NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

Technical Memorandum 33-565

Theoretical Determination of Cesiated Work Functions

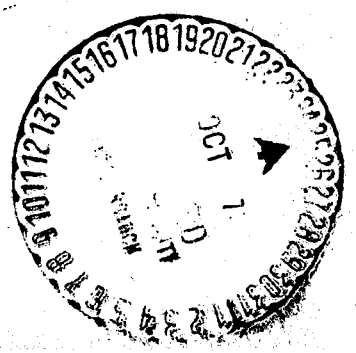
R. M. Szejn

JET PROPULSION LABORATORY

CALIFORNIA INSTITUTE OF TECHNOLOGY

PASADENA, CALIFORNIA

September 15, 1972



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16. Abstract

A computer program based on the theoretical work of Gyftopoulos, Steiner, and Levine on bimetallic systems and using a modified version of Wilkins' SIMCON subroutine SURFAS has been written for the UNIVAC 1108. This program, WFGSL, accepts the operating conditions and the physical parameters pertinent to the substrate and adsorbate and outputs the field-free work function, electron current (Richardson equation), ion current (Saha equation), and fractional substrate coverage by the adsorbate. A brief description of the theory is presented together with a program description and listing. An application of the program to a bimetallic system of cesium (adsorbate) and rhenium (substrate) is also described.

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Prepared Under Contract No. NAS 7-100
National Aeronautics and Space Administration

PREFACE

The work described in this report was performed by the Guidance and Control Division of the Jet Propulsion Laboratory.

CONTENTS

ABSTRACT

A computer program based on the theoretical work of Gyftopoulos, Steiner, and Levine on bimetallic systems and using a modified version of Wilkins' SIMCON subroutine SURFAS has been written for the UNIVAC 1108. This program, WFGSL, accepts the operating conditions and the physical parameters pertinent to the substrate and adsorbate and outputs the field-free work function, electron current (Richardson equation), ion current (Saha equation), and fractional substrate coverage by the adsorbate. A brief description of the theory is presented together with a program description and listing. An application of the program to a bimetallic system of cesium (adsorbate) and rhenium (substrate) is also described.

I. INTRODUCTION

In thermionic energy conversion, the work function of an emitter in contact with cesium vapor is a controlling parameter determining the rate of electron emission and thus converter efficiency. An understanding of the adsorbate-substrate interaction is essential to the development of more efficient thermionic converters. The theoretical work of Gyftopoulos, Steiner, and Levine (Refs. 1, 2) on the general bimetallic system evaluates the work function of this system as a function of its physical parameters.

This theory has been incorporated in a program using a modified version of Wilkins' SIMCON subroutine SURFAS (Ref. 3). The program serves as a tool in evaluating possible changes in a converter due to changes in the work function of the emitting surface with time. Although cesium is the only adsorbate considered here, the computer program is general in nature and can be used for other bimetallic systems. The program accepts the operating conditions and the physical parameters pertinent to the substrate and adsorbate and outputs the field-free work function, electron current, ion current, and fractional substrate coverage by the adsorbate. The code differs from Wilkins' program in format usage, input parameters, and output information; the printed plus graphical output facilitates the comparison of theoretical and experimental work.

II. THEORY

The work function of a pure metal can be identified with the neutral electronegativity of valence orbitals in the surface atoms. Using this approach, the work function for a bimetallic surface can be viewed as a perturbation from the bare work function. The perturbation arises from substrate-adsorbate (s-f) and adsorbate-adsorbate (f-f) interactions (Fig. 1). In an s-f bond, there is a charge transfer due to a difference in the

electronegativities of the atoms. This charge transfer results in a dipole double layer whose electrostatic potential is superimposed on the electronegativity of the substrate atoms. The perturbation which results is taken to be proportional to the electronic charge transferred, F. The f-f bond resulting from charge overlap also produces an electrostatic potential, again affecting the bare substrate electronegativity. This perturbation is assumed proportional to the amount of charge overlap, Q. Thus, our basic relation (Refs. 1, 2) is

$$\phi = \phi_s + cQ + bF, \tag{1}$$

where ϕ is the perturbed neutral electronegativity, ϕ_s is the bare substrate work function (unperturbed neutral electronegativity), and c and b are constants of proportionality. Observe that $\phi = \phi_s$ at $\theta = 0$ (θ is the fraction of substrate coverage by the adsorbate) since Q and F are zero by definition.

It is found experimentally that with a one-monolayer coverage ($\theta = 1$), the work function is approximately equal to the adsorbate work function, ϕ_f:

$$\phi = \phi_f, \quad \theta = 1. \tag{2}$$

Equation (2) is therefore a boundary condition for (1).

The quantities cQ and bF may be derived, and result in the following equations:

$$cQ = -(\phi_s - \phi_f)M, \tag{3}$$

where M is a Morse function defined by

$$M = 2 \exp\left[+ad_0(1 - \theta^{-1/2})\right] - \exp\left[+2ad_0(1 - \theta^{-1/2})\right], \tag{4}$$

where $ad_0 = 2.97$.

For bF it is found (Ref. 1) that

$$b = \frac{-0.905\ (10^{-6})\ V_f\ (R \cos \beta)\ \sigma_f \theta}{\left(1 + \frac{\alpha}{R^3}\right)\left(1 + 9\alpha\sigma_f^{3/2}\theta^{3/2}\right)}\ \text{eV/coulumb}, \tag{5}$$

$$\frac{F}{e} = \frac{(\phi_s - \phi_f)(1 - M)}{D_0\left(\frac{1 - F^2}{e^2}\right)^{-1/2} + D_1}, \tag{6}$$

where V_f is the number of bonding orbitals per adatom, R and β are defined in Fig. 2, σ_f is the adsorbate density, D_0 is the energy of the purely covalent s-f bond, and D_1 is an adjustable parameter, which may be approximated by (Ref. 1) $D_1 = 1.3\ (\phi_s + \phi_f)\ (-28.75/R)$, where R is in Å. Note that (2) is satisfied since $F(1) = 0$.

III. PROGRAM

The program logic determining the cesiated work function ϕ and the fractional coverage θ as functions of T_S and T_R consists of two nested iteration loops. The inner loop uses Newton's method to solve for F in (6), assuming an initial value of 0.5 for θ and 0.5 for F. After solving for F, the arrival rate is set equal to the desorption rate of adsorbate atoms. Since the latter quantity is a function of both ϕ and θ, it will not equal the arrival rate for the initial guess of θ. Thus θ is changed accordingly to bring the arrival and desorption rates into equality. Having adjusted θ according to the difference between the arrival and desorption rates, it is now necessary to recalculate F to determine the new value of ϕ. Figure 3 presents a flowchart of the SURFAS subroutine. Table 1 presents the input parameters. Besides the output listed in the introduction, WFGSL uses the CalComp plotter to generate a graph of ϕ versus T_S/T_R. A listing of WFGSL is given in Appendix A, along with a sample data deck and the corresponding output (Appendix B).

IV. RESULTS

The results appearing in Figs. 4-8 are for a rhenium substrate and cesium adsorbate. The physical constants that were used appear in the data deck of Appendix A. The curves in Figs. 4-7 show the dependence of the work function ϕ on the parameters ϕ_0, σ_s, σ_a, D_1 (bare work function, substrate density, adsorbate density, and adjustable parameter, respectively).

In Fig. 4, ϕ is plotted as a function of the ratio between substrate and adsorbate temperature for various values of the bare work functions. As is expected, a change in the bare work function results in a vertical displacement in the curves. Figure 5 shows the effect of varying the substrate density σ_s while keeping the adsorbate density fixed. The effect is seen to be most pronounced for smaller values of T_S/T_R, where the fractional coverage and thus the substrate-adsorbate interactions are greatest. In Fig. 6, the substrate density is held constant while the adsorbate density is varied. The curves show the strong dependence of ϕ on the adsorbate; with increasing adsorbate density the work function is lowered significantly even at high substrate-adsorbate temperature ratios. Figure 7 shows that cesiated work functions shift by a constant amount over the entire range of temperature ratios as the adjustable parameter D_1 is varied. In Fig. 8 an experimental curve (Ref. 4) is shown together with a theoretical curve generated by WFGSL. The agreement is seen to be excellent.

V. CONCLUSION

A computer program, WFGSL, based on Gyftopoulos, Steiner, and Levine's work on bimetallic systems and using a modified version of Wilkins' SIMCON subroutine SURFAS has been developed for use on the UNIVAC 1108. The program determines the field-free cesiated work function, electron current, ion current, and fractional substrate coverage by the adsorbate. The indicated relation between ϕ, the calculated work function, and the various physical input parameters is found to be consistent with expectations; agreement between experimental work functions and theoretical work functions is shown to be good.

REFERENCES

1. Gyftopoulos, Elias P., and Levine, Jules D., "Work Function Variation of Metals Coated by Metallic Films," J. Appl. Phys., 33, 67, 1962.

2. Gyftopoulos, Elias P., and Steiner, Don, "Orbital Electronegativity and Physical Properties of Bimetallic Adsorption Systems," Report on Twenty-Seventh Annual Physical Electronics Conference, Cambridge, Mass., p. 169, Mar. 1967.

3. Wilkins, D. R., SIMCON: A Digital Computer Program for Computing Thermionic Converter Performance Characteristics, AEC R&D Report GESR-2109, General Electric Co., Pleasanton, Calif., Jan. 1970.

4. Shimada, K., and Cassell, P. L., Evaluation of Converters Fueled With Uranium Nitride, Technical Memorandum 33-489, Jet Propulsion Laboratory, Pasadena, Calif., July 30, 1971.

Table 1. Input parameters

Card	Columns	Parameter	Units	Format
1	1-5	Number of independent cases ≥ 1	—	I5
	6-80	Blank		
Parameter, substrate properties				
2	1-8	Atomic weight	amu	F8.3
	9-16	Bare work function	eV	F8.3
	17-24	Full coverage work function	eV	F8.3
	25-32	Heat of sublimation	eV	F8.3
	33-40	Metallic radius	Å	F8.3
	41-48	Bond strength	—	F8.3
	49-56	Substrate atom density	$Å^{-2}$	F8.3
	57-64	Adsorption site density	$Å^{-2}$	F8.3
	65-72	D_1	eV	F8.3
	73-80	Blank		F8.3
Parameter, vapor properties				
3	1-8	Atomic weight	amu	F8.3
	9-16	Heat of sublimation	eV	F8.3
	17-24	Metallic radius	Å	F8.3
	25-32	Bond strength	—	F8.3
	33-40	Ionization potential	eV	F8.3
	41-48	Electron-neutral cross section	$Å^2$	F8.3
	49-56	Ion-neutral cross section	$Å^2$	F8.3
	57-80	Blank		F8.3

Table 1 (contd)

Card	Columns	Parameter	Units	Format
4	1-5	Number of substrate temperatures to be read	—	I5
	6-80	Blank		
5	1-10	Substrate temperature T_s	K	F10.5
	11-15	Number of adsorbate temperatures to be read in for the given substrate temperature (NURT)	—	I5
	16-80	Blank		
Parameter, NURT				
6	1-10	Adsorbate temperature T_R	K	F10.5
	11-80	Blank		

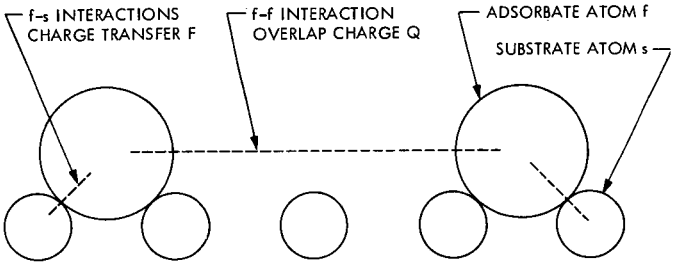

Fig. 1. Bonding interactions

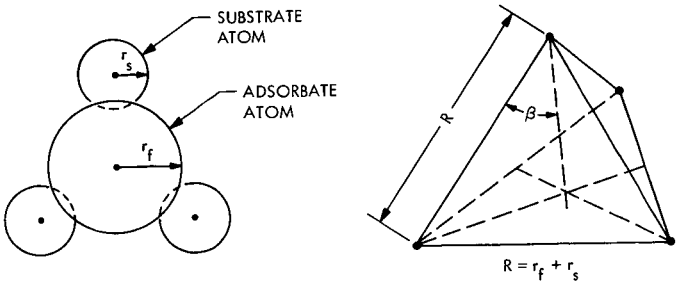

Fig. 2. Maximum ligancy on the 111-plane of tungsten

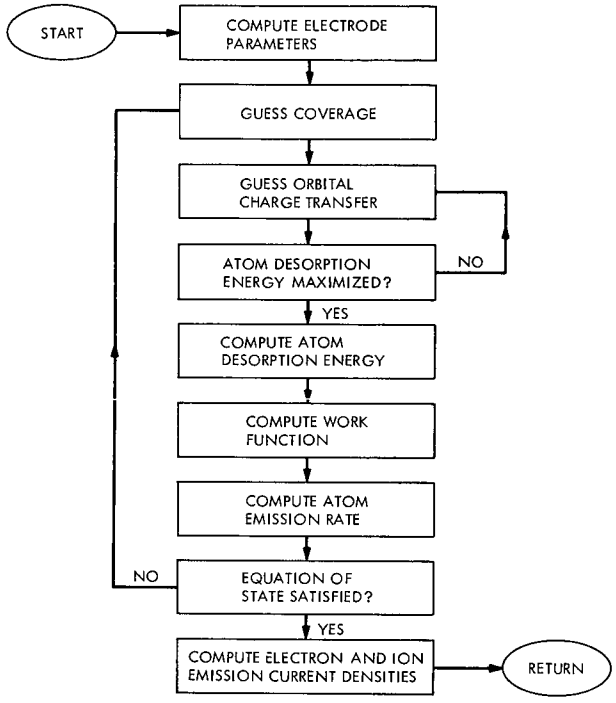

Fig. 3. SURFAS subroutine flowchart

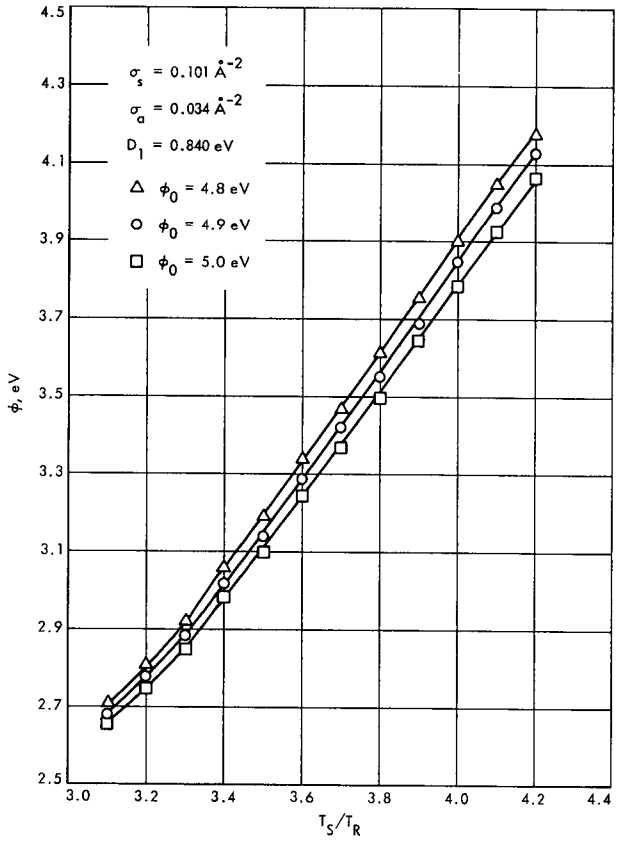

Fig. 4. Variation in ϕ (T_S/T_R, ϕ_0)

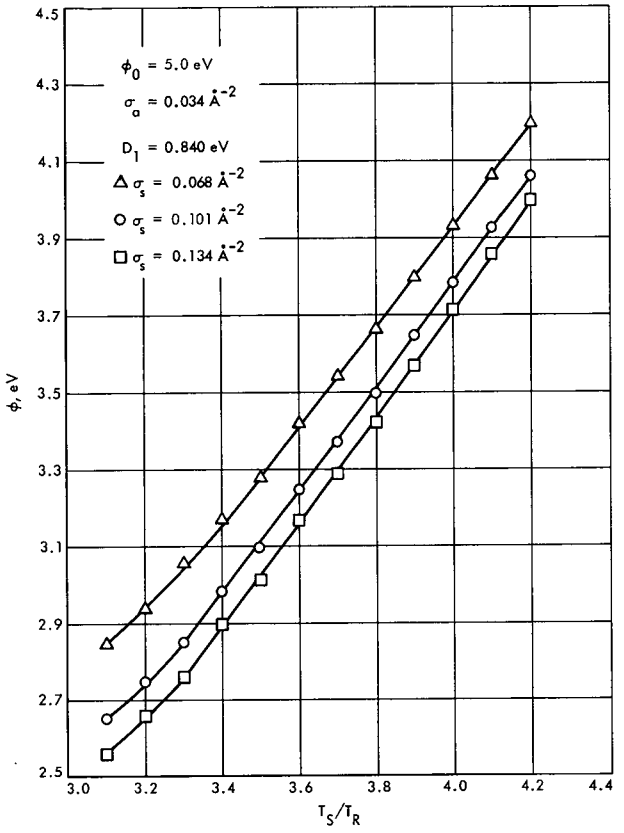

Fig. 5. Variation in ϕ (T_S/T_R, σ_s)

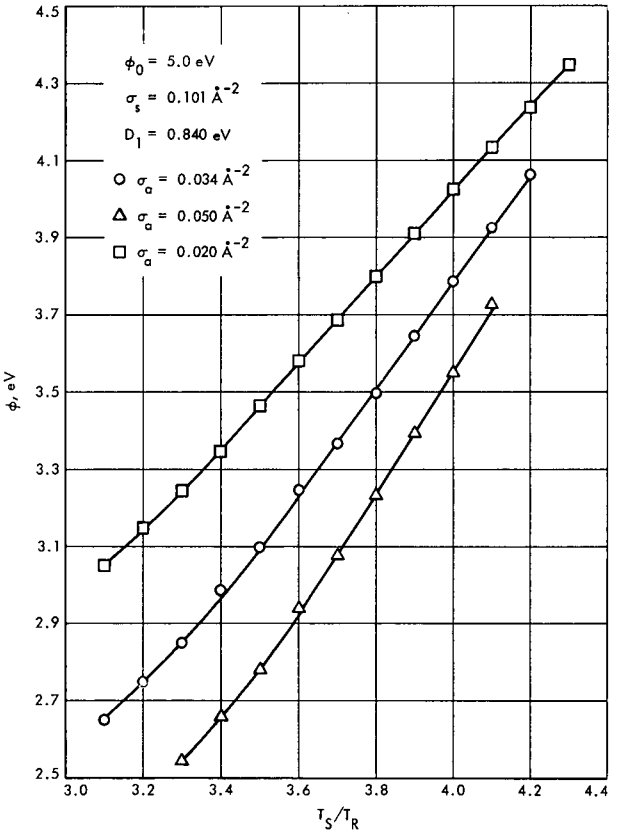

Fig. 6. Variation in ϕ (T_S/T_R, σ_a)

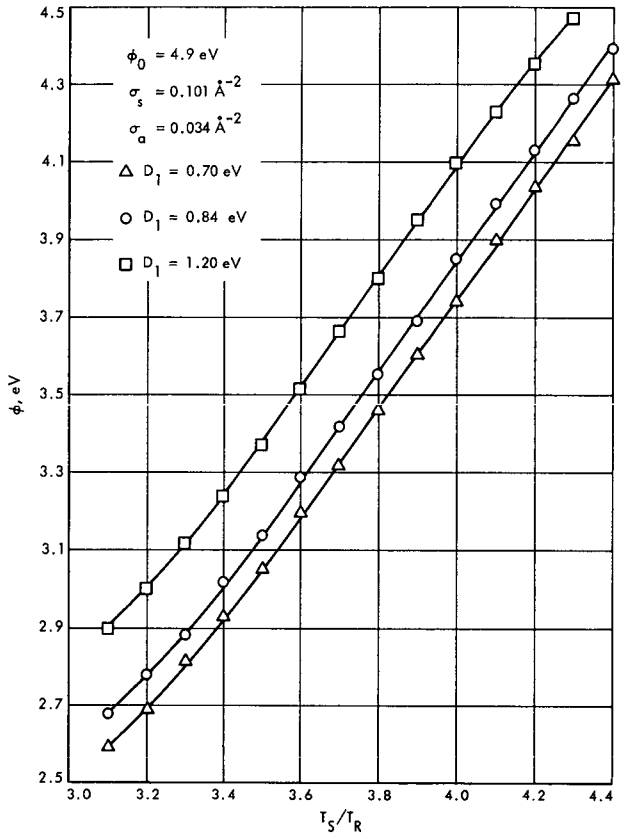

Fig. 7. Variation in ϕ (T_S/T_R, D_1)

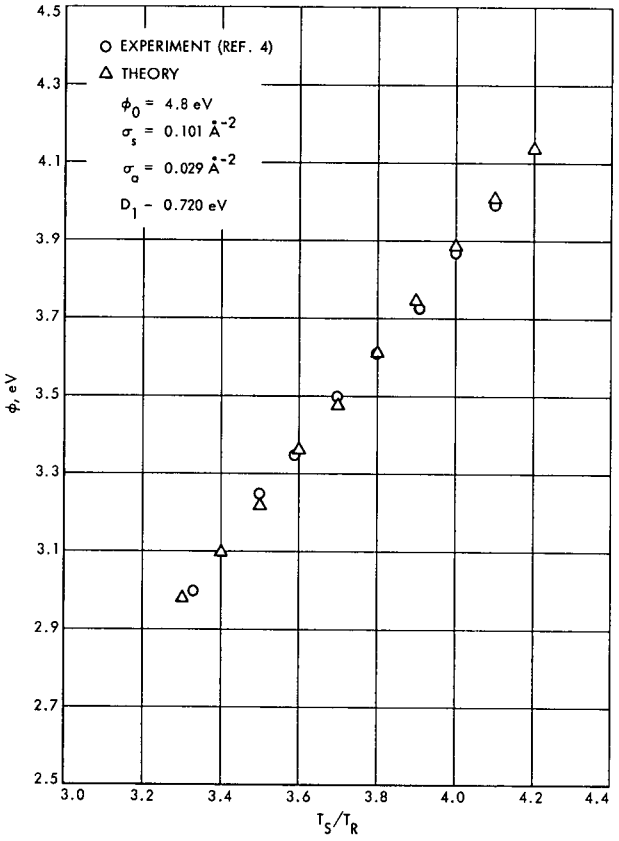

Fig. 8. Rhenium work function

APPENDIX A

PROGRAM LISTING

```
      DIMENSION RAT(10),WF(10)
      COMMON AG,HG,RG,SG,VIG,VALG,CSEG,CSIG
      REAL JO,IO
      VALG=1.0
      CALL PLOTS
      CALL PLOT(0.0,0.0,-3)
      READ(5,100) NUCS
      DO 10  NUCSN=1,NUCS
      READ(5,101) AS,FOS,FCS,HS,RS,SS,SDS,ADS,CS
      READ(5,101) AG,HG,RG,SG,VIG,CSEG,CSIG
      READ(5,100) NUSB
      WRITE(6,120)
      WRITE(6,103) AS,FOS,FCS,HS,RS,SS,SDS,ADS,CS
      WRITE(6,121)
      WRITE(6,103) AG,HG,RG,SG,VIG,CSEG,CSIG
      WRITE(6,122)
      DO 10  NUSBCN=1,NUSB
      READ(5,102) TS,NURT
      I=NURT+1
      J=I+1
      WF(I)=2.0
      WF(J)=0.25
      RAT(I)=2.0
      RAT(J)=0.25
      IF(NUSBCN.NE.1) GO TO 3
      CALL PLOT(12.0,0.0,-3)
      CALL AXIS(0.0,0.0,5HTS/TR,-5,10.0,0.0,RAT(I),RAT(J))
      CALL AXIS(0.0,0.0,12HWORK FUNCTION,13,10.0,90.0,WF(I),WF(J))
    3 CONTINUE
      DO 9  NURTCN=1,NURT
      READ(5,102) TR
      PG=2.45E+8*EXP(-8910.0/TR)/SQRT(TR)
      CALL SURFAS (AS,FOS,FCS,HS,RS,SS,SDS,ADS,CS,TS,PG,WFSO,JO,IO,DS,
     1NOCONV)
      RATIO=TS/TR
      IF (NOCONV.NE.1) GO TO 4
      WRITE (6,107) TS,TR
      WF(NURTCN) =2.0
      RAT(NURTCN)=RATIO
      NOCONV = 0
      GO TO 9
    4 CONTINUE
      RAT(NURTCN)=RATIO
      WF(NURTCN)=WFSO
      IF(NURTCN.NE.1) GO TO 5
```

```
      WRITE(6,105) TS,TR,RATIO,WFSO,JO,IO,DS
      GO TO 9
    5 CONTINUE
      WRITE(6,106) TR,RATIO,WFSO,JO,IO,DS
    9 CONTINUE
      CALL LINE(RAT,WF,NURT,1,-1,NUSBCN)
   10 CONTINUE
      CALL PLOT(10.0,0.0,999)
  100 FORMAT(I5)
  101 FORMAT(10F8.3)
  102 FORMAT(F10.5,I5)
  103 FORMAT(1H0,9F12.3)
  104 FORMAT(1H0,18H      GAP SPACING =,F5.1)
  120 FORMAT(108H0       AT WT      B W FUN      C W FUN     SUB HEAT       R
     1ADIUS     BOND STR      SUB DEN      ADS DEN     DI ENERGY)
  121 FORMAT( 94H0       AT WT     SUB HEAT       RADIUS     BOND STR      IO
     1N PCT     E-N XSEC     I-N XSEC)
  122 FORMAT( 84H0    SUB TEMP     RES TEMP        TS/TR     WORK FUN
     1    JO           IO     FRAC COV)
  105 FORMAT(1H0,5F12.3,F12.6,F12.4)
  106 FORMAT(13X,4F12.3,F12.6,F12.4)
  107 FORMAT (10X,30HSURFAS IS UNCONVERGED FOR TS =,F8.2,10H  AND TR =,F
     18.2)
      END
-FOR,IN  SURFAS
      SUBROUTINE SURFAS(AS,FOS,FOG,HS,RS,SS,SSS,ASS,C,TS,PG,FS,EES,EIS,C
     1S,NOCONV)
CSURFAS     SURFACE PHYSICS SUBROUTINE
      COMMON AG,HG,RG,SG,VIG,VALG,CSEG,CSIG
CC
C     EVALUATE ADSORPTION SYSTEM PARAMETERS
CC
      SWG = 2.0
      SDS = 100.0*SSS
      ADS = 100.0*ASS
      R = RG + RS
      COSB = SQRT(1.0 - 50.0/(SDS*R**2.0))
      ALF = RG**3 + 1.65*RS**3
      C1 = 0.905*R*ADS*COSB/(1.0+ALF/R**3)
      C2 = 9.0E-3*ALF*ADS**1.5
      C3 = C
      S = 2.0/(SG/SS + SS/SG)
      DO = SQRT(HS*HG)*S
      ARS = 0.56E+4*PG/SQRT(AG*TS)
CC
C     BEGIN SURFACE COVERAGE ITERATION
CC
```

```
      NCS = 0
      CS = 0.5
      DELCS = 0.5
   10 CONTINUE
      NCS = NCS + 1
      IF(NCS.GT.35) GO TO 45
      G = 1.0 - 3.0*CS*CS + 2.0*CS*CS*CS
      DG = -6.0*CS*(1.0-CS)
CC
C     BEGIN FRACTIONAL CHARGE TRANSFER ITERATION
CC
      NF = 0
      F=0.5
   12 CONTINUE
      NF = NF +1
      IF(NF.GT.15) GO TO 45
      FOF = DO*F/SQRT(1.0-F**2) + C3*F - (FOS-FOG)*G
      DFOF = DO/(1.0-F**2)**1.5 + C3
      FN = F - FOF/DFOF
      IF(FN.LE.0.0) FN = 0.5*F
      IF(FN.GE.1.0) FN = 0.5*(1.0+F)
      IF(ABS(FN-F).LE.0.01) GO TO 15
      F = FN
      GO TO 12
CC
C     END FRACTIONAL CHARGE TRANSFER ITERATION
CC
   15 FAS = DO*SQRT(1.0-F**2) - 0.5*C3*F**2 + F*(FOS-FOG)*G
      DFAS = F*(FOS-FOG)*DG
      FS = FOG + (FOS-FOG)*G - F*C1*CS/(1.0+C2*CS**1.5)
      FREQ1 = 2.50E-6*SQRT(FAS)*SQRT((AG+4.0*AS)/(2.0*AG*AS))/(R*COSa)
      U= 0.5*CS*DFAS/FAS
      FREQ = FREQ1*EXP(U)
      ENT = EXP(0.5*(CS/(1.0-CS)+SQRT(CS)/(1.0-SQRT(CS))))/(SQRT(1.0-CS)
     1*(1.0-SQRT(CS)))
      ERS = 1.0E+14*FREQ*SWC*ADS*CS*ENT*EXP(-11600.0*FAS/TS)
      IF(ABS((ERS-ARS)/ARS)-0.005)40,20,20
   20 IF(ERS-ARS)25,40,30
   25 DELCS = DELCS/2.0
      CS = CS + DELCS
      GO TO 10
   30 DELCS = DELCS/2.0
      CS = CS - DELCS
      GO TO 10
CC
C     END SURFACE COVERAGE ITERATION
CC
```

```
C       COMPUTE ELECTRON AND ION EMISSION CURRENT DENSITIES
CC
   40 EES = 120.0*TS**2*EXP(-11600.0*FS/TS)
      FIS = 2.598E+1*PG*EXP(11600.0*(FS-VIG)/TS)/SQRT(AG*TS/11600.0)
      GO TO 50
   45 CONTINUE
      NOCONV=1
   50 CONTINUE
      RETURN
      END
-MAP
LIB LIB*PLOTS
-XQT
    1
  186.2      4.8      1.95      8.07      1.35      2.62     0.101     0.029      0.72
  133.0     0.73      2.66       1.0      3.89      400.     570.0
    5
    2043.      3
    486.4
    475.1
    464.3
    1940.      4
    485.0
    473.2
    461.9
    451.2
    1840.0     5
    511.1
    497.3
    484.2
    471.8
    460.0
    1740.0     5
    511.8
    497.1
    483.3
    470.3
    457.9
    1660.0     7
    535.5
    518.8
    503.0
    488.2
    474.3
    461.1
    448.6
-FIN                                                                    NIF-
```

APPENDIX B

PROGRAM OUTPUT

BXGT

AT WT	B W FUN	C W FUN	SUB HEAT	RADIUS	BOND STR	SUB DEN	ADS DEN	D1 ENERGY
186.203	4.900	1.950	9.070	1.350	2.620	.101	.029	.720

AT WT	SUB HEAT	RADIUS	BOND STR	ION POT	E-N XSEC	I-N XSEC
133.300	.730	2.350	1.000	3.890	400.000	570.000

SUB TEMP	RES TEMP	TS/TR	WORK FUN	JO	IO	FRAC COV
2043.000	486.400	4.200	4.139	.031	2.714989	.1118
	475.100	4.300	4.260	.016	3.534752	.0920
	464.300	4.400	4.378	.008	4.503619	.0737
1940.000	485.000	4.000	3.885	.037	.625182	.1523
	473.200	4.100	4.017	.017	.882868	.1313
	461.900	4.200	4.148	.008	1.231198	.1104
	451.200	4.300	4.279	.003	1.728322	.0898
1840.000	511.100	3.600	3.360	.257	.058077	.2354
	497.300	3.700	3.482	.119	.078551	.2153
	484.200	3.800	3.611	.053	.110640	.1943
	471.300	3.900	3.752	.022	.157962	.1733
	460.000	4.000	3.888	.009	.247249	.1519
1740.000	511.900	3.400	3.108	.365	.009419	.2773
	497.100	3.500	3.227	.165	.012614	.2573
	483.300	3.600	3.351	.072	.017518	.2368
	470.300	3.700	3.476	.031	.024611	.2163
	457.900	3.800	3.608	.013	.036023	.1948
1660.000	535.500	3.100	2.756	1.430	.001355	.3386
	518.800	3.200	2.858	.702	.001642	.3191
	503.000	3.300	2.981	.298	.002294	.2993
	488.200	3.400	3.098	.131	.003086	.2791
	474.300	3.500	3.218	.057	.004245	.2588
	461.100	3.600	3.343	.024	.006035	.2380
	448.600	3.700	3.473	.010	.008846	.2168

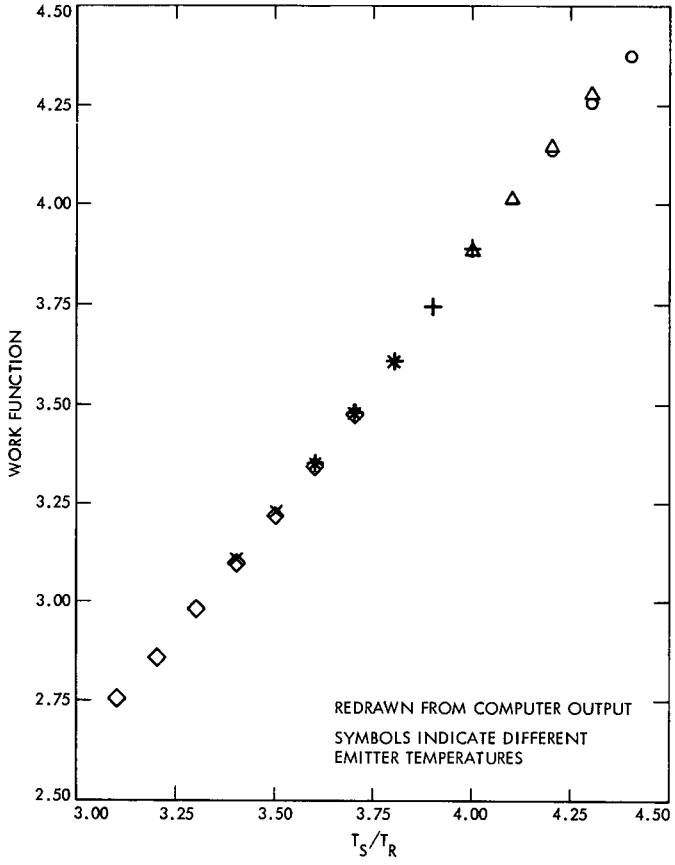

Fig. B-1. Work function vs temperature ratio

NASA — JPL — Coml., L.A., Calif.