Structural and radiation shielding simulation of $B_2O_3$–$SiO_2$–$LiF$–$ZnO$–$TiO_2$ glasses

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ABSTRACT

Penta-glasses with a $59B_2O_3$–$29SiO_2$–$2LiF$–$(10 - x)$ $ZnO$–$x$ $TiO_2$ composition with the melt-quench techniques were prepared. X-ray diffraction was used to examine the nature of fabricated glasses. The changes in the structure of fabricated glasses were studied by Fourier-transform infrared spectroscopy spectra. The molar volume of fabricated glasses reduces as the density increases. The mechanical characteristics of these glasses were evaluated. Besides, for the studied glasses, Phy-X/PSD and XCOM program were used to investigate the radiation shielding efficiency. These glasses were found to have an abnormal attenuation, structural, and density relationship. Glasses’ mass attenuation coefficient (MAC), linear attenuation coefficient (LAC), half-value layer (HVL), tenth value layer (TVL), and effective atomic number (Zeff) were designed to simulate gamma photon energies ranging from 0.015 to 15 meV. MAC values determined with Phy-X/PSD and XCOM were compared and were observed in good agreement with the other.

1 Introduction

Future trends are associated with the development of amorphous materials such as glasses for energy or scientific purposes. This means enhancing the characteristics of these glasses for mechanical and radiation shielding. Because of their unique properties, borosilicate glasses are regarded as a material treasure. High-performance borosilicate glasses have been used in a variety of requests. Because of their many applications in optoelectronics and radiation shielding, borosilicate glasses are excellent glasses [1–8]. Doping these glasses with transition metal oxides enhances their characteristics and configuration. Because of significance of transition metal-containing glasses in their implementations, these glasses are classified as commercial and scientific. Furthermore, the incorporation of $TiO_2$ into these glasses

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enhanced their mechanical, radiation, and physical characteristics. TiO$_2$-containing glasses indicated the structural units TiO$_4$, TiO$_5$, and TiO$_6$, and their characteristics are depending on the quantity of coordinates [1–8].

TiO$_2$-doped glasses to generate glasses possess unique properties that are relevant to their implementations in radiation characteristics [9–15]. To generate Li$^+$ and Na$^+$, NaF and LiF are incorporated into the glass matrix. As a result, these glasses are outstanding metal ion reagents. For a long time, because of their distinct physical properties, glasses-containing halide ions were investigated [16–18]. It is the best article for manufacturing and investigating the structural, mechanical, and shielding radiation [19] properties of 59B$_2$O$_3$–29SiO$_2$–2LiF–(10–x) ZnO–xTiO$_2$, where x (0 ≤ x ≥ 10). The novelty of this work is that the ordinary method of melt quenching has been applied to manufacture of these glasses. The fabricated glasses have being investigated by FT-IR to analyze the structure of each sample. Fabrication glasses’ mechanical properties were examined. Mass attenuation coefficient of fabricated glasses is evaluated with Phy-X/PSD and XCOM [19, 20]. We are concentrating on the investigation, and characteristics features of 29SiO$_2$–2LiF–59B$_2$O$_3$–(10-x) ZnO–x TiO$_2$, glasses, which have received little attention.

2 Methodology

The fabricated glasses in Table 1 were manufactured using the melt-quench procedure with the chemical composition 29SiO$_2$–2LiF–59B$_2$O$_3$–(10-x) ZnO–x TiO$_2$, where x : (0 ≤ x ≥ 10) mol %. SiO$_2$, LiF, H$_3$BO$_4$, ZnO, and TiO$_2$ are the start components used to generate these samples. Sigma-Aldrich Company provided all the start components. To begin, the components were heated to 650 °C for 1 h to remove H$_2$O and other impurities. The temperature was increased to 1200 °C for 45 min. The manufactured glasses were annealed at 450 °C.

XRD: The manufactured glasses were examined with a Philips X-ray diffractometer (model PW/1710).

FT-IR: Using KBr pellet technique, an infrared spectrophotometer of type JASCO, FT/IR-430 (Japan), quantified the FT-IR spectrum of these samples in the wavenumber region of 400–4000 cm$^{-1}$ at ambient temperature. To allow us to shed further light on the modifications of the various structural units, the resulting spectrum deconvoluted.

Density (ρ) of the fabricated glasses is established by the Archimedes method according to

$$\rho = \rho_0 \left(\frac{W}{W_t}\right),$$

where $W$ and $W_t$ are the weights of samples in air and fluid, respectively, the glass density is ρ and the density of toluene is $\rho_0$ (0.865 g.cm$^{-3}$) with error ± 0.001 g.cm$^{-3}$.

Molar volume $V_m$ of the fabricated glasses is established by

$$V_m = M/\rho; M \text{ is sample’s molecular weight.}$$

Ultrasonic velocities: The ultrasonic velocities were approximated using a pulse-echo method (Echo-graph model 1085). Besides the density, the velocities were used to evaluate elastic moduli [21–30].

Longitudinal waves $L = \rho v_L^2$ [30, 31]

transverse waves $G = \rho v_T^2$ [30, 31]

Young’s modulus $Y = (1 + \sigma)2G$ [30, 31]

bulk modulus $K = L - \frac{2}{3}G$ [30, 31].

Elastic moduli of fabricated samples were determined using packing density and dissociation energy [32, 33].

$$V_i = \left(\frac{2}{3}\right)N_A(mR_A^3 + nR_O^3)m^3 \cdot mol^{-1} \quad [32, 33]$$

$$G_i = \left(\frac{1}{\nu_i}\right) \sum G_iX_i \quad [32, 33],$$

where $R_m$ and $R_O$ are metallic and oxygen Pauling radii. Longitudinal waves $L = K + \left(\frac{2}{3}\right)G$; transverse waves $G = 30 \ast \left(\frac{\sqrt{\kappa}}{\sqrt{\nu}}\right)^2$; Young’s modulus $Y = 8.36V_iG_i$; bulk modulus $K = 10V_i^2G_i$; Poisson’s ratio $\sigma = \frac{1}{2} - \left(\frac{\nu_i\nu}{1 + \nu}V_i\right)$; Acoustic impedance $Z = v_I\rho$;

Micro-hardness $H = \frac{(1 - 2\nu)(\sigma)}{6(1 + \nu)}$. Debye temperature $\theta_D = \frac{1}{\pi^2 K/m}M_s$, [32, 33].

| Sample name | Chemical composition |
|-------------|----------------------|
|             | B$_2$O$_3$ | SiO$_2$ | LiF | ZnO | TiO$_2$ |
| G1          | 59        | 29      | 2   | 10  | 0       |
| G2          | 59        | 29      | 2   | 8   | 2       |
| G3          | 59        | 29      | 2   | 6   | 4       |
| G4          | 59        | 29      | 2   | 4   | 6       |
Average velocities, 

\[ M_s = \frac{1}{3} \left( \frac{4}{3} \right)^{\frac{1}{3}} \] . Thermal coefficient of expansion, 

\[ \alpha_P = 23.2 \, (\mathrm{cm} \, \mathrm{m}^{-1} \, \mathrm{C}^{-1}) \]. The volume of the oxygen molar, 

\[ V_o = \left( \frac{\mu_l}{\mu} \right) \left( \frac{1}{\sum x_i n_i} \right) \]. Density of Packing, 

\[ \text{OPD} = \left( \frac{1000 \, \text{cm} \, \mu_l^{-1}}{\mu} \right) \].

Radiation parameters have been approximated in this article using Phy-X/PSD [19]. The coefficient of mass attenuation samples 

\[ (\mu_l/\rho) = \sum_i x_i (\mu_l/\rho)_i \]. Effective atomic number 

\[ Z_{\text{eff}} = \frac{\sum_i x_i A_{i} (\mu_l/\rho)_i}{\sum_i x_i (\mu_l/\rho)_i} \]. Half and tenth value layer (HVL) and (TVL): 

\[ L = \frac{6.69}{\lambda AC} \quad \text{TVL} = \frac{2.3}{\lambda AC} \]

3 Results and discussion

3.1 XRD analysis

XRD spectrum of SiO\textsubscript{2}–LiF–B\textsubscript{2}O\textsubscript{3}–ZnO–TiO\textsubscript{2} glasses is exemplified in Fig. 1. This spectrum demonstrates no discrete lines and no sharp peaks and shows a high degree of amorphousness. The width of the halo differs significantly from G 1 to G 5, but no crystalline phase is visible in any of the manufactured glasses.

3.2 Fourier-transform infrared spectroscopy investigations

FT-IR spectra are exemplified in Fig. 2 for titanium borosilicate glasses. To obtain accurate band positions in the Fourier-transform infrared spectroscopy spectrum [34–36], a deconvoluted process is used. Residue results were plotted to get the quality in the Fourier-transform infrared spectroscopy deconvolution fitting, and the variation is less than 0.02% in the experimental and simulated graphs. The Gaussian fit of the Fourier-transform infrared spectroscopy spectrum of these glasses is exemplified in Fig. 3 and Table 2, respectively. The structural units of the network in these glasses were identified and summarized as: The band at \( \sim 444–490 \, \text{cm}^{-1} \) is correlated to the network structure’s deformation modes. Bands in the 562–570 cm\(^{-1}\) are related to \((\text{LiO}_6), (\text{ZnO}_6), \) and \((\text{TiO}_4)\) vibrations and overlap with the O–Si–O. Bands \( \sim 680–691 \, \text{cm}^{-1}\) are related to the bending vibrations of the bridging oxygen atom. Bands at \( \sim 890 \, \text{cm}^{-1}\) are assigned to different borate groups. Bands at \( \sim 1120–1044 \, \text{cm}^{-1}\) are related to B–O stretching \(\text{BO}_4\) tetrahedra vibrations. Bands \( \sim 1120–890 \, \text{cm}^{-1}\) are correlated to B–O, bridge in the \(\text{BO}_4\). The bands \( \sim 1240–1260 \, \text{cm}^{-1}\) are correlated to trigonal \(\text{BO}_3\) units of B–O bond stretching. The band at \( \sim 1360–1350 \, \text{cm}^{-1}\) is assigned to B–O stretching vibrations that mostly involve divers’ groups of connected oxygen. B–O symmetric stretching vibrations of different borate groups are due to the band at \( \sim 1650–1500 \, \text{cm}^{-1}\).

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**Fig. 1** XRD of 59B\textsubscript{2}O\textsubscript{3}–29SiO\textsubscript{2}–2LiF–(10 – \( x \)) ZnO–\( x \) TiO\textsubscript{2} glasses

**Fig. 2** FT-IR spectrum of 59B\textsubscript{2}O\textsubscript{3}–29SiO\textsubscript{2}–2LiF–(10 – \( x \)) ZnO–\( x \) TiO\textsubscript{2} glasses
With an increase in TiO\textsubscript{2} concentration, the FT-IR spectral shifts to higher wavenumbers. Also, the structure becomes more compact by incorporating TiO\textsubscript{4} species are produced. Boron transforms from BO\textsubscript{3} into BO\textsubscript{4} tetrahedra after alkali metal halide is incorporated \cite{37–39}. This increases the coherence of the glass network and the structure stiffening.

### Table 2

| Sample | C  | A  | 681.0 | 895.5 | 1041.1 | 1116.4 | 1240.7 | 1367.9 | 1525.2 | 1650.2 | BO3 | BO4 | N4 |
|--------|----|----|-------|-------|--------|--------|--------|--------|--------|--------|-----|-----|----|
| G1     | 0.8| 8.0| 17.8  | 11.9  | 3.9    | 4.4    | 28.7   | 17.1   | 7.3    | 33.6   | 33.2| 0.5 |
| G2     | 0.5| 8.1| 19.4  | 11.7  | 2.3    | 3.9    | 37.3   | 6.9    | 9.9    | 33.5   | 41.2| 0.4 |
| G3     | 0.7| 4.1| 6.0   | 17.6  | 14.9   | 3.3    | 11.2   | 21.8   | 8.8    | 11.5   | 35.8| 33.0| 0.5 |
| G4     | 4.7| 9.0| 18.6  | 10.9  | 4.0    | 4.1    | 26.7   | 14.8   | 7.3    | 33.5   | 30.7| 0.5 |
| G5     | 0.4| 1.8| 6.8   | 19.1  | 11.9   | 5.2    | 5.7    | 23.1   | 16.3   | 9.8    | 36.1| 28.7| 0.6 |
| G6     | 0.7| 8.2| 17.5  | 12.6  | 3.6    | 4.5    | 20.6   | 26.7   | 5.6    | 33.7   | 25.1| 0.6 |

### 3.3 Investigations of mechanical

Glass density raised as TiO\textsubscript{2} content increased, and molar volume reduced as Fig. 4. The density increase is ascribed to the transformation of the BO\textsubscript{3} triangles into the tetrahedral BO\textsubscript{4} of the glass network with the progressive replacement of ZnO with TiO\textsubscript{2}. Rising density results affect the stiffness of fabricated glasses network and the enhancement in inflexibility.
Enhanced density values also reflect an increase in the density of cross-links. The density increase may also be because of an increment in oxygen in the glass network. The mixed oxide effect may be contributed to a variation in density and molar volume. These results are like the FT-IR results.

The velocity of fabricated glasses TiO₂ is illustrated in Fig. 5. Velocities (vL and vT) were increased, as shown in Table 3, by an increase in TiO₂, and (vL) values higher than (vT). This increment in the estimated ultrasonic velocity is possible to explain by considering variables:

(i) Increasing TiO₂ will enhance the creation of TiO₄ and TiO₆ structural units with higher coordination in comparison with ZnO structural unit.

(ii) As a result, the polymerization of the fabricated glasses coordination number, cross-link density, and connectivity within the glass network enhanced.

(iii) Because of the increase in internal energy, the velocities were increased.

Experimentally and theoretically, elastic modules were evaluated for prepared glasses and are exemplified in Figs. 6 and 7. The elastic moduli exactly as noticed of velocities as exemplified in Figs. 6 and 7, i.e., it depends on the nature of bonds in the glass and the cross-link density [40, 41]. With the increase in TiO₂, the elastic moduli value shows an increasing trend. The rise in elastic modules as the number of coordinates raised, and bond strength of Ti–O (73 kcal/mol) is higher than Zn–O (36 kcal/mol). As the modification role of TiO₂ in the glass system, all mechanical parameters as revealed in Table 4 are increased with the increase in TiO₂ content.

3.4 Photon shielding features

Concerning mass attenuation coefficient values (MAC) are obtained by Phy-X/PSD. Because MAC is characterized by the material absorbed to attenuate radiation, a higher value indicates a more impervious shield. Phy-X/PSD and XCOM performance, MAC values concerning the energy and TiO₂ mol % are exemplified in Figs. 8 and 9. More clearly, it is shown from Figs. 8 and 9 that the impact of the glass structure has a significant impact in improving the MAC values of these glasses. The maximum value of MAC is noticed at lower energy, and as energy increment, MAC rapidly decreased. The MAC of the glasses reduces as energy increases as more photons can absorb through the sample, reducing its absorbency and reducing MAC. This sequence also shows that samples are the most efficient at lower energies. Their shielding capacity becomes less efficient when energy increases. Besides, Ti has a lower atomic number (22) than Zn (33). The atomic number correlates positively with MAC, causing the highest radiation shielding capability in the glass sample G 1. Even so, the G1 glass can be the most superior attenuation abilities to the other glasses [42–52].

As shown in Table 5, MAC values simulated with Phy-X/PSD and XCOM are compared. Our results are in excellent agreement with other values [46]. The deviations between Phy-X/PSD and XCOM were determined as Deviation % =
LAC is a useful method for calculating the glasses’ attenuation shielding efficiency, which also demonstrates the same trend as MAC. Figure 10 exemplifies LAC with the energy. The atomic number plays a significant role in improving LAC values. In general, with the decrease in atomic number LAC and MAC are decreased. The identified LAC values follow the trend for all energies. These data indicated that the LAC values are associated with the atomic number of titanates, causing G1 to be the highest LAC. It is possible to describe the reduction using the same reasoning as for MAC.

Figure 11 exemplifies HVL as energy function. HVL values increase with energy. So, G1 the most attractive shield than the other samples examined. This result suggests that, at a lower energy level, glasses are much more effective, while reducing efficiency as excess energy. Figure 12 exemplifies TVL as a photon energy function. The TVL values increased with the increment of energy in all the examined glasses. TVL was analogous with HVL values.

Figure 13 exemplifies \( Z_{\text{eff}} \) as a photon energy function. \( Z_{\text{eff}} \) symbolizes the mean atomic number, with a higher value suggesting a good shield. G1 has been reported the highest value. This behavior can be explained as Ti has a lower atomic number (22) than Zn (33). This behavior indicates that it is further needed to increase the titanate in the glasses. The photoelectric effect is strongly dependent on the atomic number; therefore, the glasses-containing Zn has a quickly reducing \( Z_{\text{eff}} \) since Zn has an atomic number of 30. The G6 glass sample shows the smallest potential for shielding than other samples.

### Table 3 The mechanical properties of prepared glasses

| Samples name | \( v_L \) (m/s) | \( V_T \) (GPa) | \( L \) (GPa) | \( G \) (GPa) | \( K \) (GPa) | \( Y \) (GPa) | \( L_M \) (GPa) | \( G_M \) (GPa) | \( K_M \) (GPa) | \( Y_M \) (GPa) |
|--------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| G1           | 5385           | 2970           | 73.66          | 22.41          | 43.78          | 57.42          | 90.9           | 71             | 37.6           | 121            |
| G2           | 5445           | 3005           | 79.16          | 24.11          | 47.01          | 61.77          | 97.6           | 80.7           | 40             | 134            |
| G3           | 5480           | 3020           | 83.48          | 25.35          | 49.68          | 65.01          | 104            | 90.1           | 42.2           | 146            |
| G4           | 5495           | 3040           | 88.77          | 27.17          | 52.55          | 69.53          | 112            | 104            | 45.1           | 164            |
| G5           | 5505           | 3060           | 93.64          | 29.93          | 55.06          | 73.86          | 120            | 118            | 48             | 182            |
| G6           | 5545           | 3070           | 98.70          | 30.25          | 58.36          | 77.39          | 127            | 131            | 50.6           | 198            |

**Fig. 6** Experimentally elastic modules of 59B\(_2\)O\(_3\)–29SiO\(_2\)–2LiF–(10 – \( x \)) ZnO–\( x \) TiO\(_2\) glasses

**Fig. 7** Theoretically elastic modules of 59B\(_2\)O\(_3\)–29SiO\(_2\)–2LiF–(10 – \( x \)) ZnO–\( x \) TiO\(_2\) glasses

\[
\left( \frac{\left( \frac{\mu}{\rho} \right)_{\text{phy}} \cdot \left( \frac{\mu}{\rho} \right)_{x \text{COM}}}{\left( \frac{\mu}{\rho} \right)_{\text{phy-x}}} \right) \times 100. \text{ The deviations acquired are small, which provides the Phy-x results.}
\]
In the current study, the fabricated glasses with the composition $59\text{SiO}_2-29\text{SiO}_2-2\text{LiF}-(10-x)\text{ZnO}-x\text{TiO}_2$ where $x = (0 \leq x \leq 10)$ conventional melt-quenching methods have been manufactured. The structure, mechanical, and shielding factors for fabricated glasses have been studied. The findings showed the resulting objects:

1. XRD analysis confirmed that the fabricated glasses are amorphous.
2. As TiO$_2$ content rises, the density of the samples raised in contrast to the molar volume reduced.
3. As TiO$_2$ concentration increased, so did the ultrasonic velocities of the fabricated glasses.
4. The Phy/X / PSD and XCOM predicted the gamma shielding characteristics of the fabricated glasses. The influence of TiO$_2$ on the shielding capacity of the fabricated glasses was investigated as: (i) MAC decreased with the increment in TiO$_2$ from 0 mol. % to 10 mol. %, (ii) The glass name G 1 has the smallest HVL and the maximum $Z_{eff}$. (iii) Lithium fluoride zinc titanate borosilicate glasses were found to have an abnormal attenuation, structural, and density relationship.

The results obtained have shown that the increase in TiO$_2$ concentration in the fabricated glasses can result in major benefits in attenuation, structural, and physical characteristics. Moreover, the fabricated glasses can be used in aircraft bodies, as a radiation shield in X-ray centers, and as house facades.
Table 5 The MAC and Div. (%) between the Phy-X/PSD and XCOM data of the glass system

| MAC G 1 | MAC G 2 | MAC G 3 | MAC G 4 | MAC G 5 | MAC G 6 |
|---------|---------|---------|---------|---------|---------|
| XCOM    | Phy-x   | Div %   | XCOM    | Phy-x   | Div %   | XCOM    | Phy-x   | Div %   | XCOM    | Phy-x   | Div %   | XCOM    | Phy-x   | Div %   |
| 8.89    | 10.33   | 13.99   | 8.03    | 9.28    | 15.55   | 7.17    | 8.22    | 12.78   | 6.31    | 7.16    | 11.89   | 5.45    | 6.10    | 10.69   |
| 4.02    | 4.73    | 15.11   | 3.62    | 4.24    | 17.18   | 3.22    | 3.75    | 14.09   | 2.82    | 3.25    | 13.33   | 2.42    | 2.76    | 12.25   |
| 1.35    | 1.61    | 16.42   | 1.22    | 1.45    | 19.15   | 1.09    | 1.29    | 15.54   | 0.96    | 1.13    | 14.96   | 0.83    | 0.96    | 14.13   |
| 0.66    | 0.80    | 16.50   | 0.61    | 0.72    | 19.20   | 0.55    | 0.65    | 15.63   | 0.49    | 0.58    | 15.00   | 0.44    | 0.51    | 14.20   |
| 0.42    | 0.49    | 15.61   | 0.39    | 0.45    | 17.85   | 0.36    | 0.42    | 14.57   | 0.33    | 0.38    | 13.89   | 0.30    | 0.34    | 13.06   |
| 0.30    | 0.35    | 14.15   | 0.29    | 0.33    | 15.74   | 0.27    | 0.31    | 12.98   | 0.25    | 0.29    | 12.27   | 0.23    | 0.26    | 11.43   |
| 0.21    | 0.24    | 10.87   | 0.20    | 0.23    | 11.50   | 0.20    | 0.22    | 9.70    | 0.19    | 0.21    | 9.02    | 0.18    | 0.20    | 8.33    |
| 0.17    | 0.19    | 8.14    | 0.17    | 0.18    | 8.30    | 0.17    | 0.18    | 7.16    | 0.16    | 0.17    | 6.62    | 0.16    | 0.17    | 6.05    |
| 0.14    | 0.14    | 4.28    | 0.14    | 0.14    | 4.22    | 0.14    | 0.14    | 3.74    | 0.14    | 0.14    | 3.50    | 0.13    | 0.14    | 3.18    |
| 0.12    | 0.13    | 2.60    | 0.12    | 0.13    | 2.49    | 0.12    | 0.12    | 2.26    | 0.12    | 0.12    | 2.08    | 0.12    | 0.12    | 1.98    |
| 0.10    | 0.11    | 1.29    | 0.10    | 0.11    | 1.23    | 0.10    | 0.11    | 1.14    | 0.10    | 0.11    | 1.07    | 0.10    | 0.11    | 1.00    |
| 0.09    | 0.09    | 0.75    | 0.09    | 0.09    | 0.71    | 0.09    | 0.09    | 0.67    | 0.09    | 0.09    | 0.62    | 0.09    | 0.09    | 0.59    |
| 0.09    | 0.09    | 0.50    | 0.09    | 0.09    | 0.47    | 0.09    | 0.09    | 0.44    | 0.09    | 0.09    | 0.42    | 0.09    | 0.09    | 0.39    |
| 0.08    | 0.08    | 0.34    | 0.08    | 0.08    | 0.32    | 0.08    | 0.08    | 0.32    | 0.08    | 0.08    | 0.30    | 0.08    | 0.08    | 0.30    |
| 0.07    | 0.07    | 0.19    | 0.07    | 0.07    | 0.18    | 0.07    | 0.07    | 0.17    | 0.07    | 0.07    | 0.16    | 0.07    | 0.07    | 0.16    |
| 0.06    | 0.06    | 0.10    | 0.06    | 0.06    | 0.11    | 0.06    | 0.06    | 0.10    | 0.06    | 0.06    | 0.09    | 0.06    | 0.06    | 0.09    |
| 0.05    | 0.05    | 0.02    | 0.05    | 0.05    | 0.03    | 0.05    | 0.05    | 0.02    | 0.05    | 0.05    | 0.02    | 0.05    | 0.05    | 0.03    |
| 0.04    | 0.04    | 0.02    | 0.04    | 0.04    | 0.01    | 0.04    | 0.04    | 0.01    | 0.04    | 0.04    | 0.00    | 0.04    | 0.04    | 0.01    |
| 0.04    | 0.04    | 0.05    | 0.04    | 0.04    | 0.05    | 0.04    | 0.04    | 0.02    | 0.04    | 0.04    | 0.02    | 0.04    | 0.04    | 0.02    |
| 0.03    | 0.03    | 0.13    | 0.03    | 0.03    | 0.10    | 0.03    | 0.03    | 0.07    | 0.03    | 0.03    | 0.07    | 0.03    | 0.03    | 0.07    |
| 0.03    | 0.03    | 0.22    | 0.03    | 0.03    | 0.19    | 0.03    | 0.03    | 0.16    | 0.03    | 0.03    | 0.15    | 0.03    | 0.03    | 0.06    |
| 0.03    | 0.03    | 0.30    | 0.03    | 0.03    | 0.25    | 0.03    | 0.03    | 0.20    | 0.03    | 0.03    | 0.15    | 0.03    | 0.03    | 0.09    |
| 0.02    | 0.02    | 0.47    | 0.02    | 0.02    | 0.39    | 0.02    | 0.02    | 0.31    | 0.02    | 0.02    | 0.23    | 0.02    | 0.02    | 0.14    |
| 0.02    | 0.02    | 0.60    | 0.02    | 0.02    | 0.51    | 0.02    | 0.02    | 0.42    | 0.02    | 0.02    | 0.32    | 0.02    | 0.02    | 0.18    |
| 0.02    | 0.02    | 0.90    | 0.02    | 0.02    | 0.77    | 0.02    | 0.02    | 0.63    | 0.02    | 0.02    | 0.44    | 0.02    | 0.02    | 0.30    |
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