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Glass Transition Temperature- and Specific Volume-Composition Models for Tellurite Glasses

September 2017

BJ Riley JD Vienna



Prepared for the U.S. Department of Energy under Contract **DE-AC05-76RL01830**

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Pacific Northwest National Laboratory Richland, Washington 99352

Summary

This report provides models for predicting specific volume and glass transition temperature of tellurite glasses. Included are the partial specific coefficients for each model, the component validity ranges, and model fit parameters.

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1.0 Introduction

Tellurite glasses are technologically important materials with many unique properties including high refractive index, high polarizabilities, low melting temperatures ($T_{\rm M} \sim 600-800^{\circ}$ C), moderate glass transition temperatures ($T_{\rm g} \sim 300-400^{\circ}$ C), high densities ($\rho \sim 5-6$ g/cm³), and a very flexible network allowing for property tailoring through changes in composition (El-Mallawany, 2002; *Handbook of Glass Data - Physical Sciences Data 15, Part B: Single-Component and Binary Non-Silicate Oxide Glasses*, 1985). The purpose of this report is to provide tellurite glass composition-property models for $T_{\rm g}$ and specific volume (v) constructed using data from the literature. Using the component coefficients for v, the ρ can be calculated through the expression $v = V/m = \rho^{-1}$ and $\rho = v^{-1}$ where V and m are volume and mass, respectively.

2.0 Methods

The datasets were assembled using SciGlass software (v7.0) where glasses with \geq 52 mass% TeO₂ were used for the T_g model and glasses with \geq 50 mass% TeO₂ were used for the *v* model. Some data from recent work were also included (Riley et al., 2017). Once the datasets were assembled, JMP® software (v13.0.0) was used to create models for each. For each dataset, components that were present in < 5 glasses were moved to an "Others" category as well as components that, through model optimization, were shown not to be statistically significant. Also, outliers were removed if residuals fell outside of approximately $\pm 5\sigma$ (i.e., absolute value of studentized residuals >5).

2.1 Glass Transition Temperature Model

For calculating $T_{\rm g}$, the following expression was used

$$T_{\rm g} = \sum_{i=1}^{N} T_{\rm g,i} g_{\rm i} \tag{1}$$

where $T_{g,i}$ is the T_g coefficient of the *i*-th component in the dataset and g_i is the mass fraction of the *i*-th component ($\sum_{i=1}^{j} g_i = 1$).

2.2 Specific Volume Model

The specific volume (v) was calculated for each glass using the following expression

$$v = \sum_{i=1}^{N} v_i g_i \tag{2}$$

where v_i is the partial specific volume of the *i*-th component. Using the value of v for a given glass, the ρ can be calculated using the expression:

$$\rho = \frac{1}{\left(\sum_{i=1}^{N} v_i g_i\right)} = \frac{1}{\nu}$$
(3)

2.3 A Note on Significant Figures

Throughout this document, a number of model coefficients and other values are reported with a higher number of figures than are statistically significant. Ideally, the appropriate number of figures to report should be evaluated in detail. However, no such evaluation was performed. We therefore suggest using all reported figures in the model coefficients for consistency.

3.0 Results

3.1 Glass Transition Temperature Model

The summary of $T_{g,i}$ coefficients for the T_g model are presented in Table 1 as well as the minimum and maximum concentrations for each component as well as the number of glasses (*N*) containing each component in the dataset. The summary of the model parameters are presented in Table 2 along with a comparison plot for the predicted and measured T_g values in Figure 1. The components that most drastically reduce the T_g were (in order) F, Li₂O, Na₂O, K₂O, and AgI, all with $T_{g,i}$ values of < -100°C. The components that most drastically increase the T_g were (in order) MgO, As₂O₃, AlN, TiO₂, Y₂O₃, LaF₃, Al₂O₃, Yb₂O₃, CaO, and Fe₂O₃, all with $T_{g,i}$ values of < 800°C.

Term	$T_{g,i}$	Min	Max	N	Term	$T_{g,i}$	Min	Max	N
Ag ₂ O	21.67121	0.0000	0.4353	90	Na ₂ O	-244.04290	0.0000	0.2093	310
AgI	-105.00570	0.0000	0.1494	6	Nb ₂ O ₅	672.53679	0.0000	0.4165	242
Al_2O_3	896.51740	0.0000	0.1251	67	NiO	693.54863	0.0000	0.1199	5
AlN	1160.29270	0.0000	0.1462	9	P_2O_5	544.26603	0.0000	0.4454	125
As_2O_3	1234.91000	0.0000	0.0500	5	PbBr ₂	178.47706	0.0000	0.3582	5
B_2O_3	731.82353	0.0000	0.3437	139	PbCl ₂	208.51647	0.0000	0.4275	30
$BaCl_2$	207.81348	0.0000	0.2922	7	PbF ₂	169.78958	0.0000	0.4095	43
BaO	491.05328	0.0000	0.3488	209	PbO	272.25955	0.0000	0.4000	115
CaO	833.30717	0.0000	0.1500	12	Rb ₂ O	-25.32185	0.0000	0.2808	9
$CdCl_2$	190.77415	0.0000	0.2438	20	Sb_2O_3	391.47863	0.0000	0.2389	15
CdF_2	152.73182	0.0000	0.1518	19	Sm_2O_3	723.02931	0.0000	0.1960	7
CeO ₂	787.99950	0.0000	0.1500	17	SrO	792.55629	0.0000	0.1500	89
Cu ₂ O	216.12003	0.0000	0.4727	9	Ta_2O_5	636.81792	0.0000	0.2943	41
Er_2O_3	661.71152	0.0000	0.2000	117	TeO_2	304.39678	0.5114	1.0000	1939
F	-1189.65300	0.0000	0.0206	11	TiO_2	1041.49940	0.0000	0.1127	49
Fe_2O_3	808.56853	0.0000	0.2021	49	Tl_2O	14.594724	0.0000	0.4701	25
Ga_2O_3	583.61413	0.0000	0.2270	34	TlF	-60.23235	0.0000	0.3749	5
Gd_2O_3	703.86665	0.0000	0.1955	6	V_2O_5	255.73055	0.0000	0.4317	216
GeO ₂	442.38509	0.0000	0.4448	101	VN	214.24536	0.0000	0.2737	5
K_2O	-146.97170	0.0000	0.2476	178	WO ₃	482.29043	0.0000	0.4468	267
La_2O_3	702.24469	0.0000	0.2662	85	Y_2O_3	988.87616	0.0000	0.0960	45
LaF ₃	936.08895	0.0000	0.1210	7	Yb_2O_3	851.90815	0.0000	0.2153	70
Li ₂ O	-488.87860	0.0000	0.1311	224	$ZnCl_2$	233.47489	0.0000	0.3827	43
LiBr	95.49429	0.0000	0.3524	14	ZnF_2	245.30147	0.0000	0.4000	36
LiCl	44.07846	0.0000	0.3733	16	Others	376.54881	0.0000	0.4713	1293
MgO	1287.08520	0.0000	0.2000	43					

Table 1. Summary of $T_{g,i}$ (°C) coefficients as well as the minimum and maximum concentration (mass fraction) for each term as well as the number of glasses (*N*) containing each component.

Parameter	Value			
R^2	0.840634			
$R^2_{adjusted}$	0.836414			
$R^2_{\rm press}$	0.828806			
RMSE	22.83494			
Press RMSE	23.3599096			
Mean of Response	319.7292			
Observations	1939			

Figure 1. Comparison of predicted and measured $T_{\rm g}$.

Table 2. Summary of results for $T_{\rm g}$ model.

3.2 Specific Volume Model

The summary of v_i coefficients for the v model are presented in Table 3 as well as the minimum and maximum concentrations for each component as well as the number of glasses (*N*) containing each component in the dataset. The summary of the model parameters are presented in Table 4 along with a comparison plot for the predicted and measured v values in Figure 2.

			e nam			uning out		lient.
vi	Min	Max	N	Term	vi	Min	Max	N
0.117748	0.0000	0.4210	85	MoO_3	0.246778	0.0000	0.4740	95
0.418729	0.0000	0.1140	61	Na ₂ O	0.484312	0.0000	0.1910	230
0.186019	0.0000	0.1010	36	NaCl	0.473941	0.0000	0.0940	5
0.404489	0.0000	0.3290	101	Nb_2O_5	0.214030	0.0000	0.4660	169
0.244366	0.0000	0.2920	25	NdCl ₃	0.098089	0.0000	0.3400	5
0.176955	0.0000	0.2110	13	P_2O_5	0.386111	0.0000	0.4450	104
0.185469	0.0000	0.4400	214	$PbBr_2$	0.165590	0.0000	0.4960	19
0.470391	0.0000	0.0440	5	PbCl ₂	0.174642	0.0000	0.4670	30
0.101682	0.0000	0.3430	108	PbF_2	0.119703	0.0000	0.4090	26
0.452587	0.0000	0.2000	22	PbO	0.107432	0.0000	0.4820	149
0.216734	0.0000	0.1520	15	$PbSO_4$	0.176297	0.0000	0.3220	13
0.137809	0.0000	0.3000	32	Rb ₂ O	0.262608	0.0000	0.2810	33
0.177973	0.0000	0.1500	23	Sb_2O_3	0.206448	0.0000	0.1690	12
0.221289	0.0000	0.2230	18	SiO_2	0.646120	0.0000	0.0870	7
0.194313	0.0000	0.4710	46	Sm_2O_3	0.156503	0.0000	0.1960	20
0.098903	0.0000	0.2000	92	SrO	0.215640	0.0000	0.1500	17
0.270236	0.0000	0.2080	12	Ta ₂ O ₅	0.152535	0.0000	0.3500	49
0.189021	0.0000	0.2810	49	TeO ₂	0.178861	0.5000	1.0000	2208
0.232000	0.0000	0.2450	14	ThO_2	0.131662	0.0000	0.2540	5
0.291046	0.0000	0.4230	37	Tl_2O	0.116919	0.0000	0.4700	41
0.075625	0.0000	0.2200	5	WO ₃	0.139667	0.0000	0.4470	420
0.478627	0.0000	0.1590	164	Y_2O_3	0.161824	0.0000	0.0730	9
0.158902	0.0000	0.2250	90	Yb ₂ O ₃	0.151827	0.0000	0.2150	55
0.685571	0.0000	0.1260	211	$ZnCl_2$	0.280899	0.0000	0.3830	66
0.491328	0.0000	0.2020	5	ZnF_2	0.189108	0.0000	0.4000	41
0.493698	0.0000	0.2130	27	ZnO	0.217218	0.0000	0.4330	264
0.475712	0.0000	0.2000	48	Others	0.308340	0.0000	0.5000	532
	vi 0.117748 0.418729 0.186019 0.404489 0.244366 0.176955 0.185469 0.470391 0.101682 0.452587 0.216734 0.137809 0.177973 0.221289 0.194313 0.098903 0.270236 0.189021 0.232000 0.291046 0.075625 0.478627 0.158902 0.685571 0.493698 0.475712	v_i Min 0.117748 0.0000 0.418729 0.0000 0.186019 0.0000 0.418729 0.0000 0.418729 0.0000 0.418729 0.0000 0.404489 0.0000 0.244366 0.0000 0.176955 0.0000 0.185469 0.0000 0.470391 0.0000 0.470391 0.0000 0.452587 0.0000 0.137809 0.0000 0.177973 0.0000 0.177973 0.0000 0.194313 0.0000 0.270236 0.0000 0.232000 0.0000 0.232000 0.0000 0.291046 0.0000 0.478627 0.0000 0.478627 0.0000 0.478627 0.0000 0.493698 0.0000 0.493698 0.0000	v_i MinMax0.1177480.00000.42100.4187290.00000.11400.1860190.00000.10100.4044890.00000.32900.2443660.00000.29200.1769550.00000.21100.1854690.00000.44000.4703910.00000.04400.1016820.00000.34300.4525870.00000.20000.2167340.00000.15200.1378090.00000.30000.1779730.00000.22300.1943130.00000.20000.2702360.00000.20000.2702360.00000.28100.2320000.00000.24500.2910460.00000.42300.0756250.00000.22500.4786270.00000.15900.1589020.00000.22500.6855710.00000.20200.4936980.00000.21300.4757120.00000.2130	v_i MinMaxN 0.117748 0.0000 0.4210 85 0.418729 0.0000 0.1140 61 0.186019 0.0000 0.1140 61 0.186019 0.0000 0.3290 101 0.244366 0.0000 0.2920 25 0.176955 0.0000 0.2110 13 0.185469 0.0000 0.4400 214 0.470391 0.0000 0.4400 214 0.470391 0.0000 0.3430 108 0.452587 0.0000 0.2000 22 0.216734 0.0000 0.1520 15 0.137809 0.0000 0.3000 32 0.177973 0.0000 0.2230 18 0.194313 0.0000 0.2000 92 0.270236 0.0000 0.2000 92 0.270236 0.0000 0.2810 49 0.232000 0.0000 0.2450 14 0.291046 0.0000 0.2250 5 0.478627 0.0000 0.2250 90 0.685571 0.0000 0.2200 5 0.493698 0.0000 0.2130 27 0.475712 0.0000 0.2000 48	v_i MinMaxNTerm 0.117748 0.0000 0.4210 85 MoO_3 0.418729 0.0000 0.1140 61 Na_2O 0.186019 0.0000 0.1010 36 $NaCl$ 0.404489 0.0000 0.3290 101 Nb_2O_5 0.244366 0.0000 0.2920 25 $NdCl_3$ 0.176955 0.0000 0.2110 13 P_2O_5 0.185469 0.0000 0.4400 214 $PbBr_2$ 0.470391 0.0000 0.4400 214 PbF_2 0.470391 0.0000 0.3430 108 PbF_2 0.452587 0.0000 0.2000 22 PbO 0.216734 0.0000 0.1520 15 $PbSO_4$ 0.137809 0.0000 0.1500 23 Sb_2O_3 0.221289 0.0000 0.2230 18 SiO_2 0.194313 0.0000 0.2000 92 SrO 0.270236 0.0000 0.2810 49 TeO_2 0.232000 0.0000 0.2200 5 WO_3 0.478627 0.0000 0.1590 164 Y_2O_3 0.478627 0.0000 0.2250 90 Yb_2O_3 0.685571 0.0000 0.2200 5 ZnF_2 0.493698 0.0000 0.2130 27 ZnO 0.475712 0.0000 0.2000 48<	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	$\begin{array}{c c c c c c c c c c c c c c c c c c c $

Table 3. Summary of v_i (cm³/g) coefficients as well as the minimum and maximum concentration (mass fraction) for each term as well as the number of glasses (*N*) containing each component.

Parameter	Value			
R^2	0.904746			
$R^{2}_{adjusted}$	0.902402			
$R^2_{\rm press}$	0.898650			
RMSE	0.007734			
Press RMSE	0.00788079			
Mean of Response	0.191562			
Observations	2208			

0.32 0.3 0.28 0.26 0.24 0.22 0.2 0.22 0.2 0.18 0.16 0.14 0.12 0.16 0.2 0.2 0.2 0.26 0.28 0.3 0.32 cm3/g Predicted RMSE=0.0077 RSq=0.90 PValue <.0001

Figure 2. Comparison of predicted and measured *v*.

Table 4. Summary of results for *v* model.

4.0 References

El-Mallawany, R. A. H. 2002. *Tellurite Glasses Handbook: Physical Properties and Data*, CRC Press LLC, Boca Raton, FL.

Handbook of Glass Data - Physical Sciences Data 15, Part B: Single-Component and Binary Non-Silicate Oxide Glasses. OV Mazurin, MV Streltsina and TP Shvaiko-Shvaikovskaya. 1985. Vol. 15B. Elsevier Science Publishing Company, Inc., New York, NY.

Riley, B. J., J. O. Kroll, J. A. Peterson, D. A. Pierce, W. L. Ebert, B. D. Williams, M. M. V. Snyder, S. M. Frank, J. L. George, and K. Kruska. 2017. "Assessment of lead tellurite glass for immobilizing electrochemical salt wastes from used nuclear fuel reprocessing." *J. Nucl. Mater.* (in press) https://doi.org/10.1016/j.jnucmat.2017.08.037.





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