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UNIVERSITY OF MICHIGAN  
ANN ARBOR

Final Report

DETERMINATION OF THE LOW-TEMPERATURE HEAT CAPACITY  
OF VITREOUS SILICON DIOXIDE, OR QUARTZ, AND OF CRISTOBALITE

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I would like to express my appreciation to the several graduate students who assisted in these measurements, to Mrs. Emilia Martin who ably handled the calculations involved, and to Dr. Alvin F. Beale, Jr., who collaborated in making the quartz measurements.

This work was favored by stimulating discussions with Professor K. Fajans and by the splendid scientific cooperation of the Owens Illinois Glass Company staff. Mr. S. W. Barber's sincere, generous, and thorough collaboration made possible the procurements of exceptionally well characterized samples, an important aspect of the measurements.

DETERMINATION OF THE LOW-TEMPERATURE HEAT CAPACITY  
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The purpose of this report is to present a brief, formal summary of the measurements on the low-temperature heat capacity of two samples of vitreous silica and samples of alpha quartz and cristobalite. The full scientific summary will be in the form of journal papers, and since this end cannot be achieved without the critical endeavors of Professor K. Fajans (at present in Europe), Mr. S. W. Barber, and other scientists, the present report is made terse and factual to minimize cost and time--and hence delay in the actual paper. It is intended essentially as an interim document for the convenience of the Owens Illinois Glass Company and for the files of the Institute. Such matters as the preparation, identification, and analysis of the samples are not detailed here since this portion of the work was performed or expedited by the Company and are already, therefore, more directly available to them.

APPARATUS

The heat-capacity determinations were made with our adiabatic calorimetric cryostat<sup>1</sup> (Laboratory Designation, Mark I) shown in Fig. 1. Liquid or solid nitrogen and liquid helium were used as the low-temperature refrigerants.

The sample was contained in the gold plated, copper calorimeter<sup>2</sup> (Laboratory Designation W-5) shown in Fig. 2. Determinations of the heat capacity were made by observing the temperature change produced by the introduction of a measured quantity of electrical energy.

Temperatures were measured with a capsule-type platinum resistance thermometer (Laboratory Designation A-1) contained in a reentrant well in the calorimeter. A 160-ohm constantan heater was wound on a cylindrical copper tube surrounding the resistance thermometer. The thermometer was calibrated against the temperature scale of the National Bureau of Standards<sup>2</sup>

from 14 to 373°K. Below 14°K the scale was obtained by fitting the equation  $R = A + BT^2 + CT^5$  to the resistance at the boiling point of helium and to the resistance and  $dR/dT$  and 14°K. It is believed that the temperature scale agrees with the thermodynamic scale within 0.1° from 4 to 14°K, within 0.03° from 14 to 90°K, and within 0.05° from 90 to 373°K.

The thermometer resistance and the power input were measured with an autocalibrated White double-potentiometer, calibrated resistances, and calibrated standard cells. An electric timer operated by a calibrated tuning fork and an amplifier was automatically started at the beginning of the heating period and stopped at the end.

#### EXPERIMENTAL CONDITIONS

The masses ( in vacuo) of the various samples run in the calorimeter are indicated below in grams:

Quartz	116.1130
Cristobalite	90.5914
1070° vitreous silica	105.6365
1300° vitreous silica	102.9248

The quartz was in the form of crystalline fragments (about 5 mesh) the cristobalite fine crystals (100 to 150 mesh), and the vitreous silica in the form of rods about 5 mm in diameter and approximately the length of the calorimeter. A pressure of 3 to 4 cm of pure helium gas at 25°C was used to provide thermal conduction within the calorimeter.

The heat capacity of the calorimeter without sample has now been determined on three separate occasions and hence may be considered well established. As a final test of the accuracy, precision, and reliability of the instrument, the heat capacity of a sample of benzoic acid provided by the National Bureau of Standards was measured. Our determinations are in excellent accord with theirs.

EXPERIMENTAL RESULTS

The measured values of the molal heat capacities are given in Tables I through IV. These data are expressed in terms of the defined thermochemical calorie, which is equal to 4.1840 absolute joules. A small correction for curvature (to convert finite increments,  $\Delta H/\Delta T$ , to the derivative,  $(\partial H/\partial T) = C_p$ , has been applied. Since the data were obtained under only a few cm of helium pressure, they approximate  $C_p$  values very closely. The heat capacity versus temperature curve for quartz is presented in Fig. 3. Within the sensitivity of the drawing, the curves for the three other samples would appear identical.

The molal heat capacities of these substances are listed at rounded temperatures in Table V. These heat-capacity values were read from a smoothed curve through the experimental points and are estimated to have a probable error of 0.1 percent above 25°K, 1 percent at 10°K, and 5 percent at 5°K. The heat capacity was extrapolated to 0°K with a Debye function and values of thermodynamic functions were computed over the entire temperature range for these substances, but only the 25°C values are presented in Table VI.

Calculations of the apparent characteristic gram atomic Debye temperature are shown in Table VII.

DISCUSSION

These new data provide definitive values of the heat capacity of these substances above 10°K. An impression of the high precision of the data on quartz relative to literature values may be obtained by reference to Fig. 4. This deviation plot indicates that our data show deviations of the order of 0.01 percent over most of the range, and that the data of Anderson<sup>3</sup> scatter considerably about our values. The older data of Nernst<sup>4</sup> show deviations of approximately 20 percent near 25°K. Koref's data also show considerable deviation. Most of Gunther's<sup>5</sup> determinations are beyond the range of the plot.

A somewhat similar deviation plot on cristobalite (Fig. 5) presents the deviation of the experimental determinations on cristobalite versus our smoothed quartz  $C_p$  curve. This plot indicates again the superior precision of our data compared with previous values. The high-temperature end of the Anderson<sup>3</sup> data, however, deviates markedly from the essentially "parallel" behaviour we find with the quartz data.

The data for the samples of vitreous silica are presented relative to quartz in Fig. 6. To emphasize the similarity of these data to those of cristobalite at low temperatures, the heat-capacity deviation curve of that substance is also included. The present samples of vitreous silica are so much better characterized than those described in the literature that the present values may be considered definitive.

Further discussion of these matters is reserved for the papers to be presented by the various authors involved.

Although we have probably carried the measurements to an adequately low temperature for thermodynamical and structural purposes, it will be interesting to determine the heat capacities to even lower temperatures, say 0.9°K, as will be possible in our Mark II cryostat now under construction.

A further matter of interest is the absolute entropy, enthalpy, and free energy of the vitreous silica samples at 0°K. These could be deduced from our data when heat-of-solution values for quartz and the several vitreous silicas are available. The measurements would require the expenditure of only several grams of the materials.

Our request of the Bureau of Standards to consider such measurements has not yet been answered. We have recently found it necessary in the course of other work to plan the construction of a calorimeter capable of handling hot aqueous HF and anhydrous HF and will consider making the above measurements ourselves. It is evident, of course, that similar measurements would prove desirable in work on the alkali oxide-silicon dioxide glass systems and indispensable in the determination of stored energy in irradiated silicon dioxide.

These measurements have demonstrated the capabilities of thermal measurements in the study of the structure of vitreous materials. It is unfortunate that it was not possible to include tridymite in the series. The discovery of new high-density silica phases produced under high pressure raises other interesting questions. Another related problem of considerable calorimetric interest is the dependence of the free energy of a vitreous phase on composition and annealing history. Presumably, in this case also thermal measurements would reveal structural details.

## REFERENCES

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3. Anderson, C. T., J. Am. Chem. Soc., 58, 568 (1936).
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5. Gunther, P., ( cf. R. Wietzel, Ztschr. Anorg. Chem. 116, 71 (1921) ).
6. Simon, F., Ann. Physik, 68, 241 (1922).
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Figure 1. Cross-sectional schematic diagram of the Mark I cryostat

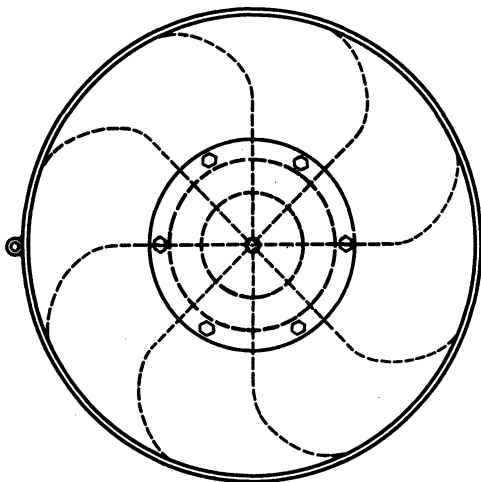
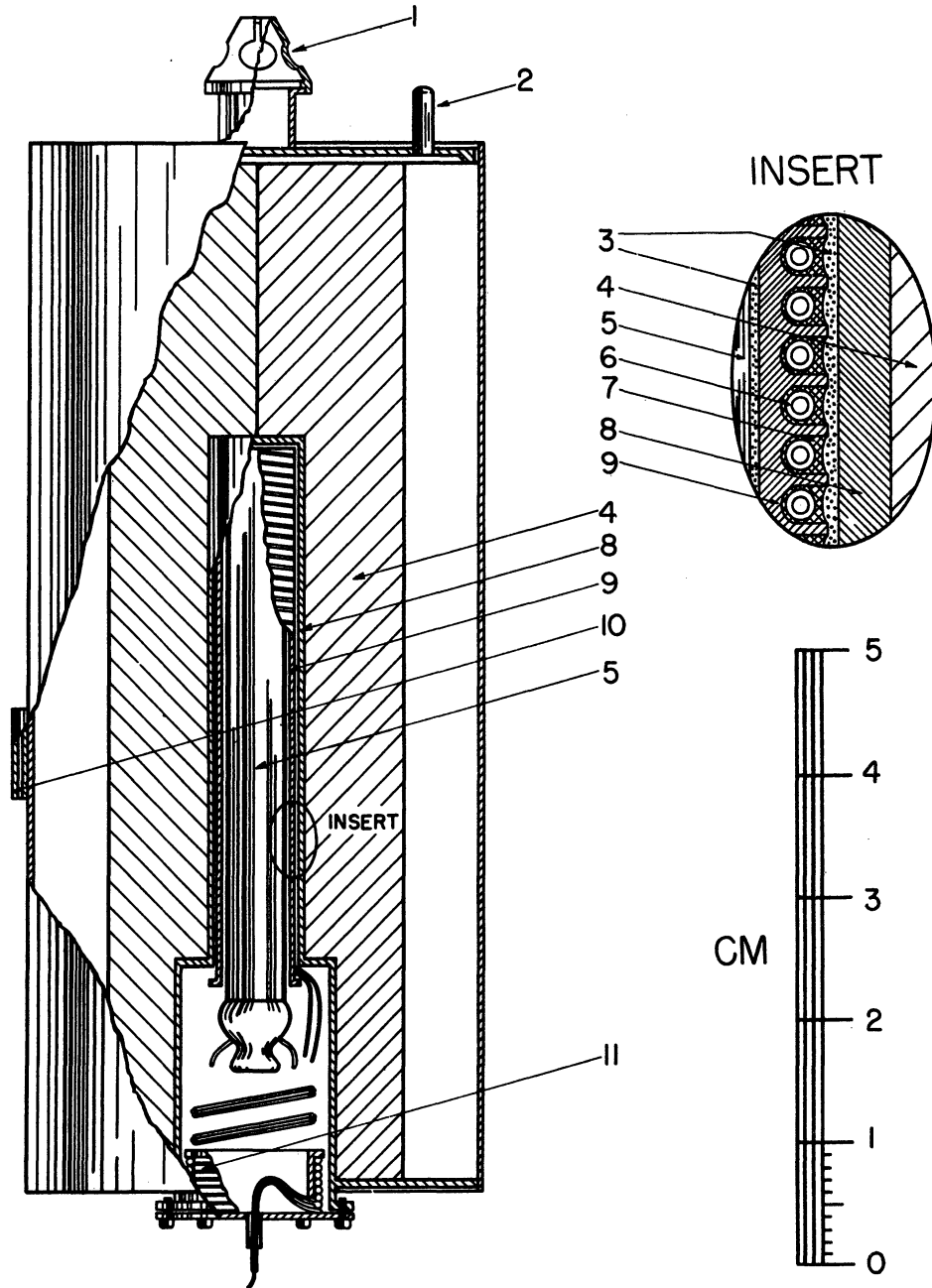
1. Helium-exit connector
2. Helium-transfer tube
3. Nitrogen-inlet and outlet connector
4. Sleeve fitting to helium-transport Dewar
5. Nitrogen filling tube
6. Helium-transfer-tube extender and cap
7. Screw fitting at inlet of helium-transfer tube
8. Brass vacuum can
9. Outer floating radiation shield
10. Nitrogen tank
11. Helium-exit tube
12. Economizer (effluent helium-vapor heat exchanger)
13. Nitrogen radiation shield
14. Helium tank
15. Bundle of lead wires
16. Adiabatic shield
17. Helium radiation shield
18. Ring for block and tackle
19. Windlass
20. Vacuum seal and terminal plate for leads
21. Head plate
22. O-Ring gasket
23. Coil spring
24. Supporting string
25. Floating ring
26. Calorimeter



Note: For Figure 1 refer to file copy.

Figure 2. Calorimeter (Laboratory Designation W-5)

1. Thermal-conductivity cone
2. Monel helium seal-off tube
3. "Lubriseal" stopcock grease
4. Copper vane
5. Leeds and Northrup platinum resistance thermometer
6. Fiberglass insulated No. 40 Advance constantan wire
7. Formvar varnish
8. Gold-plated copper heater well
9. Gold-plated copper heater sleeve
10. Differential thermocouple sleeve
11. Spool to bring leads into thermal equilibrium with calorimeter



W-5 CALORIMETER

Figure 2

Figure 3. The Molal heat capacity of quartz the experimental determinations are shown by the centers of the circles.

QUARTZ

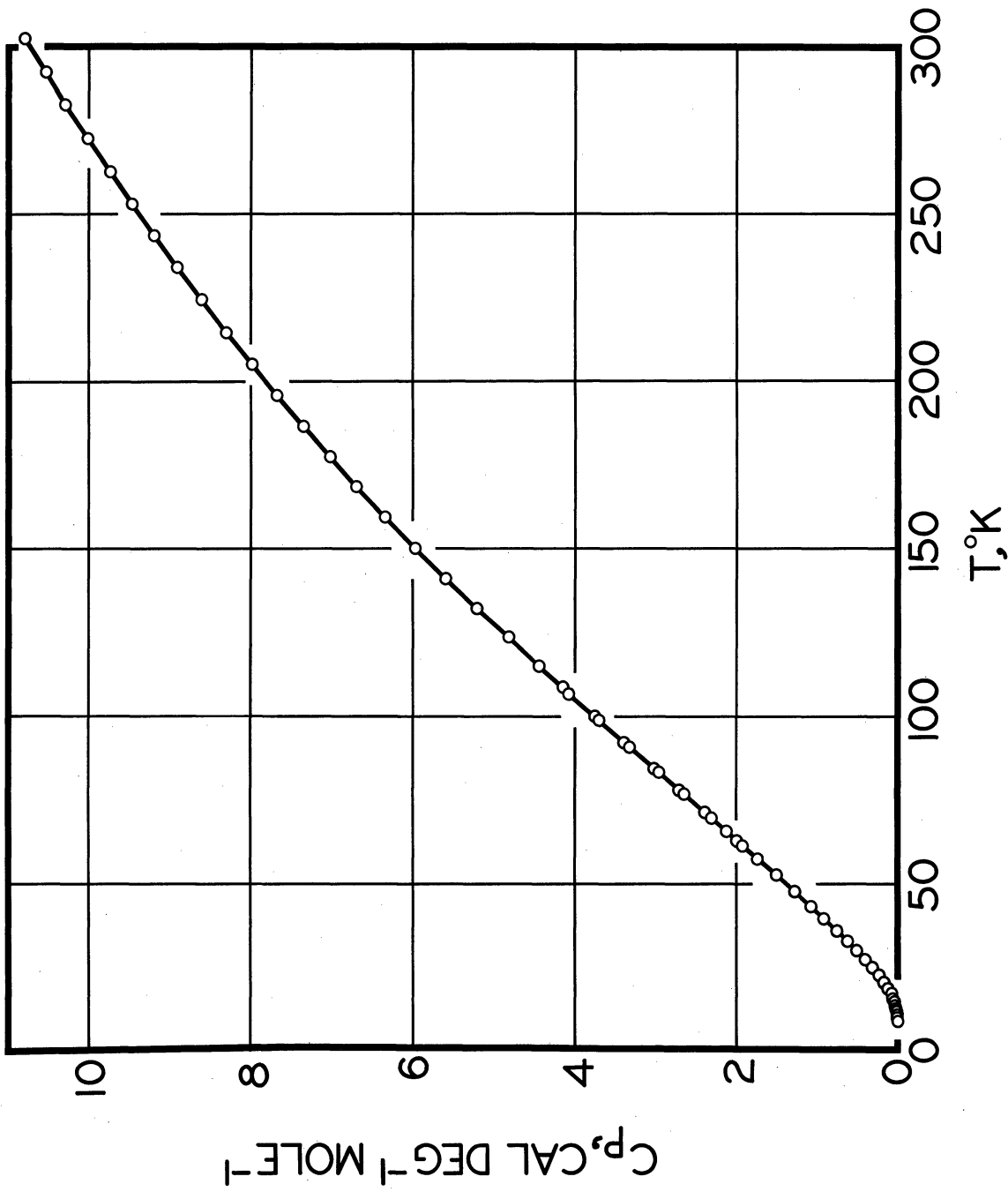


Figure 3

Figure 4. Deviation of quartz Cp data from our smoothed curve. The deviations,  $\Delta C_{pT} = (C_p \text{ individual point} - C_p \text{ smooth curve})_T$  are represented by open circles. The corresponding deviation of the data of another investigation from our smoothed curve is similarly represented, i.e.,  $\Delta C_{pT} = (C_p \text{ other investigator} - C_p \text{ our smooth curve})_T$ .

The solid circles represent the data of Anderson<sup>3</sup>, the solid triangles those of Nernst<sup>4</sup>, and the solid square those of Koref.<sup>4</sup>

Erratum: Decimal point is in error on the  $\Delta C_p$  scale. For -0.01 read -0.1, etc.

QUARTZ

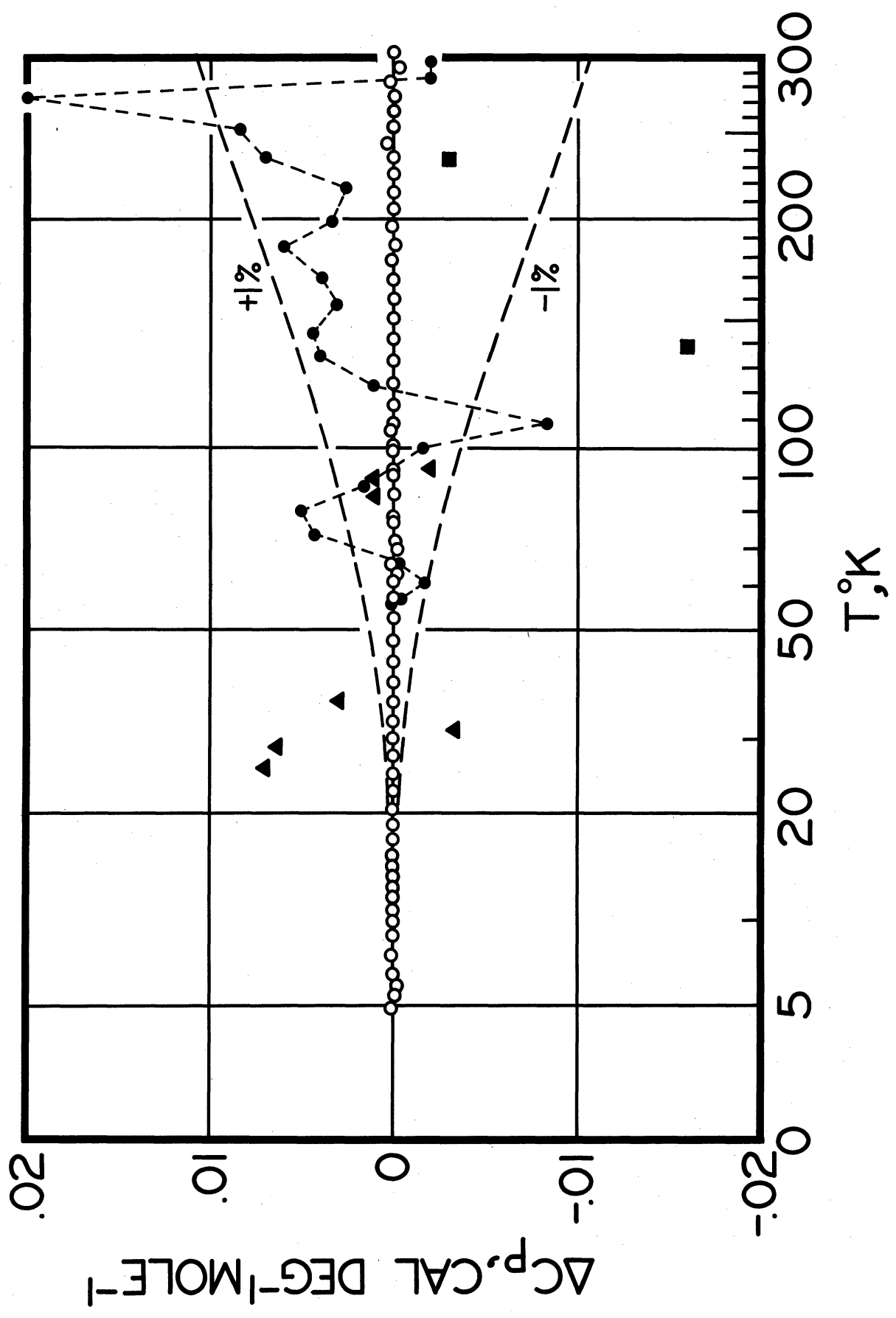


Figure 4

Figure 5. Deviation of cristobalite  $C_p$  data from our smoothed quartz curve. The deviation,  $\Delta C_{pT}$ , here represents  $(C_p \text{ cristobalite} - C_p \text{ quartz})_T$ . The open circles represent our experimental points, the solid circles the data of Simon<sup>6</sup>, and the solid triangles the data of Anderson<sup>3</sup>.



CRISTOBALITE

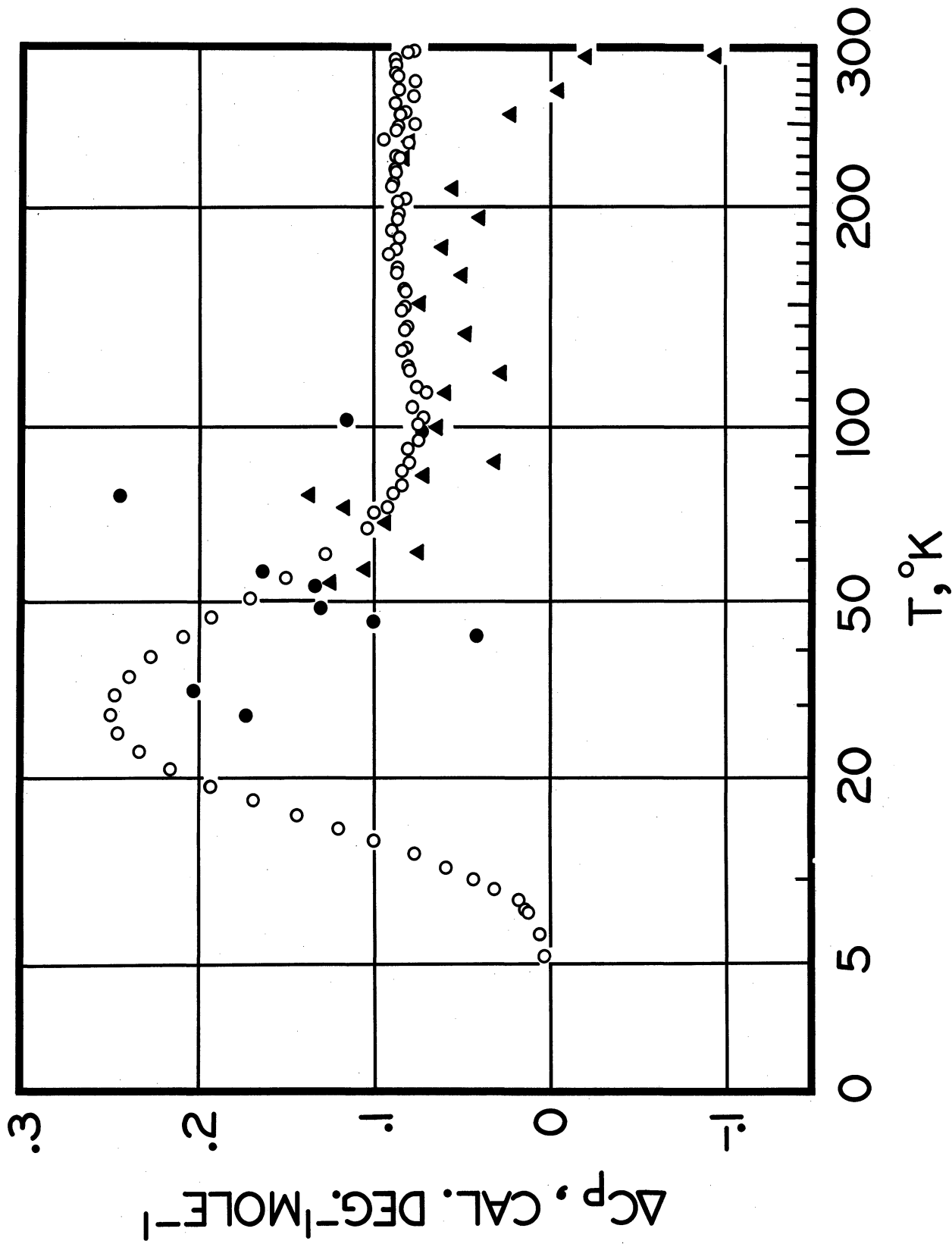


Figure 5

Figure 6. Deviation of Cp data on vitreous-silica samples from our smoothed quartz Cp curve. The deviation,  $\Delta C_{pT}$ , here represents  $(C_p \text{ vitreous silica} - C_p \text{ quartz})_T$ . The open circles represent our experimental points on 1070° vitreous silica, the open squares represent our experimental points on 1300° vitreous silica. The solid triangles present the four determinations of Simon and Lange<sup>7</sup>, the solid diamonds the data of Nernst<sup>4</sup>, and the solid circles the data of Simon<sup>6</sup>.

The dashed line represents, for comparison, the smooth-curve heat-capacity deviation of cristobalite relative to that of quartz.

VITREOUS SILICA

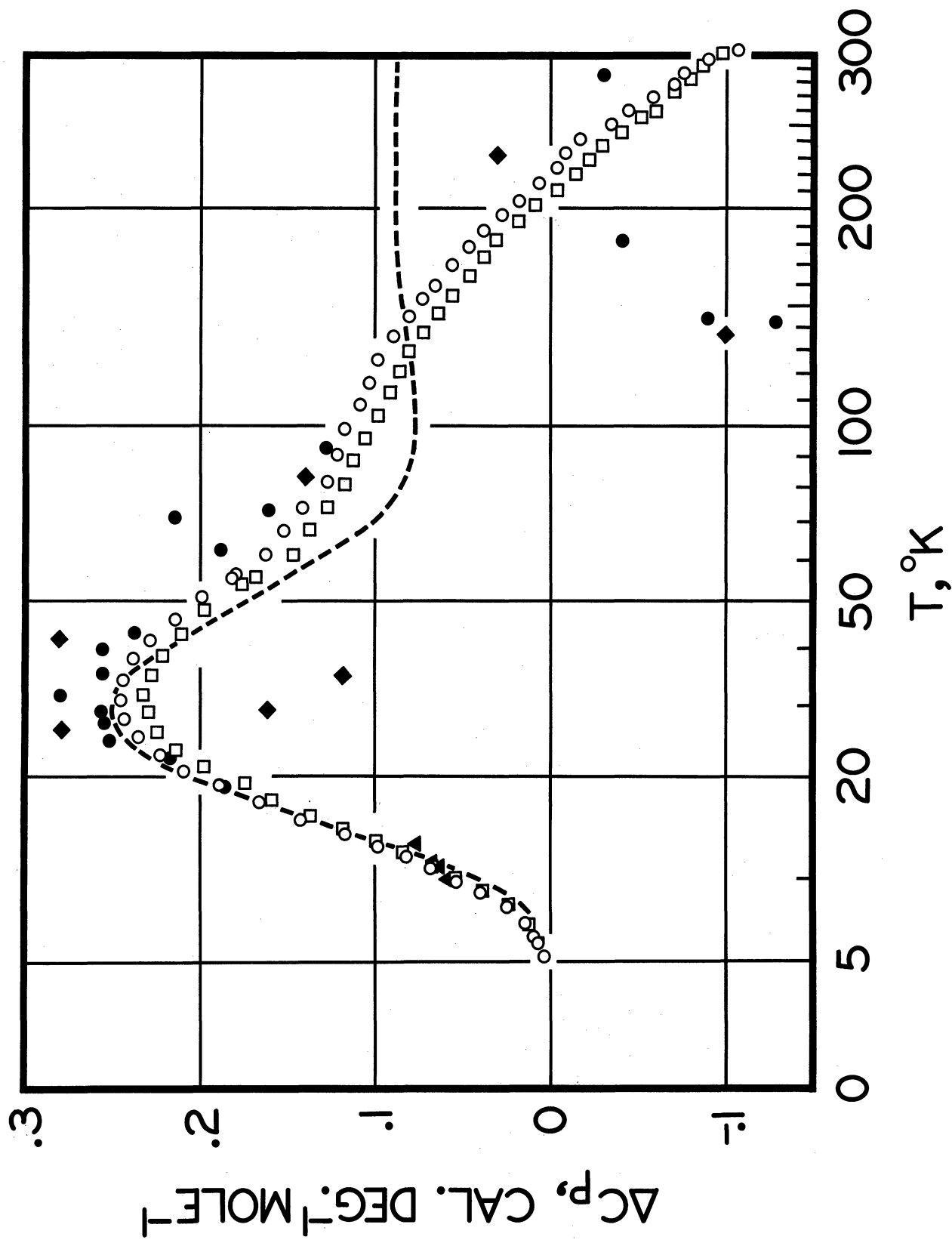


Figure 6

TABLE I

THE MOLAL HEAT CAPACITY OF QUARTZ  
(in calories per degree)

<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>S</sub></u>	<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>S</sub></u>
SERIES I					
61.32	3.064	1.924	52.53	5.069	1.508
65.60	5.466	2.127	57.42	4.725	1.735
71.42	6.188	2.393	62.85	6.123	1.993
77.63	6.218	2.690	69.50	7.169	2.301
84.44	7.402	3.025	76.72	7.251	2.646
92.02	7.755	3.383	90.70	7.962	3.321
99.80	7.802	3.736	98.70	8.013	3.684
107.57	7.727	4.095	106.84	7.881	4.063
			114.97	8.381	4.433
			123.45	8.582	4.814
SERIES II					
4.79	0.927	0.0016	132.14	8.800	5.199
5.42	0.738	0.0017	141.08	9.073	5.581
5.90	1.553	0.0020	150.18	9.120	5.961
6.50	1.398	0.0027	159.37	9.263	6.324
7.63	1.726	0.0042	168.54	9.070	6.678
8.88	1.408	0.0066	177.62	9.094	7.016
9.90	0.960	0.0099	186.71	9.071	7.336
10.70	1.096	0.0136	195.88	9.269	7.660
11.68	1.008	0.0201	205.16	9.298	7.974
12.58	0.985	0.0278	214.65	9.679	8.285
13.51	0.948	0.0371	224.38	9.781	8.591
14.46	0.979	0.0490	234.07	9.581	8.884
15.61	1.352	0.0652	243.56	9.418	9.170
17.07	1.628	0.0931	253.06	9.588	9.441
18.74	1.753	0.1297	262.81	9.916	9.718
20.62	2.019	0.1788	272.78	10.025	9.997
22.73	2.208	0.2429	282.82	10.061	10.273
25.01	2.347	0.3184	292.84	9.970	10.519
27.43	2.480	0.4063	302.77	9.801	10.771
30.00	2.675	0.5071			
32.83	2.987	0.6245			
36.00	3.339	0.7612			
39.48	3.626	0.9141			
43.29	3.984	1.084			
47.64	4.707	1.284			

TABLE II

THE MOLAL HEAT CAPACITY OF CRISTOBALITE  
(in calories per degree)

<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>S</sub></u>	<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>S</sub></u>
SERIES I					
72.65	5.117	2.550	265.39	8.874	9.876
78.40	6.391	2.819	274.17	8.710	10.117
85.13	7.061	3.143	283.86	8.661	10.350
92.72	8.121	3.492	291.71	9.067	10.584
100.51	7.456	3.844	300.95	9.414	10.806
107.71	6.950	4.177	SERIES III		
115.14	7.890	4.514			
123.08	7.990	4.881	7.75	0.555	0.0146
131.33	8.514	5.245	7.83	0.627	0.0156
140.35	9.527	5.630	8.60	0.671	0.0226
149.62	8.998	6.015	9.32	0.619	0.0379
158.83	9.425	6.388	10.09	0.831	0.0537
168.27	9.454	6.754	11.06	1.074	0.0736
177.82	9.646	7.109	12.23	1.227	0.1004
187.09	8.888	7.438	13.37	1.064	0.1339
195.92	8.771	7.748	14.48	1.139	0.1682
204.57	8.515	8.038	15.76	1.418	0.2112
213.21	8.765	8.326	17.36	1.791	0.2676
222.15	9.109	8.608	19.18	1.848	0.3342
231.31	9.211	8.892	21.14	2.059	0.4087
240.64	9.438	9.176	23.37	2.402	0.4962
250.04	9.378	9.439	25.87	2.599	0.5935
SERIES II					
130.40	8.746	5.207	28.61	2.875	0.7002
139.28	9.005	5.585	31.66	3.230	0.8220
148.06	8.562	5.950	34.96	3.365	0.9545
156.64	8.596	6.300	38.55	3.739	1.100
165.43	8.985	6.646	42.34	3.831	1.249
174.61	9.363	6.996	46.36	4.216	1.414
183.97	9.366	7.330	50.76	4.579	1.597
193.42	9.527	7.661	55.74	5.373	1.806
202.77	9.153	7.983	61.61	6.363	2.060
211.93	9.172	8.286	69.45	9.308	2.404
221.03	9.026	8.572	68.06	6.570	2.343
230.04	8.990	8.849	74.49	6.278	2.632
238.94	8.807	9.110	80.62	6.758	2.915
247.74	8.808	9.377			
256.55	8.813	9.624			

(continued)

TABLE II  
(continued)

THE MOLAL HEAT CAPACITY OF CRISTOBALITE  
(in calories per degree)

SERIES III

<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>s</sub></u>
87.56	7.126	3.251
95.25	8.238	3.597
103.58	8.422	3.981
112.35	9.130	4.386
121.65	9.470	4.812
250.43	8.644	9.440
259.14	8.776	9.693
267.98	8.900	9.937
276.86	8.871	10.178
285.75	8.925	10.424
294.59	8.753	10.656
302.19	6.474	10.832

TABLE III

THE MOLAL HEAT CAPACITY OF 1070° VITREOUS SILICA  
(in calories per degree)

<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>s</sub></u>	<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>s</sub></u>
SERIES I			SERIES II		
55.76	6.06	1.842	5.22	0.85	0.0050
61.84	6.11	2.113	5.87	0.97	0.0093
67.90	6.00	2.383	6.23	0.94	0.0118
74.43	7.05	2.677	7.04	1.07	0.0183
82.22	8.52	3.041	8.03	1.10	0.0294
90.85	8.73	3.449	9.03	0.98	0.046
99.70	8.97	3.845	9.92	0.85	0.064
108.63	8.88	4.249	10.82	0.98	0.082
117.45	8.76	4.647	11.79	0.99	0.103
126.18	8.70	5.034	12.80	1.06	0.128
134.86	8.65	5.403	13.93	1.21	0.159
143.45	8.52	5.758	15.38	1.72	0.204
152.15	8.89	6.107	17.20	1.93	0.263
161.32	9.45	6.464	19.05	1.77	0.327
170.57	9.06	6.812	20.95	2.03	0.396
179.46	8.72	7.128	23.00	2.08	0.475
188.18	8.71	7.429	25.43	2.77	0.569
197.22	9.36	7.734	28.09	2.89	0.675
206.44	9.07	8.037	31.17	3.27	0.801
215.38	8.82	8.314	34.59	3.56	0.944
224.33	9.08	8.584	38.29	3.84	1.100
233.95	9.36	8.861	42.22	4.01	1.264
242.84	9.22	9.127	46.40	4.36	1.441
251.96	9.01	9.374	51.40	5.64	1.656
261.12	9.32	9.625	57.02	5.59	1.900
270.35	9.13	9.868			
279.56	9.29	10.105			
288.59	8.79	10.338			
297.11	8.26	10.541			
30.429	6.10	10.711			

TABLE IV

THE MOLAL HEAT CAPACITY OF 1300° VITREOUS SILICA  
(in calories per degree)

<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>S</sub></u>	<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>S</sub></u>
SERIES I					
5.88	1.172	0.0091	198.16	8.856	7.748
7.01	1.349	0.0163	207.05	8.928	8.041
8.15	1.099	0.0293	215.96	8.891	8.317
9.18	1.056	0.0459	224.99	9.178	8.593
10.12	0.940	0.0657	234.21	9.263	8.866
11.12	1.130	0.0841	243.54	9.388	9.140
12.23	1.163	0.1082	252.89	9.323	9.392
13.30	1.023	0.1348	262.26	9.436	9.643
14.40	1.227	0.1659	271.66	9.364	9.881
15.72	1.467	0.2048	281.00	9.329	10.137
17.45	1.996	0.2609	290.36	9.397	10.372
19.41	2.034	0.3198	299.91	9.742	10.605
21.46	2.082	0.4030			
23.69	2.374	0.4879			
26.13	2.506	0.5822			
SERIES II					
28.99	3.222	0.6962	55.91	6.070	1.835
32.08	2.952	0.8206	62.01	6.141	2.106
35.18	3.230	0.9529	68.17	6.176	2.381
38.94	4.300	1.112	74.47	6.426	2.664
43.20	4.215	1.291	81.65	7.936	3.001
48.47	6.322	1.519	89.14	7.031	3.360
54.31	5.348	1.769	96.58	7.849	3.690
59.19	4.401		104.96	8.914	4.069
64.15	5.511		113.54	8.249	4.460
69.80	5.788	2.455	121.90	8.456	4.830
75.55	6.941	2.715	130.32	8.387	5.197
82.19	6.326	3.025	138.65	8.271	5.545
89.24	7.786	3.365	146.73	7.884	5.875
97.25	8.220	3.722	155.42	9.503	6.221
105.62	8.530	4.100	164.92	9.487	6.586
113.85	7.914	4.471	174.48	9.629	6.941
121.99	8.374	4.835	184.03	9.472	7.276
130.13	7.903	5.189	193.44	9.342	7.594
138.28	8.393	5.530	202.63	9.047	7.900
146.69	8.422	5.874	211.65	8.987	8.188
155.55	9.298	6.225	220.62	8.951	8.459
164.65	8.897	6.577	229.56	9.930	8.727
171.22	8.603	6.819	238.53	9.004	8.988
180.06	9.076	7.133	247.49	8.934	9.239
189.16	9.127	7.448	256.47	9.046	9.486



TABLE IV  
(continued)

THE MOLAL HEAT CAPACITY OF 1300° VITREOUS SILICA  
(in calories per degree)

<u>T, °K</u>	<u>ΔT, °K</u>	<u>C<sub>S</sub></u>
265.56	9.124	9.733
274.68	9.137	9.976
283.86	9.218	10.211
293.06	9.192	10.442
302.16	9.024	10.659

TABLE V

SMOOTHED HEAT CAPACITIES FOR SILICON DIOXIDE  
(in calories per degree per mole)

T, °K	Quartz	Cristobalite	Vitreous Silica	
			1070°	1300°
10	0.0105	0.044	0.066	0.062
15	0.0567	0.185	0.192	0.184
20	0.1619	0.365	0.362	0.339
25	0.3176	0.559	0.552	0.535
30	0.507	0.755	0.753	0.737
35	0.718	0.956	0.958	0.944
40	0.938	1.156	1.166	1.156
45	1.162	1.357	1.382	1.368
50	1.392	1.565	1.595	1.585
60	1.857	1.991	2.030	2.024
70	2.328	2.429	2.476	2.461
80	2.802	2.887	2.936	2.924
90	3.287	3.363	3.405	3.395
100	3.744	3.822	3.859	3.842
110	4.209	4.285	4.311	4.299
120	4.660	4.738	4.761	4.746
130	5.105	5.187	5.196	5.182
140	5.534	5.617	5.614	5.601
150	5.946	6.029	6.020	6.006
160	6.347	6.433	6.412	6.398
170	6.733	6.821	6.790	6.774
180	7.101	7.189	7.145	7.136
190	7.454	7.543	7.490	7.477
200	7.803	7.887	7.826	7.812
210	8.133	8.218	8.147	8.136
220	8.453	8.540	8.456	8.441
230	8.763	8.851	8.756	8.741
240	9.061	9.150	9.042	9.030
250	9.353	9.442	9.321	9.310
260	9.637	9.726	9.593	9.585
270	9.918	10.005	9.860	9.852
280	10.189	10.277	10.117	10.111
290	10.450	10.537	10.362	10.366
300	10.703	10.791	10.602	10.609
273.16	10.006	10.088	9.941	9.935
298.16	10.658	10.744	10.564	10.565

TABLE VI  
 MOLAL THERMODYNAMIC FUNCTIONS FOR SILICA AT 298.16°K  
 (in calories per degree)

	Literature <sup>B</sup>	This Research		
	$S^{\circ}-S_0^{\circ}$	$S^{\circ}-S_0^{\circ}$ ( $\pm 0.01$ )	$(H^{\circ}-H_0^{\circ})/T$ ( $\pm 0.006$ )	$(F^{\circ}-H_0^{\circ})/T$ ( $\pm 0.005$ )
Quartz	10.00 $\pm$ 0.10	9.880	5.543	4.337
Cristobalite	10.20 $\pm$ 0.10	10.377	5.643	4.734
1070° vitreous silica	10.3 $\pm$ 0.2	10.374	5.609	4.765

TABLE VII

APPARENT DEBYE  $\Theta$  CALCULATIONS  
 (Characteristic temperatures per gram atom of silica)

T, °K	Quartz	Cristobalite	Vitreous Silica	
			1300°	1070°
10	510.1	307.4	282.2	276.4
15	436.1	294.0	294.6	290.4
20	410.5	312.6	320.4	313.6
25	409.3	338.7	343.8	340.2
30	420.0	367.3	370.3	367.6
35	435.9	395.4	397.1	395.1
40	459.3	423.1	423.1	421.8
45	475.1	449.9	448.6	447.0
50	495.2	475.0	472.9	471.8
60	534.7	521.8	518.5	517.9
70	573.0	563.9	560.9	559.6
80	609.0	601.1	598.0	597.0
90	640.0	634.0	630.8	630.7
100	671.5	665.7	664.1	662.8
110	699.4	694.9	693.1	692.8
120	726.4	720.8	720.4	719.2
130	751.1	745.6	745.8	744.7
140	774.4	769.0	770.1	769.2
150	796.5	791.4	793.2	792.1
160	818.1	812.6	814.9	813.8
170	838.1	832.3	835.7	834.4
180	857.4	850.7	855.5	854.7
190	875.9	868.9	875.0	873.9
200	892.8	886.4	893.0	891.7
210	909.0	903.0	907.4	909.0
220	924.7	918.3	926.9	925.5
230	940.3	932.9	942.3	940.9
240	954.3	946.3	957.1	955.7
250	967.0	959.0	971.0	969.4
260	978.9	971.4	983.8	982.2
270	989.4	982.5	995.8	994.3
280	999.2	991.8	1006.9	1005.7
290	1008.5	999.6	1017.0	1016.1
300	1017.0	1006.2	1026.7	1025.4

