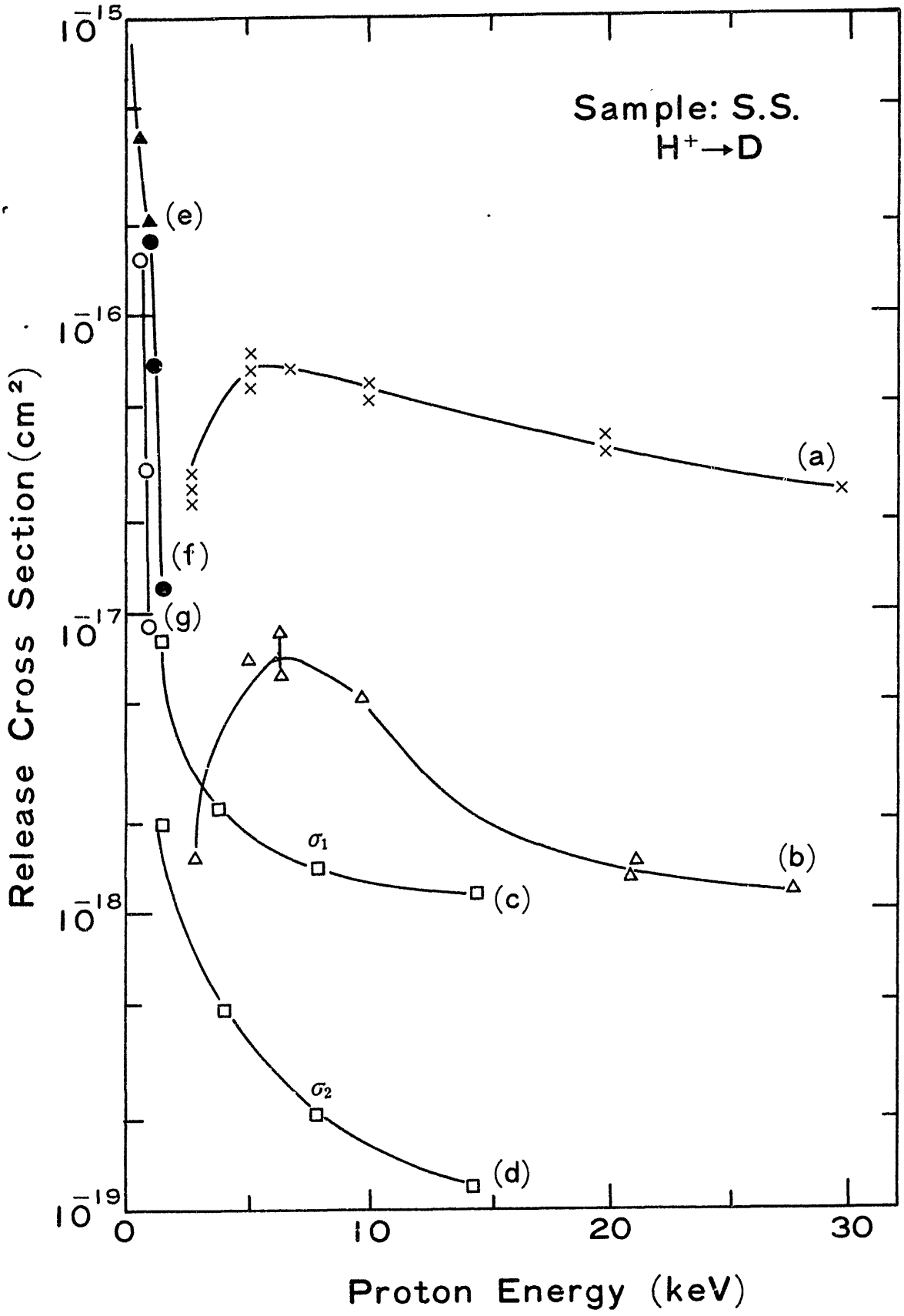
Sample: S.S.
H+→D
Release Cross Section (cm²)
Proton Energy (keV)
10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
10⁻¹⁸
10⁻¹⁹
0
10
20
30
(a)
(b)
(c)
(d)
(e)
(f)
(g)
σ1
σ2

Fig. 1

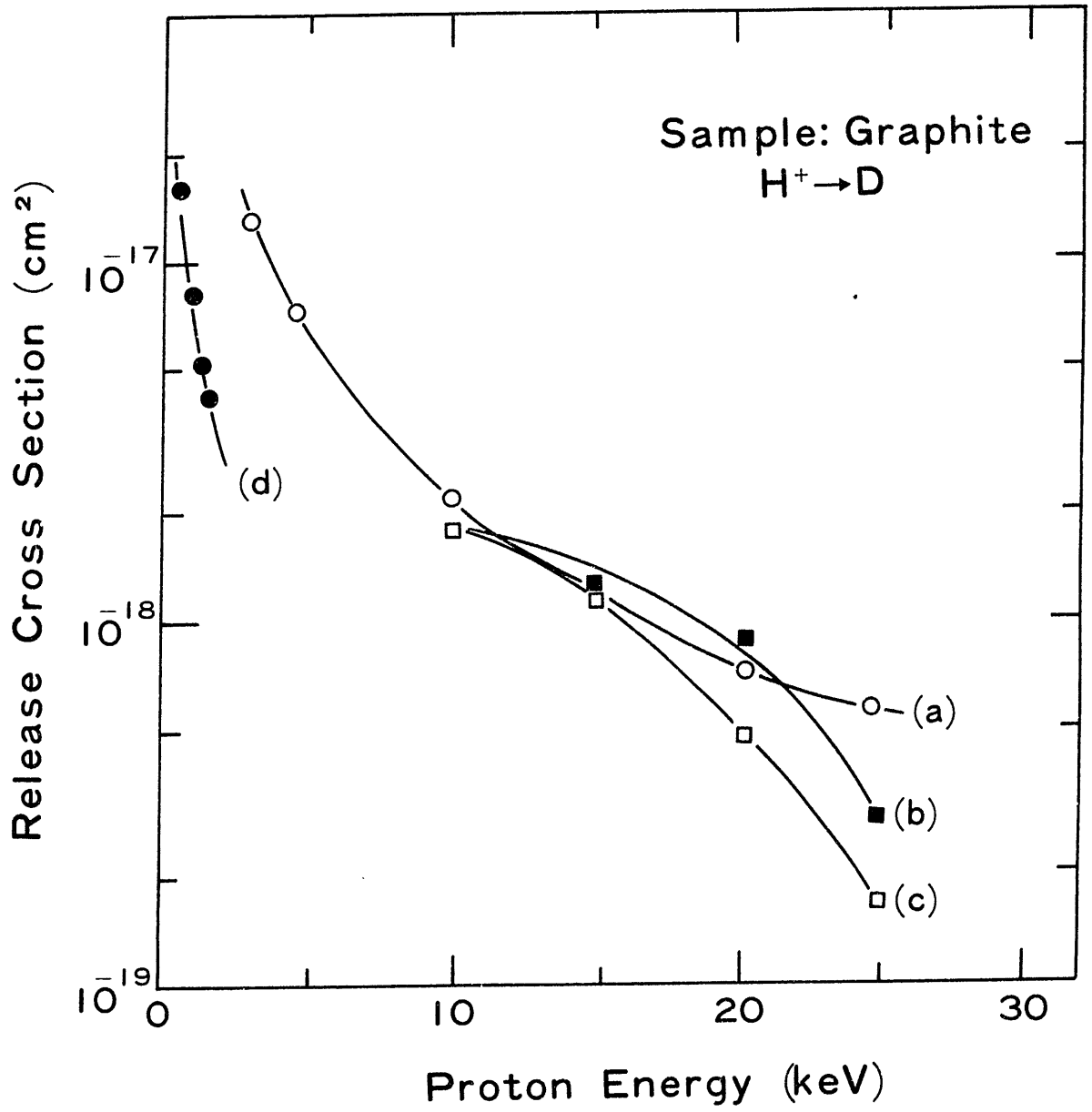
Sample: Graphite
H+→D
Release Cross Section (cm²)
10⁻¹⁷
10⁻¹⁸
10⁻¹⁹
0
10
20
30
Proton Energy (keV)
(a)
(b)
(c)
(d)

Fig. 2

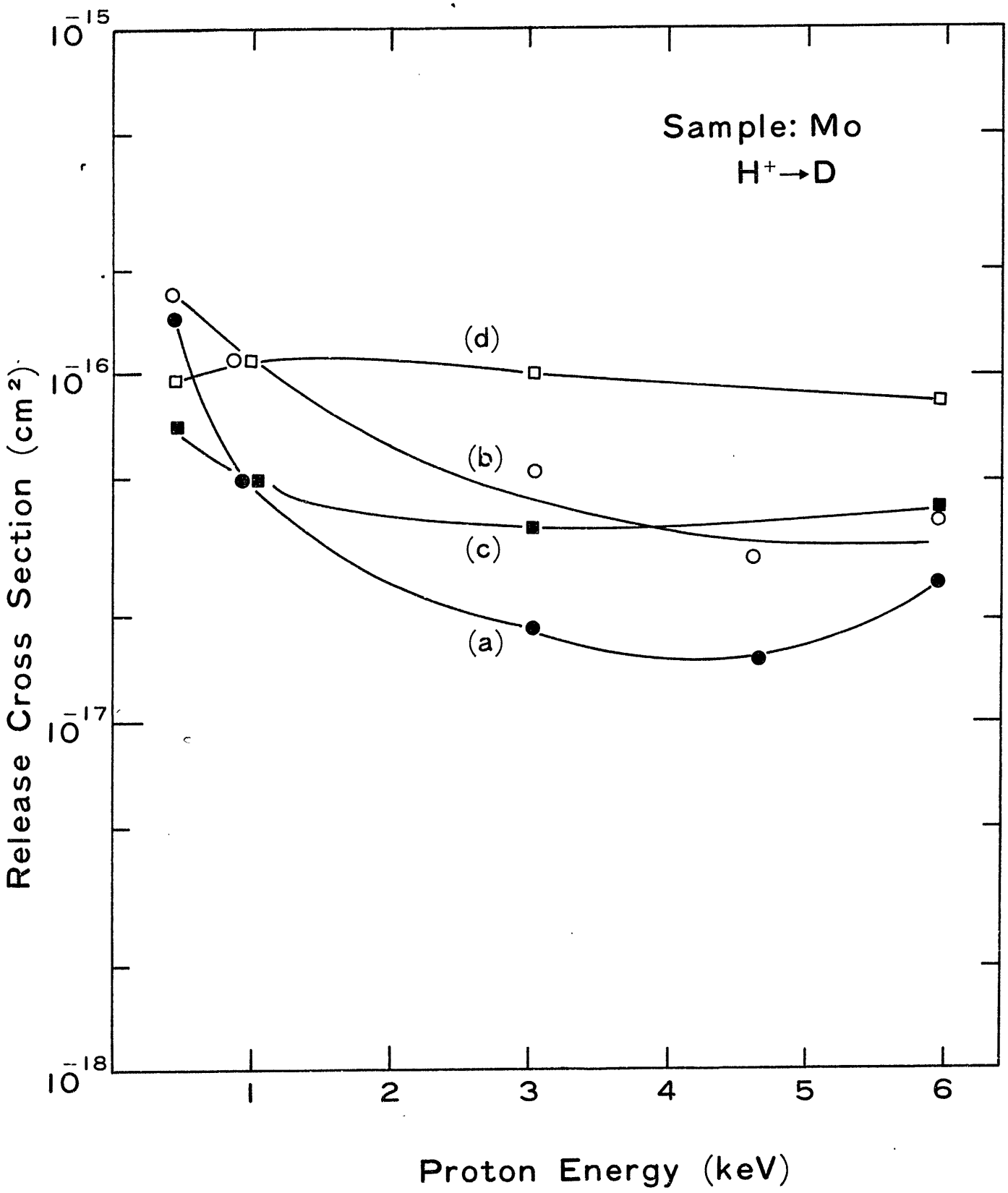
Sample: Mo
H+→D
(d)
(b)
(c)
(a)
Release Cross Section (cm²)
10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
10⁻¹⁸
1
2
3
4
5
6
Proton Energy (keV)

Fig. 3

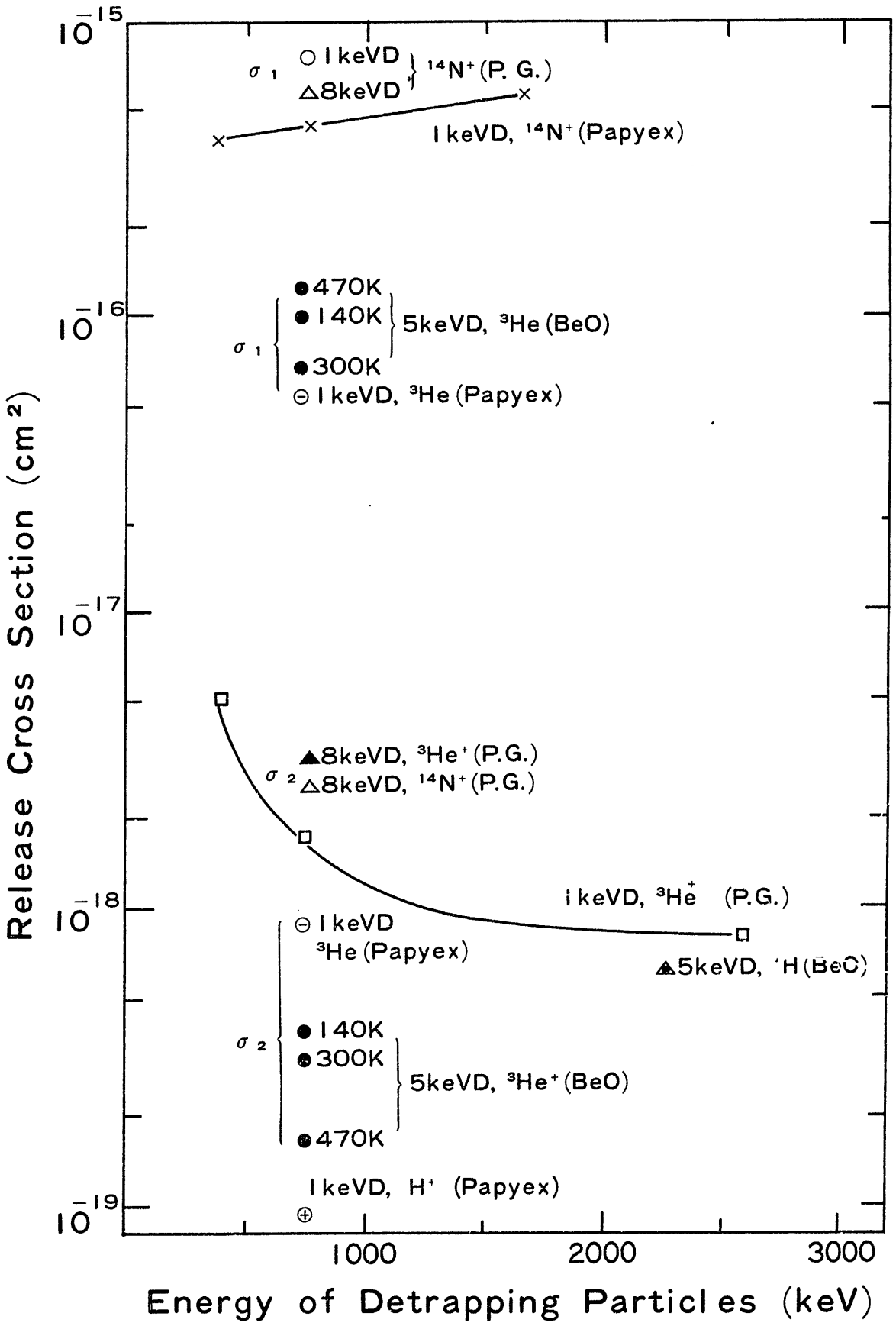
Release Cross Section (cm²)
10⁻¹⁵
10⁻¹⁶
10⁻¹⁷
10⁻¹⁸
10⁻¹⁹
σ 1
○ 1keVD
△8keVD
¹⁴N⁺ (P. G.)
1keVD, ¹⁴N⁺ (Papyex)
●470K
●140K
●300K
5keVD, ³He (BeO)
⊖ 1keVD, ³He (Papyex)
▲8keVD, ³He⁺ (P.G.)
σ 2
△8keVD, ¹⁴N⁺ (P.G.)
1keVD, ³He⁺ (P.G.)
⊖ 1keVD
³He (Papyex)
▲5keVD, ¹H (BeO)
●140K
●300K
●470K
5keVD, ³He⁺ (BeO)
1keVD, H⁺ (Papyex)
⊕
1000
2000
3000
Energy of Detrapping Particles (keV)

Fig. 4

The Chemisorption on the Carbide Surfaces

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Recently, some carbides such as SiC, TiC have received a great attention as a low-Z material in plasma device. Therefore, a lot of experiments on coating and sputtering have been carried out, but there are a little investigation of chemisorption and desorption. Here, we cite the recent literature devoted to research on the chemisorption on some carbides including SiC and TiC. Since those experiments are yet limited, at present, an individual difference of the chemisorption character on the carbide surface can not be clearly realized; however, those data suggest commom property of the carbide surface concerning to chemisorption, which is described as follows.

(1) The initial sticking probability of some gases on the carbide surface is smaller than that in the pure-metal surface. Moreover, the probability decreases rapidly with increase in uptake, an amount of adsorbates. Table 1 shows the initial sticking probability of some gases on the carbides. The values of 0.01-0.05 in Table 1 are one or two order of magnitude smaller than that of pure metals such as Ti, Ta and Mo.

Fig.1 - Fig.3 show the change in the sticking probability as a function of the uptake. The curve of the probability declines monotonically with increasing uptake. There is no constant region of the probability, which largely differs from the metal surfaces: this fact suggests that those adsorbates do not form a mobile precursor

to chemisorption on the carbide.

(2) There is a tendency that bonds of adatoms on the carbide surfaces(or the carburized surface) are weak. In fact, the binding energy of adsorbates to the carburized surface decreases depending on the degree of carburization. Consequently, the following three types of changes in chemisorption are observed, when we change the substrate from the pure metal to the metal carbide (or the carburized surface).

[2-a] In the first case, the gases that are able to be chemisorbed on the pure-metal surfaces can not be chemisorbed on the carbide surfaces (or the carburized surface). No chemisorption is found at room temperature in the following systems, atlthough it occurs on the pure metals under the same conditions.

System	Reference
H_2 / TaC (poly.)	27
H_2 and N_2/ TiC (100)	23
N ions / W_2C (poly.)	41
H_2 / W_2C (poly.)	41
H_2 / Carburized W (100)	42

[2-b] In the second case, the molecules are chemisorbed undissociatedly on the carbide surfaces (or the carburized surface), although the same molecules are dissociated on the same pure-metal surface.

System	Reference
CO/ Carburized W (100)	42
H_2O/ TiC (100)	23

[2-c] In the last case, without an apparent drastic change described above, only the binding energy of adsorbates becomes small as compared with that of the pure metal. Fig.4 shows the flash desorption spectra of CO from the three kinds of Ni surfaces: clean surface, carbide surface and graphitized surface. The desorption peak moves toward lower temperatures, when we change from the clean Ni surface to the carbide surface. The similar tendency is also observed in the case of H_2O in Fig.5.

Because the chemisorption character both on the TiC (100) surface and the carburized W surface resemble those of relatively inactive metals such as Pt, those meterials are now widely examined as a catalyst (see reference).

(3) The surface chemical composition of the carbide is apt to change by various surface treatments, because the carbides are binary compounds that have a lot of carbon defects. For example, heatings of higher than 1500°C remove preferentially the Si atoms from the SiC surface, which results in the formation of graphite layer on SiC surface. On the other hand, TiC is relatively stable against the heatings up to 1600°C under ultra hegh vacuum condition. However, oxygen atoms chemisorbed on the TiC surface are desorbed in the form of CO or CO_2 molecules at high temperature, which removes the carbon atoms from the surface. Similar phenomenon is also reported on the system of hydrogen chemisorbed on the carburized W. The desorption of the hydrogen atoms results in the decreasing in carbon content.

Some chemisorbed-oxygen atoms penetrate easily into the carbide lattice at ∿1000°C, forming a trinary compound of TiC_xO_{1-x},

since TiO has a large solubility in the TiC crystal. Those changes in surface chemical composition may affect the chemisorption character, but it is not yet clearly understood.

(4) The chemisorption character strongly depends on the carbon content of the topmost layer. With increase in the degree of carburization, an initial sticking probability of N_2, H_2 and CH_4 decreases on the Mo (100) surface. The chemisorption character of TiC single crystal differs from one crystallographic face to the other face. The (100) surface composed of nearly stoichiometric composition is relatiely inactive for the reaction with residual gases; on the contrary. the (111) surface mainly consisting of Ti atoms is very active similar to the active metals such as Ti and W. Hence, the surface property of the carbides is not due to the carbide itself, but to the surface atomic arrangement.

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Table I The sticking probability of some gases on the carbides.

System	Initial sticking probability	Reference
O_2 / SiC (polycry.)	0.01	3
H_2 / TaC (polycry.)	0.04	27
CO / TaC (polycry.)	0.05	26
O_2 / TiC (100)	0.02	23
H_2 / Carburized Mo (100)	0.02	41

Figure Captions

Fig.1 The pressure dependence of the sticking probability of hydrogen on TaC at 295K. ○ = 1.5 x 10^{-8}Torr, △ = 2.8 x 10^{-8}Torr, □ = 3.9 x 10^{-8}Torr.

Fig.2 The pressure dependence of the sticking probability of carbon monoxide (β phase). ● = 5 x 10^{-8}Torr, ■ = 7.5 x 10^{-8}Torr, □ = 1 x 10^{-7}Torr, △ = 2.5 x 10^{-7}Torr, ○ = 5 x 10^{-7}Torr.

Fig.3 The sticking probability of oxygen as a function of the normalized oxygen peak intensity.

Fig.4 The flash desorption spectra of CO from clean Ni (110), carbide and graphitized surfaces.

Fig.5 The flash desorption spectra of water from clean Ni (110), carbide and graphitized surfaces.

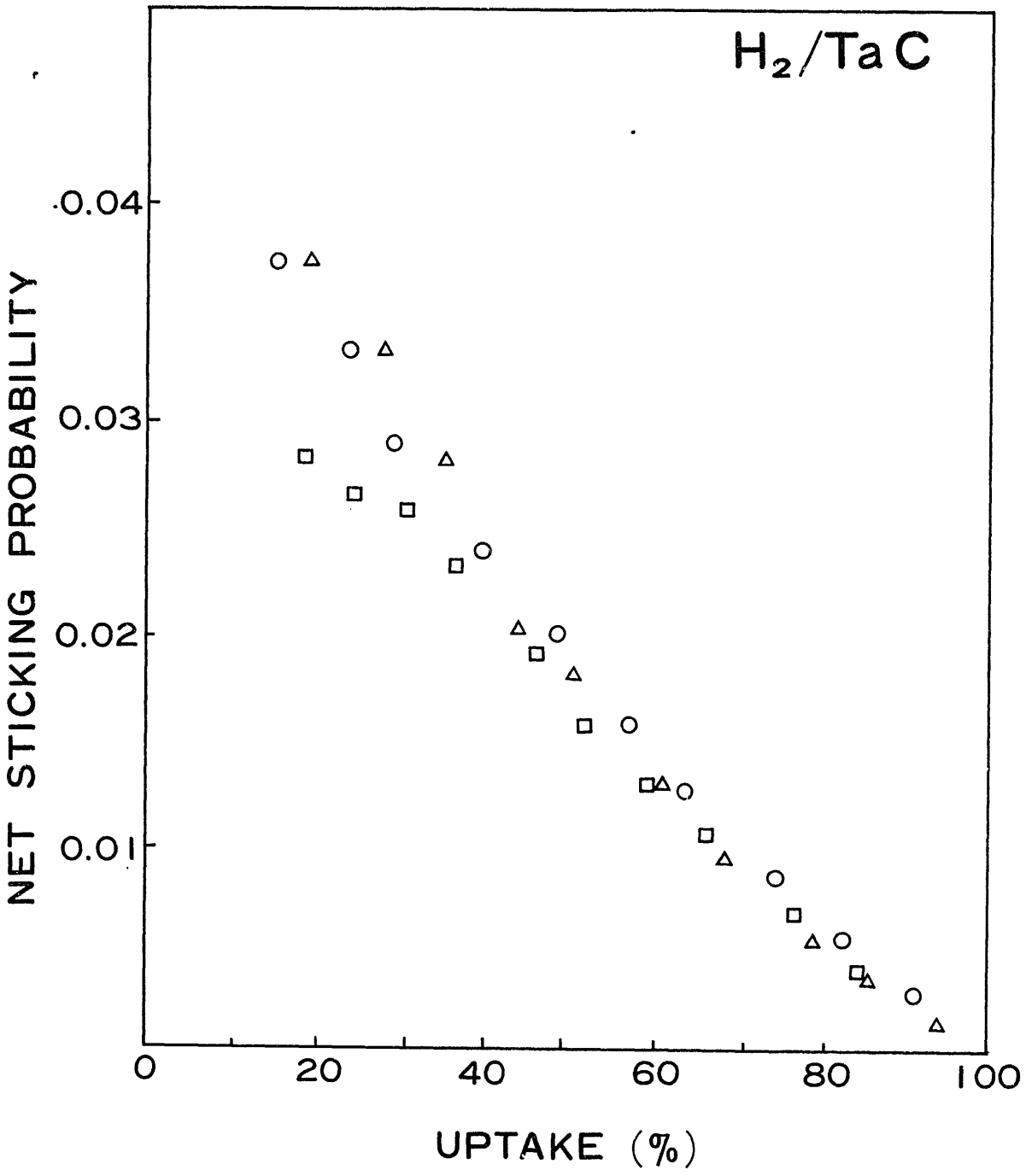
H2/TaC
NET STICKING PROBABILITY
0.04
0.03
0.02
0.01
0
20
40
60
80
100
UPTAKE (%)

Fig. 1

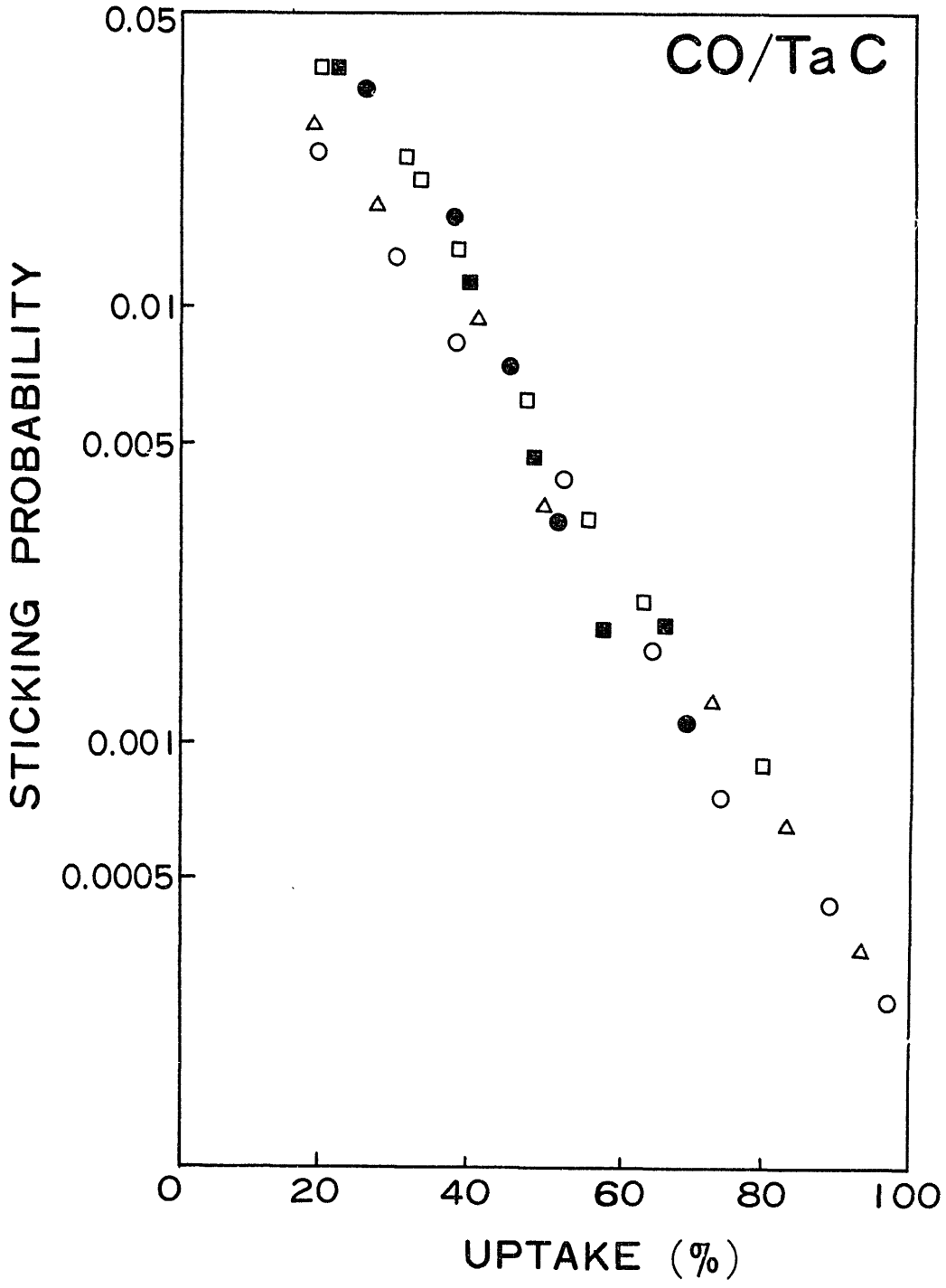
CO/Ta C
STICKING PROBABILITY
0.05
0.01
0.005
0.001
0.0005
0
20
40
60
80
100
UPTAKE (%)

Fig. 2

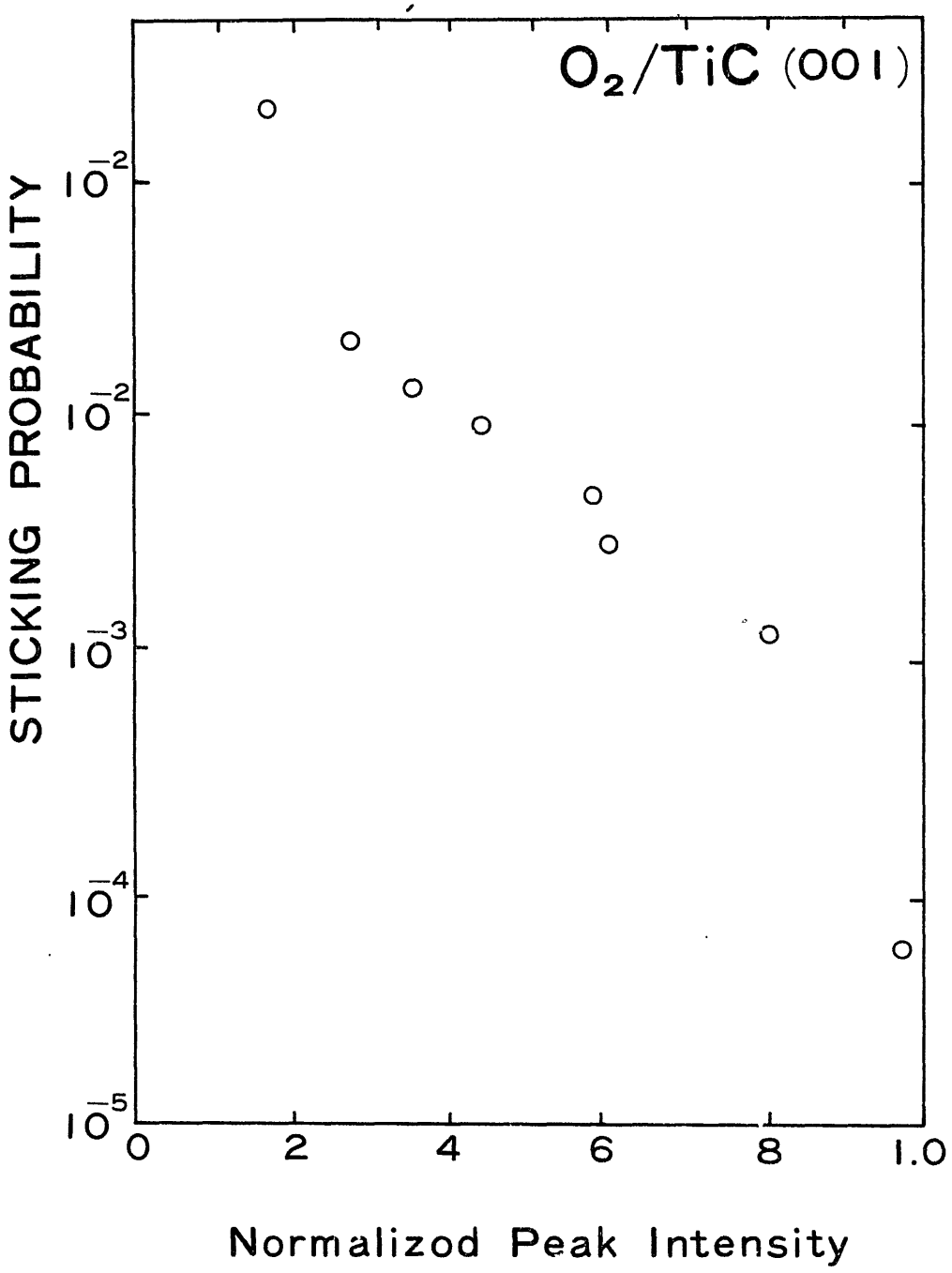
O_2/TiC (001)
STICKING PROBABILITY
10^{-2}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
0
2
4
6
8
1.0
Normalizod Peak Intensity

Fig. 3

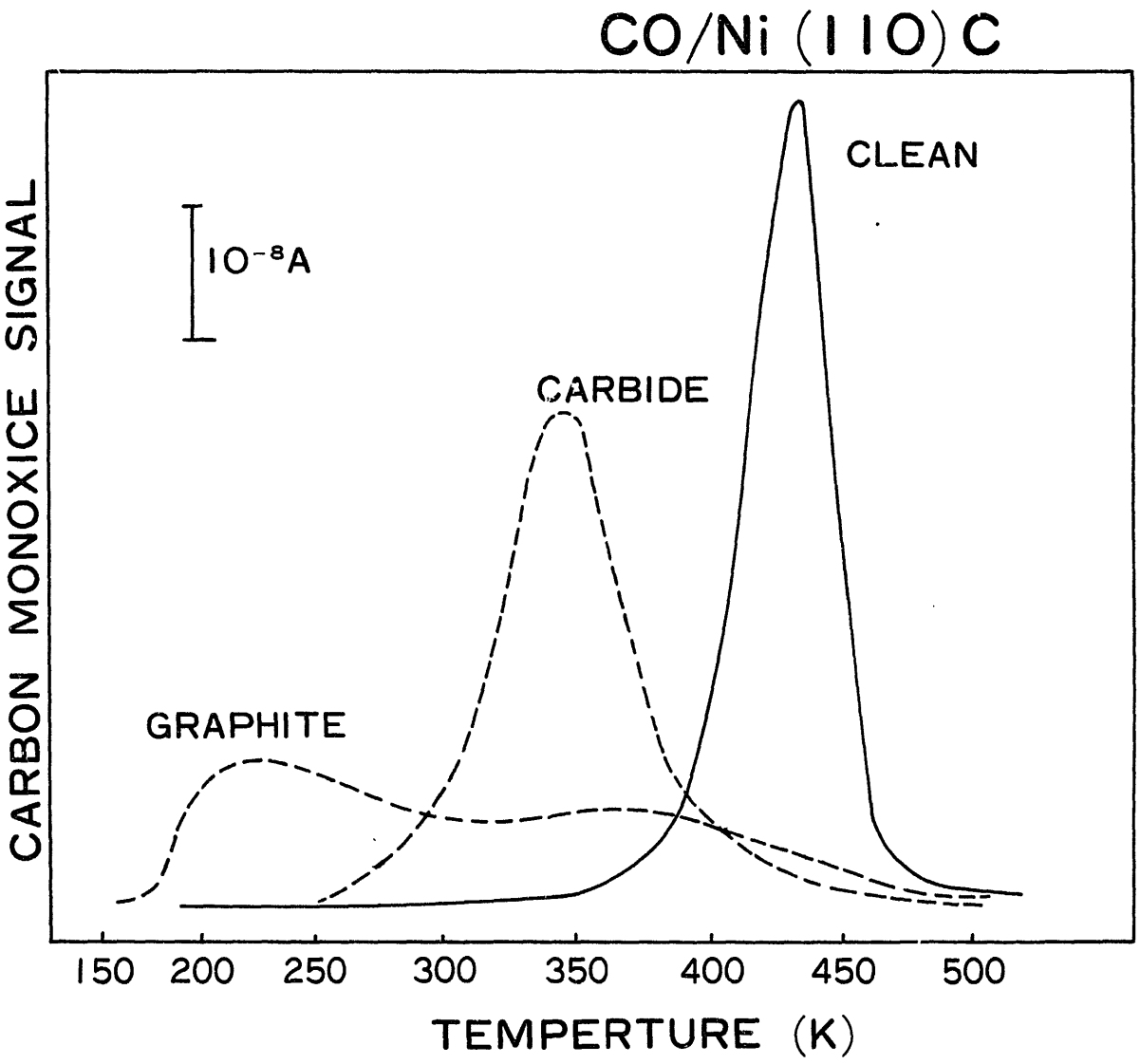
CO/Ni (1 1 0) C
CLEAN
10⁻⁸A
CARBIDE
GRAPHITE
CARBON MONOXICE SIGNAL
150 200 250 300 350 400 450 500
TEMPERTURE (K)

Fig. 4

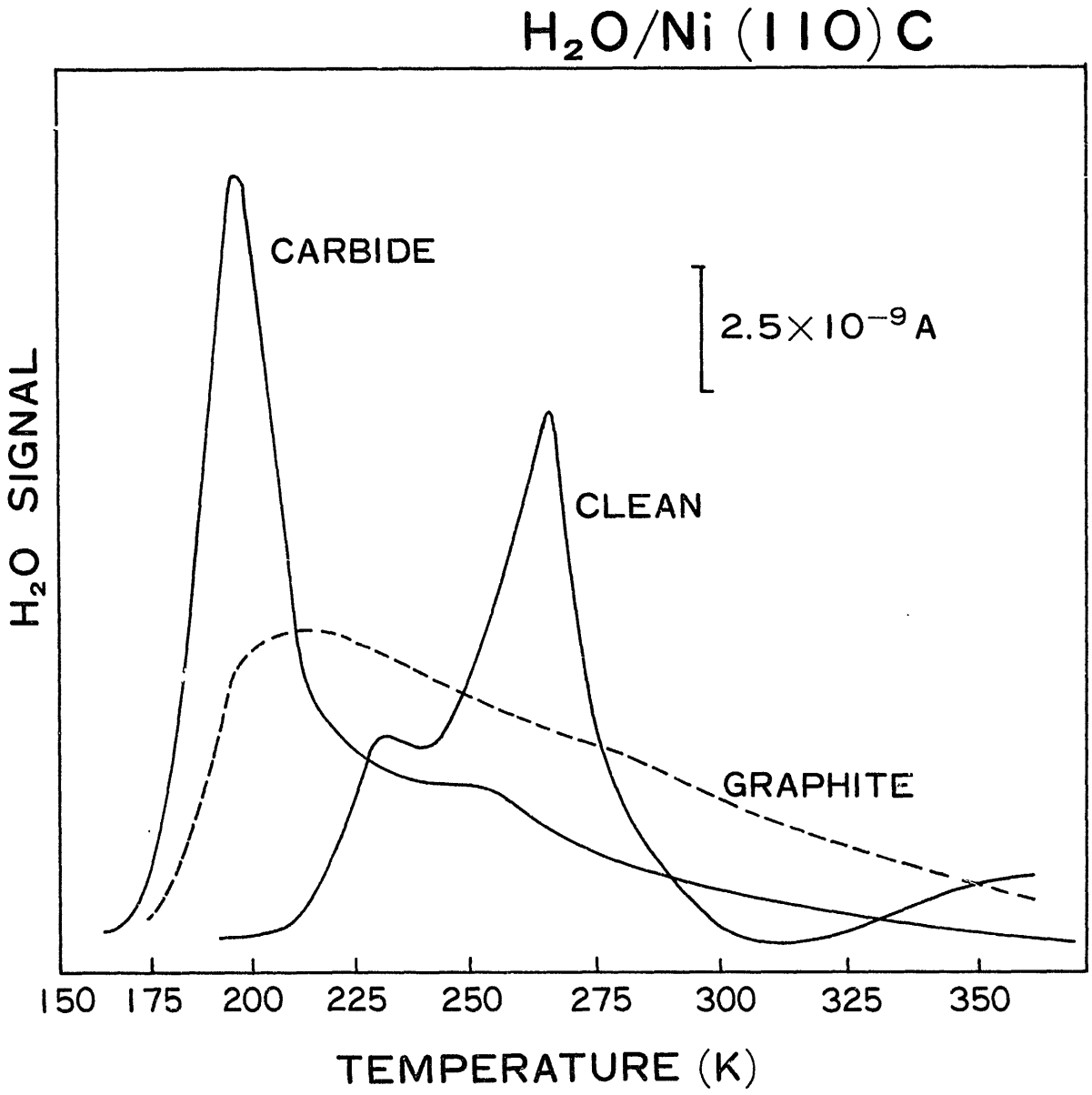
H2O/Ni (110) C
CARBIDE
2.5×10⁻⁹ A
CLEAN
GRAPHITE
H2O SIGNAL
150 175 200 225 250 275 300 325 350
TEMPERATURE (K)

Fig. 5

Present Status and Problems in the Theory of Desorption

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§1 Introduction

Although considerable progress has been made recently for their theoretical study, many of the fundamental features of the desorption phenomena are not well understood as yet. Roughly speaking, the equilibrium desorption rate of an atom from the surface with the temperature T is expressed as

$$\kappa \sim s\nu\exp(-Q/kT). \qquad (1)$$

In the above Q, ν are respectively the activation barrier height of the desorption and the number of the trial per unit time to surmount the barrier. The factor s is equal to the sticking probability for the simple one-dimensional model.[42] The quantities Q and ν are determined by the Born-Oppenheimer adiabatic potential energy surface, which can be in principle obtained by first-principle electronic structure calculations. Such calculations, however, are quite laborious and require huge computation time. Hence calculations of the adatom/surface potential energy surface have been so far performed only for simple model cluster systems.

The prefactor s is determined not only by the structure of the potential energy surface, but also by the gas-surface stochastic

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interaction. Determination of s as well as many other features of desorption phenomena which are beyond the scope of absolute rate theory are the targets of recent theoretical works. The problems investigated include

1) the transient behaviour of desorption,
2) the angle and the energy distribution of the desorbed particles,
3) the influence of macroscopic and/or microscopic properties of the substrate such as phase transition,
4) deviations from the simple Frenkel-Arrhenius type relation (1),
5) effect of non-adiabatic process on the desorption rate, such as electron-hole excitations in the substrate.

A wide variety of theoretical approaches have been reported in literatures. In the following we describe briefly the characteristics, main results and mutual relations of dominant theoretical approaches.

§2 Various theoretical approaches for desorption

2-1) Classical theory based on the thermal equilibrium hypothesis [ref.1)∿6)]

In this approach the behavior of each coordinate of motion is statistically determined by the thermal equilibrium hypothesis. Therefore the real stochastic behavior of the adparticle can not be taken into account. However the structural detail of the potential energy surface can be incorporated with this approach. A lot of works have been performed beginning from the simple linear truncated harmonic oscillator model to a realistic three demensional

system. Recently, the delay time of the molecular beam interacting with surface,[6] the effect of adsorbate lateral motion[4] and the angular distribution of the desorbed gases[1] have been investigated, as well as the deviations from the simple Frenkel Arrhenius type relation (1).

2-2) Stochastic classical trajectory approach [ref.7)∿16)]

This approach is powerful for the study of the general dynamic interactions between an adatom and a harmonic lattice system. Stochastic trajectories are obtained by the direct numerical integration of the generalised Langevin equation(GLE):

$$\ddot{X}(t) = F_R(X(t), Y(t), Z^\circ) \tag{2}$$

$$\ddot{Y}(t) = F_P(X(t), Y(t), Z^\circ) - (\Omega_P^2 - \Lambda(o))Y(t) - \Lambda(t)Y(o) - \int_{-\infty}^{t} \Lambda(t-t')Y(t')dt' + R(t) \tag{3}$$

In the above X(t); Y(t), Z^o are the coordinate of gas atom, surface atoms in the primary zone and the equilibrium positions of the atoms in the secondary zone. F_R, F_P are the forces due to adatom/surface interaction, and Ω_P^2 is the effective dynamical matrix of the primary zone. The correlation function of the stochastic force R(t) is related with the friction Kernel $\Lambda(t)$ by the following,

$$\kappa T\, \Lambda(t) = \langle R(t)R^+(o)\rangle \tag{4}$$

The key point of the trajectory calculation is that not the stochastic force R(t), but the impulse $I_n = \int_{t_{n-1}}^{t_n} R(t)\,dt$ during the integration mesh appears in the numerical integration. The random variable $\{I_n\}$ are generated by the Gaussian distribution which are determined by eq.(4). The diagonal elements of $\Lambda(t)$ are obtained by the surface phonon spectrum.

Based on this approach inelastic atom-surface scattering, sticking, desorption and the mean residence time have been investigated.[14] Typical stochastic trajectory by Shugard et al[14] are shown in Fig.2.

2-3) Approach based on Fokker-Planck equation [ref.17)∿24)]

The time evolution of the adatom/surface system is described by the distribution function F(q,p,t) of the point along the reaction path. The distribution function is determined by the Fokker-Planck equation (FPE),

$$\frac{\partial f}{\partial t} + P\frac{\partial f}{\partial q} + F(q)\frac{\partial f}{\partial p} = \eta\frac{\partial}{\partial p}(pf + MkT\frac{\partial f}{\partial p}), \qquad (5)$$

where M and F(q) are the mass and the force along the reaction path, respectively. The friction constant η is given by the correlation function of the random force, $\hat{F}$, due to the microscopic electron or spin density fluctuations of the surface,

$$M\eta \sim \mathrm{Re}\frac{1}{k_B T}\int_0^\infty d\tau \langle \hat{F}\hat{F}_0(\tau)\rangle. \qquad (6)$$

The relations (5), (6) are valid for the case that the correlation time is much shorter than the oscillation period of the adatom.

It should be noted that in the limit of very small correlation time of R(t), the GLE system (2)∿(4) is described by the similar FPE as eq.(5). It is seen by the relation (6) that η is essentially determined by the physical properties of the solid, such as the dielectric constant or spin susceptibility. Therefore anomalous enhancement of η is expected around the phase transition temperature, which affects the desorption behaviour.[17)]

The desorption rate κ tends to zero in the both limit of $\eta \to 0$, $\eta \to \infty$. Therefore substantial deviation from the Frenkel-Arrhenius relation is expected in the region of very small or large η value. For the simple one dimensional model as shown in Fig.3, κ is approximately expressed as[23)]

$$\kappa \sim (1 - e^{-B\eta Q/kT\omega_o})\left[(1 + \eta^2/4\omega_c^2)^{1/2} - \eta/2\omega_c\right] \times \omega_o \exp(-Q/kT), \tag{7}$$

where B is the numerical constant of about 3.0, Q, ω_o, ω_c are defined in Fig.3.

2-4) Random walk approach in energy space [ref.25)∿31)]

The desorption process is also formulated as the random walk problem in energy space. The basis of this approach is the master equation,

$$\frac{\partial P_n}{\partial t} = -\sum_m W_{n\to m} P_n(t) + \sum_m W_{m\to n} P_m(t) \tag{8}$$

which determines the stochastic behaviour of the adparticle in the energy space. In the above $P_n(t)$ is the probability of the

adatom existing in the vibrational level n of the potential surface, and $W_{n\to m}$ is the transition probability from the state n to m. The transition matrix element $W_{n\to m}$ due to the adatom-lattice interaction is obtained by the linear coupling approximation or by more general models.

Gortel et al[29] discussed the isothermal desorption time for a physisorption system with many bound states. Exact solutions of the master equation are given for Morse potential. Isothermal desorption times of physisorption systems correspond fairly well with experimental observations. Efrima et al [26] found that the activation energy of the desorption time is smaller than the depth of the potential energy surface. This is because the transition probabilities from the shallow vibrational level to the continuum is very large. The Frenkel-Arrhenius relation is found to hold only in a limited temperature range.

The diffusion equation in energy space, which is the classical limit of eq.(8) was also investigated by Pagni, Keck.[31] Adatom energy distributions were found to be significantly non equilibrium as opposed to the assumption of the approach 2-1).

2-5) Discussions

Each theoretical approach in previous subsections lays its emphasis on a different aspect of the desorption phenomena. For example, the approach 2-1) focusses its attention on elucidating dependence of the realistic features of the potential energy surface on the desorption rate, while the other approaches stress the stochastic process in the adatom/surface system. In the approaches 2-2) and 2-4) the coupling between the adatom and the

lattice vibration is studied, whereas the coupling between the adatom and electron density fluctuation is treated in the approach 2-3). Therefore, although the inclusion of the adatom/electron coupling as in approach 2-3) is indispensable for the chemisorption system, desorption of the physisorbed atom is conveniently studied by the approaches 2-2) and 2-4). The Brownian motion approach 2-3), as it stands, is not sufficient for the description of the desorption of the chemisorbed atom, because the energy dissipation to the lattice is ignored.

The most important quantity which cahracterizes the adsorption and desorption phenomena is the order of the kinetic energy dissipation ΔE during the oscillation period (one round trip) of the adsorbate (gas atom). For example the order of sticking probability s and desorption rate κ is roughly estimated as,

$$s \sim (1 - \exp(-\Delta E/\kappa T)) \tag{9}$$

$$\kappa \sim s\nu\exp(-Q/\kappa T) \tag{10}$$

for a simple one-dimensional model. (More detailed expressions are found in ref.46.) In Table I, magnitude of the friction constant η, the time scale of the friction kernel $\Lambda(t)$ and the order of ΔE are listed for some energy dissipation mechanisms. Since the contribution to ΔE from the lattice vibration can be the same order as that from the electron system, the both coupling mechanisms can be treated on an equal footing. Furthermore the many electron-hole excitations[32-36] would be also an important effect which dissipates the kinetic energy of

the chemisorbed atom. This effect is not incorporated with the approach 2-3). Basic mechanism of the desorption of the chemisorbed atom is not enough clarified as yet.

§3 Theory of electron stimulated (impact) desorption [ref.52) ∿72)]

Dominant features of ESD can be understood by the two-step mechanism by Menzel and Gomer[67] and Redhead[70];

i) the electron impact causes a Franck-Condon transition from the ground state of the adsorbate system to a repulsive neutral or ionic states.

ii) Radiationless transitions occur while the adatom is moving away from the surface. These transitions transfer excitation energy to the solid from the adsorbate, which falls back to its ground state without desorption in most cases.

Quantum theory of ESD was developed by Brenig.[53] It was shown within the adiabatic approximation that desorbing ions decay predominantly via antibonding neutral states. The lifetime of the antibonding neutrals[72] due to the many electron-hole excitations of transition metals were evaluated to be $\sim 10^{-15}$ sec, which corresponds well with experimental observations.

For the case of ionic adsorbates on the metal surface, the initial excitation process includes the interatomic Auger transitions.[58] In this model one of the adatom valence electron falls into the metal core hole state produced by the electron

impact. At the same time one or two other electrons are removed from the adatom. Thus produced positive adatom is strongly repelled from the surface by the Madelung potential. This model explains quite well why positive ions are desorbed from the negative chemisorption charge states, as well as the major desorption threshold energies. This mechanism is very sensitive to the coordination state of the metal atom to which the adsorbate is attached.

Some trajectory calculations[58,68] have been performed to analyze the ion angular distribution (ESDIAD) of desorbed ions. By assuming appropriate chemisorption geometries, the experimental ESDIAD is reproduced by the theoretical calculations.

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Table I Magnitude η and time scale of friction kernel $\Lambda(t)$ and order of the energy transfer per oscillation period ΔE for various dissipation mechanisms

Dissipation mechanism	lattice vibration	electron density fluctuation	many electron-hole pair excitation
order of time scale of $\Lambda(t)$	ω_D^{-1}	$(E_F/\hbar)^{-1}$	
order of magnitude of η $(\eta = \int_0^\infty \Lambda(t)dt)$	ω_D	$\frac{m_e}{M}(E_F/\hbar)$	
order of ΔE	$\hbar\omega_0 \frac{M}{M_s}(\frac{\omega_0}{\omega_D})^3$	$\eta Q/\omega_0$	$\hbar V \frac{d\varepsilon_a}{dz}/\Gamma\Big\|_{Z=Z_0}$ [38]

ω_D : Debye freq., m_e : electron mass, M : adatom mass, ω_0 : adatom vibrational frequency, E_F : Fermi energy, Q : Chemisorption energy, V,Γ : velocity and the level width of the adatom at the position Z_0, where the anion level crosses with Fermi level.

Figure Captions

Fig.1 A simple model of adatom interacting with solid surface. Only the atoms in the promary zone (shown by full circle) directly interact with the adatom. Other atoms in the solids (shown by dotted circle) are called as the secondar zone atoms.

Fig.2 An example of the trajectory in which an Ar atom impinging on W surface is trapped and desorbed.[14] Morse potential is assumed for the Ar-W interaction. The substrate temperature and the adatom initial kinetic energy are assumed as 200°K and 1.2 kcal/mol, respectively.

Fig.3 A simple one-dimensional model of the adiabatic potential energy surface for desorption. Depth of the binding energy is denoted as Q. The adom vibrational frequency in the well and the (imaginary) frequency at the barrier are assumed as ω_o, ω_c respectively.

Fig.4 Numerical result of the desorption rate κ as the function of η for the model of Fig.3. The value of κ determined by Frenkel-Arrhenius relation is shown by dashed line.[23]

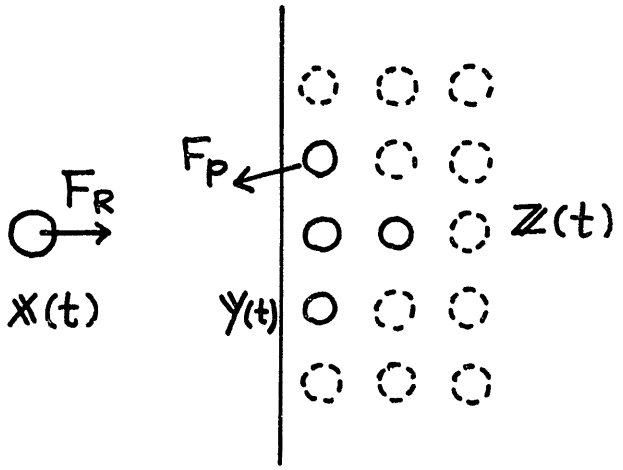
F_P
F_R
Z(t)
X(t)
Y(t)

Fig. 1

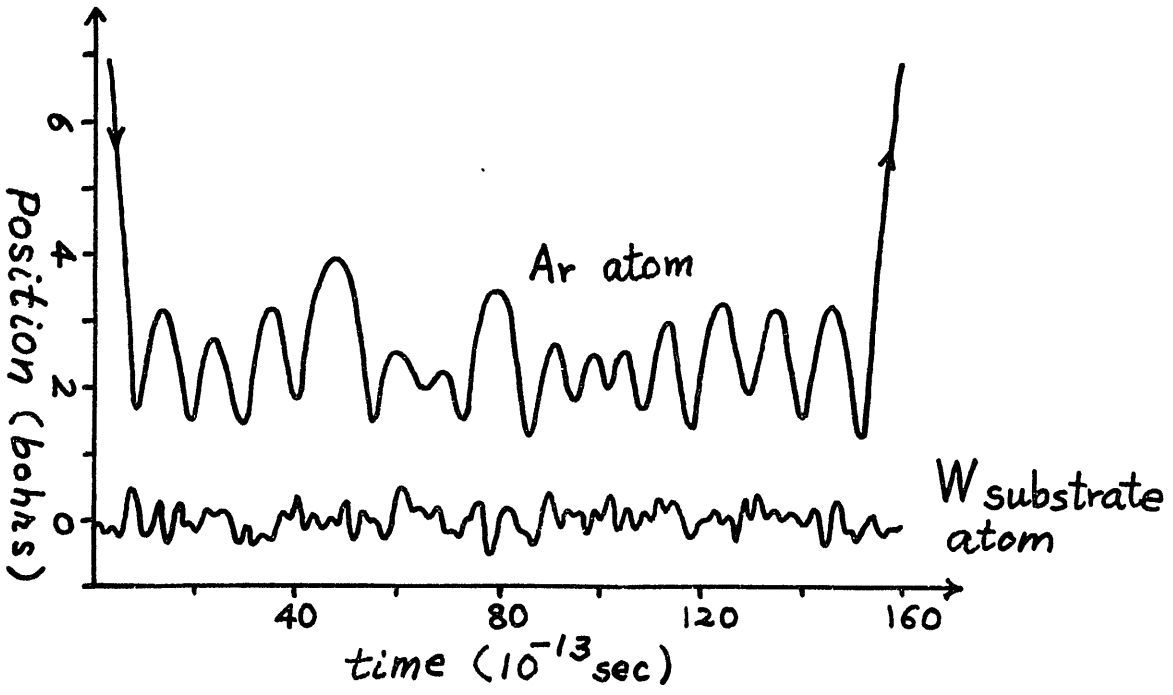
Position (bohrs)
0
2
4
6
Ar atom
W substrate atom
40
80
120
160
time (10^{-13} sec)

Fig. 2

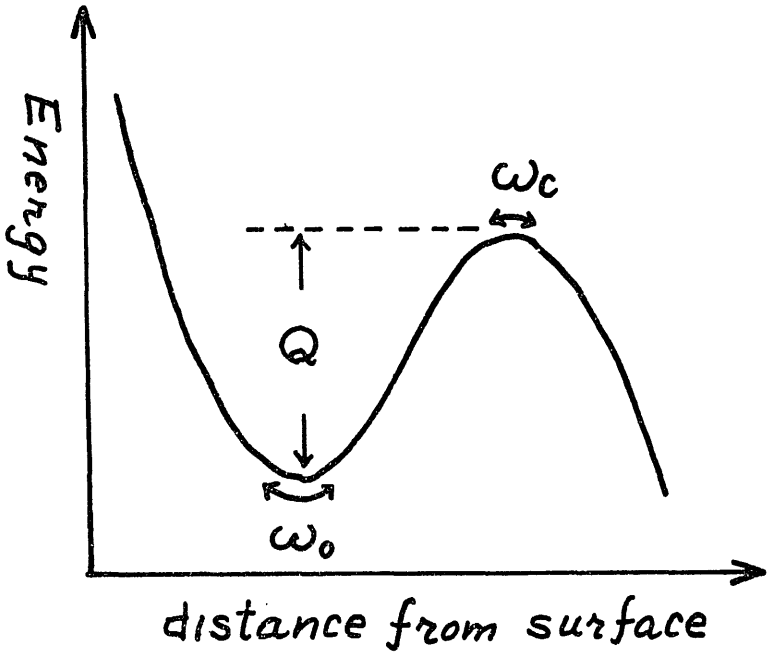
Energy
ω_c
Q
ω_o
distance from surface

Fig. 3

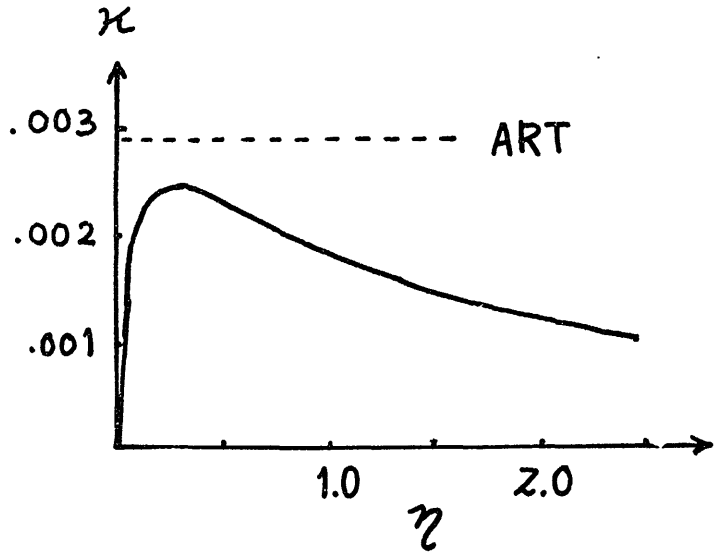
κ
.003
ART
.002
.001
1.0
2.0
η

Fig. 4

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