

INFRARED SPECTROSCOPY OF MIXED GLASS FORMER EFFECT IN BOROTELLURITE GLASSES: A REVIEW

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Abstract

Borotellurite (BT) glasses have been extensively studied since the past decades until now because of their worthy properties and for further development in the study of glass. The combination of tellurite and borate network former oxides yields improved physical, optical and electrical features of the glass. BT glass matrix consists of BO_4 , BO_3 , TeO_4 and TeO_3 units. The coordination number of tellurium was altered from four to three through the cleavage of the glassy matrix of tellurite. A review in terms of infrared spectroscopy analysis is performed for identifying the mixed glass former effect (MGFE) in BT glasses with incorporation of different types of dopants. Infrared spectroscopy (IR) revealed various functional groups that are present within the glass samples according to the types of dopants added into the glass system. The effects of different dopants of MGFE in BT glasses that observed from the IR data obtained, are further discussed.

Keyword: Borotellurite, Glass, Infrared spectroscopy, Mixed glass former effect

Introduction

Mixed glass former effect (MGFE) shows approximately intriguing characteristics such as non-linear and no additional adjustments to the glass features when the ratio of the network formers is varied (Wang et al., 2017). The introduction of tellurite into any other glass former, like B_2O_3 , is factual and empirical interest, where it may promote the development of stimulating structural units that could influence the glass network physical properties (Syed et al., 2013). Furthermore, the most stable glass is produced through the addition of borate oxide in tellurite glass and it had been extensively utilized for specific applications due to its unique properties (Azlan et al., 2019). The combination of tellurite and borate network former oxides shows a MGFE, where the conductivity of ions and glass transition temperature exhibit non-linear alterations with composition (Chatzipanagis et al., 2020). Nevertheless, the existence of tellurium oxide in the structure of borate glass had reduced its hygroscopic character. Nonetheless, it also enhances the conditions of the infrared spectroscopy (IR) transmission. The alkali metal oxides, such as potassium oxide in the borotellurite (BT) network are responsible on the modification of the main structure via the alteration of the structural units of the borate system from BO_3 to BO_4 units and the tellurite matrix from TeO_4 to TeO_3 units (Syed et al., 2013). In addition, it was also reported that the best host glasses for transition metal (TM) ions like Cr^{3+} , Cu^{2+} , Mn^{2+} , VO^{2+} , etc. are BT glasses (Chandra et al., 2020). IR is a common spectroscopic testing to identify the chemical bonds and molecular structure of organic and inorganic compounds like glasses (Chandra et al., 2020). Moreover, the properties of sample are studied through the absorbance (or transmission) spectrum and IR spectra can be obtained due to the changes of dipole moment in the material (Chandra et al.,

2020; Yadav & Singh, 2015). IR spectra exhibits the existence of numerous effective vibrational modes in the bonds like bending and stretching vibrations in distinctive structural groups that represent the glass matrix (Chandra et al., 2020). In present review article, infrared spectroscopic results for MGFE in BT glasses with incorporation of different types of dopants are summarized and discussed. Thus, it could provide a better understanding on structural properties of the glasses using infrared spectroscopy.

Infrared Spectroscopy

Borotellurite Glasses and Doped Borotellurite Glasses

FTIR studies on the binary BT glass matrix have been reported by Kaur et al. (2014) and the typical IR spectra for the glasses are shown in the **Figure 1a**. The FTIR absorption spectra present in the BT glass matrix consist of three bands in the varied wavenumber of 500-800 cm^{-1} , 800-1150 cm^{-1} and 1150-1550 cm^{-1} . These spectral bands illustrate the mode of vibration for both tellurite and borate structural units (Gupta et al., 2017). Spectral bands at about 640 cm^{-1} will normally present due to Te–O vibrations of trigonal bipyramidal units with bridging oxygen, while a sharp peak at 806 cm^{-1} depicted the presence of pure borate in the glass system (Kaky et al., 2019). The first band in the wavenumber region of 500-800 cm^{-1} is due to Te–O linkage vibrations in different Te–O units. Meanwhile, the second band, which is around 820-1140 cm^{-1} , is due to B–O linkage stretching vibrations in BO_4 units and the third band at 1150-1550 cm^{-1} is due to stretching vibration of BO_3 units (Gupta et al., 2017; Kaky et al., 2019; Kaur et al., 2014; Kaur & Khanna, 2014; Malge et al., 2020; Selvaraju & Marimuthu, 2013; Syed et al., 2013).

On the other hand, IR study on BT glasses with extra modifiers of zinc, aluminum, and alkali-alkaline was reported by Kaky et al. (2019) in the wavenumber range of 400–1500 cm^{-1} which showed various absorption bands at different wavenumber as depicted in **Figure 1b** (Kaky et al., 2019). The first region (1250-1400 cm^{-1}) was ascribed to stretching of B–O linkage in the BO_3 units, where $\sim 1400 \text{ cm}^{-1}$ bands were due to the B–O stretching of BO_3 units from different borate units, while the bands at $\sim 1340 \text{ cm}^{-1}$ were due to the transformation of some BO_4 to BO_3 units that was assigned to stretching of BO_3 units in many types of borate groups. Furthermore, the bands at $\sim 1250 \text{ cm}^{-1}$ were assigned to asymmetric stretching of B–O linkage in BO_3 units from boroxol rings. The second region (847-1063 cm^{-1}) was due to B–O stretching of BO_4 units. No sharp peak was observed at 806 cm^{-1} , which indicates that the boroxol rings of pure borate was absence in the structural units, where it may change into BO_3 and BO_4 groups (Ali et al., 2018). The commonly exist spectral bands at about 640 cm^{-1} were due to Te–O vibrations. They were seen to rise to higher frequencies of 687 cm^{-1} due to the ZnO concentrations and Al–O linkage from Al^{3+} ions will be visible in the fourfold coordination. This similar assignment of Te–O vibrations can be seen in a lower bands of $\sim 520 \text{ cm}^{-1}$ and $\sim 650 \text{ cm}^{-1}$ which were due to the MnO concentration in MnO doped BT glasses (Syed et al., 2013). Meanwhile, a band was observed at 520 cm^{-1} which is due to stretching vibration of TeO_3 units and B–O–B bending vibrations. The weak band was observed at the region at 450 cm^{-1} , where the band depicted the bending vibrations of O–B–O, Te–O–Te and Zn–O linkage vibrations from the ZnO_4 unit (Kaky et al., 2019). The bands were also assigned to Te–O–Te or Te–O–W, where similar spectral bands were also be seen in the ZnO, Li_2O and Dy_2O_3 doped BT glasses studied by Malge et al. (2020). In addition, the bands were also attributed to the alkali cations vibrations at their localized sites (Gaafar et al., 2013).

Next, Malge et al., (2020) conducted investigations on ZnO, Li_2O and Dy_2O_3 doped BT glasses by IR spectra in the range of 400 to 4000 cm^{-1} and the results are shown in the **Figure 1c** where nine spectral bands were detected for all the glass samples (Malge et al., 2020). The first band within 443–469 cm^{-1} can be ascribed to stretching vibrations of Te–O–W or Te–O–

Te bonds. Second band within $703\text{--}713\text{ cm}^{-1}$ can be assigned to stretching vibrations of Te–O–Te bonds in TeO_3 units (Selvaraju & Marimuthu, 2013). The third, fourth and fifth band were within $1043\text{--}1053\text{ cm}^{-1}$, $1181\text{--}1213\text{ cm}^{-1}$ and $1388\text{--}1393\text{ cm}^{-1}$ can be attributed to stretching vibrations of B–O linkage or bending vibrations of B–O–B linkage in the BO_4 units from tri-, tetra- and penta-borate groups, respectively. Similar assignment can be seen in a lower wavenumber region of $679\text{--}1125\text{ cm}^{-1}$ in the Sm^{3+} doped BT glasses (Selvaraju & Marimuthu, 2013). The sixth and seventh band within $2922\text{--}2927\text{ cm}^{-1}$ and at 2975 cm^{-1} respectively denoted the hydrogen bonding. Similar assignment was observed on a lower and wider wavenumber region in the Sm^{3+} doped BT glasses of $2848\text{--}2926\text{ cm}^{-1}$ (Selvaraju & Marimuthu, 2013). Bands that were detected in the wavenumber region of $3405\text{--}3415\text{ cm}^{-1}$ and $3777\text{--}3782\text{ cm}^{-1}$, were associated to hydroxyl groups due to stretching of O–H linkage. Similar assignment can be seen on a lower wavenumber region of $\sim 3200\text{ cm}^{-1}$ in the MnO doped BT glasses (Syed et al., 2013).

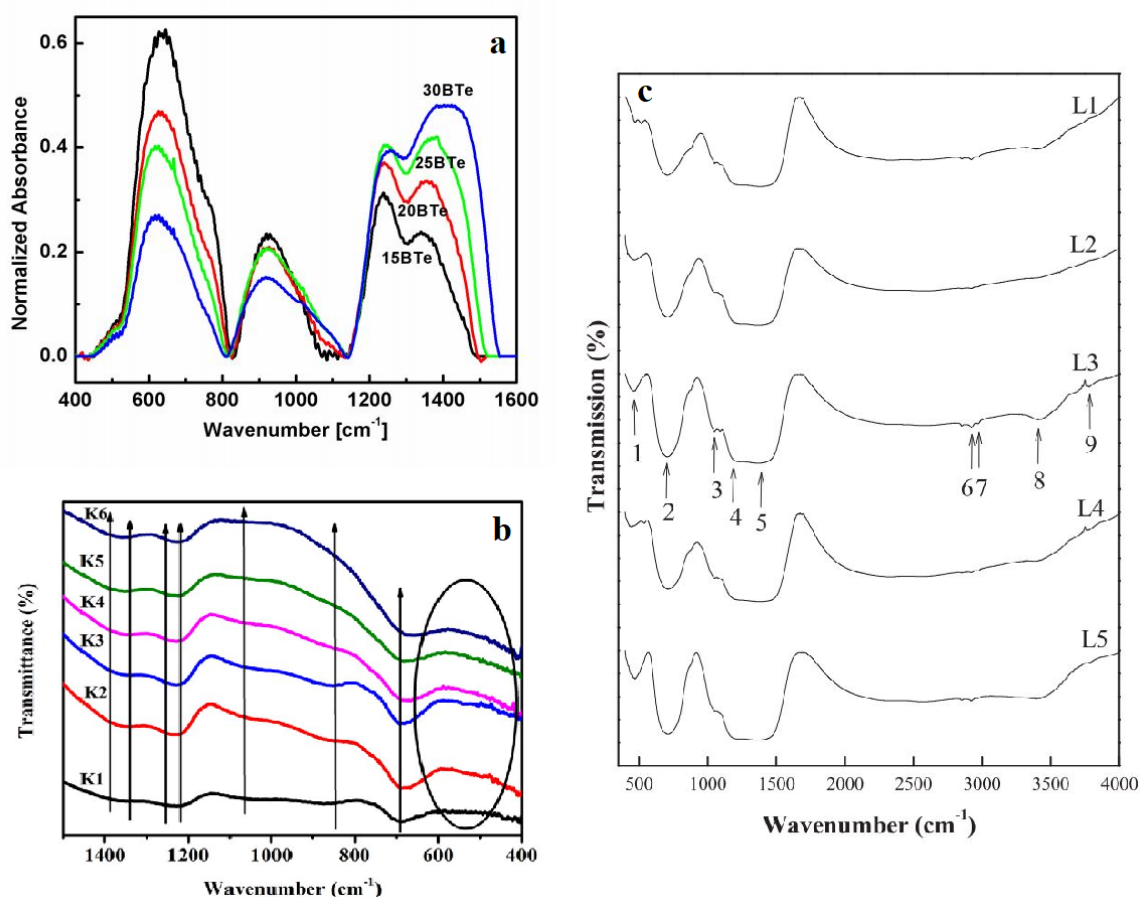


Figure 1 FTIR spectra of (a) $x\text{B}_2\text{O}_3\text{--}(100\text{--}x)\text{TeO}_2$ (Kaur et al., 2014), (b) $[(60\text{--}x)\text{B}_2\text{O}_3\text{--}(10\text{+}x)\text{TeO}_2\text{--}10\text{ZnO--}10\text{Al}_2\text{O}_3\text{--}5\text{Li}_2\text{O--}5\text{MgO}]$ (Kaky et al., 2019) and (c) $(\text{B}_2\text{O}_3)_{0.2}\text{--}(\text{TeO}_2)_{0.5\text{--}x}\text{--}(\text{ZnO})_{0.28}\text{--}(\text{Dy}_2\text{O}_3)_{0.02}\text{--}(\text{Li}_2\text{CO}_3)_x$ glass systems (Malge et al., 2020)

Lithium Borotellurite and Zinc Lithium Borotellurite Glasses

IR studies of lithium BT glass matrix that revealed different spectral bands had been reported as shown in the **Figure 2** (Chatzipanagis et al., 2020). Meanwhile, zinc doped lithium BT glasses have been studied are shown in the **Figure 3** (Naresh et al., 2019). The band $693\text{--}701\text{ cm}^{-1}$ was attributed to bending vibrations of Te–O bonds in TeO_3 and TeO_4 units. The presence of band at $759\text{--}770\text{ cm}^{-1}$ was due to bending vibrations of B–O–B linkage of BO_3 and BO_4 structural units. On the other hand, the band around $1244\text{--}1266\text{ cm}^{-1}$ was attributed

to the stretching vibrations of B–O bond from orthoborate groups. A band which presented around 1375–1450 cm^{-1} was attributed to stretching vibrations of B–O linkage in BO_3 units from different types of borate groups (Chatzipanagis et al., 2020; Naresh et al., 2019). Weak bands were revealed at 417–422 cm^{-1} because of vibrations of ZnO_4 units (Naresh et al., 2019). Bands at ~ 340 cm^{-1} (**Figure 2a**) and ~ 365 cm^{-1} (**Figure 2b**) may present due to the vibration of Li cations (Chatzipanagis et al., 2020). Yet, at a higher wavenumber of 527–545 cm^{-1} a band with similar assignment was observed (Naresh et al., 2019). Furthermore, a band in the range 3440–3455 cm^{-1} was noticed because of the hydroxyl groups (Naresh et al., 2019).

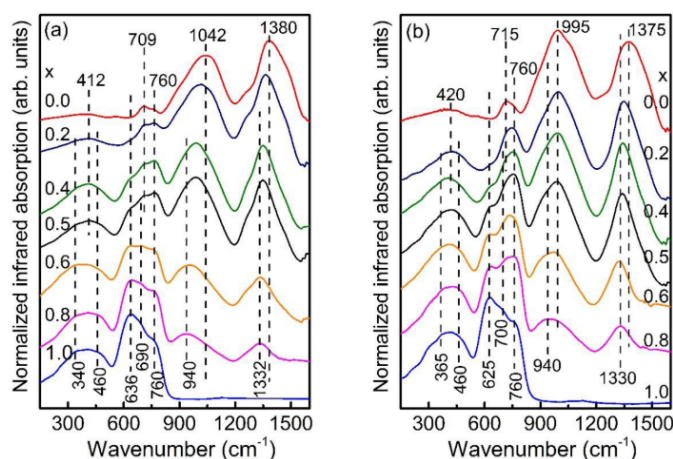


Figure 2 IR spectra of $(\text{Li}_2\text{O})_y-[(2\text{TeO}_2)_x-(\text{B}_2\text{O}_3)_{1-x}]_{1-y}$ where (a) $y=0.33$ and (b) 0.40 (Chatzipanagis et al., 2020)

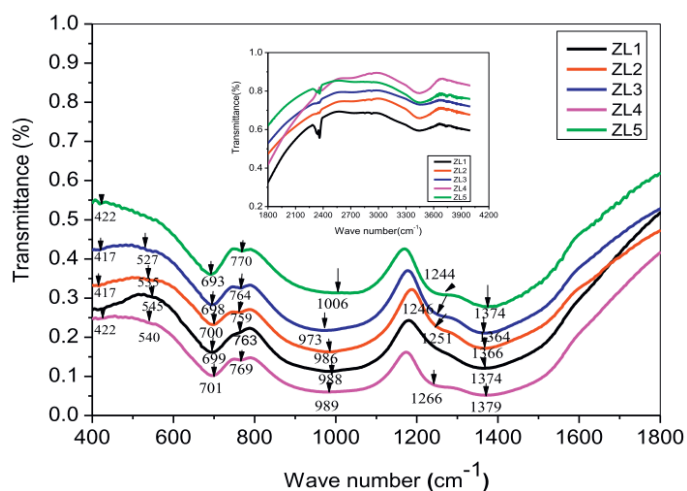


Figure 3 IR spectra of $x\text{ZnO}-(30-x) \text{Li}_2\text{O}-10\text{TeO}_2-60\text{B}_2\text{O}_3$ (Naresh et al., 2019)

Zinc Borotellurite glasses with different additives

IR spectroscopy on doped zinc borotellurite glass were reported by Halimah & Eevon (2019), Ali et al. (2018), Ami et al. (2017), Azlan et al. (2019), Hasnimulyati et al. (2016) and Anand et al. (2016). According to Halimah & Eevon (2019), the incorporation of ZnO into the glass matrix can improve the role of the glass formers and establish low rates of crystallization in the vitreous network (Halimah & Eevon, 2019). This is because the addition of ZnO altered the constructional units of borate and tellurite from trigonal to tetragonal units, which causes the presence of non-bridging oxygen (NBO) in the glass matrix. The spectral range from 400 to 1500 cm^{-1} were attributed to the tellurite and borate glass network (Halimah & Eevon,

2019). The addition of Gd_2O_3 NPs to the glass network altered the structural units from TeO_4 to TeO_3 and BO_4 to BO_3 and vice versa. The mode of vibration of Gd-O and Zn-O bonds were absent because of their role as glass modifier had broken the bond and occupied the interstitial spaces within the glass system (Azlan et al., 2019; Halimah & Eevon, 2019; Hasnimulyati et al., 2016). Dysprosium oxide mode of vibration also were not detected, which may due to small amount of dopant concentration and the instrument was not able to detect it (Ami et al., 2017). Similar result occurred for dopant of neodymium oxide in Nd_2O_3 doped zinc BT glasses (Azlan et al., 2019). The absence of Tm-O bond in Tm_2O_3 doped zinc BT glasses might referred to the same reason too (Hasnimulyati et al., 2016). The first band that had peak within $541\text{--}624\text{ cm}^{-1}$ can be ascribed to the presence of TeO_4 group, while the second band that had peak within $654\text{--}686\text{ cm}^{-1}$ can be ascribed to the presence of TeO_3 group. The stretching vibration of BO_4 can be seen within peak spectra of $880\text{--}927\text{ cm}^{-1}$ and $1046\text{--}1076\text{ cm}^{-1}$ while the stretching vibration of BO_3 can be seen within peak spectra of $1220\text{--}1227\text{ cm}^{-1}$ and $1347\text{--}1356\text{ cm}^{-1}$ (Halimah & Eevon, 2019). At a slightly higher wavenumber of 940 cm^{-1} and 1360 cm^{-1} , centered bands were observed for BO_4 and BO_3 units, respectively of Eu_2O_3 doped zinc BT glasses (Ali et al., 2018).

Lead Borotellurite Glasses

The IR spectroscopy conducted that focused on the structural changes in the BT glass system with the incorporation of metal fluorides and the spectra obtained are shown in the **Figure 4** (Chandra et al., 2020). The band at 710 cm^{-1} that depicted the existence of the vibrations of Te-O linkage in TeO_4 units and bending vibrations of BO_3 units or bending vibrations of B-O-B linkage from pentaborate group showed a reduction in intensity as $\text{CaF}_2/\text{PbF}_2$ was added. In Ca-PTBCR glasses, the substitution of TeO_2 with CaF_2 had caused the band at 920 cm^{-1} to shift to a higher wavenumber region and this may be due to the formation of BO_3 units. On the other hand, the replacement of TeO_2 with PbF_2 in Pb-PTBCR glasses causes the band to shift approaching the lower wavenumber region and this may be a result of the formation of BO_4 units. Yet in Ca-PTBCR/Pb-PTBCR glasses the band had been shifted towards lower wavenumber region and this may be a result of the formation of fluorine's linkage in the glass matrix. The band that was assigned to asymmetric vibrations of B-O linkage on BO_3 units, was detected only in the absence of CaF_2 and PbF_2 at 1310 cm^{-1} . However, the presence of CaF_2 and PbF_2 related band was absent in Ca-PTBCR and Pb-PTBCR glass samples. In addition, infrared spectroscopy wavenumber regions including its assignment and glass system related to the other doped BT glass system are shown in the **Table 1**.

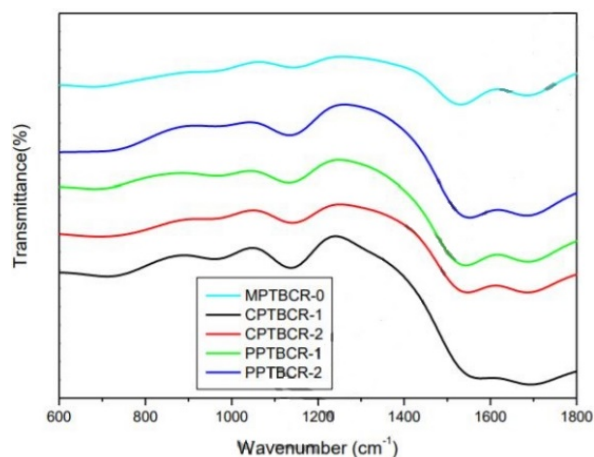


Figure 4 IR spectra of $x\text{MF}_2\text{-}30\text{PbO}\text{-(}10\text{-}x\text{)TeO}_2\text{-}59.8\text{B}_2\text{O}_3\text{-}0.2\text{Cr}_2\text{O}_3$ ($\text{M} = \text{Ca}$ and Pb ; $x = 0, 5, 10$ mole %) glass system (Chandra et al., 2020)

Table 1 FTIR band assignment in the spectra of doped BT glasses

Wavenumber (cm^{-1})	FTIR Assignment	Glass System
1518	Stretching vibration of BO_3 unit connected to large borate matrix	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-Bi}_2\text{O}_3\text{-BaO-LiF}$ (Elkhoshkhany et al., 2020)
1500–1121	B–O stretching vibrations of BO_3 units in meta-, pyro- & ortho-borate group Asymmetric stretching of borate triangles BO_3 , BO_2O with NBO Stretching vibration of BO^- bond in BO_3 unit	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-BaO}$ (Azuraída et al., 2020); $\text{TeO}_2\text{-B}_2\text{O}_3\text{-Bi}_2\text{O}_3$ (Saddeek et al., 2018); $\text{TeO}_2\text{-B}_2\text{O}_3\text{-SiO}_2\text{-Er}_2\text{O}_3$ (Umar et al., 2020); $\text{B}_2\text{O}_3\text{-TeO}_2\text{-K}_2\text{O-Al}_2\text{O}_3\text{-Eu}_2\text{O}_3$ (Van et al., 2019)
1266–1221	B–O asymmetric stretching vibrations of BO_3 units B–O bond stretching vibrations of BO_3 units from boroxyl groups and boroxol rings	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-SiO}_2\text{-Sm}_2\text{O}_3$ (Asyikin et al., 2020); $\text{TeO}_2\text{-B}_2\text{O}_3\text{-SiO}_2\text{-Er}_2\text{O}_3$ (Umar et al., 2020); $\text{B}_2\text{O}_3\text{-TeO}_2\text{-K}_2\text{O-Al}_2\text{O}_3\text{-Eu}_2\text{O}_3$ (Van et al., 2019)
~1200	Asymmetric stretching of B–O linkage from BO_4 and orthoborate	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-BaO}$ (Azuraída et al., 2020)
~1198	B–O stretching vibrations of trigonal boron units in boroxol rings	$\text{TiO}_2\text{-Bi}_2\text{O}_3\text{-B}_2\text{O}_3\text{-TeO}_2$ (Gupta et al., 2017)
1131–817	B–O stretching vibrations of BO_4 units in tri-, tetra- and penta-borate units	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-BaO}$ (Azuraída et al., 2020); $\text{B}_2\text{O}_3\text{-TeO}_2\text{-K}_2\text{O-Al}_2\text{O}_3\text{-Eu}_2\text{O}_3$ (Van et al., 2019)
840	Stretching vibrations of BO_3 units	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-BaO}$ (Azuraída et al., 2020)
~834–787	Symmetric stretching vibrations of the Te–O–Te of TeO_3 units Stretching vibration of B–O–B in B_3O_6 boroxol rings	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-Bi}_2\text{O}_3$ (Saddeek et al., 2018); $\text{TeO}_2\text{-B}_2\text{O}_3\text{-BaO}$ (Azuraída et al., 2020)
817–507	Asymmetrical stretching vibrations of Te–O linkages in TeO_4 and TeO_3 units	$\text{TiO}_2\text{-Bi}_2\text{O}_3\text{-B}_2\text{O}_3\text{-TeO}_2$ (Gupta et al., 2017); $\text{TeO}_2\text{-B}_2\text{O}_3\text{-ZnO-V}_2\text{O}_5$ (Anand et al., 2016)
800–660	B–O–B bonding in borate groups	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-SiO}_2\text{-MnO}_2$ (Zaitizila et al., 2018)
780–671	Te–O vibrations in TeO_3 groups with NBO	$\text{B}_2\text{O}_3\text{-TeO}_2\text{-K}_2\text{O-Al}_2\text{O}_3\text{-Eu}_2\text{O}_3$ (Van et al., 2019)
689–651	Te–O vibration in TeO_3 groups	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-SiO}_2\text{-Er}_2\text{O}_3$ (Umar et al., 2020)
~ 650–587	Vibrations of Te–O–Te between TeO_4 and bridging oxygen	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-BaO}$ (Azuraída et al., 2020)
650–600	Te–O linkage vibrations between TeO_4 and the bridging oxygen	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-SiO}_2\text{-Er}_2\text{O}_3$ (Umar et al., 2020)
595	Stretching vibrations of TeO_4 with BO	$\text{B}_2\text{O}_3\text{-TeO}_2\text{-K}_2\text{O-Al}_2\text{O}_3\text{-Eu}_2\text{O}_3$ (Van et al., 2019)
~575–497	Vibrations of Bi–O linkage in BiO_6 octahedral units	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-Bi}_2\text{O}_3$ (Saddeek et al., 2018)
564–504	B–O–B vibration overlap with TeO_4 polyhedron	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-BaO}$ (Azuraída et al., 2020)
~498–442	Te–O–Te or O–Te–O bond bending vibrations Stretching vibration of Ba–O linkage	$\text{TeO}_2\text{-B}_2\text{O}_3\text{-BaO}$ (Azuraída et al., 2020); $\text{TeO}_2\text{-B}_2\text{O}_3\text{-Bi}_2\text{O}_3\text{-BaO-LiF}$ (Elkhoshkhany et al., 2020)
<420	Vibration of metal cations (Li^+ , Ba^{2+})	

Conclusion

In this review article, IR results of MGFE BT glasses have been discussed through the analysis and comparison of reported research studies. At 1221–1518 cm^{-1} , 817–1200 cm^{-1} , 651–834 cm^{-1} and 650–595 cm^{-1} the various types of vibrations related to BO_3 units, BO_4 units, TeO_3 units and TeO_4 units were observed, respectively in undoped and doped BT glasses. In doped BT glasses, vibrations which were attributed to metallic cations were observed at lower wavenumber of $<450 \text{ cm}^{-1}$. On the other hand, in undoped BT glass, the sharp peak at 806 cm^{-1} was observed which indicated that the boroxol rings of pure borate presented in the glass system. Meanwhile, boroxol ring formation in doped BT glasses were not observed in IR spectra, which it could be conclude that addition of dopant in BT glasses deformed the boroxol ring.

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Conflict of interests

Authors declare that there is no conflict of interests.

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