U. S. DEPARTMENT OF COMMERCE NATIONAL BUREAU OF STANDARDS

RESEARCH PAPER RP1197

Part of Journal of Research of the National Bureau of Standards, Volume 22, April 1939

DENSITY OF SOME SODA-POTASH-SILICA GLASSES AS A FUNCTION OF THE COMPOSITION

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ABSTRACT

Certain relations between the specific volume and composition of glasses of the soda-silica series have previously been reported. Additional glasses were made in the same series and also in the potash-silica and soda-potash-silica series. The glasses were analyzed and their densities measured. Analysis of the new data suggests that the specific volume-composition relations change definitely at compositions approximating those of eutectics, instead of at those of simple molecular ratios, as previously reported.

CONTENTS

I. INTRODUCTION

The densities of 37 soda-silica glasses, containing from 13 to 50 percent of soda, were reported in a previous publication [1],[1] and certain relations between specific volume and composition were suggested. Since those data were published, many new glasses in the same range of composition have been made. These, together with all of the former glasses still available for further experimentation, constitute a new series of 50 soda-silica glasses whose densities and compositions are given in the present report. This report also includes data on 66 potash-silica glasses varying in composition from 17 to 40 percent of potash, and on 78 soda-potash-silica glasses whose compositions were distributed throughout the field between the composition-ranges of the two binary series. All the glasses were given the same heat treatment, which treatment differed materially from that described in the earlier report.

Study of the new data, which are considerably more numerous as well as more reproducible than those previously reported, indicated the desirability of some revision of the conclusions based on the previous data. In addition to presenting the new data, therefore, the present paper replaces that part of the earlier one which dealt with the soda-silica glasses.

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[1] Figures in brackets indicate the literature references at the end of this paper.

II. PREPARATION OF THE GLASSES

The reader is referred to previous publications [1, 2] for a description of the methods of preparation and analysis, and determination of density of the glasses.

"Weathering" of some of the soda-silica glasses remaining from the original series was found to have occurred when these specimens were examined for inclusion in the new series. This weathering appeared to be merely a surface effect, however, and in each case tried the original density of the glass was restored (which implied that the "interior composition" had not been changed) by removal of the surface to a depth of about 1 mm by grinding with carborundum and water. The use of water proved satisfactory provided the glasses were dried immediately after grinding. For most of the glasses the procedure was to grind off the surface twice or more immediately following heat treatment, measuring the density after each grinding, until no further change in density was noted. The densities of the few glasses which had been "reconditioned" by grinding before heat treatment were measured directly after heat treatment. These densities remained unchanged by subsequent grinding.

III. HEAT TREATMENT

In the previously reported work, each glass was held for an hour at approximately its highest "annealing temperature." Subsequent experience indicates, however, that insufficient knowledge and control of the rates of cooling from these temperatures had introduced errors which would hinder a satisfactory evaluation of the data. In an effort, therefore, to develop a method for giving reproducible and "comparable"[2] heat treatments, five different heat treatments were applied successively to each of 11 glasses (indicated by the symbol ⊙ in figure 4) representative of the entire field of compositions studied. The heat treatments (represented schematically) and the densities after each treatment (tabulated directly beneath the respective schematic curves) are given in figure 1 in the order in which the heat treatments were applied.

1. DEVELOPMENT OF FIGURE 1

The "critical temperatures" and "softening temperatures" were determined for each glass as described in a former paper [11]. The term "critical range", as used herein, may be defined as a temperature interval (for each glass) above and below which large variations in cooling rate did not appreciably affect the final, or room temperature, density. An indication of the practical limits of this range is given below in the discussion of results of heat-treating experiments.

In treatments 2, 3, and 5 the glasses were necessarily treated individually since the critical temperature, softening point, and critical range are different for each glass. The difficulties involved in the individual treatments are, however, such that the results obtained are merely indicative. First, the various critical temperatures, etc., cannot be determined with assurance; and second, the extreme nature of the quenching process is such that no single experiment could be repeated in every detail. For treatment 1, however, in which it was

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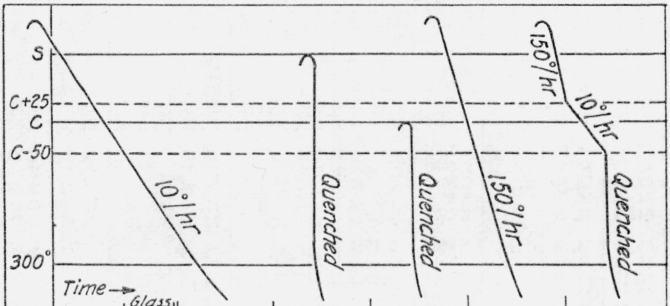
[2] The word "comparable", as used here, is intended to convey the idea of a common basis for comparison of the densities of glasses of different compositions so far as the densities may be affected by differences in heat treatment.

desired to maintain the cooling rate constant, and the same for all glasses, not only through the respective critical ranges but also for a considerable temperature interval on either side, these two difficulties were largely obviated by cooling all the glasses simultaneously in a furnace whose cooling rate was essentially constant through a temperature interval (550° to 300° C), which included by a fairly wide margin the critical range of every glass to be treated. For treatment 4 the glasses were similarly treated except that a different furnace (one having a much greater cooling rate) was used. All glasses made for this investigation were given treatment 1, whereas the 11 glasses for which data are given in figure 1 and, in addition, 14 soda-silica, 12 potash-silica, and 7 soda-potash-silica glasses were selected for treatment 4. Complete density data for these two treatments are included in table 1.



	Glass No.	1	2	3	4	5
Densities resulting from each treatment Treatments applied consecutively to the same specimen of each glass	302	2.5205	2.5167	2.5181	2.5178	2.5215
	317	2.3435	2.3379	2.3407	2.3407	2.3429
	319	2.5047	2.4982	2.5044	2.4990	2.5050
	330	2.4868	2.4827	2.4869	2.4833	2.4857
	336	2.4502	2.4458	2.4473	2.4460	2.4498
	346	2.4469	2.4429	2.4451	2.4445	2.4471
	356	2.4111	2.4084	2.4092	2.4084	2.4116
	363	2.4348	2.4299	2.4330	2.4303	2.4346
	367	2.4066	2.4005	2.4055	2.4028	2.4066
	371	2.3935	2.3866	2.3907	2.3899	2.3932
	377	2.3227	2.3205	2.3228	2.3200	2.3225

FIGURE 1.—*Schematic cooling curves for each of the 11 glasses, with densities resulting from each treatment shown at the base of the corresponding curve.*

S, softening temperature—different for each glass; *C*, critical temperature—different for each glass. The curves are not drawn to scale.

It is noteworthy that the density of a specimen after treatment does not depend upon the size of the specimen, providing size, as such, is not a factor in determining the rate of cooling (as, for instance, when the specimen is quenched in air). For example, it was found that a 46-g block of borosilicate crown glass (density approximately 2.51) and some small chips of the same material, each chip weighing about 0.2 g, had within experimental limits the same density after having been cooled simultaneously at about 70° C per hour from above the critical range. The density of the chips was measured as a composite sample. When the same specimens were quenched in air from the temperature just indicated, the large block had a considerably greater density ($\Delta = 0.0050$) than the small chips, the "free" cooling rate of the former necessarily being much less than those of the chips.

TABLE 1.—*Density and composition for 194 soda-potash-silica glasses*

Glass number[1]	Composition			Density	
	SiO_2	Na_2O	K_2O	Treatment 1	Treatment 4
	%	%	%		
1 (3)	51.62	48.38	--------	2.5676	--------
2 (4)	52.60	47.40	--------	2.5622	--------
3 (5)	53.62	46.38	--------	2.5557	--------
4 (8)	57.45	42.55	--------	2.5361	2.5325
5 (9)	58.98	41.02	--------	2.5286	2.5254
6	59.04	40.96	--------	2.5290	--------
7 (10)	59.69	40.31	--------	2.5262	--------
8 (11)	59.71	40.29	--------	2.5257	--------
9 (12)	59.97	40.03	--------	2.5236	--------
10	61.17	38.83	--------	2.5168	--------
11	61.77	38.23	--------	2.5147	--------
12	62.58	37.42	--------	2.5111	--------
13 (13)	62.77	37.23	--------	2.5092	2.5053
14	62.78	37.22	--------	2.5094	--------
15 (14)	62.86	37.14	--------	2.5084	--------
16 (15)	63.06	36.94	--------	2.5079	--------
17	63.13	36.87	--------	2.5080	--------
18	63.60	36.40	--------	2.5056	--------
19	64.00	36.00	--------	2.5028	--------
20	64.71	35.29	--------	2.4979	--------
21 (17)	65.32	34.68	--------	2.4961	2.4925
22	65.72	34.28	--------	2.4912	--------
23	66.16	33.84	--------	2.4904	--------
24 (18)	66.52	33.48	--------	2.4869	--------
25	66,58	33.42	--------	2.4859	--------
26	67.07	32.93	--------	2.4830	--------
27	67.22	32.78	--------	2.4821	--------
28	67.35	32.65	--------	2.4816	--------
29	68.10	31.90	--------	2.4770	--------
30	68.81	31.19	--------	2.4722	2.4692
31	69.10	30.90	--------	2.4706	--------
32 (20)	69.65	30.35	--------	2.4670	2.4645
33 (22)	70.44	29.56	--------	2.4623	--------
34	71.10	28.90	--------	2.4571	2.4540
35 (23)	72.15	27.85	--------	2.4519	--------
36 (24)	72.33	27.67	--------	2.4505	--------
37	73.00	27.00	--------	2.4450	2.4420
38	73.92	26.08	--------	2.4387	--------
39 (25)	74.16	25.84	--------	2.4378	--------
40 (26)	74.69	25.31	--------	2.4356	2.4332
41 (27)	75.29	24.71	--------	2.4289	2.4261
42 (28)	76.60	23.40	--------	2.4152	2.4126
43 (29)	76.65	23.35	--------	2.4152	2.4133
44 (30)	76.70	23.30	--------	2.4141	--------
45 (31)	77.85	22.15	--------	2.4031	2.4013
46 (32)	78.61	21.39	--------	2.3945	--------
47	81.54	18.46	--------	2.3677	--------
48	82.72	17.28	--------	2.3553	--------
49 (36)	85.15	14.85	--------	2.3329	2.3310
50	86.12	13.88	--------	2.3230	--------
(38)	100	--------	--------	2.203	--------
201	60.59	--------	39.41	2.4522	--------
202	60.81	--------	39.19	2.4481	--------
203	60.88	--------	39.12	2.4489	2.4433
204	61.90	--------	38.10	2.4449	--------
205	62.93	--------	37.07	2.4383	2.4326
206	63.93	--------	36.07	2.4347	--------
207	65.08	--------	34.92	2.4279	--------
208	65.92	--------	34.08	2.4242	2.4196
209	67.02	--------	32.98	2.4185	--------
210	67.24	--------	32.76	2.4166	--------
211	68.10	--------	31.90	2.4131	2.4079
212	68.60	--------	31.40	2.4108	--------
213	68.86	--------	31.14	2.4086	--------
214	69.17	--------	30.83	2.4074	--------
215	69.32	--------	30.68	2.4064	--------
216	69.50	--------	30.50	2.4046	--------
217	69.77	--------	30.23	2.4047	--------
218	70.11	--------	29.89	2.4015	--------
219	70.35	--------	29.65	2.3994	--------
220	70.58	--------	29.42	2.3984	--------
221	70.64	--------	29.36	2.3972	--------
222	70.93	--------	29.07	2.3966	--------
223	71.35	--------	28.65	2.3927	--------
224	71.45	--------	28.55	2.3940	--------
225	71.65	--------	28.35	2.3929	--------
226	71.67	--------	28.33	2.3923	2.3884
227	71.68	--------	28.32	2.3928	--------
228	72.26	--------	27.74	2.3888	--------
229	72.30	--------	27.70	2.3884	--------
230	72.33	--------	27.67	2.3891	--------
231	72.45	--------	27.55	2.3886	--------
232	72.49	--------	27.51	2.3874	--------
233	72.60	--------	27.40	2.3868	--------
234	72.74	--------	27.26	2.3862	--------
235	73.00	--------	27.00	2.3850	--------
236	73.14	--------	26.86	2.3840	--------
237	73.15	--------	26.85	2.3828	--------
238	73.25	--------	26.75	2.3830	--------
239	73.45	--------	26.55	2.3815	--------
240	73.63	--------	26.37	2.3807	--------
241	73.67	--------	26.33	2.3812	2.3767
242	74.15	--------	25.85	2.3781	--------
243	74.37	--------	25.63	2.3756	--------
244	74.89	--------	25.11	2.3728	2.3693
245	74.99	--------	25.01	2.3734	--------
246	75.00	--------	25.00	2.3728	2.3692
247	75.32	--------	24.68	2.3721	--------
248	75.49	--------	24.51	2.3701	--------
249	75.76	--------	24.24	2.3691	--------
250	76.34	--------	23.66	2.3646	--------
251	76.46	--------	23.54	2.3629	--------
252	76.57	--------	23.43	2.3636	--------
253	76.65	--------	23.35	2.3625	--------
254	76.82	--------	23.18	2.3595	--------
255	77.27	--------	22.73	2.3579	--------
256	77.74	--------	22.26	2.3529	2.3499
257	77.93	--------	22.07	2.3523	--------
258	78.74	--------	21.26	2.3470	--------
259	79.08	--------	20.92	2.3450	2.3414
260	79.22	--------	20.78	2.3430	--------
261	80.02	--------	19.98	2.3366	2.3339
262	80.58	--------	19.42	2.3336	--------
263	80.99	--------	19.01	2.3303	2.3277
264	81.00	--------	19.00	2.3314	--------
265	82.87	--------	17.13	2.3178	--------
266	82.99	--------	17.01	2.3172	--------
301	60.14	34.75	5.11	2.5198	--------
302	60.43	37.06	2.51	2.5205	2.5178
303	60.53	25.09	14.38	2.5117	--------
304	61.62	29.81	8.57	2.5097	--------

[1] Figures in parentheses are the numbers of the same glasses in the earlier density paper.

TABLE 1.—*Density and composition for 194 soda-potash-silica glasses*—Continued

Glass number	Composition			Density	
	SiO_2	Na_2O	K_2O	Treatment 1	Treatment 4
	%	%	%		
305	62.89	35.85	1.26	2.5084	--------
306	62.99	27.00	10.01	2.5004	--------
307	63.48	32.78	3.74	2.5044	--------
308	64.89	33.56	1.55	2.4968	--------
309	65.42	28.38	6.20	2.4911	--------
310	66.00	30.84	3.12	2.4886	--------
311	68.32	27.70	3.98	2.4740	2.4706
312	68.33	29.85	1.82	2.4741	--------
313	71.20	27.50	1.29	2.4564	--------
314	77.19	21.08	1.73	2.4087	--------
315	80.61	10.13	9.26	2.3630	--------
316	80.58	14.99	4.43	2.3706	--------
317	83.43	12.00	4.58	2.3435	2.3407
318	84.52	8.66	6.82	2.3275	2.3253
319	59.31	16.97	23.72	2.5047	2.4990
320	60.51	20.24	19.25	2.5048	--------
321	60.73	15.12	24.15	2.4926	--------
322	61.12	10.27	28.61	2.4825	--------
323	61.41	12.00	26.59	2.4837	--------
324	62.33	22.83	14.84	2.4982	--------
325	62.62	7.61	29.77	2.4685	--------
326	62.90	15.85	21.25	2.4826	--------
327	63.70	12.69	23.61	2.4726	--------
328	64.78	17.21	18.01	2.4757	--------
329	65.08	15.39	19.53	2.4700	--------
330	65.23	24.86	9.91	2.4868	2.4833
331	65.25	8.55	26.20	2.4542	--------
332	65.37	20.10	14.53	2.4775	--------
333	65.82	10.08	24.10	2.4572	--------
334	66.50	13.00	20.50	2.4591	--------
335	66.97	21.80	11.23	2.4700	--------
336	67.43	12.09	20.48	2.4502	2.4460
337	67.92	10.01	22.07	2.4436	--------
338	68.02	15.09	16.89	2.4544	--------
339	68.12	6.93	24.95	2.4384	2.4337
340	69.98	24.60	5.42	2.4603	2.4573
341	69.94	8.08	21.98	2.4300	--------
342	70.61	15.18	14.21	2.4409	--------
343	70.60	12.90	16.50	2.4348	--------
344	70.76	20.03	9.21	2.4464	--------
345	70.87	10.04	19.09	2.4301	--------
346	72.30	25.20	2.50	2.4469	2.4445
347	71.75	7.04	21.21	2.4191	--------
348	72.77	10.45	16.78	2.4176	--------
349	72.92	15.27	11.81	2.4268	--------
350	73.28	12.69	14.03	2.4181	--------
351	73.66	7.51	18.83	2.4050	--------
352	73.60	22.55	3.85	2.4360	--------
353	73.90	16.63	9.47	2.4218	--------
354	74.76	23.99	1.25	2.4312	--------
355	75.12	12.57	12.31	2.4048	--------
356	75.38	14.80	9.82	2.4111	2.4084
357	75.52	7.75	16.73	2.3943	2.3912
358	75.87	10.21	13.92	2.3968	--------
359	76.30	16.79	6.91	2.4063	--------
360	78.94	9.27	11.79	2.3764	--------
361	61.54	5.04	33.42	2.4653	--------
362	64.14	2.09	33.77	2.4422	--------
363	66.44	3.30	30.26	2.4348	2.4303
364	68.73	4.40	26.87	2.4258	--------
365	69.11	1.54	29.35	2.4140	--------
366	70.75	5.25	24.00	2.4191	--------
367	71.00	2.55	26.45	2.4066	2.4028
368	71.28	0.96	27.76	2.3999	--------
369	71.61	5.35	23.04	2.4145	--------
370	73.50	2.19	24.31	2.3898	--------
371	74.35	4.44	21.21	2.3935	2.3899
372	75.48	3.37	21.15	2.3797	2.3764
373	75.62	5.12	19.26	2.3851	--------
374	76.05	1.03	22.92	2.3696	--------
375	79.30	4.55	16.15	2.3581	--------
376	81.05	5.24	13.71	2.3467	2.3443
377	83.17	2.16	14.67	2.3227	2.3200
378	84.69	5.27	10.04	2.3197	--------

The size of the specimen does, however, affect the accuracy to which the density may be measured. In general, the sizes of the specimens were such that the experimental error in density varied from about ±0.0002 to ±0.0004. The 11 specimens used in the several heat-treating experiments, however, were so selected as to be of nearly the same size, in order that the results of the quenching experiments might be comparable.

2. RESULTS OF HEAT-TREATING EXPERIMENTS

It will be noted that for any given glass the two densities resulting from treatments 1 and 5, respectively, are essentially the same, although the cooling rates were alike only through a temperature interval between approximately 25° C above to 50° C below the critical temperature. It may be said, therefore, that this interval includes the critical range as defined above. Although no attempt was made to determine the limits for this range more closely, it is interesting to note that a considerable increase in the density of a given glass over that found by quenching from the softening temperature (treatment 2) was obtained merely by heating the glass rapidly to the critical temperature and immediately quenching it in air (treatment 3). In 3 of the 11 cases, the densities increased enough to equal those resulting from treatment 1.

From a comparison of the results of treatments 4 and 1, respectively, it was found that the difference between the corresponding specific volumes (V_4 and V_1) is a linear function of the percentages of soda (B) and potash (E) (no serious error was introduced in assuming the factor for silica (A) to be zero). Specifically, for each glass

$$V_4 - V_1 = (1.6B + 2.5E) \times 10^{-5}$$

The individual deviations from this expression were found not to exceed the errors incident to the measurement of specific volume.[3] Thus, it follows (provided the cooling rate is constant, and the same for all the glasses, through their respective critical ranges) that the actual rate used does not determine the character of the surface showing the relationship between specific volume and composition, nor establish the existence or location of "critical points" such as those noted in the following sections. For if

$$V_1 = f_1(A,B,E),$$

then

$$V_4 = f_1(A,B,E) + (1.6B + 2.5E) \times 10^{-5},$$

and at any given composition the rate of change of slope is the same for both surfaces, as the second derivatives are identical. Since the two cooling rates used are wholly arbitrary, it is reasonable to infer that a similar relation would hold had some other rates been chosen.

For the purpose of correlating specific volume with composition, the specific volumes used were those resulting from treatment 1.[4]

IV. EVALUATION OF THE DATA

In figure 2, specific volume, V $(=1/D)$, is plotted as a function of silica content for the potash-silica (A) and soda-silica glasses (B), as well as for a short series of lithia-silica glasses (C) discussed in the next section. For a closer study of these data, in order to determine the type of function which would best represent them, the points were replotted as in figure 3, the ordinates (ΔV) representing, for each binary series, the departures of the observed V-values from the respective straight lines a and b, indicated in figure 2.

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[3] Specific volume is, of course, determined from the same measurements as density—weight and volume. In figure 1, and also in table 1, density, being the more familiar quantity, is recorded rather than specific volume.

[4] The densities of the soda-silica glasses reported by Morey and Merwin [3], which were "annealed to maximal density", correspond very closely to those obtained from treatment 1, except in the very low soda range of composition. In this range the densities of our glasses were lower. After table 1 was prepared, the 14 soda-silica glasses used in treatment 4 (see table 1) were re-treated by the same procedure, except that a cooling rate of about 2° per hour was used. The resulting densities were as follows:

(4) Devitrified.	(21) 2.4992.	(34) 2.4587.	(41) 2.4307.	(45) 2.4059.
(5) 2.5312.	(30) 2.4752.	(37) 2.4468.	(42) 2.4168.	(49) 2.3336.
(13) 2.5120.	(32) 2.4697.	(40) 2.4381.	(43) 2.4179.	

As was expected, these densities are greater than those resulting from treatment 1. The new densities, although remaining lower than those of Morey and Merwin for glasses containing less than about 23 percent of soda, were somewhat higher than theirs (in the order of about 0.003) for glasses containing more than about 27 percent of soda.

Certainly none of the simpler types of curves would be adequate for representing these data since, from figure 3, the best fit appears to be provided for each series by a line undergoing a "deflection" (that is, a rapid, although perhaps "continuous," alteration in direction) through each of two narrow ranges of composition, while remaining relatively straight outside these ranges.[5] For the purpose of simplicity in expressing this fact mathematically, it was decided that three straight lines could be fitted to the points of each binary series, the intersections of these lines representing the deflections in the "true" function—in whatever way that function should "properly" be expressed.

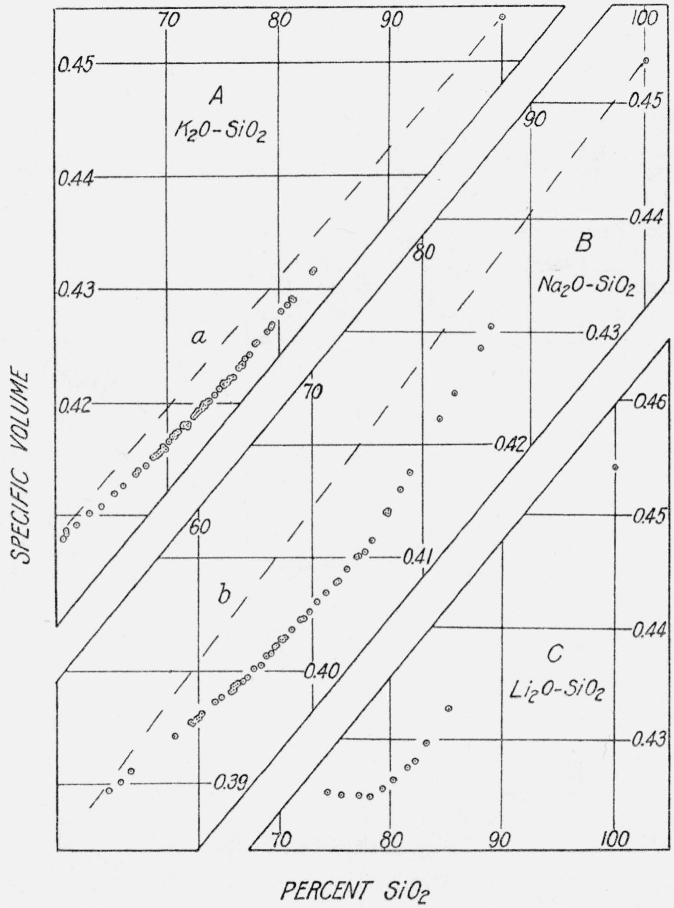

Figure 2.—*Specific volume of A the potash-silica, B the soda-silica, and C the lithia-silica glasses plotted against the percentage of silica.*

The dashed lines in figure 3 for each series represent the maximum expected deviation on either side of the "best-fitting" straight lines (the constants of which were evaluated as part of the correlation of the ternary relations described below), taking into account only the esti-

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[5] In the previously mentioned report by Morey and Merwin the "smooth curve," which they have drawn through their density-composition (and also their refractive index-composition) points, exhibits similar characteristics—a fact made sharply evident by plotting the first derivative of their curve.

mated maximum error in making the chemical analyses (expressible as ±0.2 percent SiO_2), and in the measurement of specific volume (±0.0001). The fact that only three or four points in each series lie outside these limits is further indication that the method of heat treatment is reproducible.

To determine the relation between specific volume and composition of the soda-potash-silica glasses, their compositions were plotted on a triordinate diagram (fig. 4). Then several lines of constant V (specific volume) were obtained by first interpolating graphically each selected constant V value between several appropriate pairs of neighboring points, and then sketching a smooth line through each of the sets of interpolated points representing the same V value. The trend and separation of the constant V lines thus drawn revealed that the over-all surface, representing the variation in specific volume with composition, divided itself into several relatively flat regions of different slopes. Accordingly, each region was treated arbitrarily as a

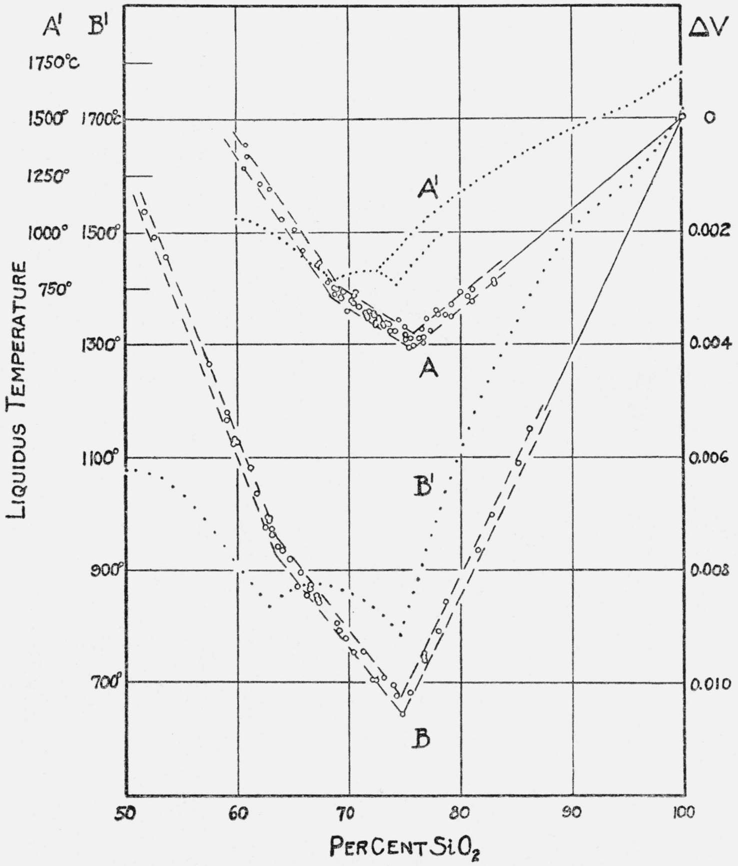

FIGURE 3.—*Departures* (ΔV) *of the observed specific volumes of A the potash-silica, and B the soda-silica glasses from the corresponding straight lines a and b in figure 2.*

A' and B' are the corresponding liquidus curves, taken from references [3] and [4].

plane of the form $V=aA+bB+eE$, in which A, B, and E are the percentages of silica, soda, and potash, respectively, and a, b, and e are constants which are different for each region. The constants were evaluated for each region by the method of least squares, using the foregoing graphic representation as a guide for the appropriate grouping of the points. The data for the two binary series were, of course, also included in these calculations, but the points are not recorded on the triordinate diagrams. The constants thus derived are shown in table 2 and are given to 5×10^{-7} in order to avoid numerical errors in the "computed specific volumes" as great as the error in measuring specific volume. The statistical error in the constants themselves is, of course, much greater than 5×10^{-7}.

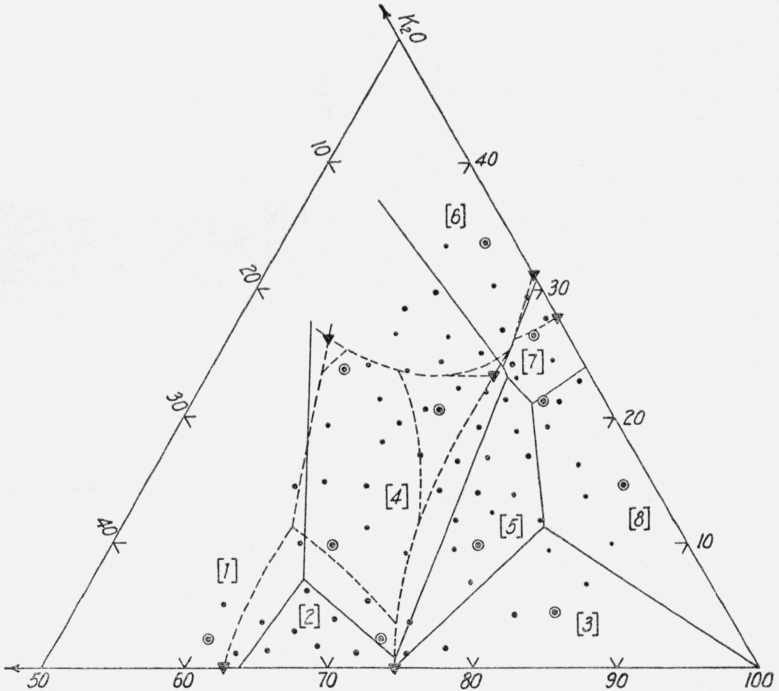

FIGURE 4.—*Compositions of the soda-potash-silica glasses and the boundaries (solid lines) of the various planes (numbered as in table 2) representing the surface on which the specific volumes lie.*

Dotted lines are the boundary curves of the phase-equilibrium diagram. Points in circles identify the glasses used in the heat-treating experiments outlined in figure 1.

Simultaneous solutions, by pairs, of the equations of adjacent planes yielded the lines of intersection of the planes (shown by heavy lines, fig. 4).

TABLE 2.—*Numerical value of constants for the different regions*

Constant×100	Region→1	2	3	4	5	6	7	8
a	0.428 00	0.437 10	0.454 00	0.441 05	0.448 35	0.441 65	0.447 85	0.454 00
b	.348 60	.332 65	.282 95	.320 55	.299 60	.290 85	.273 20	.265 15
e	.358 90	.339 15	.308 00	.350 75	.334 00	.356 25	.342 50	.322 85

It is not the authors' contention, however, that physical significance is necessarily to be attached to the system of planes as derived, nor that the individual deviations represent merely experimental or random errors. On the contrary, if, by an examination of the magnitude and distribution of plus and minus departures from the calculated surface (recorded in fig. 5), the reader is enabled to form some conception of what the "true" surface would be, making due allowance for experimental error, then the solution here presented has fulfilled its chief purpose. If the latter viewpoint is taken, then the fact that a different number of parameters or some other grouping of the points would lead to a different statistical solution becomes of minor importance, since the discrepancies between two such solutions would be compensated for by a correspondingly different pattern of departures. That is, the "true" surface conceivably would be indicated in either case. To the authors, the calculated surface here presented is a very good approximation to the "true" surface in view of the comparative absence of systematic deviation.

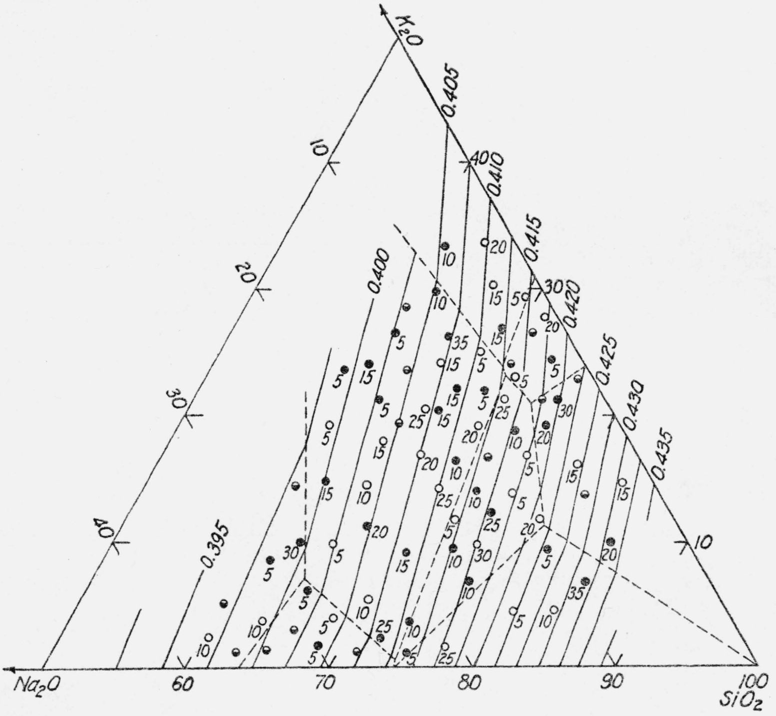

FIGURE 5.—*Relation between specific volume and composition of some soda-potash-silica glasses.*

Observed specific-volume values lie on (◒), above (●), or below (○), the computed surface, by the amounts indicated, times 10^{-5}. Density deviations equal approximately six times these amounts.

V. DISCUSSION

If the specific volume-composition relations are compared to the liquidus curves of the appropriate equilibrium diagrams presented by Kracek [4, 5, 6, 7], one finds a surprising, if not significant, correspondence. In the case of the soda-silica data (*B*, fig. 3), the two deflections occur at or near the eutectic compositions (See *B'*, fig. 3, at 63 and 75 percent SiO_2). In the potash-silica data (*A*, fig. 3), the deflection at 69 percent SiO_2 adequately reflects the eutectic at this composition (see *A'*, fig. 3). The other deflection occurs at about 76 percent SiO_2, and appears to correspond, not to the "true" eutectic (which is at about 72 percent SiO_2), but to the minimum point along the metastable cristobalite liquidus curve, which point is found under conditions more nearly comparable to the glassy state than those under which the "true" eutectic composition is found.

Concerning the soda-potash-silica data, the deflections (represented as lines of demarcation, fig. 4) correspond roughly to the boundary curves of the equilibrium diagram (dotted lines, fig. 4) connecting the eutectic compositions (small triangles, fig. 4) or other invariant points.

The plot of the specific volume-composition data thus far obtained on the lithia-silica glasses (*C*, fig. 2) is included here only for comparative purposes, since this phase of the work is far from complete. The two deflections—at about 78 and 82 percent SiO_2—are, however, the most pronounced observed for any series of glasses studied at this Bureau. The deflection at 82 percent SiO_2 corresponds to the eutectic at that composition, but the equilibrium diagram does not indicate a "critical composition" at 78 percent SiO_2.

A series of 10 boric oxide-silica glasses was reported by Cousens and Turner [8]. They plotted density as a function of composition and concluded that these data should be represented by a "smooth curve" showing no deflections—an apparent contradiction to the interpretation just given concerning our own data. It appears, however [9], that the eutectic, if any, in the boric oxide-silica system would occur very near to 100 percent B_2O_3, so that the existence of a corresponding deflection in the density-composition line could not easily be demonstrated, even if one were expected. Of interest, furthermore, is the fact that, when the specific volumes instead of the densities of their glasses are plotted against the compositions, the points are found to lie upon a single *straight* line connecting the specific volume of vitreous silica (0.4540) with that of vitreous B_2O_3, using for the latter the value 0.5440 as computed from the density reported by the same laboratory in a subsequent paper [10].[6]

VI. SUMMARY

The foregoing presentation may be briefly summarized as follows:

1. Data are presented on the density and composition of 194 glasses in the ternary system soda-potash-silica.

2. A reproducible and, it is felt, consistent method of heat treatment is described which, when applied simultaneously to all the glasses, results in the cooling of each glass at the same rate through its own "critical range" as defined.

3. A study of the data presented does not indicate that there is a simple relationship between specific volume and composition, but that the relation is best expressed by a system of planes in a three-dimensional triordinate diagram whose intersections correspond fairly well to the boundary curves and eutectic compositions indicated by

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[6] In that paper, the value reported in the earlier paper [8] was said to have been erroneous.

the equilibrium diagram of Kracek. The correspondence is very close for the soda-silica and potash-silica data, but it is only approximate for glasses containing all three constituents.

4. Data on the specific volume of 10 lithia-silica glasses are presented graphically.

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