PNNL-26760

Pacific Northwest NATIONAL LABORATORY

Proudly Operated by Battelle Since 1965

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

September 2017

BJ Riley
JD Vienna

U.S. DEPARTMENT OF ENERGY

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

PACIFIC NORTHWEST NATIONAL LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC05-76RL01830

Printed in the United States of America

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information,
P.O. Box 62, Oak Ridge, TN 37831-0062;
ph: (865) 576-8401
fax: (865) 576-5728
email: reports@adonis.osti.gov

Available to the public from the National Technical Information Service
5301 Shawnee Rd., Alexandria, VA 22312
ph: (800) 553-NTIS (6847)
email: orders@ntis.gov <http://www.ntis.gov/about/form.aspx>
Online ordering: http://www.ntis.gov

This document was printed on recycled paper.

(8/2010)

PNNL-26760

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

BJ Riley
JD Vienna

September 2017

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

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.

Acknowledgments

Pacific Northwest National Laboratory (PNNL) is operated by Battelle Memorial Institute for the DOE under contract DE-AC05-76RL01830. This work was supported by the DOE Office of Nuclear Energy (DOE-NE).

Contents

Figures

Tables

1.0 Introduction

Tellurite glasses are technologically important materials with many unique properties including high refractive index, high polarizabilities, low melting temperatures (T_M ~ 600–800°C), moderate glass transition temperatures (T_g ~ 300–400°C), high densities (ρ ~ 5–6 g/cm^3), 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_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 ≥ 52 mass% TeO_2 were used for the T_g model and glasses with ≥ 50 mass% TeO_2 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 ±5σ (i.e., absolute value of studentized residuals >5).

2.1 Glass Transition Temperature Model

For calculating T_g, the following expression was used

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

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 ($\sum_{i=1} g_i = 1$).

2.2 Specific Volume Model

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

$$v = \sum_{i=1}^{N} v_\mathrm{i} g_\mathrm{i} \tag{2}$$

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:

$$\rho = {}^{1}\!\big/\!\left(\sum_{i=1}^{N} v_\mathrm{i} g_\mathrm{i}\right) = {}^{1}\!/\!_{v} \tag{3}$$

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, Li_2O, Na_2O, K_2O, and AgI, all with $T_{g,i}$ values of < -100°C. The components that most drastically increase the T_g were (in order) MgO, As_2O_3, AlN, TiO_2, Y_2O_3, LaF_3, Al_2O_3, Yb_2O_3, CaO, and Fe_2O_3, all with $T_{g,i}$ values of > 800°C.

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.

Term	$T_{g,i}$	Min	Max	N	Term	$T_{g,i}$	Min	Max	N
Ag_2O	21.67121	0.0000	0.4353	90	Na_2O	-244.04290	0.0000	0.2093	310
AgI	-105.00570	0.0000	0.1494	6	Nb_2O_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_2O	-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_2O	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_2O	-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 2. Summary of results for T_g model.

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

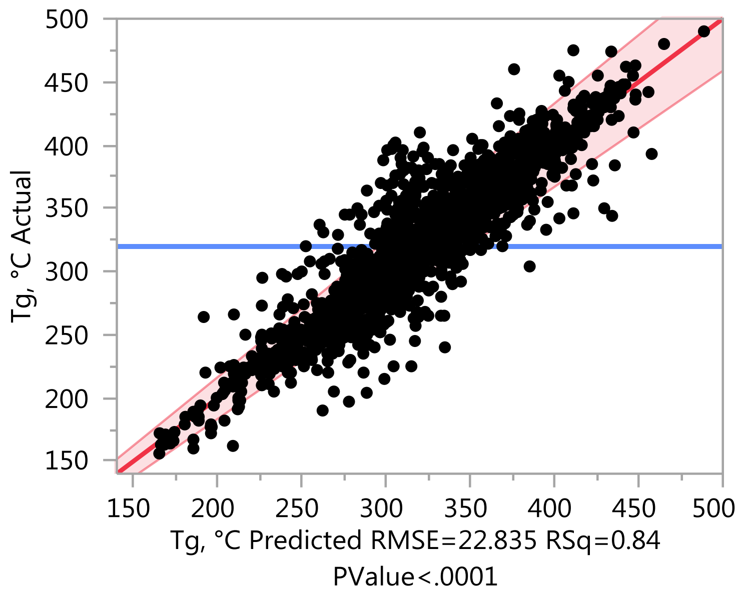

Figure 1. Comparison of predicted and measured T_g.

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.

Table 3. Summary of v_i (cm^3/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.

Term	v_i	Min	Max	N
Ag_2O	0.117748	0.0000	0.4210	85
Al_2O_3	0.418729	0.0000	0.1140	61
As_2O_3	0.186019	0.0000	0.1010	36
B_2O_3	0.404489	0.0000	0.3290	101
$BaCl_2$	0.244366	0.0000	0.2920	25
BaF_2	0.176955	0.0000	0.2110	13
BaO	0.185469	0.0000	0.4400	214
BeO	0.470391	0.0000	0.0440	5
Bi_2O_3	0.101682	0.0000	0.3430	108
CaO	0.452587	0.0000	0.2000	22
CdF_2	0.216734	0.0000	0.1520	15
CdO	0.137809	0.0000	0.3000	32
CeO_2	0.177973	0.0000	0.1500	23
Cs_2O	0.221289	0.0000	0.2230	18
CuO	0.194313	0.0000	0.4710	46
Er_2O_3	0.098903	0.0000	0.2000	92
Fe_2O_3	0.270236	0.0000	0.2080	12
Ga_2O_3	0.189021	0.0000	0.2810	49
Gd_2O_3	0.232000	0.0000	0.2450	14
GeO_2	0.291046	0.0000	0.4230	37
In_2O_3	0.075625	0.0000	0.2200	5
K_2O	0.478627	0.0000	0.1590	164
La_2O_3	0.158902	0.0000	0.2250	90
Li_2O	0.685571	0.0000	0.1260	211
LiBr	0.491328	0.0000	0.2020	5
LiCl	0.493698	0.0000	0.2130	27
MgO	0.475712	0.0000	0.2000	48

Term	v_i	Min	Max	N
MoO_3	0.246778	0.0000	0.4740	95
Na_2O	0.484312	0.0000	0.1910	230
NaCl	0.473941	0.0000	0.0940	5
Nb_2O_5	0.214030	0.0000	0.4660	169
$NdCl_3$	0.098089	0.0000	0.3400	5
P_2O_5	0.386111	0.0000	0.4450	104
$PbBr_2$	0.165590	0.0000	0.4960	19
$PbCl_2$	0.174642	0.0000	0.4670	30
PbF_2	0.119703	0.0000	0.4090	26
PbO	0.107432	0.0000	0.4820	149
$PbSO_4$	0.176297	0.0000	0.3220	13
Rb_2O	0.262608	0.0000	0.2810	33
Sb_2O_3	0.206448	0.0000	0.1690	12
SiO_2	0.646120	0.0000	0.0870	7
Sm_2O_3	0.156503	0.0000	0.1960	20
SrO	0.215640	0.0000	0.1500	17
Ta_2O_5	0.152535	0.0000	0.3500	49
TeO_2	0.178861	0.5000	1.0000	2208
ThO_2	0.131662	0.0000	0.2540	5
Tl_2O	0.116919	0.0000	0.4700	41
WO_3	0.139667	0.0000	0.4470	420
Y_2O_3	0.161824	0.0000	0.0730	9
Yb_2O_3	0.151827	0.0000	0.2150	55
$ZnCl_2$	0.280899	0.0000	0.3830	66
ZnF_2	0.189108	0.0000	0.4000	41
ZnO	0.217218	0.0000	0.4330	264
Others	0.308340	0.0000	0.5000	532

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

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

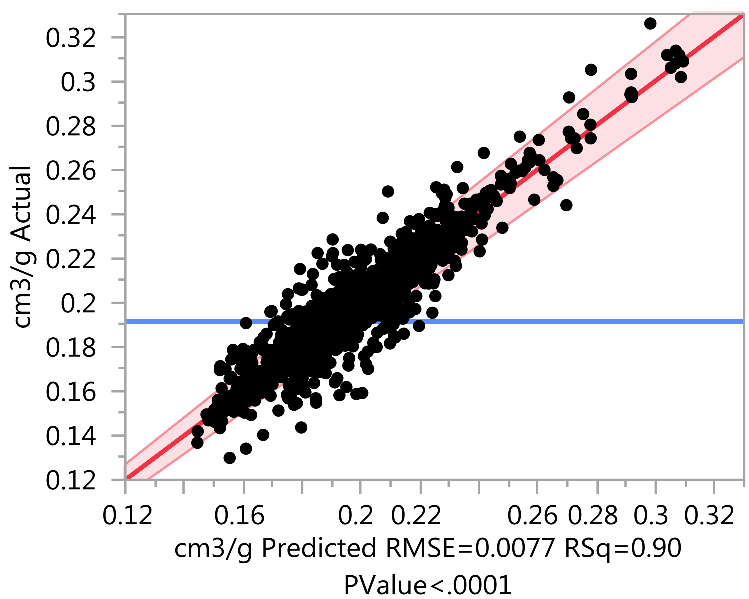

Figure 2. Comparison of predicted and measured *v*.

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.

Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by **Battelle** *Since 1965*

U.S. DEPARTMENT OF
ENERGY

902 Battelle Boulevard
P.O. Box 999
Richland, WA 99352
1-888-375-PNNL (7665)

www.pnnl.gov