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Accurate Determination of Nonlinear Refractive Index for Some Binary Glass Systems

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Abstract: Problem statement: This study investigates the relation between the nonlinear optical susceptibility ($\chi^{(3)}$) and the ratio (α_1/α_{o-2}), then the nonlinear index determination for some binary glass systems. Recently suitable relationship between the oxidation polarizability and the nonlinear optical properties was established for simple oxides glasses. So in this study more attention and investigation is paid to the trend of that relation quantitatively as possible. **Approach**: The idea of the present research is how to obtain the non linear refractive index as function of ratio (α_1/α_{o-2}), as we know the cation polarizability α_l can be determined easy and α_{o-2} also can be obtained according the energy gap or the real refractive index. Consequently a proposed scale has been established between the nonlinear optical susceptibility ($\chi^{(3)}$) and the ratio (α_1/α_{o-2}) for some binary glass systems. **Results**: A comparison between our obtained results and the empirical method of Boling shows that there is a good agreement between them. **Conclusion:** The advantages of the suggested method fix on the expectation of the value ($\chi^{(3)}$) without the complex and sensitivity experimental set-up.

Key words: Nonlinear refractive index, cation polarizability, oxide ion polarizability, nonlinear optical susceptibility, accurate determination

INTRODUCTION

Near resonance, the imaginary part of the susceptibility ($\chi^{(3)}$) becomes important. One may argue that a part of the light beam will be absorbed near the resonance (Varshneya, 1993). As a result that the redistribution of the electron energy levels depending upon the intensity of the beam according to the following equation of refraction (Varshneya, 1993):

$$n(\omega) = n_0 + \gamma I = n_0 + n_2 \langle E^2 \rangle \quad (1)$$

Where:
n_0 = The real index of refraction
γ = The non-linear coefficient
n_2 = The non-linear refractive index

Also E is the rms of the electric field of the incident light beam.

The absorption frequency would then shift toward the incident beam, since the frequency shifts.

Boling *et al*. (2003) had estimated γ or n_2 by suggesting an experimental formula as an approximation method as follows:

$$\gamma = \frac{k'(n_0 - 1)(n_0 + 1)}{n_0 \upsilon_0 [1.52 + \frac{(n_0 + 1)(n_0^2 + 2)}{6n_0})} \quad (2)$$

where, k' is a constant nearly $2.8*10^{-18}$ m^2/W, ν_d is the abbi number.

On the basis of a physical nature between the cation and the oxide ion polarizability, the nonlinear optical susceptibility has been obtained as a function of the ratio $\frac{\alpha_1}{\alpha_{o^{-2}}}$. There is no systematic data about the correlation between the nonlinear optical properties and $\frac{\alpha_1}{\alpha_{o^{-2}}}$.

MATERIALS AND METHODS

The theoretical idea of this research study is that the nonlinear optical susceptibility $\chi^{(3)}$ increases with presence of the no bridging oxygen atoms in the system of glass and decreases as the system tends to the metallization state.

Theory: Optical devices, which are currently being investigated for use in computational systems and telecommunications, require materials that are characterized with large optical nonlinearity (Zhao *et al*., 2007). These applications require that the optical properties of the active material change in the response to an applied optical field, the magnitude of this effect being characterized by a single term called the third order susceptibility tensor, $\chi^{(3)}$ the latter can be thought of as a coefficient in a power series expansion of the relationship between the applied electric field E and the polarization p (Varshneya, 1993). However the importance of the determination of the nonlinear coefficient is now necessary.

One of the most important properties of materials, which are closely related to their applicability in the field of optics and electronics, is the electronic polarizability of ions (Chimalawong *et al*., 2010).

Optical nonlinearity of a material is caused as it exposes to intense light beams, hence the nonlinear response of a material is controlled by the electronic polarizability (Jackson, 2003).

Accordingly, materials of high nonlinear coefficient have to be discovered on the basis of correlation of nonlinear response with some electronic properties which can be explained easily (Rekha and Ramalingam, 1970).

In this research study, the relationship between the nonlinear optical susceptibility index $\chi^{(3)}$ and the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ has been investigated for some binary glass systems; hence the nonlinear refractive index n_2 can be estimated according to the following Equation (Varshneya, 1993):

$$n_2 = 12\chi / n_0 [\chi^{(3)}] \text{ in (esu)} \quad (3)$$

RESULTS

Tables: 1-10 illustrate the behavior of the optical susceptibility with the ratio $r = \frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ in the cases under study, in tabular shape for many binary systems under study.

Table 1: Illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3)}$ Bi_2O_3 –B_2O_3 (Dimitrov and Komatsu, 1999)

$r = \frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.435	0.511	0.609	0.689	0.787
$\chi^{(3)}*10^{-14}$ in (esu)	31.9	32.5	51.5	63.4	81.3

Note: The ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ increases with increasing Bi_2O_3 conten

Table 2: Illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3)}$ in case of Sb_2O_3 –B_2O_3 (Dimitrov and Komatsu, 1999)

$r = \frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.177	0.253	0.364	0.452	0.573	0.573	0.632	0.647	0.703
$\chi^{(3)}*10^{-14}$ in (esu)	4.96	10.6	16.4	30.2	35.6	44	51.5	65.4	76.2

Note: The ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ increases with increasing Sb_2O_3 content.

Table 3: Illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3)}$ in case of La_2O_3-B_2O_3 (Dimitrov and Komatsu, 1999)

$r = \frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.3045	0.3474	0.3915
$\chi^{(3)}*10^{-14}$ in (esu)	31.9	32.5	51.5

Note: The ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ increases with increasing La_2O_3 content

Table 4: Illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3)}$ in case of Li_2O-TeO_2 (Katagiri *et al*., 1994)

$r = \frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.511	0.546	0.571	0.606
$\chi^{(3)}*10^{-14}$ in (esu)	33.00	42.000	46.000	52.000

Note: the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ increases with increasing Li_2O content

Table 5: illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3)}$ in case of PbO-SiO_2 (Katagiri *et al*., 1994)

Ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.77	0.969	1.136	1.325	1.380
$\chi^{(3)}*10^{-14}$ in (esu)	7.10	12.700	18.100	23.000	31.100

Note: The ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ increases with increasing PbO content

Table 6: Illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3)}$ in case of PbO-B_2O_3 (Kim *et al*., 1993)

Ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.81	1.08	1.32	1.48
Nonlinear optical susceptibility $\chi^{(3)}$	5.90	9.70	12.30	21.40

Note: The ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ increases with increasing PbO content

Table 7: Illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3)}$ in case of WO_3-TeO_2 (Kim *et al*., 1993)

Ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.619	0.556	0.486
Nonlinear optical susceptibility $\chi^{(3)}$	142.000	148.000	159.000

Note: The ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ increases with decreasing WO_3 content

Table 8: Illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3)}$ in case of Ag_2O-TeO_2 (Katagiri *et al*., 1994)

$R = \frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.801	0.843	0.88
$\chi^{(3)}*10^{-14}$ in (esu)	120.000	119.000	110.00

Note: The ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ increases with decreasing Ag_2O content

Table 9: Illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3}$ in case of MO_3-TeO_2 (Kim *et al*., 1993)

Ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.649	0.567	0.514
$\chi^{(3)}*10^{-14}$ in (esu)	67.000	68.000	69.000

Note: The ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ decreases with increasing MO_3 content

Table 10: Illustrates the calculated values of the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ corresponding $\chi^{(3)}$ in case of PbO-TiO_2 (Sugimoto *et al*., 1993)

Ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$	0.435	0.536	0.300
$\chi^{(3)}*10^{-14}$ in (esu)	57.000	110.000	140.000

Note: The ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ increases with decreasing PbO content systems

DISCUSSION

It is obvious with respect to Fig. 1-6 that the nonlinear optical susceptibility $\chi^{(3)}$ increases with increasing the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ for almost binary oxide glass systems, but in some oxide glass systems the $\chi^{(3)}$ decreases with increasing $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$. To explain the above behavior, Dimitrov and Saka (1996) have established that the nonlinear refractive increases with increasing linear refractive index and decreasing energy gap of the oxides. Also the heavy metal content such as Bi_2O_3 contributes to the polarizability of the glass system, consequently the nonlinear coefficient increases due to the high polarizability of the cations of the all molecule and easy distortion of the electron density, when applying strong electromagnetic field. At the same time there is a contribution from the oxide ion because of the high electron donor ability of the oxide ion. So the valence band is composed of O_2p orbitals, this means higher optical transition probability and increase in $\chi^{(3)}$.

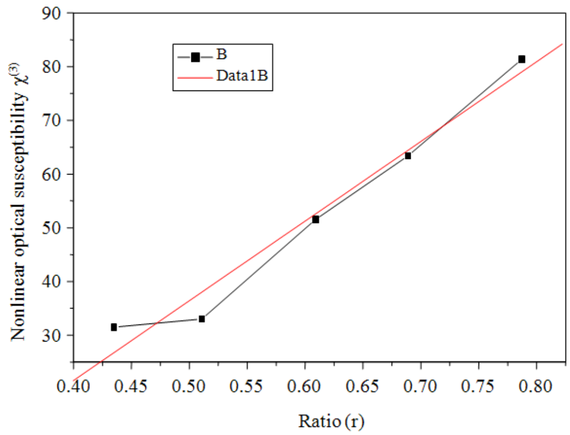

Fig. 1: Shows the relationship between the ratio r and the optical susceptibility $\chi^{(3)}$ in case of Bi_2O_3-B_2O_3 system

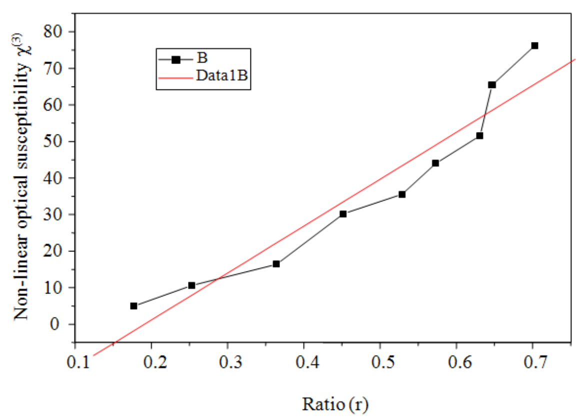

Fig. 2: Shows the relationship between the ratio r and $\chi^{(3)}$ in case of sb_2O_3-B_2O_3 system

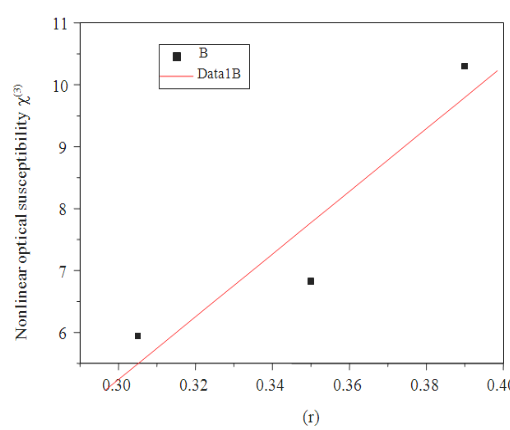

Fig. 3: Shows the relationship between the ratio r and the $\chi^{(3)}$ in case of La_2O_3-B_2O_3

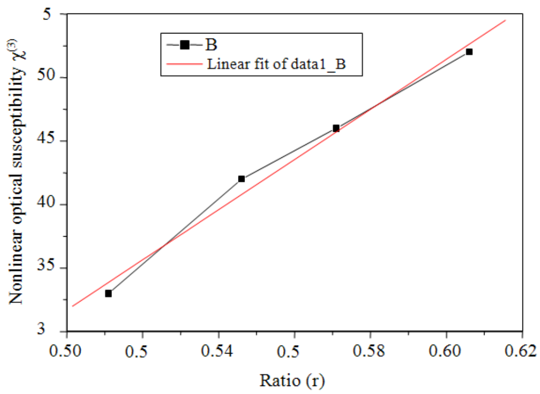

Fig. 4: Shows the relationship between the ratio in case of Li_2O_3-TeO_3 system

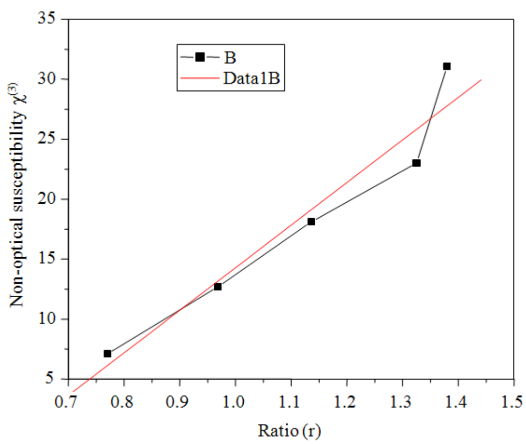

Fig. 5: Shows the relationship between the ratio r and the optical susceptibility $\chi^{(3)}$ in case of Li_2O_3-TeO_3 system

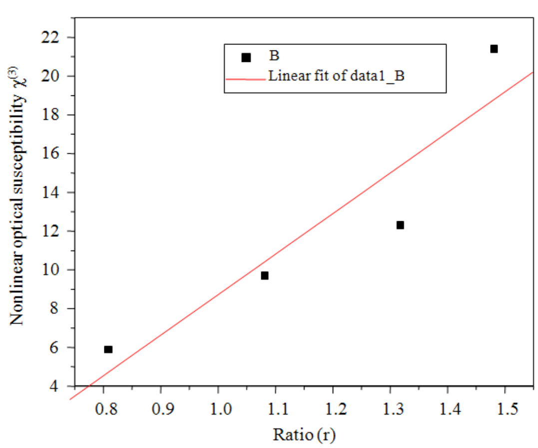

Fig. 6: Shows the relationship between the ratio in case of PbO-TiO_2 system

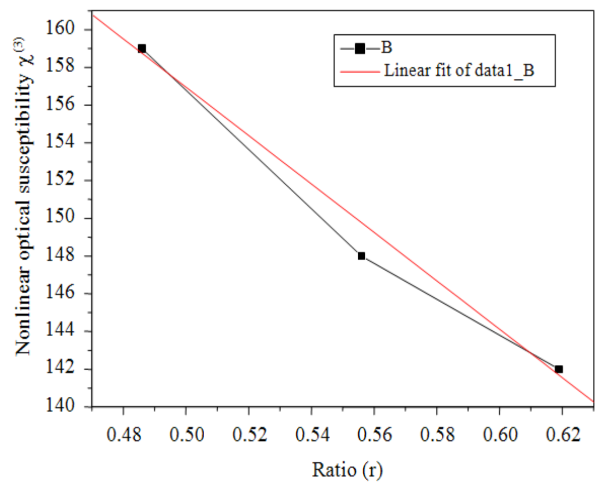

Fig. 7: Shows the relationship between the ratio r and the optical susceptibility $\chi^{(3)}$ in case of Wo_3-TeO_2 system

According to Dimitrov *et al*. (1996) in Lorentz-Lorentz equation $R_m/V_m \approx 1$ corresponds to the metallization of covalent solid materials, where R_m and V_m are the molar refraction and volume respectively.

Dimitrov *et al*. (1996) suggestion was a good correlation between the energy gap of the oxides and their molar refraction, also (Dimitrov and Sakka, 1996) have been reported a data of $M(E_g)$ and non-linear optical susceptibility $\chi^{(3)}$. It was found that the oxides with large non-linear refractive posses a metallization factor (criterion) smaller than the oxide glasses contain alkaline and alkaline-earth oxides such as B_2O_3 and SiO_2, the two later oxides have a small non-linear optical susceptibility $\chi^{(3)}$. With respect to Fig. 7-9, although the optical susceptibility $\chi^{(3)}$ has a large value i.e., high non-linear refractive index, but as the Ag_2O, Mo_3, Wo_3 oxides content increases in the glass systems, $\chi^{(3)}$ decreases with increasing the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ in this case. It can be explained as the relationship between non-linear optical susceptibility $\chi^{(3)}$ and the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ is inversely proportional when the metallization process arises in glass system. Also with respect to Fig. 10 which illustrates the relationship between the ratio $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$ and $\chi^{(3)}$ for PbO-TiO_2 system although PbO in the previous cases is nearly directly proportional, but in Fig. 10 is inversely proportional with increasing PbO

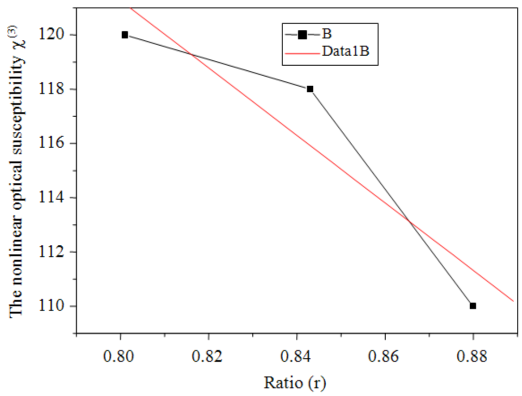

Fig. 8: Shows the relationship between the ratio r and the optical susceptibility $\chi^{(3)}$ in case of Ag_2O-TeO_2 system

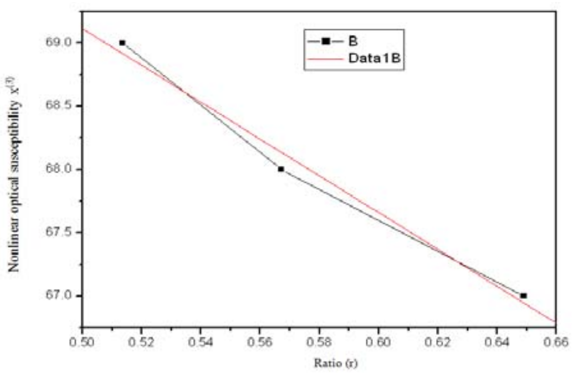

Fig. 9: Shows the relationship between the ratio r and the optical susceptibility $\chi^{(3)}$ in case of Mo_3-TeO_2 system

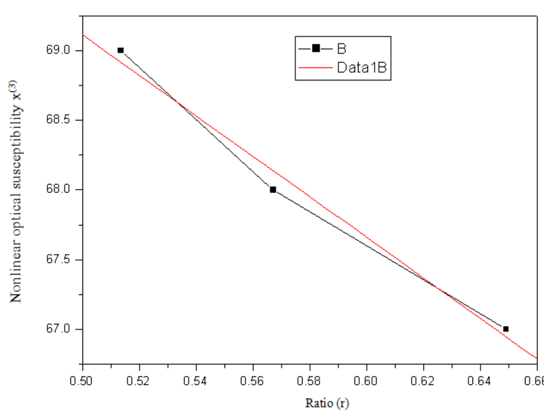

Fig. 10: Shows the relationship between the ratio r and the optical suscpetebility $\chi^{(3)}$ in case of PbO-TiO_2

content, this because of the metallization criterion decreasing i.e., the glass tends to be metal.

Finally, it is obvious that a general formula can be suggested in order to correlate the non-linear optical susceptibility $\chi^{(3)}$ and the ratio $\frac{\sum \alpha_i}{\alpha_{o^{-2}}}$ as follows:

$$\chi^{(3)} = A\,\frac{\sum \alpha_i}{\alpha_{o^{-2}}} + B \qquad (4)$$

where, A and B are constants based on the type of glass system, they can be determined from the slope of line represents the relationship between $\chi^{(3)}$ and $\frac{\sum \alpha_i}{\alpha_{o^{-2}}}$.

Table 11 summarizes the values of the constants A, B and the correlation factor R for different above binary glass systems respectively. The values of the constants of the proposed formula which correlates $\chi^{(3)}$ with $\frac{\sum \alpha_i}{\alpha_{o^{-2}}}$.

As an application of the above suggested method in case of Bi_2O_3–B_2O_3 glass system, the nonlinear refractive index calculated by Boling formula is of order $2.5*10^{-19}$ m^2/W. At the same time $n_2= 2.71*10^{-19}$ m^2/W according to the suggested method (where $n_0 = 2$ in this case).The difference percentage nearly ± 7%, i think that error is suitable for such measurements.

CONCLUSION

A quantitative relation between the nonlinear optical susceptibility $\chi^{(3)}$ and the ratio $\frac{\sum \alpha_i}{\alpha_{o^{-2}}}$ is to be expected. This is because the oxide ion represents the ability to donate negative charge when combined with a suitable cation.

Table 11 can help us and the interesting peoples in the field of studying non-linear optical properties for obtaining the corresponding values of $\chi^{(3)}$ for each individual content of binary glasses; hence n_2 can be determined easily.

The above study is a trial of treatment the relationship between the nonlinear optical susceptibility and the ratio of the value of cation to the oxide ion polarizability for binary glass systems with a simulating method.

Table 11: Summarizes the values of the constants A, B and the correlation factor R for different above binary glass systems respectively. The values of the constants of the proposed formula which correlates $\chi^{(3)}$ with $\frac{\sum \alpha_1}{\alpha_{o^{-2}}}$.

No of sys	1	2	3	4	5	6	7	8	9	10
A	148.3300	128.3700	50.6400	197.4090	35.4990	20.9270	-128.3600	-124.930	-14.560	-234.820
B	-37.7800	-24.5400	-9.9510	-67.0030	-21.2170	-12.2090	220.7400	221.100	76.390	215.500
R	0.9842	0.9612	0.9339	0.9929	0.9685	0.9281	0.9905	0.932	0.993	0.924

R: is the correlation factor of fitting of data for all above glass systems

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