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The simulation of gold adsorption by carbon using a film diffusion model

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```
C SUBROUTINE TO RECEIVE THE OPERATING PARAMETERS OF A ADSORPTION
C BATCH TEST AND CALCULATE THE CONSTANTS IN THE DE
C
      TYPE * *
      TYPE *************************************************
      TYPE *****       OPERATING       PARAMETERS       *****
      TYPE *************************************************
      TYPE **
C DEFINE ALL VARIABLES TO DOUBLE PRECISION
      DOUBLE PRECISION A1,A2,A3,A4,FB,S0,C0
      DOUBLE PRECISION SM,CM,FA,DP,VS,AC
C DEFINE SUBROUTINE
      SUBROUTINE DATAIN(A1,A2,A3,A4,FB,S0,C0)
C ACCEPT DATA
      ACCEPT "MASS OF SOLUTION                          KG ",SM
      ACCEPT "INITIAL SOLUTION CONCENTRATION         MG/KG ",S0
      ACCEPT "MASS OF CARBON                            KG ",CM
      ACCEPT "INITIAL CARBON   CONCENTRATION         MG/KG ",C0
      ACCEPT "DIAMETER OF CARBON PARTICLES               M ",DP
      ACCEPT "FREUNDLICH CONSTANT (NUMBER)                 ",FA
      ACCEPT "FREUNDLICH CONSTANT (POWER)                  ",FB
C CALCULATE OTHER VARIABLES IE SOLN VOLUME & CARBON AREA
      VS=SM/1000
      AC=CM*5.615*(EXP(-435.8*DP))
C CALCULATE CONSTANTS IN DE
C DS/DT = KL ( A1.S + A2 ( A3 + A4.S ) ** B )
      A1=-AC/VS
      A2=(AC*FA)/VS
      A3=C0+(SM*S0)/CM
      A4=-SM/CM
C STORE DATA INPUT
      LD=16
      OPEN 16,"DATINP"
      WRITE FREE (16) "MASS OF SOLUTION                    KG ",SM
      WRITE FREE (16) "INITIAL SOLUTION CONCENTRATION   MG/KG ",S0
      WRITE FREE (16) "VOLUME OF SOLUTION               M**3 ",VS
      WRITE FREE (16) "MASS OF CARBON                     KG ",CM
      WRITE FREE (16) "INITIAL CARBON   CONCENTRATION   MG/KG ",C0
      WRITE FREE (16) "DIAMETER OF CARBON PARTICLES        M ",DP
      WRITE FREE (16) "AREA OF CARBON                   M**2 ",AC
      WRITE FREE (16) "FREUNDLICH CONSTANT (NUMBER)         ",FA
      WRITE FREE (16) "FREUNDLICH CONSTANT (POWER)          ",FB
      WRITE FREE (16) " A1=-AC/VS                           ",A1
      WRITE FREE (16) " A2=(AC*FA)/VS                       ",A2
      WRITE FREE (16) " A3=C0+(SM*S0)/CM                    ",A3
      WRITE FREE (16) " A4=-SM/CM                           ",A4
      CLOSE 16
C END
      RETURN
      END
```

```
C SUBROUTINE TO CALCULATE THE VALUE OF THE SOLUTION AT A STEP
C SIZE VIA A RUNGE KUTTE METHOD
C
C DEFINE VARIABLES TO DOUBLE PRECISION
      DOUBLE PRECISION K0,K1,K2,K3,A1,A2,A3,A4,SRK,SCH
      DOUBLE PRECISION RC,SS,DD,FB
C DEFINE SUBROUTINE
      SUBROUTINE RUNKUT(A1,A2,A3,A4,SS,SCH,RC,FB)
C
C RUNGE KUTTE NUMERICAL METHOD
C
      SRK=SCH
C CHECK VALUE OF A3+A4*SRK TO SEE IF THE VALUE IS NEAR ZERO
      DD=A3+A4*SRK
      IF (DD.LT.1) GO TO 10
      K0=SS*RC*(A1*SRK+A2*((A3+A4*SRK)**FB))
      SRK=SCH+K0/2
      K1=SS*RC*(A1*SRK+A2*((A3+A4*SRK)**FB))
      SRK=SCH+K1/2
      K2=SS*RC*(A1*SRK+A2*((A3+A4*SRK)**FB))
      SRK=SCH+K2
      K3=SS*RC*(A1*SRK+A2*((A3+A4*SRK)**FB))
      GO TO 20
10    K0=SS*RC*A1*SRK
      SRK=SCH+K0/2
      K1=SS*RC*A1*SRK
      SRK=SCH+K1/2
      K2=SS*RC*A1*SRK
      SRK=SCH+K2
      K3=SS*RC*A1*SRK
20    SCH=SCH+((K0+K1*2+K2*2+K3)/6)
C END
      RETURN
      END
```

```
C SUBROUTINE TO CALCULATE THE SUM OF SQUARES GIVEN THE PREDICTED
C AND THE OBSERVED VALUES
C
C SET VARIABLES TO DOUBLE PRECISION
      DOUBLE PRECISION SCO(20),SCP(20),TIM(20),SP(6000),SS,S0
C DEFINE SUBROUTINE
      SUBROUTINE DETSSQ(SCO,N,SSQ,TIM,SP,SS,S0)
      SCP(1)=S0
      DO 20 I=2,N
      K=TIM(I)/SS
      SCP(I)=SP(K)
 20   CONTINUE
      SSQ=0
      DO 10 I=1,N
      SSQ=SSQ+((SCO(I)-SCP(I))**2)
 10   CONTINUE
C END
      RETURN
      END
```

APPENDIX C

VALUES OF PHYSICAL PARAMETERS

VALUES OF PHYSICAL PARAMETERS

During the manipulation of the data, values must be assigned to the variables in the equations. The values, or methods of obtaining the values, for each variable are listed below.

MASS OF CARBON

Moisture trapped in the carbon granules because of the pore structure and activity of the carbon, can give rise to errors in the determination of the carbon mass. Therefore the percentage moisture content of a carbon sample must be determined prior to determining its mass.

MASS OF SOLUTION

Solution is measured by its volume. The mass of the solution can be obtained by assuming its density to be 1000 kg m^{-3}.

LINEAR VELOCITY

The linear velocity is defined as the superficial velocity of solution through an empty column, and is measured by means of a stopwatch and measuring cylinder. The linear velocity is then

$$\text{linear velocity} = \frac{\text{flowrate}}{\text{cross sectional area of column}}$$

VISCOSITY AND DENSITY

As clear solutions were used, both the viscosity and density were assumed to be those of water:

density	=	1000 kg m^{-3}
viscosity	=	0,001 kg m^{-1} s^{-1}

DIFFUSIVITY

The diffusivity of the aurocyanide ion is not known, no mention of it being found in the literature. Whatever the actual value may be, it would be a constant for all adsorption tests. The diffusivity is incorporated in the Sherwood number and the Schmidt number. Provided that a reasonable figure is attached to the diffusivity, and kept constant for all tests, its correctness or otherwise will have no effect on the outcome of the proposed correlations.

Considering that the diffusivity of hydrogen in water is 5×10^{-9} m^2 s^{-1}, that of chlorine $1,4 \times 10^{-9}$, and that of the organic molecule lactose $0,5 \times 10^{-9}$, a figure of 10^{-9} m^2 s^{-1} for gold cyanide seems reasonable.

VOIDAGE

The voidage of a column is the fraction of volume not occupied by the carbon particles. The following test was undertaken on the three carbon size fractions to determine the voidage of wet settled carbon: water was put into a measuring cylinder and its volume noted.

Moist carbon was then added and allowed to settle. Two readings were then taken, firstly, the volume of the wet settled carbon, and, secondly, the volume of the carbon and the water. The actual volume of the carbon is then the difference in the volume of the water before and after the addition. Therefore the interstitial volume is the volume of the wet settled carbon minus the actual carbon volume, and the voidage of wet settled carbon can be calculated. If the packing remains consistent, this value should be constant and not dependent on particle size. It was determined that the voidage of wet settled carbon is 0,57. Coulson[29] gives the voidage of triangular pyramids as 0,52.

If the bed is fluidized, the voidage changes. Two measurements are then required to determine the change in voidage, namely, the height of the wet settled bed and the height of the fluidized bed. The voidage can then be determined:

$$\text{voidage} = \frac{\text{Volume of fluidized bed - volume of carbon}}{\text{Volume of fluidized bed}}$$

$$= \frac{\pi r^2 H_f - \pi r^2 H_s (1 - \epsilon_B)}{\pi r^2 H_f}$$

$$= \frac{H_f - H_s (1 - \epsilon_B)}{H_f}$$

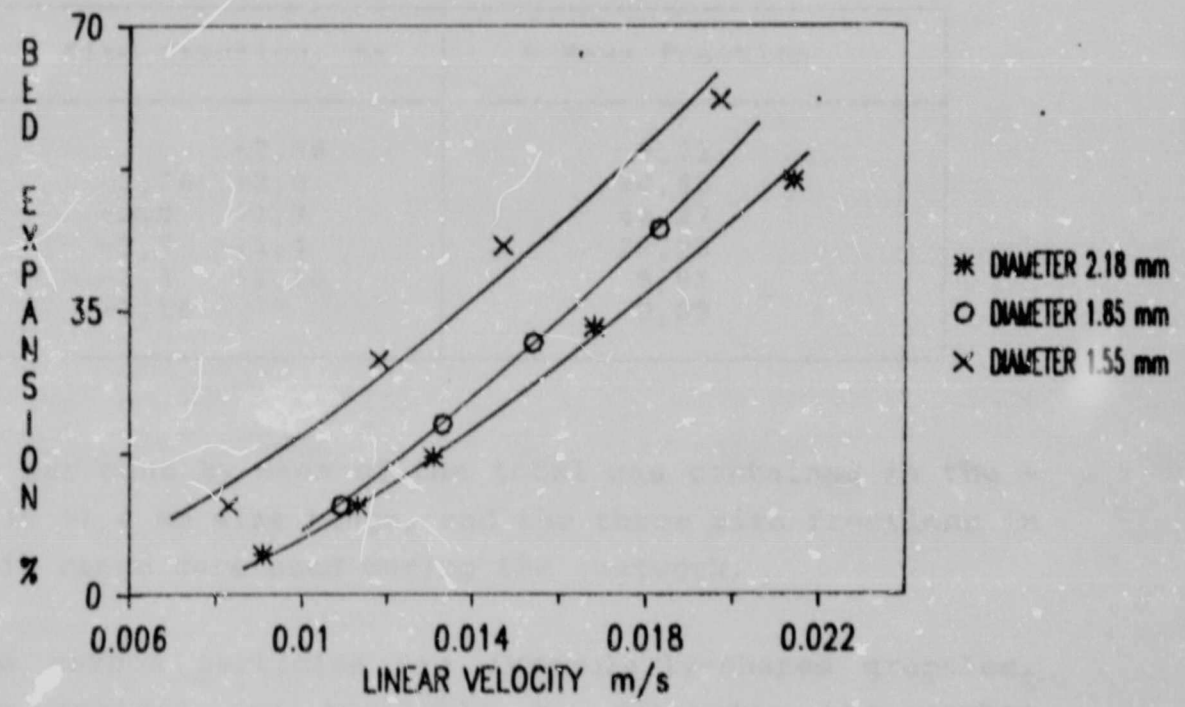
where H_f = Height of fluidized bed
H_s = Height of wet settled bed
ϵ_B = Voidage of the wet settled bed.

Fluidization tests on all three carbon size fractions were undertaken in the column by varying the linear velocity and measuring the height of the fluidized bed.

A plot of the bed expansion versus the linear velocity for the three carbon size fractions is shown in Figure C1, where bed expansion is defined as

$$\text{Bed expansion} = 100 \left(\frac{H_f - H_s}{H_s} \right)$$

FIG C1 BED EXPANSION CURVES



PARTICLE DIAMETER

A sample of Le Carbone G210 AS virgin carbon was screened over a series of screens with apertures of 2,36 mm, 2,0 mm, 1,7 mm, 1,4 mm and 1,18 mm respectively. The per cent mass fraction in each size is shown below in Table C1.

TABLE C1: Mass distribution of carbon in size fractions

Size fraction, mm		% Mass fraction
	+2,36	3,21
-2,36	+2,0	24,43
-2,0	+1,7	42,27
-1,7	+1,4	23,28
-1,4	+1,18	5,91
-1,18		0,89

90 per cent by mass of the total was contained in the -2,36 +1,4 mm size range, and the three size fractions in this range were used during the testwork.

The carbon particles are irregularly-shaped granules. Two options are available to determine the carbon particle sizes.

* The first is the less scientific but easier method. The carbon particle diameter is taken to be the mean of the apertures of the two screens. Hence, the carbon in the size fraction -2,36 +2,0 mm is considered to have a particle diameter of 2,18 mm.

* The second method is to inspect the carbon by means of an image analyser, which is capable of giving ferret diameters and shape factors, as well as many other variables. Although available, the facility of an image analyser was not used on this project due to the problem of sample preparation. If carbon granules are laid on a flat surface, they will orientate themselves in such a manner as to show their maximum surface area. This is undesirable, as the images thus analysed would give diameters greater than they actually are. An alternative would be to set the carbon in a transparent block and cut sections from it. An understanding of stereology is required to analyse these two-dimensional planes and obtain three-dimensional data (Oosthuyzen[30]).

It was decided to define the particle diameter by means of the first method for the following reasons:

* The inherent difficulties in obtaining a realistic value via the image analyser.

* Using the mean of the apertures through which the carbon was screened requires the minimum of equipment, enabling the proposed dimensionless number correlations to be used without the need for an image analyser or an understanding of stereology. This facilitates the transfer of technology.

AREA OF CARBON

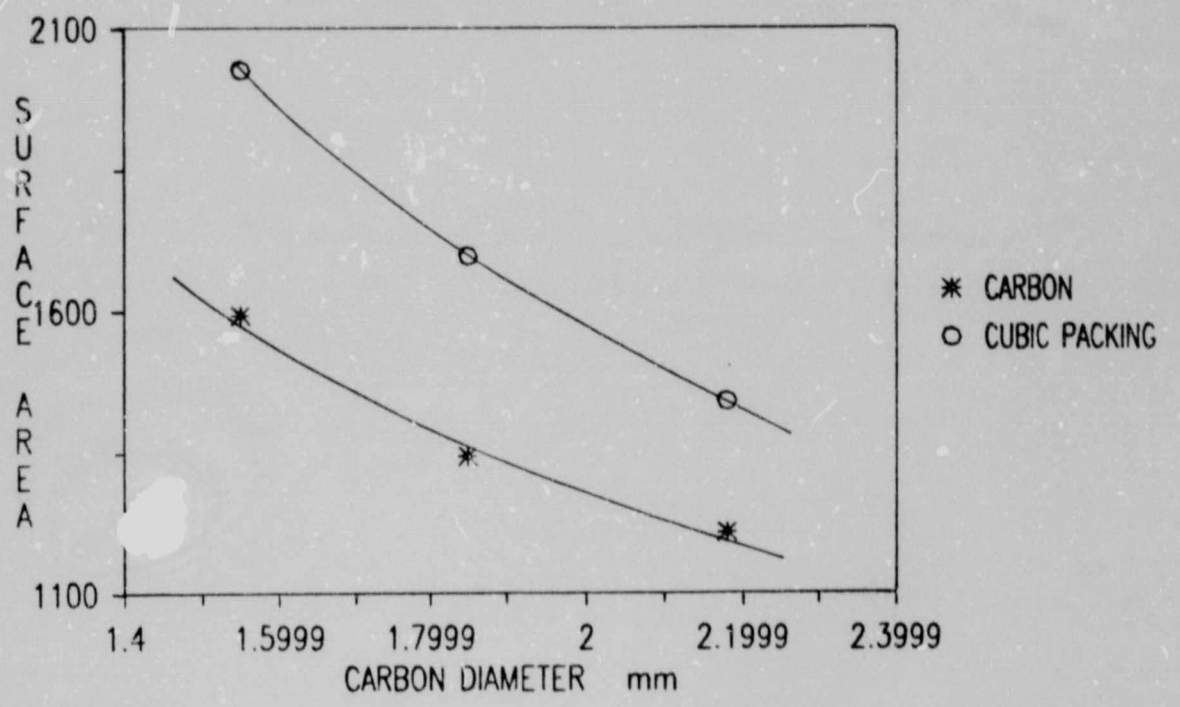
The method of ascertaining the area of the carbon in the three size fractions was as follows: water and carbon were added to a measuring cylinder and mixed. The carbon was allowed to settle and its wet-settled volume noted. The carbon was then dried and its mass taken. A sample of the dried carbon was taken, its mass determined, and the number of particles in the sample counted. The variables are the wet settled density and the number of particles per mass or volume. If the particles are assumed to be spherical and of a diameter the mean of the screen apertures through which they were sized, the surface area per mass or volume can be calculated. The data from these determinations are given in Table C2. A plot of the surface area of the carbon per cubic metre against the radius was drawn. Also plotted on the graph is the curve for surface area per cubic metre versus radius for perfect spheres in cubic packing (voidage 0,48). The plots, shown in Figure C3, follow a similar pattern, producing a degree of confidence in the assumptions.

As the number of granules per cubic metre is known, the volume of granules per cubic metre can be calculated. This figure provides an alternative method of obtaining the voidage.

TABLE C2 Results of tests on density and number of particles per volume or mass

Carbon size fraction	mm	-2,36 +2,0	-2,0 +1,7	-1,7 +1,4
Particle diameter	m	0,002 18	0,001 85	0,001 55
Particle radius	m	0,001 09	0,000 925	0,000 775
Wet settled density	kg m^{-3}	556	481	543
Number of particles per volume	No. m^{-3}	81 000 000	125 000 000	211 000 000
Number of particles per mass	No. kg^{-1}	146 000	260 000	388 000
Area of particles per volume	m^2 m^{-3}	1 209	1 344	1 593
Area of particles per mass	m^2 kg^{-1}	2,198	2,444	2,896

FIG C2 SURFACE AREA OF CARBON



APPENDIX D

PROGRAM TO CALCULATE THE DIMENSIONLESS NUMBERS,

AND TO CHECK THE FIT OF DIFFERENT CORRELATIONS

PROGRAM TO CALCULATE THE DIMENSIONLESS NUMBER AND TO CHECK THE FIT OF DIFFERENT CORRELATIONS

This Appendix outlines the method of checking the fit of different dimensionless number correlations. In Section 1.5 the literature was reviewed regarding the linking of the mass transfer coefficients to the physical parameters of the system via dimensionless number correlations, and six general forms of the equations were identified. The equations link the Sherwood number to the Reynolds number and the Schmidt number, and are listed as equations 1-1 to 1-6. Linear plots are required to test which of the six equations best fit the experimental data. The linear plots were shown in Table 4.4. The program at the end of this Appendix follows the following pattern:

1 The data from each batch test are put into a data file as

$$k_l \quad d_p \quad D \quad \eta \quad \rho \quad u \quad \epsilon$$

2 The data are read and the dimensionless numbers calculated and outputted. The numbers calculated are

$$Re = \frac{d_p \cdot u \cdot \rho}{\eta}$$

$$Sh = \frac{k_l \cdot d_p}{D}$$

$$Sc = \frac{\eta}{D \cdot \rho}$$

3 The values for the required linear plots for the six equations were calculated, namely

$\ln (Re)$	versus	$\ln ((Sh - 2)/(Sc^{0,33}))$
$\ln (Re)$	versus	$\ln ((Sh \cdot \epsilon)/(Sc^{0,33}))$
$\ln ((1 - \epsilon)(Re))$	versus	$\ln ((Sh \cdot \epsilon)/(Sc^{0,33}))$
$\ln ((1 - \epsilon)(Re))$	versus	$\ln ((Sh - 2)/(Sc^{0,33}))$
$\ln (Re)$	versus	$\ln (Sh/Sc^{0,33})$
$\ln ((1 - \epsilon)(Re))$	versus	$\ln (Sh/Sc^{0,33})$

These values are put into six files as x ; y.

4 The slope, intercept and correlation coefficients for the six sets of x, y data are calculated by means of the equations from Draper and Smith[28] and outputted as

slope =

$$\frac{\Sigma x \cdot y - ((\Sigma x \cdot \Sigma y)/N)}{\Sigma x^2 - (\Sigma x)^2/N}$$

intercept =

$$\frac{\Sigma y - \text{slope } \Sigma x}{N}$$

correlation coefficient =

$$\frac{\Sigma \left(x - \frac{\Sigma x}{N}\right)\left(y - \frac{\Sigma y}{N}\right)}{\left[\Sigma \left(x - \frac{\Sigma x}{N}\right)^2\right]^{0,5}\left[\Sigma \left(y - \frac{\Sigma y}{N}\right)^2\right]^{0,5}}$$

```
C PROGRAM TO CALCULATE THE FIT OF DIFFERENT MASS TRANSFER CORRELATIONS
C
C TYPE TO SCREEN THE OPERATION OF THE PROGRAM
      TYPE 'DATA INPUTTED TO PROGRAM IN FORM'
      TYPE ' MTC , PART DIA , DIFF , VIS , DENS , LIN VEL , VOID '
      TYPE '
      TYPE 'THE PHYSICAL PARAMETERS ARE ACCEPTED'
      TYPE 'THE DIMENSIONLESS NUMBERS ARE CALCULATED '
      TYPE 'AND TYPED ON THE SCREEN'
      TYPE '
      TYPE 'THE FOLLOWING RELATIONSHIPS ARE CALCULATED'
      TYPE 'AND PUT INTO FILES AS DESIGNED'
      TYPE 'RELATIONSHIP NO 1    LN(Re)         Vs  LN(Sh-2/Sc**0.33) '
      TYPE 'RELATIONSHIP NO 2    LN(Re)         Vs  LN(Sh*e/Sc**0.33) '
      TYPE 'RELATIONSHIP NO 3    LN(Re*(1-e))   Vs  LN(Sh*e/Sc**0.33) '
      TYPE 'RELATIONSHIP NO 4    LN(Re*(1-e))   Vs  LN(Sh-2/Sc**0.33) '
      TYPE 'RELATIONSHIP NO 5    LN(Re)         Vs  LN(Sh/Sc**0.33)   '
      TYPE 'RELATIONSHIP NO 6    LN(Re*(1-e))   Vs  LN(Sh/Sc**0.33)   '
      TYPE 'FOR ALL RELATIONSHIPS THE IS SLOPE=q  INTERCEPT=LN(p) '
      TYPE '
      TYPE 'THE SLOPE INTERCEPT AND CORRELATION COEFFICIENT ARE THEN'
      TYPE 'CALCULATED FOR THE SIX RELATIONSHIPS'
      TYPE 'THE SLOPE INTERCEPT AND CORRELATION COEFF ARE THEN TYPED'
C ACCEPT AND OPEN INPUT AND OUTPUT FILES
      CALL IAF01 (16,'INPUT DATA FILE          : $')
      CALL IAF01 (17,'OUTPUT DATA FILE  NO 1   : $')
      CALL IAF01 (18,'OUTPUT DATA FILE  NO 2   : $')
      CALL IAF01 (19,'OUTPUT DATA FILE  NO 3   : $')
      CALL IAF01 (20,'OUTPUT DATA FILE  NO 4   : $')
      CALL IAF01 (21,'OUTPUT DATA FILE  NO 5   : $')
      CALL IAF01 (22,'OUTPUT DATA FILE  NO 6   : $')
C ACCEPT NO OF DATA POINTS
      ACCEPT 'NO OF DATA POINTS INPUT FILE  ',PD
C LOOP FOR CALCULATION OF DIMENSIONLESS NUMBERS AND RELATIONSHIPS
      DO 32 I=1,PD
C READ INPUT DATA
      READ FREE (16) CKL,DP,DF,VI,DE,VE,VO
C CALCULATE Sh Sc Re NUMBERS
      SH=(CKL*DP)/DF
      SC=VI/(DF*DE)
      RE=(DP*VE*DE)/VI
C TYPE TO SCREEN DIMENSIONLESS NUMBERS
      TYPE 'TEST NUMBER        ',I
      TYPE 'SHERWOOD NUMBER    ',SH
      TYPE 'SCHMIDT  NUMBER    ',SC
      TYPE 'REYNOLDS NUMBER    ',RE
      TYPE '                   '
C CALCULATE LN RELATIONSHIPS AS STATED ABOVE
      SHLN1=ALOG(((SH-2)/(SC**0.3333)))
      SHLN2=ALOG(((SH*VO)/(SC**0.3333)))
      SHLN3=ALOG((SH/(SC**0.3333)))
      RELN1=ALOG(RE)
      RELN2=ALOG((RE*(1-VO)))
C OUTPUT DATA INTO DESIGNED FILES
      WRITE FREE (17) RELN1,SHLN1,'&'
      WRITE FREE (18) RELN1,SHLN2,'&'
      WRITE FREE (19) RELN2,SHLN2,'&'
      WRITE FREE (20) RELN2,SHLN1,'&'
      WRITE FREE (21) RELN1,SHLN3,'&'
```

```
      WRITE FREE (22) RELN2,SHLN3,'&'
C CLOSE LOOP
 32   CONTINUE
C REWIND DATA FILES TO BEGIN LINEAR REGRESSION
      REWIND 17
      REWIND 18
      REWIND 19
      REWIND 20
      REWIND 21
      REWIND 22
C
C LINEAR REGRESSION
C
C THE SUMS ARE OBTAINED IN LOOPS AND THE SLOPE INTERCEPT CORRELATION
C COFF CALCULATED BY THE FOLLOWING EQUATIONS   NB (=SUM
C SLOPE=((XY-((X.(Y)/N)/((X**2-((X)**2-((X)**2/N)
C INTERCEPT=(Y-SLOPE.(X/N
C CORR COFF=((X-(X/N)(Y-(Y/N)/(((X-(X/N)**2)**.5)(((Y-(Y/N)**2)**.5)
C
C DIMENSION ARRAYS
      DIMENSION X(25),Y(25)
C READ DATA IN LOOP TO OBTAIN SUMS
      DO 100 I=1,PD
      READ FREE (17) X(I),Y(I)
 100  CONTINUE
C CALL SUBROUTINE TO CALCULATE THE SLOPE INTERCEPT & CORR COEFF
      CALL LINREG(PD,X,Y,SLO,P,CC)
C TYPE TO SCREEN REGRESSION DATA
      TYPE 'RELATIONSHIP NO 1   LN(Re)       Vs  LN(Sh-2/Sc**.33)'
      TYPE '                    IE    Sh=2+p(Re**q)(Sc**.33)'
      TYPE ' p ',P,' q ',SLO,' CC ',CC
      TYPE '    '
C READ DATA IN LOOP TO OBTAIN SUMS
      DO 110 I=1,PD
      READ FREE (18) X(I),Y(I)
 110  CONTINUE
C CALL SUBROUTINE TO CALCULATE THE SLOPE INTERCEPT & CORR COEFF
      CALL LINREG(PD,X,Y,SLO,P,CC)
C TYPE TO SCREEN REGRESSION DATA
      TYPE 'RELATIONSHIP NO 2   LN(Re)       Vs  LN(Sh*e/Sc**.33)'
      TYPE '                    IE    Sh=p/e(Re**q)(Sc**.33)'
      TYPE ' p ',P,' q ',SLO,' CC ',CC
      TYPE '    '
C READ DATA IN LOOP TO OBTAIN SUMS
      DO 120 I=1,PD
      READ FREE (19) X(I),Y(I)
 120  CONTINUE
C CALL SUBROUTINE TO CALCULATE THE SLOPE INTERCEPT & CORR COEFF
      CALL LINREG(PD,X,Y,SLO,P,CC)
C TYPE TO SCREEN REGRESSION DATA
      TYPE 'RELATIONSHIP NO 3   LN(Re)(1-e)  Vs  LN(Sh*e/Sc**.33)'
      TYPE '                    IE    Sh=p*(1-e)**q/e(Re**q)(Sc**.33)'
      TYPE ' p ',P,' q ',SLO,' CC ',CC
      TYPE '    '
C READ DATA IN LOOP TO OBTAIN SUMS
      DO 130 I=1,PD
      READ FREE (20) X(I),Y(I)
 130  CONTINUE
C CALL SUBROUTINE TO CALCULATE THE SLOPE INTERCEPT & CORR COEFF
```

```
      CALL LINREG(PD,X,Y,SLO,P,CC)
C TYPE TO SCREEN REGRESSION DATA
      TYPE 'RELATIONSHIP NO 4   LN(Re)(1-e)  Vs  LN(Sh-2/Sc**.33)'
      TYPE '                    IE    Sh=2+p((1-e)Re**q)(Sc**.33)'
      TYPE ' p ',P,' q ',SLO,' CC ',CC
      TYPE '    '
C READ DATA IN LOOP TO OBTAIN SUMS
      DO 140 I=1,PD
      READ FREE (21) X(I),Y(I)
 140  CONTINUE
C CALL SUBROUTINE TO CALCULATE THE SLOPE INTERCEPT & CORR COEFF
      CALL LINREG(PD,X,Y,SLO,P,CC)
C TYPE TO SCREEN REGRESSION DATA
      TYPE 'RELATIONSHIP NO 5   LN(Re)        Vs  LN(Sh/Sc**.33)'
      TYPE '                    IE    Sh=p(Re**q)(Sc**.33)'
      TYPE ' p ',P,' q ',SLO,' CC ',CC
      TYPE '    '
C READ DATA IN LOOP TO OBTAIN SUMS
      DO 150 I=1,PD
      READ FREE (22) X(I),Y(I)
 150  CONTINUE
C CALL SUBROUTINE TO CALCULATE THE SLOPE INTERCEPT & CORR COEFF
      CALL LINREG(PD,X,Y,SLO,P,CC)
C TYPE TO SCREEN REGRESSION DATA
      TYPE 'RELATIONSHIP NO 6   LN(Re)(1-e)  Vs  LN(Sh/Sc**.33)'
      TYPE '                    IE    Sh=p((1-e)Re**q)(Sc**.33)'
      TYPE ' p ',P,' q ',SLO,' CC ',CC
      TYPE '    '
C END PROGRAM
      STOP
      END
```

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