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CALCULATED PHYSICAL PROPERTIES
OF SRP WASTE GLASSES USING FRIT 131

Introduction and Summary

Several physical properties of SRP waste glass which are important for melter design have not been determined experimentally. In this report, the heat capacity, thermal conductivity, density, and refractive index are detailed for the various types of waste (ie. Composite, High-Fe, and High-Al) plus frit 131. Included is a discussion of the calculation methods and a listing of waste compositions in both weight and mole percent.

Discussion and Results

Composition

The composition of SRP synthetic waste and of frit 131 are specified in DPST-80-607 (John Plodinec, 10/24/80)[1]. From these,

the glass compositions were calculated in both weight percent and mole percent. It was assumed that all of the coal in the sludge would be burned and removed as off-gas, and that the molecular weight of decomposed Zeolite was the same as the average molecular weight of the glass. (This led to a simple trial and error solution where the average molecular weight is assumed.) The following formula apply.

I. For weight fraction.

Basis = 100g. of frit + sludge.

$$X_a = \frac{(Y_f Z_{af} + Y_s Z_{as})}{(1.0 - 1.0\ (Y_s)(Z_{cs}))} \qquad (1)$$

where:

X_a = weight fraction of compoennt a in glass.

Y_f = weight fraction of frit in glass.

Y_s = weight fraction of sludge in glass.

Z_{af} = weight fraction of component a in frit.

Z_{as} = weight fraction of component a in sludge.

Z_{cs} = weight fraction of coal in sludge.

II. For mole fraction:

Basis: 1.0g of glass.

$$\eta_i = \frac{N_i}{N_t} \qquad (2)$$

$$N_i = 1.0g \quad (\frac{Xi}{Mi}) \qquad (3)$$

$$N_t = \sum_{i=1}^{n} \frac{X_i}{M_i} \qquad (4)$$

where:

η_i = mole fraction of component i.

N_i = moles of i in one gram of glass.

N_t = total moles in one gram of glass.

X_i = weight fraction of component i in glass.

n = total number of components.

TABLE 1 - COMPOSITIONS OF SRP WASTE GLASS

Component	Composite (TDS-3A)		High Iron		High - Al.	
	Wt. Frac.	Mole Frac.	Wt. Frac.	Mole Frac.	Wt. Frac.	Mole Frac.
SiO_2	0.422	0.4670	0.418	0.466	0.423	0.4596
B_2O_3	0.104	0.0990	0.104	0.1001	0.104	0.0975
Na_2O	0.134	0.1440	0.143	0.1546	0.140	0.1474
Li_2O	0.040	0.0890	0.040	0.0897	0.040	0.0874
CaO	0.011	0.0130	0.012	0.0143	0.003	0.0035
MgO	0.014	0.023	0.014	0.0230	0.014	0.0227
TiO_2	0.007	0.0058	0.007	0.0059	0.007	0.0057
La_2O_3	0.004	0.0008	0.004	0.0008	0.004	0.0008
ZrO_2	0.004	0.0022	0.004	0.0022	0.004	0.0021
Fe_2O_3	0.142	0.0592	0.177	0.0743	0.041	0.0168
MnO_2	0.041	0.0314	0.012	0.0093	0.034	0.0255
Zeolite	0.031	0.0299	0.029	0.0290	0.031	0.0302
Al_2O_3	0.029	0.0192	0.004	0.0026	0.148	0.0948
NiO	0.017	0.0151	0.030	0.0269	0.006	0.0052
Na_2SO_4	0.002	0.00937	0.002	0.0009	0.002	0.0009

Heat Capacity

Several methods for estimating the heat capacity of glasses are available.[2] Of those considered, the method of Scharp and Gunthner was chosen due to it's wide temperature range (0 to 1300°C) and it's high accuracy. Agreement of ± 1 % is common if factors are available for all of the component oxides. Unfortunately, only 94% of the waste glass could be considered. The method is summarized below.

$$C_{p\ mean.} = \left(\frac{at + c}{0.00146t + 1}\right) \qquad (5)$$

$$C_{p\ true} = \left(\frac{at + C_{p\ mean}}{0.00146t + 1}\right) \qquad (6)$$

$$a = \frac{\sum_{i=1}^{n} a_i X_i}{\sum_{i=1}^{n} X_i} \qquad (7)$$

$$c = \frac{\sum_{i=1}^{n} c_i X_i}{\sum_{i=1}^{n} X_i} \qquad (8)$$

Where:

$C_{p\ mean}$ = Mean heat capacity referenced to $0^{\circ}C$ $(\frac{cal}{gm^{\circ}C})$

$C_{p\ true}$ = True heat capacity $(\frac{cal}{gm^{\circ}C})$

t = Temperature, $^{\circ}C$

X_i = weight fraction of component i.

a_i & c_i = factors for component i.

n = number of oxides for which factors are available.

Values of a_i & c_i are listed in Appendix A. Using equations 5 - 8, values of $C_{p\ mean}$ and $C_{p\ true}$ were calculated. They are presented in Table 2.

Density

The densities of SRP wastes have been measured at low temperature.[3] In addition, the linear expansion has been measured up to the "melting point" of the glass.[4] Figures 1-3 show this expansion as a function of temperature. The linear coefficient of expansion is defined as $\alpha = \frac{1}{L}\frac{DL}{dT}$. From the graphs, it is evident that α is nearly constant in the low temperature range, and then takes a sharp increase at approximately 450 C. This point is called the transition temperature, Tr, and is given on the plots as GTT. Past Tr, α is again constant. The "tail" in Figures 1-3 is due to the inability of the dilatometer to operate with molten sam-

TABLE 2. HEAT CAPACITIES FOR SRP WASTE GLASS WITH FRIT 131

Temperature °C	Composite TDS-3A		High-Fe		High-Al	
-	C_{pm} $\frac{Cal}{gm\,^0C}$	C_{pt} $\frac{Cal}{gm\,^0C}$	C_{pm}	C_{pt}	C_{pm}	C_{pt}
0	0.186	0.186	0.168	0.168	0.173	0.173
100	0.213	0.237	0.198	0.224	0.201	0.225
200	0.234	0.271	0.221	0.262	0.223	0.261
300	0.251	0.296	0.239	0.289	0.240	0.286
400	0.264	0.314	0.254	0.309	0.254	0.305
500	0.276	0.328	0.267	0.324	0.266	0.319
600	0.285	0.338	0.277	0.335	0.275	0.330
700	0.293	0.346	0.286	0.345	0.284	0.339
800	0.300	0.353	0.294	0.348	0.291	0.346
900	0.307	0.359	0.301	0.358	0.298	0.352
950	0.309	0.361	0.304	0.361	0.300	0.354
1000	0.312	0.363	0.307	0.363	0.303	0.356
1025	0.313	0.364	0.308	0.364	0.304	0.357
1050	0.315	0.365	0.309	0.365	0.306	0.358
1075	0.316	0.366	0.311	0.366	0.307	0.359
1100	0.317	0.367	0.312	0.367	0.308	0.360
1125	0.318	0.368	0.313	0.368	0.309	0.361
1150	0.319	0.369	0.314	0.369	0.310	0.362
1175	0.320	0.369	0.316	0.370	0.312	0.363
1200	0.321	0.370	0.317	0.371	0.313	0.364
1250	0.323	0.372	0.319	0.373	0.315	0.365
1300	0.325	0.373	0.321	0.374	0.317	0.366

ples. According to Dr. L. D. Pye of Alfred University, it is reasonable to assume that α for the "liquid" glass is the same as α for the "solid" glass above the transition temperature.[5] This provides a route for calculating the glass density up to operating temperatures, as outlined below.

1. Obtain α from thermal expansion plot. (α for T_o up to T_r = LOW CTE, α for T_r up to operating temperature = HIGH CTE)

2. Calculate volumetric coefficient of expansion;

   $\beta m \cong 3\alpha$

3. Calculate the denisty at $T = To + \Delta T$ from:

$$\beta_m = \frac{\rho_o^2 - \rho^2}{2\rho_o\rho} \quad \frac{1}{\Delta T}$$

Where: ρ_o = Density at T_o

ρ = Density at T

$\Delta T = (T - T_o)\ ^0C$

β_m = mean coefficient of volumetric expansion, $1/^0C$.

The density is calculated up to T_r using $\rho_o = \rho_{expt.}$ and α = LOW CTE. Past T_r, ρ is calculated using $\rho_o = \rho_r$ and α = HIGH CTE. Unfortunately, data for high-Al and high-Fe wastes are available only for 20% loading. However, comparison of expansion data for composite waste at 20% and 28% loading shows a difference of 5% in α for $T<T_r$; and 15% for $T>T_r$. In

light of the small changes in density, this difference may be neglected. The results of these calculations are given in Table 3.

TABLE 3: DENSITIES OF SRP WASTE GLASSES

TEMP. °C	TDS-3A g/cm³	High Fe 8/cm³	High Al gm/cm³
20 C	2.750	2.820	2.60
100	2.743	2.813	2.593
200	2.735	2.804	2.585
300	2.726	2.795	2.577
400	2.718	2.786	2.569
* $T_r \simeq 450$	2.713	2.781	2.565
500	2.672	2.721	2.520
600	2.575	2.607	2.432
700	2.481	2.499	2.348
800	2.391	2.396	2.266
900	2.305	2.298	2.188
1000	2.223	2.204	2.113
1025	2.202	2.181	2.094
1050	2.183	2.159	2.076
1075	2.163	2.137	2.058
1100	2.143	2.115	2.040
1125	2.124	2.093	2.023
1150	2.105	2.072	2.006
1175	2.086	2.051	1.988
1200	2.068	2.031	1.971

* Since T_r is different for each type, its exact value is omitted in this table. See Figures 1-3 for listings. (under GTT=)

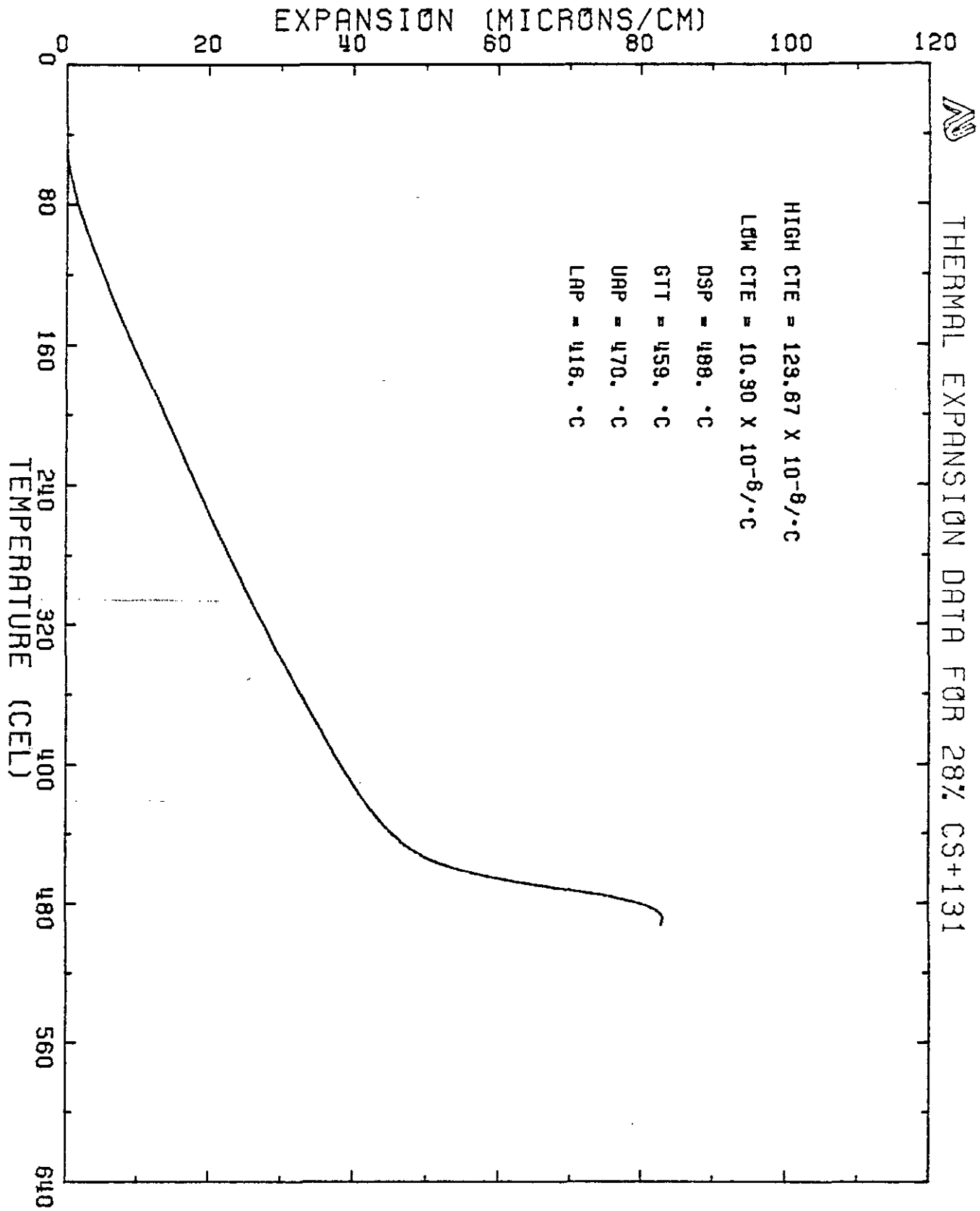
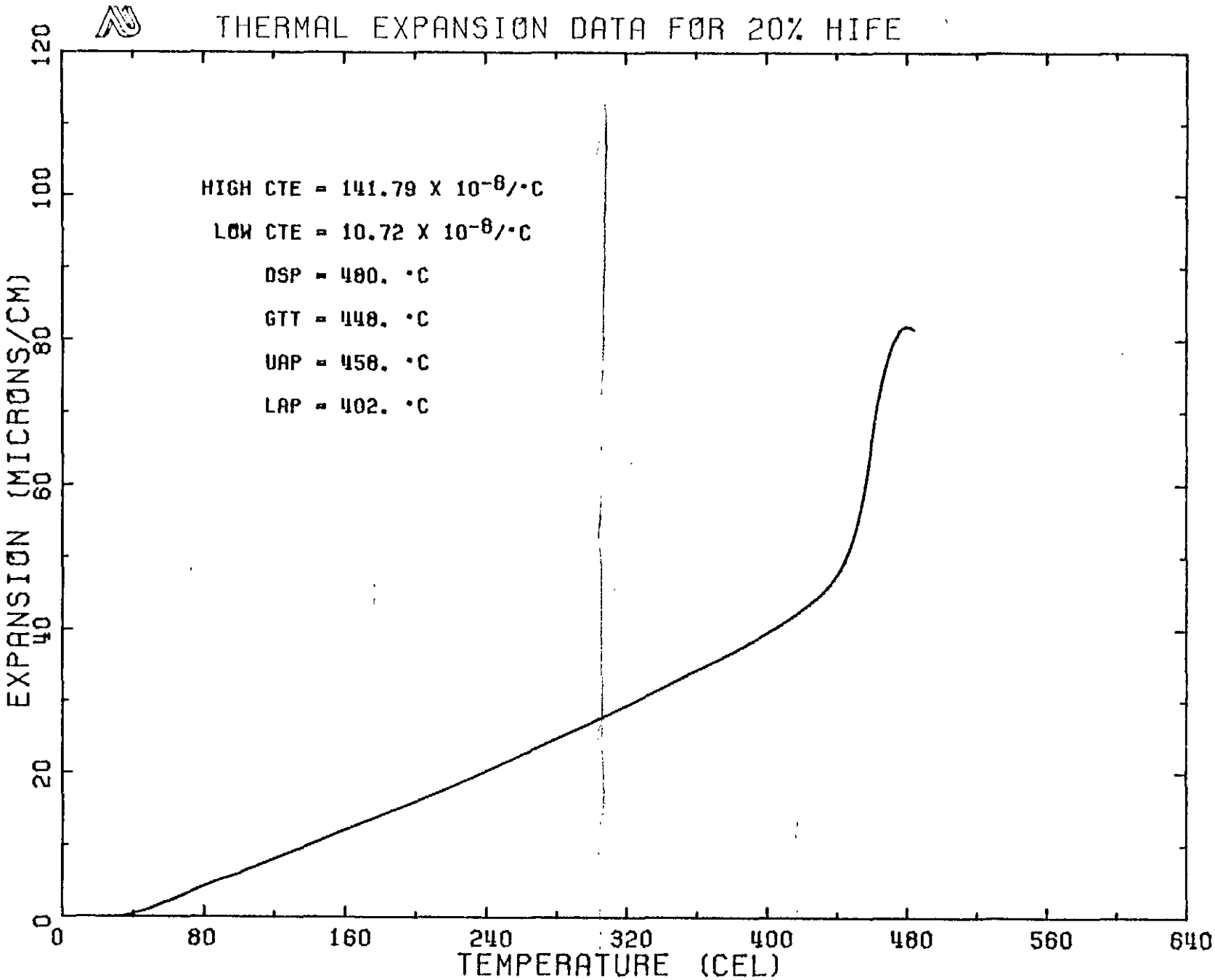

THERMAL EXPANSION DATA FOR 28% CS+131
HIGH CTE = 129.67 X 10-6/°C
LOW CTE = 10.90 X 10-6/°C
DSP = 488. °C
GTT = 459. °C
UAP = 470. °C
LAP = 416. °C
EXPANSION (MICRONS/CM)
0
20
40
60
80
100
120
TEMPERATURE (CEL)
0
80
160
240
320
400
480
560
640

FIG. I


THERMAL EXPANSION DATA FOR 20% HIFE
HIGH CTE = 141.79 X 10^-6/°C
LOW CTE = 10.72 X 10^-6/°C
DSP = 480. °C
GTT = 448. °C
UAP = 458. °C
LAP = 402. °C
EXPANSION (MICRONS/CM)
0
20
40
60
80
100
120
0
80
160
240
320
400
480
560
640
TEMPERATURE (CEL)

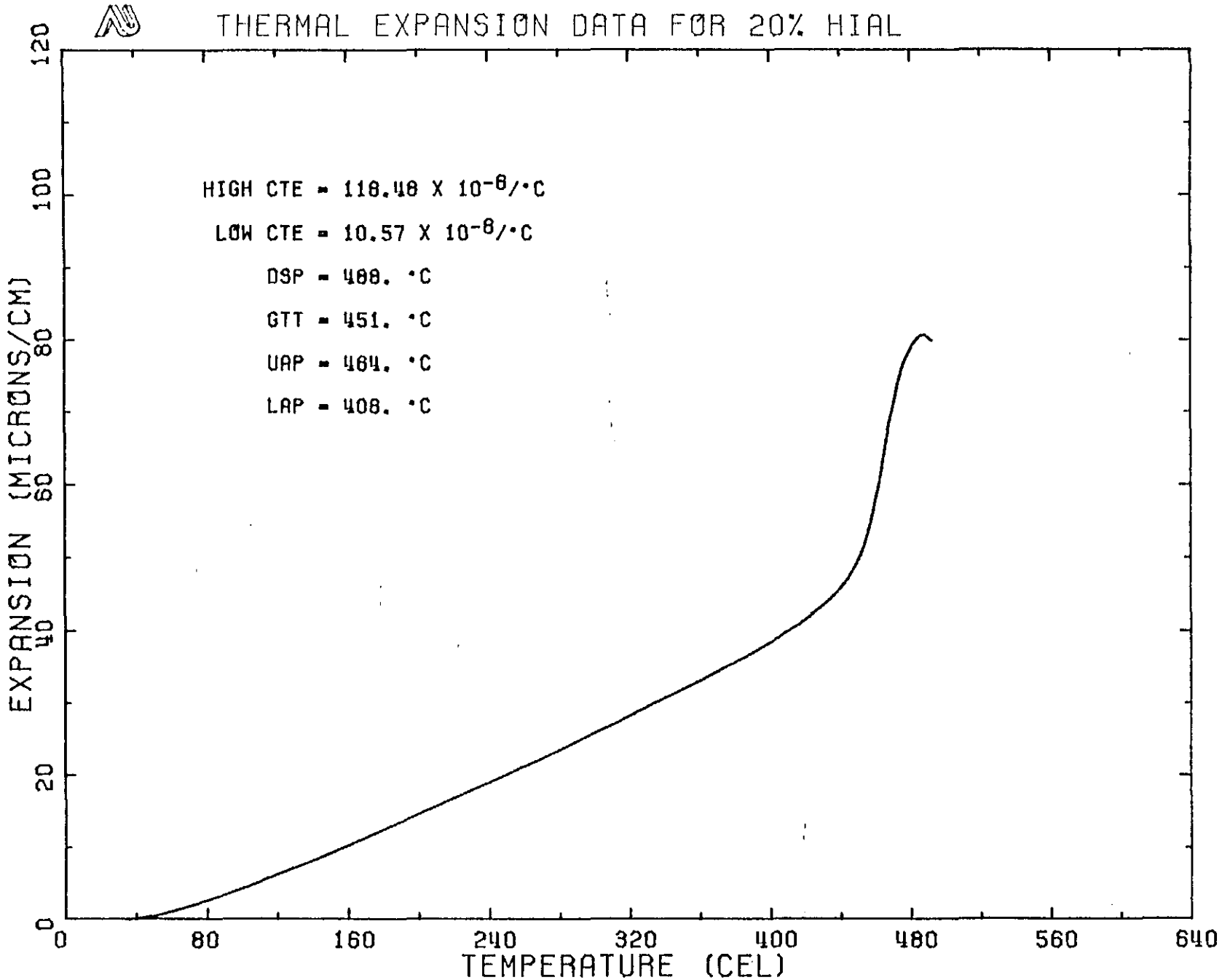

THERMAL EXPANSION DATA FOR 20% HIAL
HIGH CTE = 118.48 X 10-6/·C
LOW CTE = 10.57 X 10-6/·C
DSP = 488. ·C
GTT = 451. ·C
UAP = 464. ·C
LAP = 408. ·C
EXPANSION (MICRONS/CM)
0
20
40
60
80
100
120
0
80
160
240
320
400
480
560
640
TEMPERATURE (CEL)

FIG 3

Index of Refraction

The refractive index is a measure of the degree to which light is bent when passing through a material. It is rigorously defined as

$$\eta_\lambda = \frac{\sin \theta_1}{\sin \theta_2} \qquad (12)$$

Where

η_λ = index of refraction for light at a wave length of λ.

θ_1 = angle between incident beam and a normal plane.

θ_2 = angle between refracted beam and a normal plane. (See Figure 4)

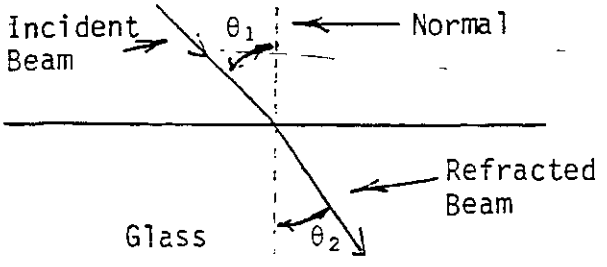


FIGURE 4

η is generally a weak function of λ and a very weak function of temperature. (approximately 1% change over 1000°C)[6]

The method of Huggins[7] was used to estimate η for SRP waste glasses due to the fact that coefficients are available for most components in SRP waste glass. The following formula is used.

$$\eta_D = 1 + \rho \frac{\sum_{i=1}^{n} \eta_{Di} X_i}{\sum_{i=1}^{n} X_i} \qquad (13)$$

Where

η_D = index of refraction at λ = 4860 Å *

X_i = weight fraction of component i

η_{Di} = coefficient for component i (See Appendix A)

ρ = density of the glass

n = number of components for which factors are available

The method is complicated by the fact that B_2O_3 exists in two forms, BO_4 (TETRA) and BO_3, and η_{Di} is different for each. The ratio of BO_4 to BO_3 was estimated by the method of Demkina[8] and is given in Appendix B. The following values of η_D were calculated from equation 13.

TABLE 4: INDEX OF REFRACTION FOR SRP WASTE GLASSES

TYPE OF WASTE	INDEX OF REFRACTION AT λ = 4860 Å
COMPOSITE TDS-3A	1.662
HIGH - Fe	1.6971
HIGH - Al	1.577

* See note on η_D in section on calculation of K_{rad} for significance of wavelength.

Thermal Conductivity

The thermal conductivity of a substance is a measure of the heat transferred through a substance by conduction, and is defined by Fourier's law:

$$q = k\nabla T \qquad (14)$$

where

q = heat flux (typically BTU/hr ft^2)

k = thermal conductivity - BTU/ hr ft^2 ^{0}F/ft

∇T = gradient of the temperature field (^{0}f/ft)

While k is defined in terms of heat transfer by conduction, it is common practice to represent heat transferred by radiation in terms of a "radiant conductivity". K_{rad}. Thus, the effective thermal conductivity is given by:

$$K_{eff} = K_{rad} + K_{true} \qquad (15)$$

Tooley[9] states that for optically thick glass (ie. radiant transmission is small).

$$K_{rad} = \frac{16\sigma\eta^2 T^3}{3a} \qquad 16)$$

Where: σ = Stephan-Boltzman constant

η = index of refraction*

T = absolute temperature

a = absorption coefficient = 1/F

F = mean free path

* Only η at λ = 4860 Å was available. The variation of η with λ will cause a small amount of error

Data is available for F as a function of Fe_2O_3 concentration and temperature.[10] Jenkins[11] fit the temperature data to the equation.

$$F = Gt^2 + Ht + J^{\dagger} \qquad (17)$$

Where:

t = temperature, ^0C

G, H, and J = Constants for a given Fe_2O_3 concentration.

G, H, & J were plotted against % Fe_2O_3 (See Appendix A for graphs). The values for SRP waste glass are summarized in Table 5.

TABLE 5

TYPE OF WASTE	G	H	J
COMPOSITE TDS-3A	-2.350 E-7	6.550 E-4	-0.3325
HIGH IRON	-2.09 E-7	5.83 E-4	-0.300
HIGH - Al	-4.75 E-7	13.09 E-4	-0.687

The radiative conductivity may now be calculated from

$$K_{rad} = 1290.2\ \sigma\ \eta^2\ (t + 273.15)^3\ F\ U(F) \qquad (16B)$$

Where

$$\sigma = 1.356 \times 10^{-2}\ \frac{CAL}{cm^2 sec^1 K^4}$$

t = temperature, ^0C

† This is an empirical fit, and will give negative values at low temperature. This is handled mathematically by the inclusion of the unit step function. (See Equation 16B).

$F = f(t)$ (Eqn. 17)

$U(F)$ = Unit step function

$U = 0$ for $F \leq 0.0$

$U = 1$ for $F > 0.0$

K_{rad} = BTU/hr ft^2 ^{0}F/ft

Results are given in Table 7.

The true thermal conductivity is estimated in the following manner.

$$K_{true} = \frac{241.9 \sum_{i=1}^{\eta} a_i X_i}{\sum_{i=1}^{\eta} X_i} \qquad (18)$$

where:

K_{true} = thermal conductivity - BTU/hr-ft^2 ^{0}F/ft

a_i = factor for ith oxide.

X_i = weight fraction of ith oxide

η = number of components for which factors are available.

Values of a_i are listed in Appendix A for 0°C and 100°C. K_{true} was calculated at these temperatures and is listed below.

TABLE 6

TYPE OF WASTE	K_{true} @ 0°C	K_{true} @ 100°C
COMPOSITE TDS-3A	0.4893	0.5616
HIGH - Fe	0.4670	0.5479
HIGH - Al	0.5389	0.5655

It was assumed that K_{true} is a linear function of temperature, since only two points are available.

$$K_{true}(t) = K_t(0^\circ C) + \left(\frac{K_{t100} - K_{to}}{100-0^\circ C}\right) t \qquad (19)$$

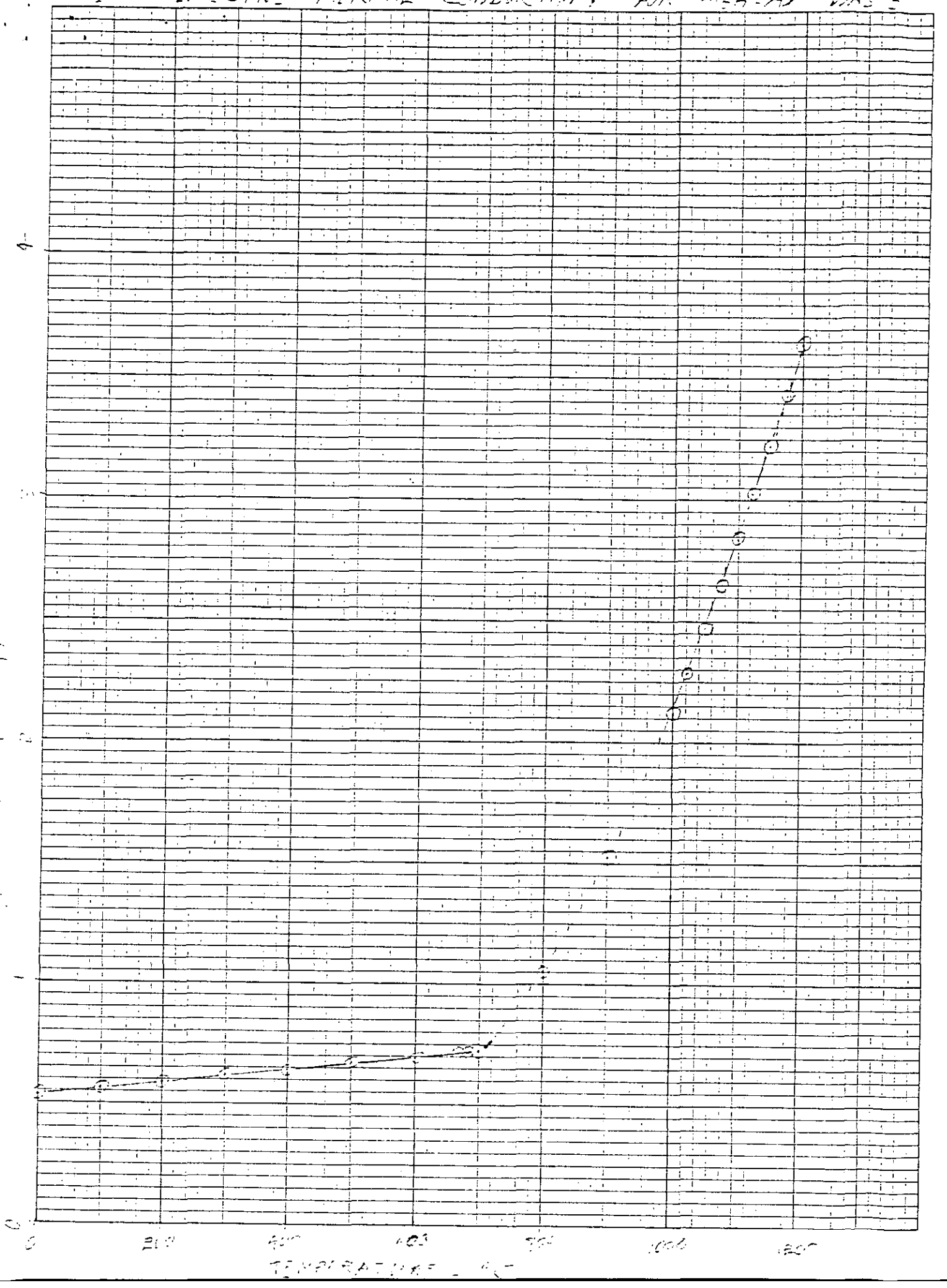
Equations 16 and 19 are then combined to give K_{eff}. The results are listed in Table 7 and plotted in figures 5-7. Note the change in functionality at $\simeq 700^\circ C$ when F becomes positive and radiation becomes a factor.

Conclusions

Due mainly to the lack of empirical coefficients for all of the glass components, the accuracy of many of the methods used herein is questionable. While this is unfortunate, no better data will be available until at least August of 1981, when a study of the high temperature glass properties is completed by Dr. L. D. Pye of Alfred University. Until then, care should be taken to include a sufficiently large safety factor when using these data for design purposes.

JPM:dhw

EFFECTIVE THERMAL CONDUCTIVITY FOR HIGH-AS WASTE



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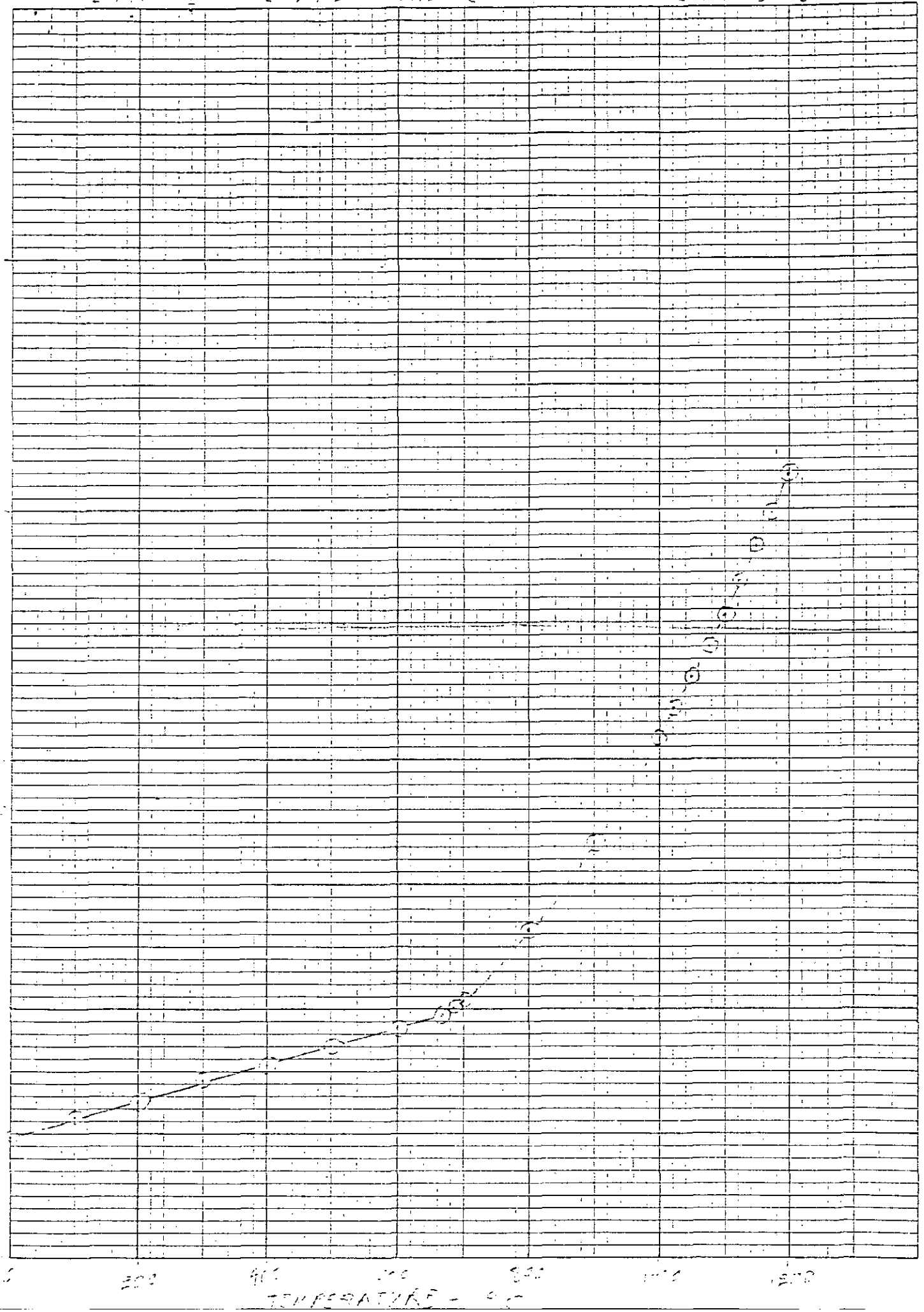

EFFECTIVE THERMAL CONDUCTIVITY FOR HIGH-Fe WASTE

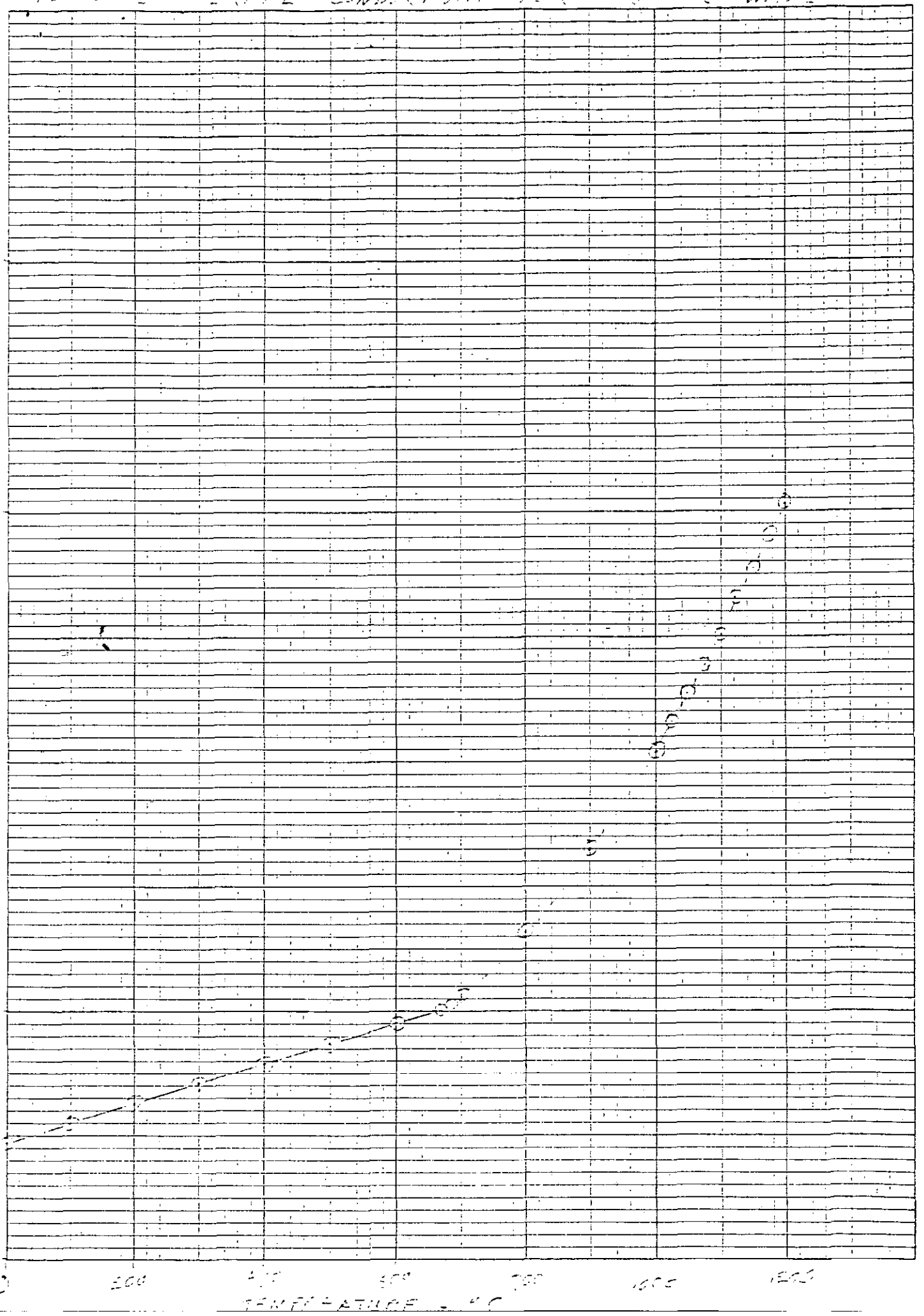

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TABLE 7: K_{rad}, & K_{eff} VERSUS TEMPERATURE AND COMPOSITION - BTU/hr ft^2 ^{0}F/ft

TEMP °C	COMPOSITE WASTE		HIGH-Fe		HIGH-Al	
--	K_{rad}	K_{eff}	K_{rad}	K_{eff}	K_{rad}	K_{eff}
0	0	0.4893	0	0.4670	0	0.5389
100	0	0.5616	0	0.5479	0	0.5655
200	0	0.6339	0	0.6288	0	0.5921
300	0	0.7062	0	0.7097	0	0.6187
400	0	0.7785	0	0.7906	0	0.6453
500	0	0.8508	0	0.8715	0	0.6719
600	0	0.9231	0	0.9524	0	0.6985
670	3.5 E-3	0.9737	0	1.009	0	0.7171
685	--	1.0105	5.70 E-3	1.026	0	0.7211
700	4.83 E-2	1.044	2.64 E-2	1.060	1.62 E-3	0.7251
800	2.45 E-1	1.313	2.03 E-1	1.317	3.02 E-1	1.054
900	5.20 E-1	1.660	4.51 E-1	1.646	7.47 E-1	1.525
1000	8.73 E-1	2.085	7.69 E-1	2.045	1.32	2.12
1025	9.72 E-1	2.200	8.60 E-1	2.156	1.48	2.29
1050	1.08	2.33	9.54 E-1	2.270	1.65	2.47
1075	1.18	2.45	1.05	2.39	1.83	2.65
1100	1.30	2.58	1.15	2.51	2.01	2.84
1125	1.41	2.71	1.27	2.64	2.19	3.03
1150	1.53	2.85	1.37	2.77	2.38	3.22
1175	1.65	2.99	1.48	2.90	2.58	3.43
1200	1.78	3.14	1.59	3.03	2.78	3.64

APPENDIX A

CONTENTS:

Factors for Heat Capacity	Page 1
Factors for Index of Refraction	Page 2
Coefficients for Mean Free Path	Page 3
Factors for True Thermal Conductivity	Page 7

FACTORS FOR USE IN THE HEAT CAPACITY EQUATIONS (7 & 8)

OXIDE	a_i	C_i
SiO_2	4.68 E-4	0.1657
B_2O_3	5.98 E-4	0.1935
Na_2O	8.29 E-4	0.2229
Li_2O *	1.24 E-3	0.4792
CaO	4.10 E-4	0.1709
MgO	5.14 E-4	0.2142
TiO_2 **	--	--
La_2O_3 **	--	--
ZrO_2 **	--	--
Fe_2O_3 *	5.40 E-4	0.1374
MnO_2 *	5.60 E-4	0.1384
Zeolite **	--	--
Al_2O_3	4.53 E-4	0.1765
NiO **	---	--
Na_2SO_4 **	--	--

* Values estimated from comparison with another empirical method.

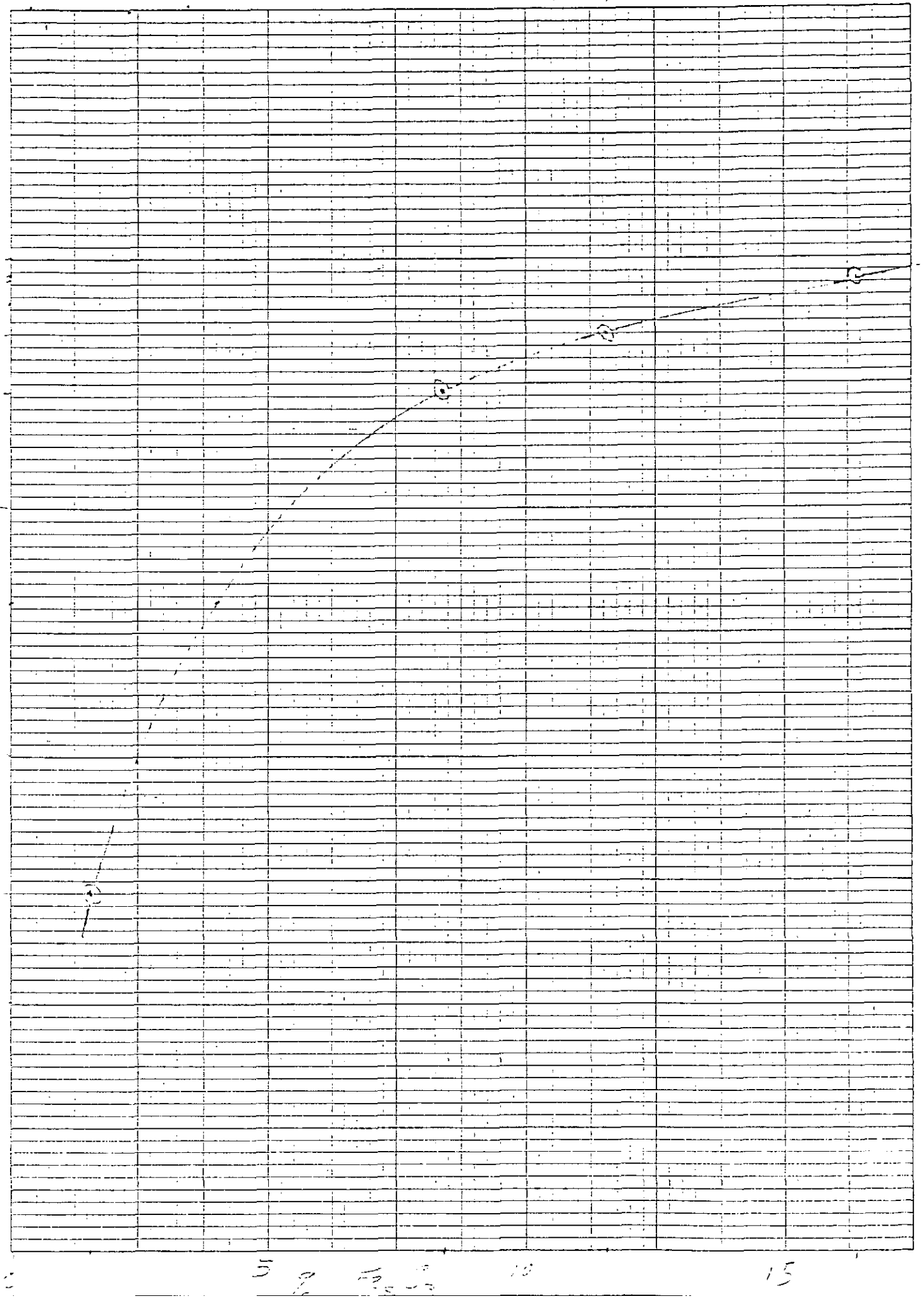
** No data available

FACTORS USED IN EQUATION 13 TO CALCULATE THE INDEX OF REFRACTION.

OXIDE	η_{Di}	$R_i \times 10^2$
SiO_2	0.2083	3.33
B_2O_3 (BO_4)	0.2150	4.3079
B_2O_3 (BO_3)	0.2530	4.3079
Na_2O	0.1941	1.6131
Li_2O	0.3080	3.3470
CaO	See below	1.7832
MgO	0.2100	2.48
TiO_2	0.3130	2.5032
La_2O_3	0.1460	0.9207
ZrO_2	--	--
Fe_2O_3	0.3920	1.8785
MnO_2	--	--
Al_2O_3	0.2038	2.9429
NiO	--	--
Na_2SO_4	--	--

$$\eta_{Di} \text{ for CaO} = 0.2257 + 0.4770 \times 10^{-4} \left(\frac{X\ CaO}{\Sigma R_i X_i} \right)$$

COEFFICIENT FOR CALCULATION OF MEAN FREE PATH



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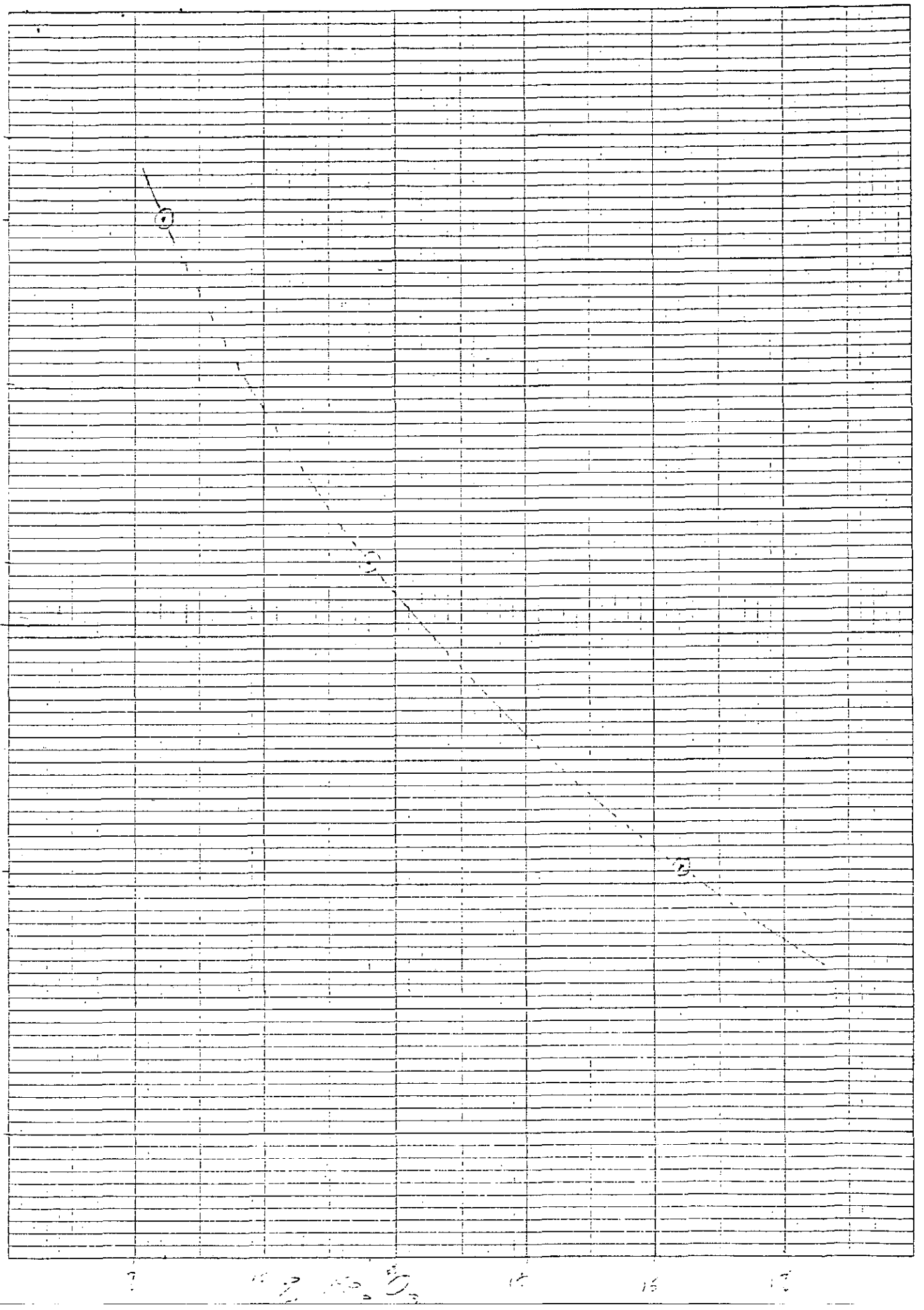
COEFFICIENT FOR CALCULATION OF MEAN FREE PATH



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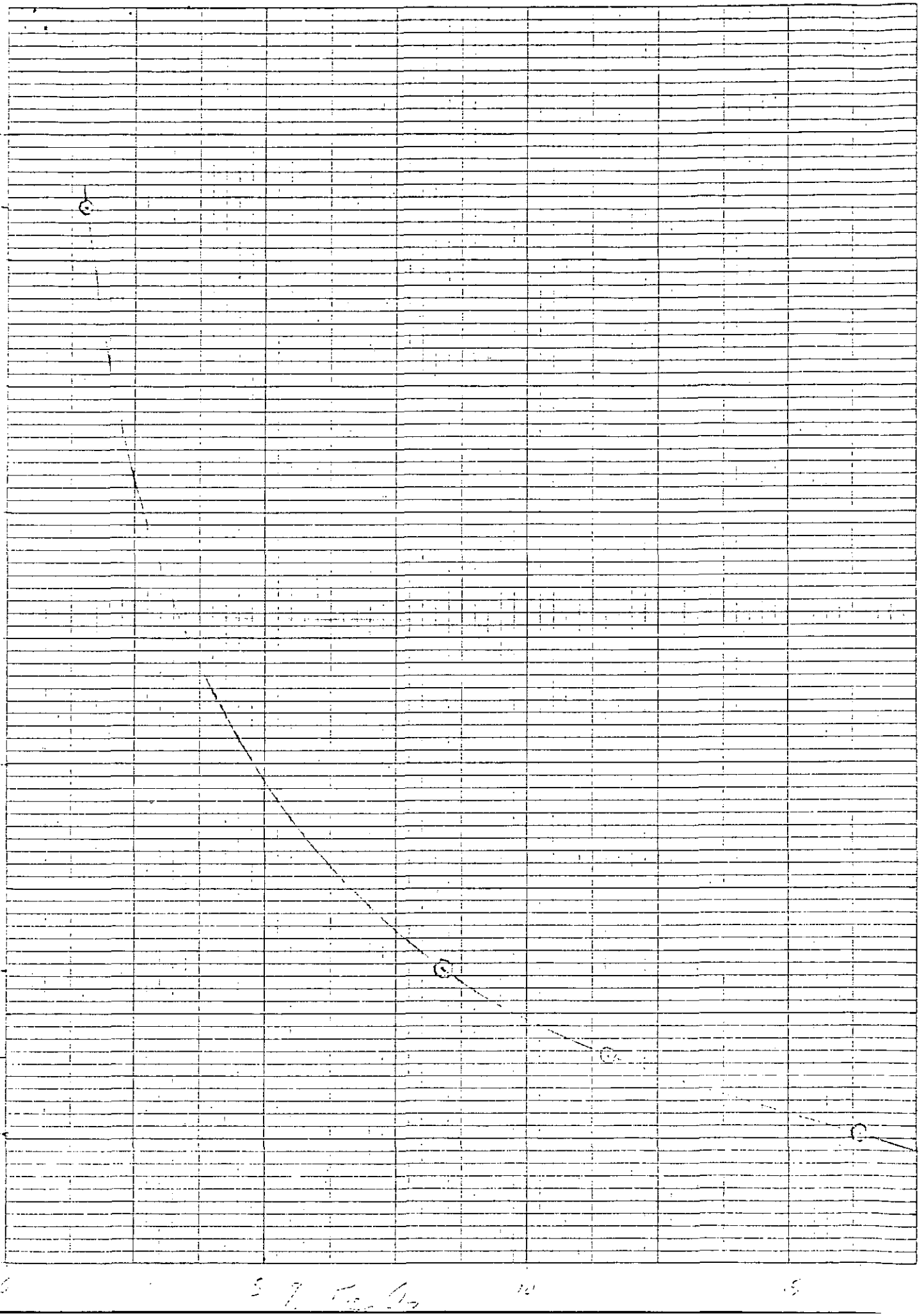
COEFFICIENT FOR CALCULATION OF MEAN FREE PATH



DIETZGEN CORPORATION
MADE IN U.S.A.
NO. 340R-10 DIETZGEN GRAPH PAPER
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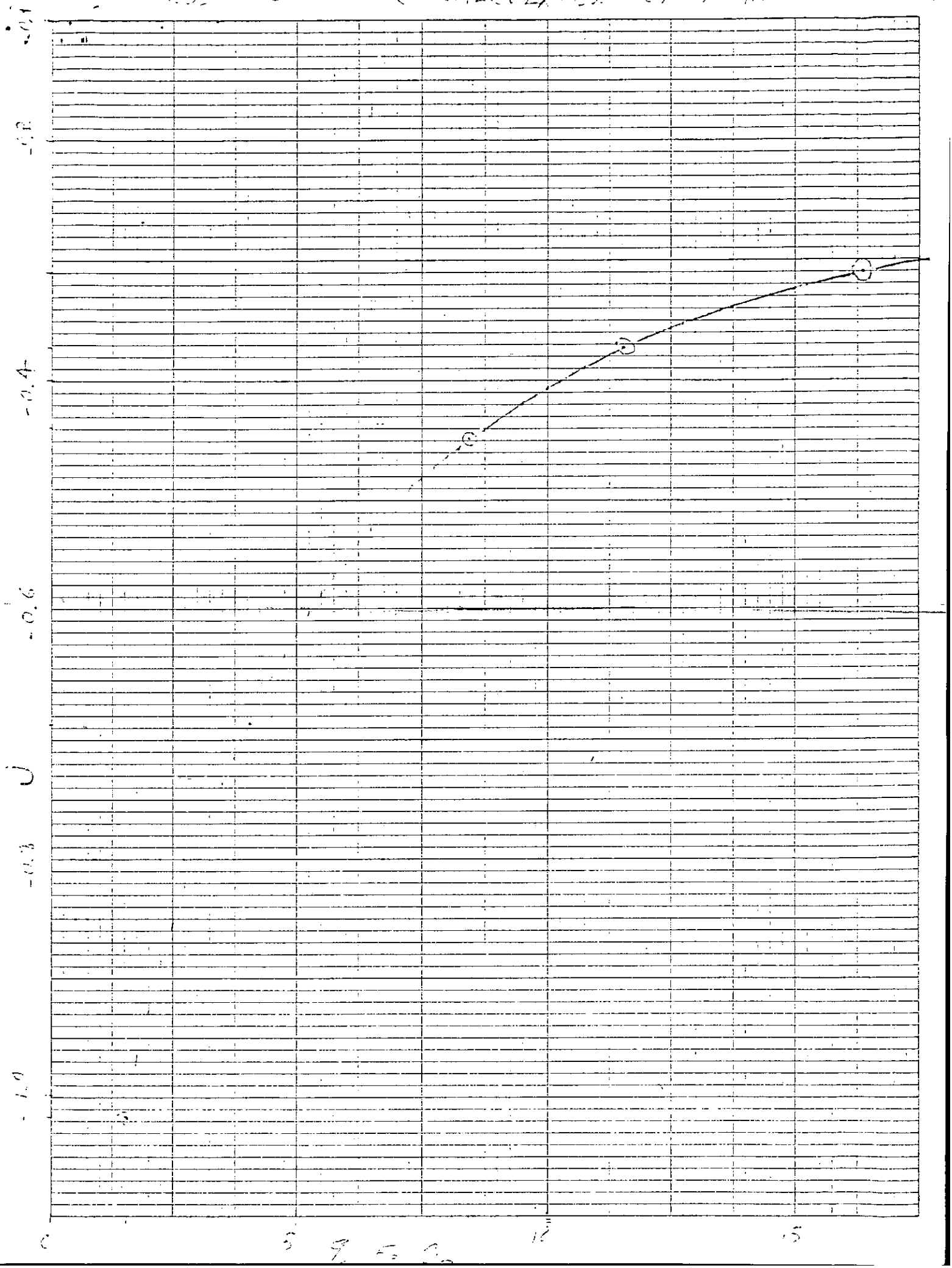

-0.4
-0.6
0
5
10
15
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FACTORS FOR ESTIMATING K_{true}[14]

OXIDE	a_i - 0°C	a_i - 100°C
SiO_2	3.07 E-3	3.44 E-3
Na_2O	-1.27 E-3	-0.67 E-3
B_2O_3	1.59 E-3	2.49 E-3
CaO	3.17 E-3	2.39 E-3
Fe_2O_3	1.90 E-3	1.73 E-3
Al_2O_3	3.72 E-3	2.14 E-3

APPENDIX B

CONTENTS:

Determination of B_2O_3 coordination by the method of Demkina[15].

In order to determine the amounts of B_2O_3 present in the form of B_2O_4-I (BO_4 tetrahedra) or B_2O_3-II (BO_3), Demkina proposed the following rules.

(1) First, the free silicone concentration, ΔS_iO , and the "oxygen number" are calculated.

$$\Delta SiO = \eta_{SiO_2} - 0.5\,(\eta_{ZrO} + \eta_{MgO}) + \eta_{BaO} + \eta_{CaO} + 2.0\,(\eta_{B_2O_3} + \eta_{pbO} + Na_2O) + 4\eta_{K_2O}$$

and

$$"O" = \frac{(\eta_{pbO} + \eta_{BaO} + \eta_{CaO} + \eta_{K_2O} + \eta_{Na_2O} - \eta_{Al_2O})}{\eta_{B_2O_3}}$$

where:

ΔSiO_2 = "free silicone concentration. (ie., the excess SiO_2 over the amount needed to form silicates of other cations present in the glass).

η_i = mole fraction of component i.

"O" = Oxygen number.

(2) When "O" $\geq$ 1.2 and $\Delta SiO_2 \geq 0$. all B_2O_3 is present in BO_4 form.

(3) When "O" < 1.2; the fraction $b_4 = BO_4/(BO_4 + BO_3)$ is given by

$$b_4 = (\text{"O"} - 0.20)$$

(4) When "O" $\geq$ 1.2 but $\Delta SiO_2 < 0$, $b_4 = (1/3)$.

Values of "O", ΔSiO_2, and b_4 for SRP waste glass are given below.

TYPE OF WASTE	"O"	ΔSiO_2	b_4
COMPOSITE	1.39	0.511	1.0
HIGH - Fe	1.64	0.980	1.0
HIGH - Al	0.5759	0.9416	0.3759

1. M. J. Plodinec, *Physical Properties of Glasses for Large-Scale Vitrification Tests.* Internal report, DPST-80-607, Savannah River Laboratory, E. I. DuPont de Nemours & Co., Aiken, S.C. (October 24, 1980).

2. M. A. Matveev, G. M. Matveev, B. N. Frenkel, *Calculation and Control of Electrical, Optical, and Thermal Propertie of Glass*, Ordentlich, Holon Israel, p 50 (1975).

3. Ibid, 1

4. L. D. Pye.

5. L. D. Pye, Personal Conversation, November 20, 1980.

6. F. V. Tooley, *The Handbook of Glass Manufacture*, Books fo Industry, Inc., New York N.Y., Vol. II. p 969 (1979).

7. Ibid, 2, p.41.

8. Ibid, 2, p.32.

9. Ibid, 6, Vol I, p. 219 (1974).

10. Ibid, 9.

11. W. J. Jenkins, *Models of Physical Properties of SRP Wast Glass*, Internal Report, DPSTD-77-13-3, Savannah River La E. I. DuPont de Nemours & Co., Aiken, S.C. Appendix 15,4

12. Ibid, 2, p.50.

1. M. J. Plodinec, *Physical Properties of Glasses for Large-Scale Vitrification Tests.* Internal report, DPST-80-607, Savannah River Laboratory, E. I. DuPont de Nemours & Co., Aiken, S.C. (October 24, 1980).

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12. Ibid, 2, p.50.