

The effect of some glass components on the viscosity of glass

The aim of this investigation was to evaluate the effects of the most common glass components on glass viscosity and to be able to calculate the viscosity-temperature curve of a planned glass composition.

These calculations are based on three different independently performed experimental series. The first investigation was composed of the most common soda-lime glass components: SiO_2 — Al_2O_3 Na_2O — K_2O — CaO — MgO and the results will be published elsewhere (1).

The second investigation was aimed to establish the effect of the molecular relationship between Na_2O , K_2O and Li_2O in a basic household-type glass on viscosity and other different properties.

The third series was aimed to investigate the effects of Li_2O , BaO , ZnO , PbO and B_2O_3 .

All the compositional changes were calculated on a molecular basis and in relation to the silica content.

The glass batches giving 2000 g of glass were mixed in a porcelain ball mill, melted in platinum lined ceramic crucible in an electric furnace at 1400—1425° C overnight, poured out, mixed and remelted for good homogeneity.

Viscosity values were determined in the range of 10^2 — 10^7 poise in a rotating cylinder viscometer described elsewhere (1).

The viscosity-temperature curves were calculated by the Fulcher-Tamman equation:

$$T = T_0 + \frac{B}{\log \eta + A}$$

Where: T = temperature in °C
 $\log \eta$ = log poise viscosity
 B , A and T_0 = constants

The experimentally determined viscosity values were smoothed by an iterative least-squares method to the Fulcher equation and the standard deviations for each viscosity-temperature curve were less than 1,0° C.

Experimental Results

As the calculations are based on different experimental series, there is no randomization or any balance in the material of the investigation. This is a serious bias, but to construct a single balanced experimental plan for 10 different components on several levels should practically be impossible. E. g. 10 components on three levels each, gives 59049 experiments. The only practical way was to make 2—3 experiments for every component and balance the experiments with the results of the first series, which was a statistically correct, balanced investigation. This method puts very high demand on good reproducibility and high accuracy on the viscosity measurements, but as the results proved, no severe loss of accuracy has resulted.

The glass compositions were calculated on a molecular basis as moles per mole of silica.

The effect of B_2O_3 was measured on two levels and the results did not fit into the pattern. It was obvious that more points are necessary for the evaluation of the B_2O_3 -effect and therefore it was decided to make a separate investigation for this, which will be published separately.

The molecular compositions and the experimentally determined Fulcher-constants are shown on Table 1. The compositions no. 1—10 comes from the series for the investigation of Li_2O , BaO , ZnO and

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Table 1. Glass compositions expressed in moles, and corresponding Fulcher-constants.

No	SiO ₂	Al ₂ O ₃	Na ₂ O	CaO	Li ₂ O	BaO	ZnO	PbO	K ₂ O	MgO	B	A	T _o
1	1.0	0.010	0.1900	0.1800							3838.7	1.448	302.8
2	1.0	0.010	0.1850	0.1800	0.0050						3908.5	1.485	290.9
3	1.0	0.010	0.1750	0.1800	0.0150						3997.5	1.510	272.5
4	1.0	0.010	0.1600	0.1800	0.0300						4025.0	1.509	258.3
5	1.0	0.010	0.1900	0.1300		0.0500					4058.4	1.561	253.7
6	1.0	0.010	0.1900	0.0800		0.1000					3937.3	1.464	251.7
7	1.0	0.010	0.1900	0.1500			0.0500				4083.6	1.568	273.3
8	1.0	0.010	0.1900	0.1000			0.1000				4366.4	1.668	246.9
9	1.0	0.010	0.1900	0.1550				0.0250			3758.4	1.363	286.2
10	1.0	0.010	0.1900	0.1300				0.0500			4013.6	1.501	252.3
11	1.0	0.005	0.0415	0.1107	0.0191	0.0170	0.0423	0.0200	0.0995		4965.0	1.877	203.4
12	1.0	0.005	0.0364	0.1107	0.0000	0.0170	0.0423	0.0200	0.1236		4895.2	1.874	239.2
13	1.0	0.005	0.0253	0.1107	0.0233	0.0170	0.0423	0.0200	0.1114		5005.2	1.890	204.4
14	1.0	0.005	0.0602	0.1107	0.0277	0.0170	0.0423	0.0200	0.0722		4946.7	1.844	180.9
15	1.0	0.005	0.0546	0.1107	0.0126	0.0170	0.0423	0.0200	0.0928		4901.6	1.848	209.4
15	1.0	0.005	0.0500	0.1107	0.0000	0.0170	0.0423	0.0200	0.1100		4864.8	1.854	233.1
17	1.0	0.005	0.0889	0.1107	0.0000	0.0170	0.0423	0.0200	0.0711		4778.9	1.796	215.5
18	1.0	0.005	0.0661	0.1107	0.0190	0.0170	0.0423	0.0200	0.0749		4899.9	1.831	193.3
19	1.0	0.005	0.0611	0.1107	0.0094	0.0170	0.0423	0.0200	0.0896		4871.2	1.839	212.4
20	1.0	0.010	0.1900	0.1800					0.0400		3861.8	1.489	280.4
21	1.0	0.010	0.1900	0.1800					0.0800		3791.8	1.513	268.2
22	1.0	0.030	0.1900	0.1800							3974.7	1.433	298.6
23	1.0	0.045	0.1900	0.1800							4005.3	1.410	303.1
24	1.0	0.010	0.1850	0.1800							3964.5	1.473	292.4
25	1.0	0.010	0.1750	0.1800							4028.3	1.490	292.4
26	1.0	0.010	0.1600	0.1800							4123.6	1.515	292.4
27	1.0	0.010	0.1900	0.1300							4154.0	1.401	262.8
28	1.0	0.010	0.1900	0.0800							4368.9	1.328	235.6
29	1.0	0.010	0.1900	0.1500							4065.7	1.427	274.6
30	1.0	0.010	0.1900	0.1000							4288.3	1.364	245.0
31	1.0	0.010	0.1900	0.1800							3933.3	1.465	292.4
32	1.0	0.010	0.1900	0.1550							4044.0	1.433	277.6
33	1.0	0.010	0.1900	0.1300							4155.2	1.402	262.8
34	1.0	0.010	0.1900	0.1800						0.0300	4114.5	1.624	281.6
35	1.0	0.010	0.1900	0.1800						0.0450	4204.5	1.707	276.3

PbO, — the compositions no. 11—19 from the series for the investigation of the relationship between Li₂O, Na₂O and K₂O and no. 20—35 are "dummies" calculated from the soda-lime glass series.

The dependence of the Fulcher constants (B, A and T_o) on the glass components was calculated by multiple regression analysis and resulted in three equations:

$$\text{"B"} = 2830.0 A - 6802.0 N - 4566.7 C - 611.6 L - 3622.9 Ba + 1121.1 Z - 3650.3 Pb - 1519.0 K + 6590.0 M + 5991.5.$$

$$\text{"A"} = 1.29799 A - 1.37112 N + 1.22366 C + 0.90151 L + 1.73879 Ba + 3.13803 Z + 1.32178 Pb + 0.90479 K + 5.63513 M + 1.50848.$$

$$\text{"T}_o\text{"} = 308.48 A - 53.25 N + 584.23 C - 1515.11$$

$$L + 95.40 Ba - 49.03 Z - 207.86 Pb - 253.60 K - 410.44 M + 195.29.$$

where:

$$A = \frac{Al_2O_3}{SiO_2}, N = \frac{Na_2O}{SiO_2}, C = \frac{CaO}{SiO_2}$$

$$L = \frac{Li_2O}{SiO_2}, Ba = \frac{BaO}{SiO_2}, Z = \frac{ZnO}{SiO_2}$$

$$Pb = \frac{PbO}{SiO_2}, K = \frac{K_2O}{SiO_2} \text{ and } M = \frac{MgO}{SiO_2}$$

calculated in moles.

For practical calculations the equations has been recalculated into weight relationships, where the factors are calculated for the weight of the components per weight of SiO₂, or percentage of component per percentage of SiO₂. The equations are the following:

Table 2. Compositions in weight percentages, determined and calculated temperatures and deviations at log η 2.0, 4.0 and 6.0 viscosity levels.

No	Weight %										°C at log η 2.0			°C at log η 4.0			°C at log η 6.0		
	SiO ₂	Al ₂ O ₃	Na ₂ O	CaO	Li ₂ O	BaO	ZnO	PbO	K ₂ O	MgO	Determ.	Calc.	Δ T	Determ.	Calc.	Δ T	Determ.	Calc.	Δ T
	1	72.41	1.23	14.19	12.17							1416.1	1423.7	7.6	1007.4	1009.3	1.9	818.2	817.3
2	72.55	1.23	13.85	12.19	0.18						1412.4	1421.5	9.1	1003.5	1006.1	2.6	813.1	813.3	0.2
3	72.84	1.24	13.15	12.24	0.54						1411.5	1417.5	6.0	998.1	1000.0	1.9	804.9	805.5	0.6
4	73.26	1.24	12.09	12.31	1.09						1405.3	1410.8	5.5	988.9	990.4	1.5	794.3	793.5	-0.8
5	68.40	1.16	13.41	8.30		8.73					1398.4	1404.3	5.9	988.5	990.0	1.5	795.5	797.3	1.8
6	64.82	1.10	12.70	4.84		16.54					1388.4	1385.2	-3.2	972.3	971.0	-1.3	779.2	777.4	-1.8
7	70.39	1.19	13.80	9.85			4.77				1417.8	1419.6	1.8	1006.7	1008.5	1.8	812.9	814.4	1.5
8	69.36	1.18	13.59	6.47			9.39				1437.3	1435.8	-1.5	1017.2	1014.7	-2.5	816.3	813.2	-3.1
9	68.94	1.17	13.51	9.97				6.40			1403.8	1409.8	6.0	987.0	993.5	6.5	796.7	800.4	3.7
10	65.79	1.12	12.89	7.58				12.22			1398.7	1395.9	-2.8	981.9	977.6	-4.3	787.4	783.5	-3.9
11	66.89	0.57	2.86	6.91	0.64	2.90	3.83	4.97	10.43		1484.0	1490.8	6.8	1048.2	1049.2	1.0	833.7	832.8	-0.9
12	65.87	0.56	2.47	6.81	0.00	2.86	3.77	4.89	12.77		1502.8	1503.8	1.0	1072.6	1069.0	-3.6	860.9	854.7	-6.2
13	66.71	0.57	1.74	6.89	0.77	2.89	3.82	4.96	11.65		1491.4	1484.7	-7.2	1064.2	1047.5	-16.7	838.8	831.3	-7.5
14	67.76	0.57	4.21	7.00	0.93	2.94	3.88	5.03	7.67		1467.8	1465.6	-2.2	1027.4	1029.5	2.1	811.5	816.1	4.6
15	66.90	0.57	3.77	6.91	0.42	2.90	3.83	4.97	9.73		1483.2	1483.4	0.2	1047.6	1048.2	0.6	834.0	834.9	0.9
16	66.19	0.56	3.41	6.84	0.00	2.87	3.79	4.92	11.42		1495.4	1498.2	2.8	1064.1	1063.9	-0.2	852.5	850.7	-1.8
17	67.12	0.57	6.16	6.94	0.00	2.91	3.85	4.99	7.48		1474.4	1481.1	6.7	1040.0	1048.8	8.8	828.8	839.1	10.3
18	67.48	0.57	4.60	6.97	0.64	2.93	3.87	5.01	7.93		1472.3	1471.7	0.6	1033.6	1036.6	3.0	819.0	824.1	5.1
19	66.89	0.57	4.22	6.91	0.31	2.90	3.83	4.97	9.40		1481.3	1483.9	2.6	1046.7	1049.3	2.6	833.8	836.7	2.9
20	69.27	1.18	13.58	11.64				4.34			1387.3	1384.4	-2.9	984.0	983.4	-0.6	796.1	796.5	0.4
21	66.38	1.13	13.01	11.15				8.33			1347.6	1345.8	-1.8	956.0	957.7	1.7	772.9	775.8	2.9
22	72.69	1.23	13.87	12.21							1433.9	1431.6	-2.3	1016.8	1014.9	-1.9	822.9	821.6	-1.3
23	73.23	1.24	13.22	12.30							1446.7	1447.2	0.5	1026.2	1026.0	-0.2	830.2	830.3	0.1
24	74.07	1.26	12.23	12.44							1465.5	1470.4	4.9	1040.1	1042.7	2.6	841.1	843.2	2.1
25	74.95	1.27	14.69	9.09							1484.2	1482.2	-2.0	1031.9	1030.6	-1.3	824.1	823.3	-0.8
26	77.66	1.32	15.22	5.80							1544.1	1543.8	-0.3	1053.0	1053.0	0	829.5	829.9	0.4
27	73.91	1.25	14.49	10.35							1461.0	1458.2	-2.8	1023.8	1021.8	-2.0	822.0	820.7	-1.3
28	76.55	1.30	15.00	7.15							1519.7	1518.7	-1.0	1044.4	1043.9	-0.5	827.3	827.2	-0.1
29	72.41	1.23	14.19	12.17							1427.5	1423.5	-4.0	1012.1	1009.1	-3.0	819.3	817.1	-2.2
30	73.66	1.25	14.44	10.66							1455.3	1452.4	-2.9	1021.8	1019.7	-2.1	821.6	820.2	-1.4
31	74.95	1.27	14.69	9.09							1484.2	1482.2	-2.0	1031.9	1030.6	-1.3	824.1	823.3	-0.8
32	70.68	3.60	13.85	11.87							1456.2	1455.1	-1.1	1030.3	1029.4	-0.7	833.3	832.9	-0.4
33	69.43	5.30	13.61	11.66							1477.7	1478.6	0.9	1043.5	1044.4	0.9	843.7	844.6	0.9
34	71.37	1.21	13.99	11.99				1.44			1417.0	1412.9	-4.1	1013.2	1010.4	-2.8	821.3	819.0	-2.3
35	70.86	1.20	13.89	11.91				2.14			1411.7	1407.6	-4.1	1013.5	1010.7	-2.9	822.1	819.8	-2.3

Table 3.

Viscosity $\log \eta$	Effect of 1 kg component per 100 kg silica in \pm °C									
	SiO ₂	Al ₂ O ₃	Na ₂ O	CaO	MgO	Li ₂ O	K ₂ O	BaO	ZnO	PbO
2.0	4.8	9.1	-15.3	-12.0	-5.1	-39.9	-6.3	-5.9	-5.5	-4.6
3.0	3.5	7.0	-12.7	-7.2	-0.7	-36.8	-4.9	-4.2	-3.1	-3.5
4.0	2.8	5.9	-10.8	-4.3	+0.8	-35.0	-4.1	-3.1	-1.8	-2.8
5.0	2.2	5.1	-9.5	-2.5	1.2	-34.0	-3.6	-2.4	-1.2	-2.4
6.0	1.7	4.6	-8.4	-1.3	1.1	-33.4	-3.3	-2.0	-0.9	-2.1
7.0	1.4	4.2	-7.6	-0.2	0.8	-32.9	-3.1	-1.7	-0.7	-1.9

"B" = 1667.7 A — 6594.1 N — 4892.8 C — 1230.0 L — 1419.6 Ba + 827.8 Z — 982.7 Pb — 968.8 K + 9822.6 M + 5991.5.

"A" = — 0.76490 A — 1.32922 N + 1.31105 C + 1.81302 L + 0.68133 Ba + 2.31718 Z + 0.35584 Pb + 0.57710 K + 8.39936 M + 1.50848.

"T₀" = 181.79 A — 5162 N + 625.95 C — 3047.00 L + 37.38 Ba — 36.21 Z — 55.96 Pb — 161.75 K — 611.78 M + 195.29.

To test the goodness of these equations, in *Table 2* are shown the compositions in weight percentages, determined and calculated temperatures for three viscosity levels and differences between determined and calculated values.

The standard deviations for $\log \eta$: 2.0, 4.0 and 6.0 levels are:

$$\sigma_{2.0} = 4.10 \text{ } ^\circ\text{C}$$

$$\sigma_{4.0} = 2.97$$

$$\sigma_{6.0} = 3.19$$

which are quite satisfactory for all practical purposes.

As it is difficult to see the effect on viscosity from the Fulcher constants, the effect of the increase by 1 kg of a component per 100 kg of silica on temperature at different viscosity levels in a basic soda-lime glass composition has been calculated and is shown on *Table 3*.

The basic glass composition, and its viscosity-temperature values are:

SiO ₂	73.0 %	$\log \eta$ °C
Al ₂ O ₃	0.5 "	2.0 1417.0
Na ₂ O	14.0 "	3.0 1165.9
CaO	12.5 "	4.0 1006.5
		5.0 896.4
		6.0 815.8
		7.0 754.2

Summarizing the effects of the different glass components on viscosity, the following general rules can be established:

1. The alkali-oxides decrease viscosity and tend to make the glass "longer". This is most characteristic for lithium oxide. Lithia is the most effective viscosity-decreasing component, its effect is more than twice as large as that of sodium oxide. Nevertheless, the use of lithium oxide is determined by its aptitude to increase the tendency to devitrification and its high price. The viscosity-decreasing effect of potassium oxide is about one half of that of sodium oxide.
2. The divalent oxides: lime has a strong viscosity decreasing effect only in the high temperature range, increasing lime content makes the glass "short". Magnesia has an effect similar to that of lime, but the viscosity decrease is much less compared with lime.
The viscosity-decreasing effect of BaO, ZnO and PbO is almost identical and about the same degree as the effect of potassium oxide.

(1) Viscosity-Temperature Relations in the Glass system SiO₂ — Al₂O₃ — Na₂O — K₂O — CaO — MgO in the Compositional Range of Technical Glasses. To be published in *Glass Technology*. T. Lakatos, L.-G. Johansson, B. Simingsköld.