journal

J. Am. Ceram. Soc., 90 [8] 2622–2625 (2007)
DOI: 10.1111/j.1551-2916.2007.01751.x

Global Model for Calculating Room-Temperature Glass Density from the Composition

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A large silicate glass density database including 6719 compositions, obtained from SciGlass© and other published sources, was systematically analyzed for agreement among room-temperature density data reported in the literature in 1030 different studies from various investigators. It was found that the literature data agree very well. Using just 101 of the initial 6719 degrees of freedom, 99.8% of the measured density data variance could be reproduced by an empirical polynomial model. The standard error of the developed model was 0.019 g/cm^3.

I. Introduction

Since the beginning of scientific research on glass in the 19th century by Schott in Jena, Germany,[1] thousands of room-temperature density data were collected. The SciGlass© Information System[2] contains the largest collection of those data (more than 52 000 in 2006), plus 18 procedures for density calculation from the chemical glass composition. Glass density is important in the industry for quality control because it is relatively easy to measure very accurately, reflecting the compositional constancy of the produced glass. Other areas where the density is important include weight reduction of glass components for glass transport and application, e.g., for flat-panel displays. In 2003, Priven and Mazurin[3] compared various methods for density estimation, reporting estimation errors of 0.038–0.11 g/cm^3 for glasses containing more than 50 mol% silica. Some important models were also summarized by Scholze (p. 204).[4] A statistical analysis of the available silicate glass density data in SciGlass© has not been performed. It is the objective of this paper to complete this analysis for (1) determining the agreement between various investigators, (2) reducing the density estimation errors given by Priven and Mazurin,[3] (3) detecting interactions of glass components that affect the density.

II. Model Development

The source data and all source data references used in this work are listed in the SciGlass database[2] and are available in detail from the author.[5] The glass composition basis used was mole percent (mol%) of oxides. The concentrations of some transition metals in varying oxidation states such as Fe_xO_y were combined by addition into the most common oxidation state, e.g., Fe_2O_3. The compositions used were limited to silicate glasses with concentrations of 40%–87% mol% SiO_2, further limits are given in Table I, and a long list of additional validity limits regarding component combinations are available from the author.[5] The glass compositions in mole percent were employed as independent variables for least-square linear regression modeling, while the densities were chosen as model responses. The equation allowing density estimation is a common third-order polynomial

$$\text{Density} = b_o + \sum_{i=1}^{n}\left[b_i C_i + \sum_{k=i}^{n}\left(b_{ik} C_i C_k + \sum_{m=k}^{n} b_{ikm} C_i C_k C_m\right)\right] \quad (1)$$

where the densities are expressed in g/cm^3, the b-values are the model coefficients in Table II, the C-values are the glass component concentrations in mol% excluding silica, and n is the total number of glass components excluding silica. Equation (1) with the density as immediate response was used to facilitate model application. From the thermodynamic standpoint, only molar volumes but not densities of ideal mixtures are additive; however, glasses are nonideal mixtures and empirically no improvement in accuracy is expected based on a model with the molar volume as response.[6] Strongly correlated terms in Eq. (1) with an absolute of "Pearson's r" higher than 0.8 or with insufficient support were excluded from the calculation. Coefficients with a significance of at least 95% were selected by mixed forward selection and backward elimination considering the coefficient hierarchy. Outlying data were deleted based on standardized or externally studentized residuals higher than three. A validation procedure was performed by examining high Cook-values individually, by calculating R^2 (predicted), and by determining R^2 (validation), and by comparing the model standard error with the experimental error (taking into account incidental interlaboratory differences) displayed in Fig. 1. The error within one arbitrary laboratory or the average error of several individual laboratories cannot be considered as the global interlaboratory measurement error determined in Fig. 1. R^2 (validation) was obtained by sorting all source data after increasing responses, selecting each fifth point to form an independent validation dataset, and basing the validation model on the remaining 80% of the data. Further details of the statistical data analysis procedure are described elsewhere.[7,8] Model calculations can be performed conveniently in a program connected to this study,[5] including error estimation.

III. Modeling Results

Table II lists all model coefficients. The error and significance of the coefficients can be derived from the t-values in Table III that equal the quotient of the considered coefficient and its error. The inverse information and Pearson's correlation matrices are also provided.[5] The total number of analyzed source data equaled 7509. After stepwise exclusion of 790 composition-density sets, the final model contained 6719 data, while the number of coefficients was 101, resulting in a degree of freedom of 6618. The density minimum, average, maximum, and standard deviation in g/cm^3 among the considered 6719 data were 2.110, 2.631, 6.810, and 0.469, respectively. The outliers are given on the author's website.[5] They represent 10.5% of the initial data of

H. Gan—contributing editor

Manuscript No. 22728. Received January 24, 2007; approved March 19, 2007.
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Table I. Concentration Distributions Among Source Data (Comp, Glass Component; Avg, Average Concentration in Mol%; Max, Maximum Concentration in Mol%; SD, Standard Deviation of all Concentrations in Mol%; #, Number of Concentration Values Above 0), Concentration Minima Always Zero Except for SiO_2 With 40 Mol%

Comp	Avg	Max	SD	#	Comp	Avg	Max	SD	#
Ag_2O	0.000	0.078	0.003	51	Nb_2O_5	0.000	0.006	0.000	53
Al_2O_3	2.810	20.000	4.783	2907	Nd_2O_3	0.004	1.006	0.033	243
As_xO_y	0.001	0.240	0.013	80	NiO	0.018	2.921	0.110	319
B_2O_3	2.219	23.097	4.825	1410	P_2O_5	0.014	1.888	0.097	376
BaO	0.511	20.000	2.440	665	PbO	1.758	60.000	6.855	854
Bi_2O_3	0.000	0.312	0.004	2	PdO	0.000	0.071	0.004	133
Br	0.000	0.065	0.001	4	Pr_xO_y	0.000	0.012	0.001	202
CaO	4.441	40.000	8.145	2550	Rb_2O	0.000	0.033	0.002	171
CdO	0.008	1.699	0.059	246	Re_xO_y	0.000	0.033	0.002	95
Ce_xO_y	0.003	0.909	0.030	272	Rh_xO_y	0.000	0.024	0.002	207
Cl	0.012	3.238	0.102	221	RuO_2	0.001	0.139	0.010	270
Co_xO_y	0.000	0.194	0.005	13	Sb_xO_y	0.002	5.000	0.064	94
Cr_2O_3	0.003	0.561	0.019	407	SeO_2	0.000	0.268	0.007	22
Cs_2O	0.001	0.095	0.005	210	SiO_2	68.036	87.100	9.756	6719
CuO	0.002	0.233	0.016	190	Sm_2O_3	0.000	0.018	0.001	197
F	0.086	18.288	0.644	388	SnO_2	0.000	0.072	0.002	58
Fe_xO_y	0.250	10.000	1.064	687	SO_3	0.013	1.984	0.067	429
Ga_2O_3	0.000	0.184	0.002	1	SrO	0.324	20.000	2.009	450
Gd_2O_3	0.000	0.025	0.000	3	TeO_2	0.000	0.091	0.003	64
H_2O[†]	0.019	9.711	0.365	42	ThO_2	0.001	1.228	0.031	12
I	0.000	0.066	0.001	116	TiO_2	0.414	20.000	2.295	438
K_2O	3.023	30.000	6.067	2334	Tl_2O_3	0.000	0.032	0.001	5
La_2O_3	0.005	1.109	0.047	321	U_xO_y	0.002	2.065	0.043	17
Li_2O	1.877	30.000	5.600	916	V_2O_5	0.000	2.130	0.029	3
MgO	1.641	30.000	4.695	1388	WO_3	0.000	0.063	0.002	7
Mn_xO_y	0.023	4.988	0.204	340	Y_2O_3	0.000	0.027	0.002	200
Mo_xO_y	0.004	0.883	0.034	268	ZnO	0.422	20.000	2.204	434
Na_2O	11.899	40.460	9.981	5009	ZrO_2	0.152	10.000	0.836	434

[†]High concentrations of H_2O are introduced under elevated pressure.

7509 and were distributed unevenly in the multidimensional composition space. For example, more of the compositions containing PbO appeared as outliers (24.6% out of 1135 compositions containing PbO), compared with compositions containing all other components. The model standard error was 0.0191 g/cm^3, with R^2, R^2 (adjusted), and R^2 (validation) values of 0.9984, and with an R^2 (predicted) of 0.9983.

IV. Discussion

To better understand the model application a simple calculation example is given here: a binary sodium silicate glass containing 20 mol% Na_2O would have an estimated density of $2.121560704+0.018129123\times 20-0.000264838\times 20^2+0.000001614\times 20^3 = 2.391$ g/cm^3, where 95 out of 100 independently prepared and measured samples would fall in an error range of 2.391 ± 0.008 g/cm^3 (95% confidence interval in mass production),[5] assuming a perfectly constant glass composition. For comparison, 45 literature data from SciGlass[2] give a value of 2.388 g/cm^3 with a standard deviation of 0.014 g/cm^3, which relates well to the model estimation. A plot of all 6719 measured versus estimated density values is provided in Fig. 2. The model standard error of 0.0191 g/cm^3 is placed within the estimated experimental error range of 0.007–0.026 g/cm^3, taking into account incidental interlaboratory differences, as shown in Fig. 1. This is a sign of a well-developed model that does not contain too many or too few terms and where outliers were excluded accordingly. The relative ratio of 10.5% outliers in the global model can be considered as equal to the 11.1% outliers in the binary system SiO_2–Na_2O given in Fig. 1.

The high coefficient of determination, R^2, shows an excellent model fit, i.e., 99.84% of the measured density variance is explained by the model. Glass densities at room temperature can be described well over relatively wide composition areas using polynomial functions, which possibly is a sign for constant stoichiometrical groupings in glass as discussed by Markova *et al.*[9] It is assumed that phase separated, crystallized, rapidly quenched (Scholze[4], p. 211), or difficult to prepare samples preferably appeared as outliers because unusual composition-density changes may occur. For example, some lead silicate glasses are subjected to evaporation during melting and phase separation during cooling,[1] which may explain why as much as 24.6% of them appeared as outliers. It was not the subject of this work to investigate systematic differences between experimental data from selected laboratories to the majority of all other experimental data, but it is planned in a later study because it would allow for reduced error of model estimations. The good agreement between R^2 and R^2 (adjusted) indicates that the model does not include insignificant variables, while the agreement between R^2, R^2 (predicted), and R^2 (validation) shows homogeneous leverage, i.e., most data contributed evenly to the model result in Table II. The standard error of 0.0191 g/cm^3 in this study demonstrates an at least twofold improvement of the model error compared with earlier work.[2–4] In addition, the presented model covers a wider glass composition area than that of many previous papers[2] with high accuracy.

All model coefficients in Table II represent interactions with the main glass former silica that was excluded in Eq. (1), e.g., the terms Na_2O and $(Na_2O)^2$ used for fitting the simple density curve in Fig. 1 do not reflect the density behavior of sodium oxide alone but of sodium oxide in interaction with silica. For an accurate interpretation of the coefficients, the correlation matrix[5] must be considered. None of the variables are perfectly statistically independent, i.e., all variables interfere mutually. It will be hardly possible in future to de-correlate all variables

Table II. Coefficients of Glass Density Model at Room Temperature, Considering Validity Limits[5]

Variable	Coefficient	Variable	Coefficient	Variable	Coefficient
Intercept	2.121560704	CdO	0.052945783	$K_2O \times MgO$	−0.000337747
Al_2O_3	0.010525974	La_2O_3	0.10643194	$K_2O \times CaO$	−0.000349578
$(Al_2O_3)^2$	−0.000076924	Nd_2O_3	0.090134135	$K_2O \times SrO$	−0.000425589
B_2O_3	0.00579283	NiO	0.024289113	$K_2O \times BaO$	−0.000392897
$(B_2O_3)^2$	0.000129174	ThO_2	0.090253734	$Al_2O_3 \times CaO$	−0.000102444
$(B_2O_3)^3$	−0.000019887	U_xO_y	0.063297404	$Al_2O_3 \times PbO$	−0.000651745
Li_2O	0.012848733	Sb_xO_y	0.044258719	$Al_2O_3 \times TiO_2$	−0.000563594
$(Li_2O)^2$	−0.000276404	SO_3	−0.044488661	$Al_2O_3 \times BaO$	−0.000273835
$(Li_2O)^3$	0.00000259	F	0.00109839	$Al_2O_3 \times SrO$	−0.000177761
Na_2O	0.018129123	Cl	−0.006092537	$Al_2O_3 \times ZnO$	−0.000109968
$(Na_2O)^2$	−0.000264838	Remainder[†]	0.02514614	$Al_2O_3 \times ZrO_2$	−0.002381651
$(Na_2O)^3$	0.000001614	$Na_2O \times K_2O$	−0.000395491	$Na_2O \times PbO$	−0.000036455
K_2O	0.019177312	$Na_2O \times Li_2O$	−0.00031449	$Na_2O \times TiO_2$	−0.00014331
$(K_2O)^2$	−0.000319863	$K_2O \times Li_2O$	−0.000329725	$Na_2O \times ZnO$	−0.000155275
$(K_2O)^3$	0.00000191	$Na_2O \times B_2O_3$	0.000242157	$Na_2O \times ZrO_2$	−0.000126728
MgO	0.01210604	$K_2O \times B_2O_3$	0.000259927	$Na_2O \times Fe_2O_3$	−0.000371343
$(MgO)^2$	−0.000061159	$Li_2O \times B_2O_3$	0.000106359	$K_2O \times PbO$	−0.000525213
CaO	0.017992367	$MgO \times B_2O_3$	−0.000206488	$K_2O \times TiO_2$	−0.000386587
$(CaO)^2$	−0.00005478	$CaO \times B_2O_3$	−0.000032258	$K_2O \times ZnO$	−0.000329812
SrO	0.034630735	$PbO \times B_2O_3$	−0.000186195	$CaO \times PbO$	−0.00084145
$(SrO)^2$	−0.000086939	$Fe_2O_3 \times B_2O_3$	−0.000720268	$ZnO \times Fe_2O_3$	−0.001536804
BaO	0.049879597	$ZrO_2 \times B_2O_3$	−0.000697195	$Na_2O \times K_2O \times B_2O_3$	−0.000032967
$(BaO)^2$	−0.000168063	$Al_2O_3 \times B_2O_3$	−0.000735749	$Na_2O \times MgO \times CaO$	−0.000009143
ZnO	0.025221567	$Li_2O \times Al_2O_3$	−0.000116227	$Na_2O \times MgO \times Al_2O_3$	−0.000012286
$(ZnO)^2$	0.000099961	$Na_2O \times Al_2O_3$	−0.000253454	$Na_2O \times CaO \times Al_2O_3$	−0.000005106
PbO	0.070020298	$K_2O \times Al_2O_3$	−0.000371858	$Na_2O \times CaO \times PbO$	0.000100796
$(PbO)^2$	0.000214424	$MgO \times CaO$	0.000057248	$K_2O \times MgO \times CaO$	−0.00001217
$(PbO)^3$	−0.000001502	$MgO \times Al_2O_3$	0.000167218	$K_2O \times MgO \times Al_2O_3$	−0.000041908
Fe_xO_y	0.036995747	$MgO \times ZnO$	0.000220766	$K_2O \times CaO \times Al_2O_3$	−0.000012421
Mn_xO_y	0.016648722	$Li_2O \times CaO$	−0.00008792	$K_2O \times CaO \times PbO$	0.000125759
TiO_2	0.018820343	$Na_2O \times MgO$	−0.000300745	$MgO \times CaO \times Al_2O_3$	−0.000011236
ZrO_2	0.043059714	$Na_2O \times CaO$	−0.000228249	$CaO \times Al_2O_3 \times Li_2O$	−0.000016177
$(ZrO_2)^2$	−0.000779078	$Na_2O \times SrO$	−0.00023137	$Al_2O_3 \times B_2O_3 \times PbO$	0.000030116
Ce_xO_y	0.061277268	$Na_2O \times BaO$	−0.000171693		

[†]The remainder includes traces of: Ag_2O, Bi_2O_3, Br, Co_xO_y, Cr_2O_3, Cs_2O, CuO, Ga_2O_3, Gd_2O_3, I, MoO_3, Nb_2O_5, PdO, Pr_xO_y, Rb_2O, Re_xO_y, Rh_xO_y, RuO_2, SeO_2, Sm_2O_3, SnO_2, TeO_2, Tl_2O_3, WO_3, Y_2O_3.

completely because it would require a very high number of well-planned experiments. It is recommended to consider the model coefficients in this paper as preliminary findings until further experimental data become available. Nevertheless, as long as all model validity limits[5] are followed, mentioned in the section above about model development, accurate estimations are possible. Considering these precautions, influences of specific glass components and component interactions on the density can be derived from Table II. As expected, most interaction coefficients are negative, i.e., the simultaneous presence of several components leads to a less-efficient packing in the glass network structure,[10] thereby decreasing the density. At first sight it appears contradictory to the statement that at constant total alkali content experimental data[2] and the model estimations in this study[5] show a higher density of ternary mixed-alkali silicate glasses, compared with corresponding binary alkali silicate systems. This contradiction will be resolved in a planned forthcoming publication, which also includes a more detailed discussion of the relation between the glass composition and its room-temperature density.

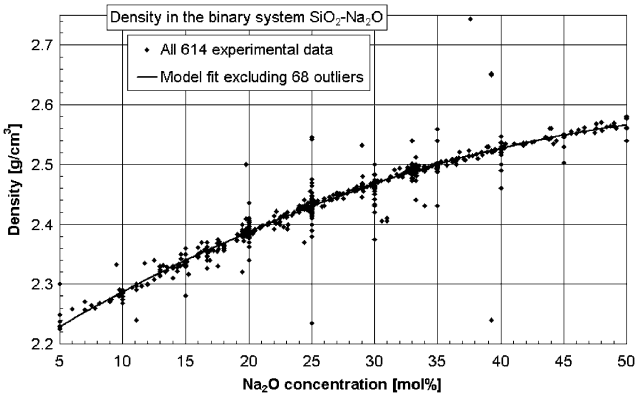

Fig. 1. Density curve (polynomial fit according to Eq. (1)) in the binary system SiO_2–Na_2O, standard error excluding outliers: 0.007 g/cm^3, standard error including outliers: 0.026 g/cm^3.

V. Summary

Statistical analysis of 6719 published room-temperature density data of glasses from the SciGlass© database[2] and other published sources provides proof of an excellent agreement between the literature data from 1030 different studies. A model for estimating the density with a standard error of 0.019 g/cm^3 was developed, improving the estimation error at least twice compared with earlier publications. The influences of glass components and component interactions could be quantified empirically.

Table III. *t*-Values of Coefficients From Table II

Variable	*t*-value	Variable	*t*-value	Variable	*t*-value
Intercept		CdO	7.011	$K_2O \times MgO$	−18.418
Al_2O_3	35.881	La_2O_3	16.834	$K_2O \times CaO$	−34.280
$(Al_2O_3)^2$	−6.656	Nd_2O_3	7.001	$K_2O \times SrO$	−18.544
B_2O_3	10.463	NiO	5.680	$K_2O \times BaO$	−18.824
$(B_2O_3)^2$	1.774	ThO_2	9.574	$Al_2O_3 \times CaO$	−11.081
$(B_2O_3)^3$	−7.846	U_xO_y	9.093	$Al_2O_3 \times PbO$	−29.653
Li_2O	23.128	Sb_xO_y	12.027	$Al_2O_3 \times TiO_2$	−13.715
$(Li_2O)^2$	−5.937	SO_3	−8.720	$Al_2O_3 \times BaO$	−7.738
$(Li_2O)^3$	2.432	F	2.201	$Al_2O_3 \times SrO$	−6.156
Na_2O	56.619	Cl	−2.263	$Al_2O_3 \times ZnO$	−4.348
$(Na_2O)^2$	−17.402	Remainder[†]	4.158	$Al_2O_3 \times ZrO_2$	−10.478
$(Na_2O)^3$	6.324	$Na_2O \times K_2O$	−28.880	$Na_2O \times PbO$	−3.128
K_2O	45.971	$Na_2O \times Li_2O$	−23.294	$Na_2O \times TiO_2$	−11.074
$(K_2O)^2$	−10.065	$K_2O \times Li_2O$	−19.077	$Na_2O \times ZnO$	−9.626
$(K_2O)^3$	2.386	$Na_2O \times B_2O_3$	23.393	$Na_2O \times ZrO_2$	−3.508
MgO	40.234	$K_2O \times B_2O_3$	17.703	$Na_2O \times Fe_2O_3$	−9.014
$(MgO)^2$	−6.155	$Li_2O \times B_2O_3$	4.749	$K_2O \times PbO$	−34.147
CaO	96.518	$MgO \times B_2O_3$	−5.685	$K_2O \times TiO_2$	−19.858
$(CaO)^2$	−13.586	$CaO \times B_2O_3$	−2.092	$K_2O \times ZnO$	−18.139
SrO	61.850	$PbO \times B_2O_3$	−12.927	$CaO \times PbO$	−52.594
$(SrO)^2$	−3.289	$Fe_2O_3 \times B_2O_3$	−13.172	$ZnO \times Fe_2O_3$	−3.920
BaO	107.615	$ZrO_2 \times B_2O_3$	−8.399	$Na_2O \times K_2O \times B_2O_3$	−20.018
$(BaO)^2$	−7.455	$Al_2O_3 \times B_2O_3$	−40.312	$Na_2O \times MgO \times CaO$	−4.827
ZnO	53.540	$Li_2O \times Al_2O_3$	−9.456	$Na_2O \times MgO \times Al_2O_3$	−5.083
$(ZnO)^2$	4.253	$Na_2O \times Al_2O_3$	−25.772	$Na_2O \times CaO \times Al_2O_3$	−5.525
PbO	200.039	$K_2O \times Al_2O_3$	−16.801	$Na_2O \times CaO \times PbO$	18.847
$(PbO)^2$	13.588	$MgO \times CaO$	6.104	$K_2O \times MgO \times CaO$	−3.871
$(PbO)^3$	−7.616	$MgO \times Al_2O_3$	9.425	$K_2O \times MgO \times Al_2O_3$	−6.635
Fe_xO_y	39.942	$MgO \times ZnO$	10.536	$K_2O \times CaO \times Al_2O_3$	−3.624
Mn_xO_y	11.568	$Li_2O \times CaO$	−7.155	$K_2O \times CaO \times PbO$	8.678
TiO_2	66.704	$Na_2O \times MgO$	−24.909	$MgO \times CaO \times Al_2O_3$	−10.395
ZrO_2	28.056	$Na_2O \times CaO$	−33.107	$CaO \times Al_2O_3 \times Li_2O$	−5.000
$(ZrO_2)^2$	−4.531	$Na_2O \times SrO$	−11.108	$Al_2O_3 \times B_2O_3 \times PbO$	8.488
Ce_xO_y	7.529	$Na_2O \times BaO$	−11.019		

[†]The remainder includes traces of: Ag_2O, Bi_2O_3, Br, Co_xO_y, Cr_2O_3, Cs_2O, CuO, Ga_2O_3, Gd_2O_3, I, MoO_3, Nb_2O_5, PdO, Pr_xO_y, Rb_2O, Re_xO_y, Rh_xO_y, RuO_2, SeO_2, Sm_2O_3, SnO_2, TeO_2, Tl_2O_3, WO_3, Y_2O_3.

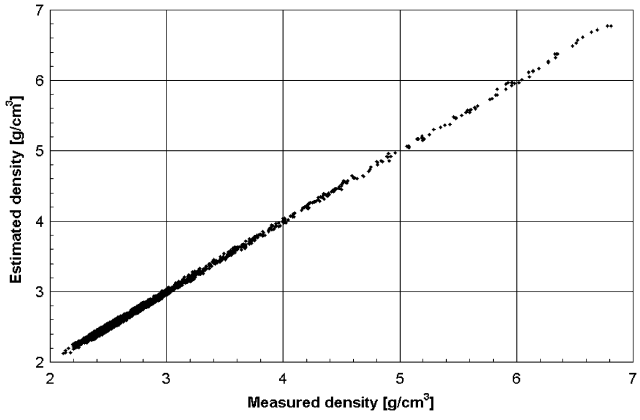

Fig. 2. Plot of 6719 measured versus estimated density values according to the model in Table II.

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