Recent innovations in properties of nanostructured glasses and composites

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ABSTRACT

The thrust to invent new materials and technology has been ever increasing due to technological challenges in progressive world. Among new materials, nanomaterials possess superior optical, electrical, magnetic, mechanical, and thermal properties, which have made them suitable for a multitude of applications. The present review paper deals with recent advances in properties of nanostructured glasses and composites in terms optical, electrical, mechanical and thermal properties. A brief discussion has been done on fabrication method of nanostructured glasses. The review of optical properties shows that nanostructured glasses show both direct and indirect band gap and this tuning of band gap depends on extent of nano structuring of samples. The electrical properties also show enhancement in electrical conductivity on nano-structuring of glasses compared to their bulk counterparts. The changes in mechanical and thermal properties of nanostructured glasses and composites are attributed to many microstructural features like grain size and shape, their distribution, pores and their distribution, other flaws/defects and their distribution, surface condition, impurity level, second phases/dopants, stress, duration of its application and temperature effect on the samples. Literature reports that nano structuring leads to enhanced phonon boundary scattering which reduces the thermal conductivity.

1. Introduction

In the rapidly advancing world, it is important to study new materials for new properties and applications. Many new materials have been discovered which show interesting properties for scientific applications [1–11]. Among these materials, nanostructured materials have emerged as boon to mankind. In the past decade, many researchers have invested...
their time and efforts to study the exciting properties of materials by manipulating their nanostructures. Nanostructured materials having the solid form are most common nano-materials. Solid nanostructured materials are roughly classified into four categories i.e. powder, fiber, film, and bulk depending on their appearance. They can be Zero-dimensional nanostructures (0D), One-dimensional nanostructures (1D), Two-dimensional nanostructures (2D) and Three-dimensional nanostructures (3D). Zero-dimensional (0D) nanostructures have all three dimensions quantized to nanoscale and are often called quantum dots. In 1D nanostructures, only two dimensions are restricted to nanoscale (e.g. nanowires). In 2D nanostructures, one dimension is quantized to nanoregime (e.g. nanofilms) while 3D nanostructures do not have any quantization at all [12]. These are also known as bulk nanomaterials. Many studies and reviews are focussed on study of these 3D, 2D, 1D and 0D materials [13–19]. The literature [20–25] reports that apart from dimensionality control from 0D to 3D, properties of nanomaterials can also be controlled in terms of their pore size. They are microporous, mesoporous or may have macropores and can be tailored to modify their performance. They are used in rechargeable batteries, gas sensing, supercapacitors, solar cells, electro-/photocatalysis etc. Many studies have been focussed on the various properties and applications of nano-crystalline materials [26–47]. Some amorphous materials such as glasses and polymers have also contributed substantially to the area of nanotechnology [48–55] and have proved themselves promising in terms of applications. Amorphous alloys are structurally disordered with only short-range order in contrast to crystalline materials, which have long range order with repeating unit cell. Amorphous alloys do not have regular arrangement of atoms, i.e. periodicity, and hence also termed as non-crystalline. These amorphous or non-crystalline materials are sometimes referred as “glasses”. While “non-crystalline” is a generic term that describes a solid material having no crystallinity, a ‘glass’ is usually formed by continuous cooling from the liquid. Also, glass is characterized by the presence of a glass transition temperature, $T_g$. All non-crystalline materials have one feature in common; their structures are such that the sub-unit arrangements can get tangled so easily and so completely in liquid state that they are almost impossible to untangle them once the material is solidified [56–59]. Crystals are well known for their anisotropic nature, where properties in different directions differ considerably. But, amorphous materials particularly glasses have proved interesting due to their varied properties and are widely used as a storage media in optical recording for data storage devices. Glasses are mainly categorized as metallic glasses, chalcogenide glasses and oxide glasses. Metallic glasses are also referred as amorphous metals. These glasses have non-crystalline structure with disordered atomic arrangement. Metallic glasses were first produced by rapidly solidifying metallic melts at cooling rates of about $10^6$K/s [60]. These glasses were produced in different sizes and shapes (ribbons, wires, powders, etc.) by various techniques. Moreover, metallic glasses were found to have excellent mechanical, chemical, and magnetic properties. Some of these techniques have been described in [61–63]. Some of the metallic glasses are Ti–Cu, Ti–Be, Zr–Cu, Zr–Ni, Zr–Ni–Cu etc. Besides metallic glasses, chalcogenide glasses have gained much importance. These glasses contain one or more of any of the chalcogen atoms of group VI, Sulphur, Selenium and Tellurium. Since these glasses contain at least one chalcogen element, they have been have given a name ‘chalcogenide glasses’. Kolomiets [64] suggested that chalcogenide glasses behave as semiconductors and have band gap lying in the range 1 ~ 3 eV. Ioffe and Regel [65] justified the occurrence of band gap in amorphous glasses by the fact that the band gap in these glasses depends on the existence of short- range order of the lattice. Further, semiconducting behaviour of
these glasses is attributed to the presence of covalent bonds existing in these glasses. In chalcogenide glasses, the covalent bonded atoms are arranged with order extending up to the third or fourth nearest neighbors in an open network. Therefore, these glasses are also called network glasses. Examples of chalcogenide glasses are Se-Te, Se-Te-Bi, Se-Te-Sb, Se-Ge-Sb, Se-Ge-Sn etc. Third type of glasses is oxide glasses. These are the oldest glasses and are particularly based on silica. Oxides glasses generally show insulating behavior and possess large band gap ($\sim 10$ eV in case of SiO$_2$). Some of the examples of oxide glasses are Bi$_2$O$_3$–SiO$_2$, Bi$_2$O$_3$–B$_2$O$_3$, Bi$_2$O$_3$–P$_2$O$_5$, B$_2$O$_3$–Li$_2$O–ZnO–V$_2$O$_5$, Li$_2$Si$_2$O$_5$ etc.

The nanostructurization of glasses is extremely important as it will open new doors of applications with changes in electrical, optical, mechanical, thermal and other properties of these materials. But, fabrication of these glasses at nanoscale is not easy as atomic manipulation is meaningless in a disordered structure, whereas, in crystalline materials, fabrication of atomically flat surfaces can be done with ease by controlling atoms one by one. Moreover, over the crystalline materials, the nanostructurized glasses yield a greater variety of properties as the bonding constraint arising from crystalline unit cells does not exist in these glasses. In the past decade, many new developments have been done to study the properties of nanostructurized glasses. These glasses have completely different structure than their bulk counterparts. Furthermore, it has also been found that these nanostructurized glasses in some cases, show medium-range ordered structure rather than short range order. Therefore, it is evident that by controlling nanostructurization formation, properties of these glasses can be tuned as per requirement. In nanostructurized glasses, free volume also plays an important role. Free volume of nanoglasses is more than bulk quenched glasses. Free volume affects the structure and properties of these glasses and is also affected by temperatures or stresses. Nanoglasses have enhanced free volume, therefore, glass transition, plastic deformation, structure and properties of nanoglasses are greatly altered. These alterations have evolved entirely new interest in these nanostructurized glasses as new properties make these glasses potential candidate for many applications [66–75].

The nanoglasses are found to show two glassy nanostructures such as glassy core and glass-glass interface. Nano-glasses are solid materials composed of nanometer-sized crystalline or nanometer-sized glassy regions connected by crystal/crystal or glass/glass interfaces. As the atomic structures of these interfaces differ from the ones in the building blocks, the atomic and electronic structures of these interfaces differ from the ones in the building blocks. The properties of nanoglasses are different from the conventional glass of the same composition. This has grown interest in researchers to study and modify the properties of glass through nano-structurization. One such technique is plastic deformation, which facilitates the different atomic configurations other than the un-deformed glass at the nanoscale.

The present review article reports the new developments done in various properties of nanostructurized glasses and composites. Some of the recent research on optical, electrical, mechanical and thermal properties of these materials have been discussed.

2. Preparation of nanostructurized glasses

Many nanostructure materials have been synthesized for various applications in the past years. Several methods are [76–85] available in the literature for preparation of nanomaterials. Among these existing methods, inert-gas consolidation (IGC) method is one of the important methods for synthesis of nanoparticles [77]. This method was introduced by
Gleiter and coworkers [78]. In this method, the bulk source material is typically vaporized into a low-density gas under high vacuum of nearly 10^6 torr, by resistive heating. The vapors of heated sample migrate upwards where the substrate is placed. The substrate is cooled, generally by continuous flow of liquid nitrogen so that by combination of convection and diffusion, sample get deposited on cooled substrate. The cooling of substrate results into rapid decrease in the equilibrium vapor pressure leading to high supersaturation [79]. It is well known that at high supersaturation, the vapors rapidly nucleate, thereby forming very large numbers of extremely small particles lying in nanodimension. The growth of nuclei’s is assisted by Brownian coagulation [80]. The literature reports the synthesis of several types of nanomaterials using this method [51, 58, 66–70, 81, 82].

The nanostructured glasses are prepared mostly by inert gas condensation or physical vapor deposition method and thin films of the samples are obtained. Figure 1 shows the schematic diagram of thermal evaporator used for preparation of nanostructured thin films of glassy samples. This arrangement was used by Tripathi et al. [87] and Deepika et al. [88] for fabrication of Se-Te nanostructured thin films. Nanostructures were grown on thin films by pouring liquid nitrogen over the substrate during the film deposition. The arrangement in the vacuum chamber consisted of a molybdenum boat which carries source material and a pre-cleaned glass substrate, which was pasted using a double-sided tape on the circular steel plate located at a vertical distance of 30 cm from source material. The vacuum in the chamber was maintained at 10^{-5} torr. After attaining the desired vacuum, films were deposited by passing the LT current to the electrodes. During this process, simultaneously, liquid nitrogen was poured over the circular steel plate through a gas inlet. This was done to limit the growth of particles and achieve nanostructure in thin films.

Besides IGC method, nanoglass films have been recently synthesized by a variety of methods, such as magnetron sputtering [89–92], multiphase pulse electrodeposition [93], and the combination of electron beam and thermal evaporation [94].

Figure 1. Schematic diagram of thermal evaporator [86] (reprinted with permission from Springer Nature, Copyright 2018).
3. Properties

3.1. Optical properties

From atoms and molecules to crystals and bulk components, optical materials are naturally structured at different scales. The interaction of materials with optical waves and photons is strongly dependent on the structure, which can then be used to control light field distribution and light propagation. For semiconductor nanoparticles, electron confinement also induces a modulated absorption spectrum. The refractive index is then modified. The bandgap of the material is changed because of the discretization of the electron energy, which can be controlled by the nanometers size particles. The nanostructured glasses especially nano-chalcogenides show a dramatic change in their optical and electronic properties on reduction of size below the excitonic size [95,96]. Depending on the deposition technique used, the micro/nanostructure of thin films can be more or less complex. It is well known that low energy evaporation condensation techniques based on e-beam gun or on heated crucible give columnar thin films for most of the materials [97]. The refractive index of thin films of nanostructured glasses is generally smaller than the refractive index of the bulk material. This is attributed to the lacunar structure of these films [98]. In particular columnar thin films have a packing density that can be much smaller than one. The packing density is given by the ratio of the volume occupied by bulk material over the total volume of the layer [99,100].

3.1.1. Determination of optical properties

The optical study of nanostructured films is carried out using absorption, reflection and transmission spectra recorded on UV-Vis spectrophotometer at room temperature.

3.1.1.1. Calculation of refractive index and film thickness. Swanepoel [101] suggested a theory based on transmittance to obtain the various optical constants of the thin film sample. According to his theory, transmittance (T) is function of $\alpha$, $\lambda$, $n$, $n_s$ and film thickness $d$ and is given by

$$T = \frac{Ax}{B - Cx \cos \phi + Dx^2}$$

where

$A = 16n^2n_s$, $B = (n + 1)^2(n + n_s^2)$, $C = 2(n^2 - 1)(n^2 - n_s^2)$, $D = (n - 1)^2(n - n_s^2)$, $\phi = 4\pi nd/\lambda$, and $x$ is the absorbance given by $x = \exp(-\alpha d)$. 

Equation (1) is proposed for homogeneous thin film of thickness $d$ and complex refractive index $n_c = n - ik$, where $n$ is refractive index and $k$ is extinction coefficient. The substrate is considered to have thickness of several order of magnitude larger than $d$ with refractive index $n_s$. The refractive index of surrounding air is taken as 1. Taking all the multiple reflections at three interfaces into account, in case of $k^2 \ll n^2$, Equation (1) can be employed to thin film samples. The values of the transmission at the maxima and minima of the interference fringes can be obtained from Equation (1) by setting the interference condition $\cos \phi = +1$ for maxima ($T_M$) and $\cos \phi = -1$ for minima ($T_m$).

On the basis of Manifacier et al. [102] idea of creating upper and lower envelopes of the interference fringes, Swanepoel proposed the approximate value of refractive index ($n_1$) of the film in the spectral region of medium and weak absorption by the expression

$$n_1 = \left[ N + (N^2 - n_s^2)^{1/2} \right]^{1/2}$$

where,
$$N = \frac{2n_s(T_M-T_m)}{T_MT_m} + \frac{n_s^2 + 1}{2}$$  \hspace{1cm} (3)

$T_M$ and $T_m$ are the maximum and minimum values of the upper and lower envelopes at a certain wavelength and $n_s$ is the refractive index of the glass substrate. The refractive index of the glass substrate ($n_s$) has been calculated from the well known relation [103]

$$n_s = \frac{1}{T_s} + \left(\frac{1}{T_s^2} - 1\right)^{1/2}$$  \hspace{1cm} (4)

where $T_s$ denotes the transmittance spectra of the glass substrate.

If $n_a$ and $n_b$ are the refractive indices at two adjacent maxima (or minima) at wavelengths $\lambda_1$ and $\lambda_2$ respectively, then formula for film thickness is given by [104–106],

$$d_1 = \frac{\lambda_1\lambda_2}{2(\lambda_1 n_b - \lambda_2 n_a)}$$  \hspace{1cm} (5)

These values are denoted as $d_1$. The $d_1$ values are obtained at different wavelengths and average value is noted as $\overline{d_1}$. The above equation is derived from the basic equation of interference fringes,

$$2nd = m_o \lambda$$  \hspace{1cm} (6)

where the order number $m_o$ is an integer for maxima and a half integer for minima. The $\overline{d_1}$ and $n_1$ values are then used to calculate $m_o$ for different maxima and minima using Equation (6). The accurate value of film thickness ($d_2$) can now be calculated by taking the exact value of $m$ associated with each maxima or minima. The average value of $d_2$ is denoted as $\overline{d_2}$. Now, the value of $\overline{d_2}$ and $m$ can be used for each wavelength to obtain improved value of refractive index $n_2$.

### 3.1.1.2. Determination of optical band gap.

The absorption spectra can be used to determine the absorption coefficient ($\alpha$) using the relation [106]

$$\alpha = \frac{1}{d} \ln \left(\frac{1}{X}\right)$$  \hspace{1cm} (7)

Here, $d$ is the thickness of the film and $X$ is the absorbance. Absorption coefficient can also be employed to determine the nature and value of the energy band gap. The relation between the absorption coefficients ($\alpha$) and the incident photon energy ($h\nu$) is given by [107]:

$$(\alpha h\nu) = A (h\nu-E_g)^n$$  \hspace{1cm} (8)

where $A$ is a constant and $E_g$ is the energy band gap of the material and exponent $n$ depends on the type of transition. The values of $n$ for direct allowed, indirect allowed transition and direct forbidden, are given as $1/2$, $2$ and $3/2$ respectively. The energy band gap is calculated from the absorption spectra by plotting a graph between $(\alpha h\nu)^{1/n}$ and $h\nu$. The extrapolation of the straight line to $(\alpha h\nu)^{1/n} = 0$ axis gives the value of the energy band gap.

Considerable research has been done on the optical properties of nanostructured glasses in last decade. Khan et al. [70] studied optical properties of a-Se$_{70}$Te$_{30}$ nanorods. Their thin films contained nanorods of diameter 30 to 80 nm and length of orders of few hundreds of nanometers. They reported indirect band gap for the samples. Al-Agel [108] studied optical and structural phenomenon in as-deposited thin films composed of aligned
nanorods of amorphous $\text{Se}_x\text{Te}_{100-x}$ ($x = 3$, 6, 9, and 12) glasses. On the basis of optical absorption data, they observed a direct optical band gap in their samples. This observation of a direct optical band gap in these nanorods is interesting as chalcogenides normally show an indirect band gap, and due to this reason, these materials could not become very popular for semiconducting devices. Figure 2 shows FESEM images of $a$-$\text{Se}_x\text{Te}_{100-x}$ thin films. It is evident from these images that $\text{Se}_x\text{Te}_{100-x}$ thin films contain high yield of short and perfectly aligned nanorods. The diameter of these nanorods was reported between 10 and 20 nm, whereas the length is in the order of several hundred nanometers. Figure 3 shows the plot showing the variation of $(\alpha h\nu)^2$ with photon energy ($h\nu$) for $a$-$\text{Se}_x\text{Te}_{100-x}$ films composed of aligned nanorods. Their studies reveal that due to this size reduction, the optical properties show a dramatic change and the optical band gap becomes direct. The values of optical constants (refractive index and extinction coefficient) are also enhanced significantly as compared to results from their previous report [109]. This enhancement in the value of optical band gap and optical constants are attributed to the phenomena of size effect. Salah et al. [110] studied the optical properties of thin films of $\text{Te}_9\text{Se}_6$ nanoparticles synthesized using physical vapor condensation technique at different Ar pressures. SEM images (Figure 4) display uniform nanoparticles in these films where the size increases from ~12 to about 60 nm by decreasing Ar pressure from 667 to 267 Pa. These as-grown thin films were found to have direct band gaps, whose value decreases with increasing particle size. Argon pressure was found to be effective for controlling the particle size. The sizes of the grown nanoparticles were

Figure 2. FESEM images of thin films composed of $a$-$\text{Se}_3\text{Te}_{97-x}$ aligned nanorods [108] (reprinted with permission from Springer Nature, Copyright 2013).
significantly reduced by increasing Ar pressure. These results might be useful for development of optical disks and other semiconducting devices based on these controlled direct band gap. Alvi [111] studied the optical properties of Se77Sb15Ge8 nano-structured thin films. The average particle size lies in the range 30–60 nm. The detailed examination of the data of optical absorption points out that the optical transition is indirect in nature. The optical band gap is viewed around 1.53 eV. In their another study [112], they studied the optical properties of Bi doped Se-Te nanostructured thin films. The band gap of the samples was indirect and was found to increase on nanostructure formation. This was due to decrease in disorderliness and defects in the system. Further, they reported that large refractive index and small extinction coefficient of these films makes them suitable for antireflection multi layers.

Authors have also carried out considerable work on nanostructured glasses. They have studied the effect of nanorods formation on optical properties of Se-Te-Bi films [113]. For
nanostructured Se-Te-Bi system, the band gap values were found to be low as compared to bulk samples [114]. Their results [113] show that energy band gap decreases on increase in Bi concentration in Se-Te-Bi nanostructured system. This was attributed to the presence of large number of unsaturated bonds and structural defects in the system. The higher concentration of defect states increases the number of localized states which eventually narrows the optical band gap. Figure 5 shows transmission spectra of thin films of Se$_{80-x}$Te$_{20}$Bi$_x$ (0 $\leq x \leq 12$) containing nanorods and Figure 6 shows the plot of ($\alpha$hv)$^{1/2}$ versus hv for thin films of Se$_{80-x}$Te$_{20}$Bi$_x$ (0 $\leq x \leq 12$) glasses. The band gap was found to be indirect and lower than corresponding bulk sample of nearly same composition. The other studies of authors in this field include optical characterization of nanostructured Ge$_{1-x}$Sn$_x$Se$_{2.5}$ ($x = 0, 0.3, 0.5$) films [86], optical properties of nanostructured Se$_{58}$Ge$_{39}$Pb$_3$ and Se$_{58}$Ge$_{36}$Pb$_6$ thin films [115] and structural and optical study of nanostructured Se$_{80-x}$Te$_{20}$Sb$_x$ (0 $\leq x \leq 12$) thin films [116].

The most recent studies in optical properties were carried out on the plasmon field enhanced light-induced changes in amorphous Ge$_{24}$Se$_{76}$ layer incorporating Au nanoparticles [117]. The presence of gold nanoobjects in amorphous chalcogenide/gold heterostructure influences the optical and structural properties, as well as volume changes of the material. Their results show that superposition of the localized plasmon field of the nanoparticles with the electromagnetic field of incident photons, during the irradiation enhances the light-induced transformations. In another study, a new approach for nano-structuring of glassy Selenium (g-Se) using silver nanoparticles (AgNPs) as precursor has been discussed by Amit Kumar et al. [118]. They synthesized the nano-structured g-Se with the help of silver nanoparticles. It was found that the addition of AgNPs in glassy Se creates fine nano-wires of trigonal Selenium (t-Se) in the glass matrix of g-Se. Further, they have studied electrical, mechanical, and thermal properties of the Ag doped glassy nanocomposite material using current-voltage characteristics, d.c. conductivity, micro-hardness, and kinetic parameters of glass/crystal phase transformation. Their calorimetric results depict that the glass-forming ability and thermal stability of g-Se/AgNPs nanocomposite is greater than Se$_{98}$Ag$_2$ glass. Further, the nanocomposite sample was found to be harder than the Se$_{98}$Ag$_2$ alloy which is confirmed by the values of their Vickers hardness.
number. Furthermore, their results show that d.c. conductivity of g-Se/AgNPs nanocomposite sample is increased appreciably as compared to g-Se98Ag2 sample.

Besides nanostructured chalcogenide glasses discussed above, some studies are also reported on optical properties of nanostructured metallic and oxide glasses. Ning Wang et al. [119] prepared the 3D nanostructure rutile TiO2 photocatalyst by dealloying method using Cu60Ti30Y10 amorphous ribbons as precursors. Their morphological images show that the synthesized rutile TiO2 nano-material presents homogeneous distributed 3D nano-flowers structure, which is composed of large quantities of fine rice-like nanorods (40–150 nm in diameter and 100–250 nm in length). For the sake of convenience, Cu60Ti30Y10, Cu60Ti30Al10, Cu70Ti30 and Cu60Ti30Sn10 amorphous ribbons are labelled as the CTY, CTA, CT and CTS sample, respectively, while Cu60Ti30Y10 amorphous ribbons subjected to immersion in 14.4 M HNO3 aqueous solution at 343 K for 3, 24 and 72 h are denoted as the CTY-3, CTY-24 and CTY-72 sample, respectively. Besides, the sample prepared by dealloying Cu70Ti30 in 14.4 M HNO3 solution at 343 K for 72 h is denoted as the CT-72 sample. Figure 7 shows the SEM images of the samples: (a) CTY-3; (b) CTY-24; (c) CTY-72; (d) CT-72. The photocatalytic activities of the samples were evaluated by degrading rhodamine B (RhB) dye (10 mg/L) under the irradiation of both simulated visible light (>420 nm) and ultraviolet (UV) light (=365 nm). The results of their study show that the photocatalytic activity of rutile TiO2 prepared by dealloying Cu60Ti30Y10 amorphous ribbons is higher than those of commercial rutile and the sample synthesized by dealloying Cu70Ti30 precursors. Min Zhou et al. [120] fabricated nanoporous structures through chemical dealloying from quaternary Zr65Cu17.5Fe10Al7.5 amorphous ribbons. A three-dimensional bi-continuous nanoporous copper (NPC) structure was formed on Zr65Cu17.5Fe10Al7.5 ribbon alloys having the pore sizes of 44.91 nm, 52.60 nm and 57.37 nm, respectively, after immersion in 0.02 M HF solution for 15 h, 20 h, and 25 h. The results show that the pore size of NPC structure increases with increasing the dealloying time. Figure 8 displays the nanoporous structure of Zr65Cu17.5Fe10Al7.5 amorphous
Figure 7. SEM images of the samples: (a) CTY-3; (b) CTY-24; (c) CTY-72; (d) CT-72 [119] (reprinted with permission from Elsevier, Copyright 2018).

Figure 8. FESEM morphologies of Zr$_{65}$Cu$_{17.5}$Fe$_{10}$Al$_{7.5}$ amorphous ribbons after dealloying in (a) 0.02 M, (b) 0.035 M, (c) 0.05 M HF solutions for 5 h [120] (reprinted with permission from Elsevier, Copyright 2017).
Abhiram Jagannathan et al. [121] studied nonlinear optical properties of gold nanoparticles doped fluoroborate glasses for optical limiting applications. High resolution-transmission electron microscopic (HR-TEM) measurements further confirmed the existence of spherical and ellipsoidal Au NPs (Figure 9). They reported that optical bandgap values were red shifted for samples doped with gold nanoparticles and having more concentration of lithium content. These sample show better nonlinear refractive index and nonlinear absorption coefficient. Weldegrum et al. [122] studied hybrid ZnO nanowires (ZnO-NWs)/metallic nanotube arrays (MeNTA) structure. MeNTA was formed using sputter-deposition of five different metallic glasses. Optical properties were studied and results confirm the formation of nanowires having the hexagonal wurtzite structure. Also, they show strong UV emissions, which can be used in optoelectronics, sensors, piezoelectric-nanogenerator, solar cells and battery applications.

3.2. Electrical properties

Electrical properties of the materials are important for selection of material for a device. The electrical properties of the nanostructured glasses and composites have led to several applications in electronic and opto-electronic devices. In contrast to crystalline solids, the theory of electronic band structure and the mechanism of electron transport in semiconducting glasses are not well understood. It is generally agreed, however, that the band structure in glasses is not drastically different from that of analogous ordered structures and that the forbidden gap may persist [123]. The major change on transitions from a crystalline to disordered structure is the introduction of ‘tails’ of localized state in the forbidden gap [124]. Two types of conduction processes are possible in amorphous solids. An electron in a localized state can make a transition directly to a neighbouring localized state. At each transition energy is exchanged with the lattice through interaction with phonons; (hopping conduction). On the other hand, an electron (or hole) can be excited to the conduction band (or valence band) where it can drift in the same way as in crystalline semiconductors and be subjected to the same type of scattering and trapping mechanisms. In addition, the drift mobility will be affected by the disappearance of periodicity and because of the high density of localized state the mobility may become severely trap

Figure 9. HRTEM image confirming uniform distribution of Au NP’s in Glass (a)HRTEM image of 2 nm Au NP’s depicts the inter planar distance of Au [121] (reprinted with permission from Elsevier, Copyright 2020).
limited. The section below discusses the basic methods to study the electrical properties of nanostructured glasses.

3.2.1. Determination of electrical properties

Electrical properties of nanostructured glasses largely depend on defects, grain size, density distributions and doped impurities. It is reported that nanostructures of glasses comprise of nanometer-sized glassy clusters connected by glass-glass interface of reduced density, reduced nearest-neighbour atoms and different electronic structure. These changes in atomic and electronic structures lead to entirely different electrical properties of the system compared to bulk glasses of the same composition [125]. These interesting properties have led their applications in nanoelectronic devices, optical memory devices, fabrication of nanomemories etc. [126–128].

3.2.1.1. Calculation of dc electrical conductivity. To determine the dc electrical conductivity, the I-V curves of the nanostructured samples are recorded on Keithley electrometer and are linearly fitted. The slope of the curves is used to evaluate the dc electrical conductivity using the following relation [129,130]:

$$\sigma_{dc} = \frac{L}{RA}$$

where, R denotes resistance, L and A are thickness and cross-sectional area of the sample respectively. A graph is plotted between 1000/T versus ln $\sigma_{dc}$ for the samples. The plots are fitted to straight line and slope of the straight line gives activation energy of conduction ($\Delta E$) while the intercept gives pre-exponential factor ($\sigma_o$). The dc electrical conductivity is dependent on temperature through the Arrhenius equation given by [131,132]

$$\sigma_{dc} = \sigma_o \exp \left[ \frac{-\Delta E}{KT} \right]$$

where, $\Delta E$ is activation energy of conduction, $\sigma_o$ represents the pre-exponential factor and K is Boltzmann constant.

In the low temperature region (303–343 K), dependence of electrical conductivity on temperature could be studied using Mott’s three-dimensional variable range hopping model (3D VRH) given by [130]

$$\sigma = \sigma_o \exp \left[ \frac{-B}{T^{3/4}} \right]$$

The slope of the plot gives the value of B and intercept $\sigma_o$ is pre-exponential factor. The value of B [130] is equivalent to

$$B^4 = T_o \frac{\lambda \sigma^5}{kN(E_f)}$$

Here, $\sigma^{-1}$ denotes spatial extent of the wave function $\exp(-\sigma R)$ associated with the localized states, $\lambda$ is a dimensionless constant (about 18), N(E_f) is the density of localized states near the fermi level, T_o is degree of disorder and k is Boltzmann constant.

The value of pre-exponential factor is given by [131,132]

$$\sigma_o' = 3e^2\gamma \left( \frac{N(E_f)}{8\pi\alpha kT} \right)^{1/2}$$

where $\gamma$ is Debye frequency (about $10^{13}$ Hz) [133] and e is the electronic charge.
3.2.1.2. Solving simultaneously Equations (3) and (4) gives.

\[
\alpha = 22.52\sigma_0 B^2 \text{cm}^{-1}
\] (13)

\[
N(E_f) = 2.12 \times 10^9 \left(\sigma_0\right) \text{cm}^{-3}\text{eV}^{-1}
\] (14)

The values of \(N(E_f)\) indicate that number of states available near the fermi level for conduction. The values of \(\alpha\) and \(N(E_f)\) can be used to determine hopping range (R) and hopping energy (W) using the following relations [131,132]

\[
R = \left[\frac{9}{8\pi\alpha N(E_f) kT}\right]^{1/4}
\] (15)

\[
W = \frac{3}{4\pi R^3 N(E_f)}
\] (16)

The values of R and W are also helpful in ascertaining the conduction mechanism taking place in glassy systems.

Some of the studies carried out on electrical properties of nanostructured glasses are discussed. Alvi [111] studied the electrical properties of Se\(_{77}\)Sb\(_{15}\)Ge\(_8\) nano-structured thin films. The dc activation energy (\(\sigma_{dc}\)) in nano-structured Se\(_{77}\)Sb\(_{15}\)Ge\(_8\) thin films was calculated from the temperature dependence and was found to be 1.23 eV. Moreover, \(\sigma_{dc}\) increases exponentially with temperature, which demonstrates that the conduction in these glassy alloys takes in the course of an activated procedure and also indicates the semiconducting nature of these alloys. Figure 10 displays the temperature dependence of \(\sigma_{dc}\) of nano-structured Se\(_{77}\)Sb\(_{15}\)Ge\(_8\) thin film. Figure clearly indicates that with increasing temperature, \(\sigma_{dc}\) increases exponentially with temperature. Mohd. Nasir and M. Zulfequar [134] have studied space charge limited conduction (SCLC) on the fabricated thin films of Se\(_{95-x}\)S\(_x\)Zn\(_5\) (0.2 \(\leq x \leq 10\)) in temperature range 313–353 K. They calculated the dc conductivity and activation energy and it was found to decrease and increase respectively, with the increase of sulfur concentration. They observed an ohmic and non-ohmic behaviour both in the low field (\(\leq 10^3\) V/cm) and a non-ohmic behaviour is observed due to predominant charge injection from the electrode gap to the samples at high field (\(\sim 10^4\) V/cm). Further, they reported that dc conductivity decreases due to
increase of nanocrystalline phase in Se$_{95}$S$_x$Zn$_{5}$ alloys. Khan et al. [66] studied the effect of composition on electrical properties of thin films of amorphous Ga$_x$Se$_{100-x}$ nanorods. TEM image of these nanorods is presented in Figure 11. It is clear from the image that the diameter of the nanorods varies from 140 to 180 nm, and the length is of several hundreds of nanometers. DC electrical conductivity of deposited thin films of a-Ga$_x$Se$_{100-x}$ nanorods is measured as a function of temperature range from 298 to 383 K. An exponential increase in the dc conductivity is observed with the increase in temperature, suggesting thereby a semiconducting behavior. Figure 12 shows the temperature dependence of dc conductivity in the temperature range (298–383 K) at various concentration of Ga of thin films of a-Ga$_x$Se$_{100-x}$ nanorods. The estimated value of activation energy decreases on incorporation of dopant (Ga) content in the Se system. The calculated value of pre-exponential factor ($\sigma_0$) is of the order of $10^{-1}$ cm$^{-1}$, which suggests that the conduction takes place in the band tails of localized states. It is suggested that the conduction is due to thermally assisted tunnelling of the carriers in the localized states near the band edges. In another communication, Khan et al. [135] studied electrical transport properties of films
of Se$_{87}$Te$_{13}$ nanorods. The dc conductivity for these nanorods as studied as a function of over a temperature range of 500–100 K. Both thermally activated process and variable range hopping mechanism were reported responsible for electrical transport process.

The authors have studied the electrical conduction mechanism in several nanostructured glasses. Electrical properties of films of Se$_{80-x}$Te$_2$Bi$_x$ (0 ≤ $x$ ≤ 12) glassy alloys [131] has been studied. The TEM results (Figure 13) indicate the formation of nanorods within the films. The electrical conductivity of the samples was studied using Keithley electrometer in the temperature range 303–373 K. Figure 14 shows the variation of dc electrical conductivity with temperature for Se$_{80-x}$Te$_2$Bi$_x$ (0 ≤ $x$ ≤ 12) films. The results show that conduction in these samples takes place via thermally assisted tunnelling and variable range hopping of charge carriers corresponding to higher and lower temperature ranges respectively. Figure 15 shows the plot of ln $\sigma$/T with T$^{-1/4}$ for Se$_{80-x}$Te$_2$Bi$_x$ (0 ≤ $x$ ≤ 12) films in the temperature range 303–343 K.

It can be noticed that conductivity increases sluggishly with temperature, which suggest that conduction takes place via variable range hopping (VRH). Authors have also studied the electrical properties of Se$_{80-x}$Te$_2$Sb$_x$ (0 ≤ $x$ ≤ 12) nanorods as a function of temperature and composition [132]. Two conduction mechanisms were observed in the Se$_{80-x}$Te$_2$Sb$_x$ (0 ≤ $x$ ≤ 12) system. In higher temperature range (343–373 K), conduction takes place through thermally assisted tunnelling of charge carriers while in the temperature

**Figure 13.** TEM images of nanostructured thin films of Se$_{80-x}$Te$_2$Bi$_x$ (0 ≤ $x$ ≤ 12) glassy alloys. (a) Se$_{80}$Te$_{20}$ (b) Se$_{77}$Te$_{20}$Bi$_{3}$ (c) Se$_{74}$Te$_{20}$Bi$_{6}$ (d) Se$_{71}$Te$_{20}$Bi$_{9}$ (e) Se$_{68}$Te$_{20}$Bi$_{12}$ [132] (reprinted with permission from Canadian Science Publishing, Copyright 2019).
range 303–343 K, variable range hopping of charge carriers contributes to conduction. Moreover, it was found that nanostructure formation augments the electrical conductivity by order of $10^4$ as compared to its bulk counterparts.

### 3.3. Mechanical properties

Mechanical properties of a material are largely dependent on the structural arrangement of atoms and nature of bonds between them. The addition of impurities and presence of defects affect the mechanical properties of a material. Some of the mechanical properties of solids are elasticity and plasticity, stress, strain and elastic moduli. Mechanical properties of solids elaborate the characteristics such as the resistance to deformation and their strength. These properties play an important role in determining potency of solids in various applications [136–142].

The development of nanostructured materials is now raising the question on change of different properties as the microstructural scale is reduced to nanometer dimensions. Among potential applications of nanostructured materials, the design to achieve optimum mechanical properties is a common concern. The mechanical strength ($\sigma$) of crystalline
materials is believed to be largely controlled by the grain size \( (d) \), and is generally described by the Hall-Petch relationship \([143]\)

\[
\sigma = k d^{-1/2} + \sigma_0 \quad (17)
\]

As the structural scale reduces to the nanometer range, new mechanisms may come into play due to small dimensions. In addition, the intrinsically high interface to-volume ratio of the nanostructured materials may enhance interface-driven processes to extend the strain to failure and plasticity. The mechanical properties of nanoparticles allow their novel applications in many fields, e.g. surface engineering, tribology and nanomanufacturing/nanofabrication. Understanding some basic mechanical properties of nanoparticles, such as the hardness and the elastic modulus, will aid a lot in the proper design of particles in specific applications, as well as evaluating their roles and action mechanisms \([144–146]\). It has been pointed out that \([147–149]\) many microstructural features like grain size and shape, their distribution, pores and their distribution, other flaws/defects and their distribution, surface condition, impurity level, second phases/dopants, stress, duration of its application and temperature affect the mechanical properties of nanocrystalline \((n-)\) materials. For crystalline metal nanoparticles, dislocations inside the particles have been demonstrated as one of the factors contributing to the change in the mechanical behaviour of nanoparticles, which is in contrast to the traditional view that no dislocation is present in crystalline nanoparticles. Apart from dislocations or defects, the changes of the lattice strain and the bond energies of nanoparticles to the compressive stress are another cause for the strengthening and weakening of the mechanical properties of nanoparticles \([147–149]\). Very few studies have been carried out on mechanical properties of nanostructured glasses. Vella et al. \([150]\) studied the mechanical properties of nanostructured amorphous CuNb, FeB, and FeTi metal multilayer thin films. The hardness of amorphous metal multilayered films was investigated by nanoindentation. Bilayer material systems of amorphous CuNb, FeB, and FeTi were produced by dc sputtering on sapphire substrates to a total thickness of 1 µm. The bilayer periods \((A)\) ranged from 2 to 50 nm. The hardness behaviour is in contrast with the behaviour of crystalline multilayered films, which generally display significant enhancements as the bilayer period is decreased below 10 nm. The lack of a significant hardness variation in the amorphous films strongly suggests that dislocation-mediated mechanisms do not govern inhomogeneous flow in amorphous metals. Xuning Xue et al. \([151]\) studied the microhardness of chalcogenide glasses (ChGs) and glass–ceramics of the Sn-Sb-Se Ternary System. The mechanical property of ChGs was investigated in terms of microhardness \((HV)\), and the relationship between HV and mean coordination number \((MCN)\) (related to the chemical order of the ChG network) was studied. The microhardness of ChGs of the Sn-Sb-Se \((SSS)\) ternary system was investigated. Microhardness rapidly increased as the MCN of the SSS ChGs approached 2.4, signifying the transition of the SSS glass network from under-cross-linked floppy to over-constrained rigid state. Kancherla et al. \([152]\) studied the mechanical properties of glass fabric composite with surfactant treated zirconia nanoparticles. The optimal dispersion of yttria stabilized zirconia \((YSZ)\) nanoparticles in matrix-fabric system was carried out. The study reports that the dispersion of as low as 3 wt% of YSZ is found to be optimal for the developed processing condition. The enhancement is due to two-fold effects, one is the nano-cluster segregation by surfactant and the other one is an optimal interfacial stress/fracture modification in the matrix-fabric system via optimal length-scale effects of nano-composite with respect to the fabric interface. Saida et al. \([153]\) studied the nanostructured materials based on Zr-Al-Ni-Cu metallic glasses. They found that the nanoscale structure control in metallic glasses exhibits
unique mechanical properties such as a high strength and good ductility. They investigated the transformation from the glassy state to the nanoscale primary phase in the $\text{Zr}_{65}\text{Al}_{17.5}\text{Ni}_{10}\text{Cu}_{17.5-x}\text{Pd}_x$ ($x = 0, 5, 10$ and $17.5$) metallic glasses. The nucleation and grain growth kinetics were examined and it was found that it was easy to control the nanostructure in the Zr-based BMGs by the Pd addition and it was also possible to improve the mechanical properties by the nanoparticle precipitation. Chen et al. [89] studied formation and properties of Au-based nanograined metallic glasses. Gold-based nanograin glass (NGMG) exhibits a heterogeneous granular structure but possesses a glassy nature. The smallest particle size of the NGMG can be less than $10 \text{ nm}$. NGMG sustains the good mechanical properties of metallic glasses, showing a high hardness of $\sim 5.3 \text{ GPa}$. In addition to the nanostructure-induced catalytic functionality, this material sustains the good mechanical properties of a metallic glassy structure, showing a high hardness and a low elastic modulus. Yvonne Ritter et al. [154] studied structure, stability and mechanical properties of internal interfaces in Cu$_{64}$Zr$_{36}$ nanoglasses by MD simulations. Their results provide evidence for the existence of glass–glass interfaces, which are characterized by a defective short-range order and an excess free volume of about $1–2\%$. Jian et al. [155] studied the strength, hardness and ductility of Cu$_{64}$Zr$_{36}$ nanoglasses via embedded nanocrystals. They reported that with an increasing fraction of nanocrystalline grains, the tensile strength of the composite is enhanced, while its ductility decreases. Further, they inferred that for an appropriate fraction of nanocrystalline grains, a good balance among strength, hardness and ductility can be realized, which is useful for the synthesis of novel nanograin glass/crystalline composites with high strength, high hardness and superior ductility.

### 3.4. Thermal properties

Thermal properties play an important role in determining the application of material [156–159]. Thermal properties include knowledge of flow of heat from one place to another. Several methods have been studied for heat flow in materials [160–163]. For heat flow in crystalline nanomaterials, energy carriers are mainly crystal vibrations or phonons. These phonons travel through lattice vibrations and have characteristic lengths of the order of nanometers. The thermal properties of material are characterized by thermal conductivity ($k$), specific heat ($c_p$) and thermal diffusivity ($\alpha$). These properties are greatly affected by intrinsic electron–phonon and phonon–phonon scatterings, grain boundaries, surface interactions, grain size and temperature [164]. In nanostructured glasses, motion of phonons is hindered due to disordered structure. The primary means of thermal transport in these glasses are disordered diffusion between high-energy localized vibrational modes (LVMs). The thermal conductivity ($k$) of nanostructured glasses mainly depends on disorderliness and temperature of material [165]. In crystalline nanomaterials, both electrons and phonons play vital role in transfer of heat energy while in amorphous nanostructured glasses, phonons are primary energy carriers responsible for thermal transport [165]. Moreover, in nanostructured materials, the mean free path (MFP) of phonons and the wave vectors has dimensions of nanoscale [166]. Therefore, MFP distribution of phonons in nanostructured glasses play an important key to understand the thermal transport characteristics of phonons as carriers of thermal energy. But, thermal conductivity is significantly reduced compared to bulk material, due to scattering of nanomaterials at the interface.

For amorphous nanomaterials like nanostructured glasses, the dominant factor affecting thermal conductivity is the degrees of disorder ($\epsilon$). Degrees of disorder ($\epsilon$) is a strong
indicator of the thermal transport properties of materials. A very low value of $\varepsilon$ indicates an ordered structure and has extremely high value of thermal conductivity ($k$) up to 5300 W/m K [165] whereas, a high $\varepsilon$ correspond to disordered structure. For such a material, the $k$ is close to that of an insulator typically below 1 W/m K [165]. However, if the $\varepsilon$ is quite high then, the phonon velocity and polarization direction inside the structure is not useful to evaluate the thermophysical properties. Moreover, it is found that the heat conduction mainly depends on both phonons and diffusions for value of $\varepsilon$ lying between 0–1. Therefore, for amorphous nanostructures such as nanoglasses having higher degree of disorder, the thermal conductivity ($k$) is said to consists of two parts [166]:

$$k = k_P + k_D$$  \hspace{2cm} (18)

where $k_P$ denotes the contribution by phonons from propagating modes, and $k_D$ represents the contribution by diffusions from nonpropagating modes given by the Allen–Feldman (AF) theory [167,168]. The contributions due to phonons ($k_p$) has been obtained using the specific heat ($c_p$) in different modes and vibrational density of states (VDOS) of phonons [166]. For calculation of $k_p$, it was assumed that the system is isotropic and has a single polarization, so that mode properties are only related to $\omega$ [167,168]:

$$k_p = \frac{1}{V_s} \int_{0}^{\omega_{\text{cut}}} D_\omega C(\omega) \alpha(\omega) d\omega$$  \hspace{2cm} (19)

where $V_s$ is the volume of system, $\omega_{\text{cut}}$ denotes the cutoff frequency of phonon, $D(\omega)$ is vibrational density of states, $C(\omega)$ is mode specific heat capacity and $\alpha(\omega) = \frac{s^2}{3} \tau(\omega)/3$ is mode thermal diffusivity of the vibration mode related to $\omega$. Phonon lifetime is represented by $\tau(\omega)$ and $s$ is the sound speed. The phonons contribute to transport when the system frequency is less than $\omega_{\text{cut}}$, while diffusions contribute at higher frequencies. For phonons, the density of states (DOS) of the material can be evaluated using the Debye model [166]:

$$D(\omega) = \frac{3V \omega^2}{2\pi^2 S^3}$$  \hspace{2cm} (20)

The quantum expression for the $C(\omega)$ is given by [166]:

$$C(\omega) = k_B \left[ \frac{\hbar \omega / 2k_B T}{\sinh(\hbar \omega /2k_B T)} \right]^2$$  \hspace{2cm} (21)

where $k_B$ denotes the Boltzmann constant. Similarly, the contribution of diffusions to k can be obtained from the C in different modes and $\alpha$ [167,168]:

$$k_D = \frac{1}{V} \sum_{\omega > \omega_{\text{cut}}} C(\omega_i) \alpha(\omega_i)$$  \hspace{2cm} (22)

where vibration frequency of the $i^{th}$ localized mode is $\omega_i$, $C(\omega_i)$ denotes the diffusion isobaric specific heat capacity, and $\alpha(\omega_i)$ is the diffusion diffusivity of the $i^{th}$ localized mode. The AF theory [167] can be employed to determine the thermal diffusivity ($\alpha$) of the localized mode using the relation

$$\alpha(\omega_i) = \frac{\pi V^2}{\hbar^2 \omega_i^2} \sum_{j \neq i} |M_{ij}|^2 \delta(\omega_i - \omega_j)$$  \hspace{2cm} (23)
where $M_{ij}$ is the thermal coupling between $i^{th}$ modes and $j^{th}$ modes, which can be calculated by the modal frequencies and the spatial overlap of mode vectors.

The determination of thermal conductivity ($k$) depends on many factors. Some of the factors are discussed below.

i. **Effect of grain size:** The effect of grain size ($L$) is not negligible in nanostructured glasses. Grain size determines the grain boundary scattering rate, increasing grain size helps in reducing the scattering [168]. Less scattering results in high thermal conductivity.

ii. **Effect of grain boundary:** By definition, the grain boundary is the interface between the grains with the same structure and orientation. Generally, nanocrystalline (NC) materials are comprised of grains having size in nanoregime (1–10 nm). These grains cover more than 50% of NC materials and the average grain size of grain boundaries is found to be inversely proportional to fractional grain size [169]. The small grain size and large specific surface area brings significant change in thermo-physical properties of NC materials as compared to bulk crystals. The effect of phonon grain boundary scattering on thermal properties of nanostructured materials has been studied widely to study thermoelectric applications of these materials. The grain boundaries present in NC materials can effectively scatter short-wave phonons, thereby reducing the lattice thermal conductivity of the material in the low temperature region. It has also been reported [170] that thermal conductivity due to grain boundary scattering is an index of the cohesive energy of the grain boundaries.

iii. **Effect of temperature:** Temperature plays an important role in determining thermal conductivity of nanostructure materials as temperature has direct effect on thermo-physical properties especially at high temperatures. The study on effect of temperature reveals that thermal conductivity changes with change in temperature and potential application of nanostructure materials can be adjusted by controlling temperature of system.

iv. **Effect of defects:** Defects are unavoidable in a material. Defects can be in any form like vacancies, impurity atoms, or rough edges. The phonon zero acoustic mode plays a leading role in thermal transport at low temperatures. Further, the stronger the zero acoustic mode scattering, the smaller is the effect of defects on thermal conductivity ($k$). This is consequence of additional phonon scattering caused due to the localization of phonons near the defect. Therefore, controlling defects and their density together with temperature affect the thermal conductivity of nanostructured materials [171,172].

Beside thermal conductivity, thermal diffusivity ($\alpha$) is another important parameter to study thermal property of a material. Thermal diffusivity ($\alpha$) refers to rate of heat transfer in a material. Thermal diffusivity is independent of the amount of energy during heat conduction. It is the rate of diffusion from the hot end to the cold end in the material, and has a transient value. Thermal conductivity ($k$) and thermal diffusivity ($\alpha$) are interconnected and the $k$ of the material can be determined by measuring the $\alpha$ and vice versa. The relation between $k$, density ($\rho$) and specific heat ($c_p$) is given by [163]:

$$\alpha = \frac{k}{\rho \cdot c_p}$$

(24)

From above relation, it is evident that the higher value of $\alpha$ indicate stronger ability of the medium to balance its internal temperature. Therefore, $\alpha$ is an indicator of the ability
of the medium to propagate energy through the material. In other words, \( \alpha \) describes the rate of heat transfer from heat source to heat sink. The specific heat capacity \( (c_p) \) mentioned in Equation (23) consist of phonon specific heat capacity and electron specific heat capacity. In some nanomaterials, \( c_p \) is primarily dominated by phonons whereas, in some other materials, electron specific heat capacity plays a major role. The relationship between time \( (t) \) and temperature \( (T) \) with thermal diffusivity \( (\alpha) \) is given by [163]:

\[
\frac{\partial T}{\partial t} = \alpha \Delta^2 T
\]  

Figure 16. Thermal conductivity vs average grain size for fully dense samples 550 (99%), 144 (99%), and 76 (99%) at three temperatures [174] (reprinted with permission from American Chemical Society, Copyright 2011).

Substantial work has been done on the thermal properties of nanocrystalline materials. However, work on nanostructured glasses is scanty and done in recent years. Dong et al. [173] proposed a theoretical model for describing effective thermal conductivity (ETC) of nanocrystalline materials on the basis of grain size, single crystal thermal conductivity, single crystal phonon mean free path (PMFP), and the Kaptiza thermal resistance. It has been demonstrated that with increasing grain size, both grain boundaries (GBs) and size effects become weaker, while size effects have stronger effect on thermal conductivity than GBs effects. Wang et al. [174] studied the thermal Conductivity of nanocrystalline silicon. The effect of grain size and temperature on thermal conductivity has been studied. Figure 16 shows \( k \) as a function of \( D_{\text{avg}} \) for the fully dense samples \( (\rho > 99\%) \) at three temperatures. At low \( T \), the power law is \( k \propto D^1 \), as expected for a system dominated by boundary scattering, while for higher \( T \) and larger grains the \( k(D) \) power law becomes weaker due to the increasing importance of impurity /defect scattering also called umklapp scattering. Soyez et al. [175] also corroborated that increasing the nanoparticle size is another effective way to boost \( k \). They studied grain-size-dependent thermal conductivity of nanocrystalline yttria-stabilized zirconia (YSZ) films grown by metal-organic chemical vapor deposition. They stated that reduced thermal conductivity in nanocrystalline coatings arises due to enhanced phonon scattering and presence of numerous closely spaced grain boundaries. Figure 17 shows the thermal conductivity for both a 13 nm grain-sized YSZ film and an alumina substrate as a function of temperature. The results indicate that over the narrow measured temperature range, the YSZ thermal conductivity
is temperature independent, while the Al$_2$O$_3$ thermal conductivity decreases significantly with increasing temperature. Further, they reported that significant reduction in thermal conductivity in these films compared to that of bulk YSZ is observed for grain sizes smaller than the phonon mean-free path. The authors also reported presence of porosity in the samples, however, the amount of porosity was not observed to vary significantly with grain size. Their results conclude that the observed changes in thermal conductivity are more likely attributable to the effects of grain size than to porosity. Mishra and Kumar [176] have studied the thermal transport properties of Se$_{95-x}$Sn$_5$Bi$_x$ ($x = 0, 4, 8$) chalcogenide alloys. Transmission electron microscopy (TEM) demonstrates the formation of nano-rods and clusters of nano-particles. It was observed that thermal conductivity ($\lambda$) shows a decreasing trend with the inclusion of Bi in Se–Sn system. Thermal diffusivity ($\chi_e$) and volumetric heat capacity ($Q_{Cp}$) show the optimum value for $x = 4$. Thermal transport parameters remain almost temperature independent. They stated that nano structuring leads to enhanced phonon boundary scattering which reduces the thermal conductivity. Also, the combined effect of increased boundary phonon scattering and scattering from defects, contributes to the further reduction of thermal conductivity. Figure 18 shows the variation of effective thermal conductivity with temperature for different composition of Bi. The authors have also calculated the phonon mean free path ($\tau$) using the relation:

$$\tau = \frac{3}{\chi} / v_s$$

(26)

where $\chi$ is thermal diffusivity and $v_s$ is velocity of sound (mean speed of carriers in a material), in case of chalcogenide glasses which is $\approx - 2000$ m/s [177]. Anupama Viswanathan et al. [178] studied the concentration dependent thermal diffusivity of Ge$_{25}$Se$_{65}$Sb$_{10}$ and Ge$_{25}$Se$_{60}$Sb$_{10}$Ag$_{5}$ chalcogenide glass nanocolloids. Nanocolloids of five different concentrations from each composition were prepared by dissolving finely powdered bulk glasses in organic solvent ethylenediamine (EDA). Enhancement in thermal diffusivity was observed with increase in solute concentration and was attributed to increased number of localised defects states, phonon scattering at particle liquid interface and Brownian motion of the particles. Maryam Mohri et al. [92] investigated the effect of deposition conditions on the microstructure and thermal stability of Ti-Zr-Cu-Pd nanoglass thin films. Nano-grained metallic glass Ti-Zr-Cu-Pd thin films were deposited at room temperature on Si substrate by using direct current (DC) magnetron sputtering.
with an alloy target. Nanometer-sized glassy clusters were observed in the thin films deposited at higher Ar pressure. Average grain size was noted to decrease while the roughness of the thin films was increased. All of these observations seem to indicate that the sputtering pressure is the dominating factor controlling the microstructure and thermal stability of the sputtered films.

Now, most recently, machine learning and artificial intelligence has offered a promising path to leverage existing datasets and infer data-driven models that, in turn, can be used to accelerate the discovery of new materials [179–190]. In the field of glasses also, Han Liu et al. [191] have presented a review of machine learning applied to glass science. As a notable success, machine learning modelling techniques have been used to accelerate the design of Corning® Gorilla® glasses [192]. Therefore, in the coming years we can expect new modelling and simulation-based research in glass science.

4. Conclusions

A comprehensive review has been carried out on optical, electrical, mechanical and thermal properties of nanostructured glasses and composites. The easiest method to fabricate nanostructured glasses is inert gas consolidation (IGC) method. The optical properties of these glasses can be studied using absorption, transmission and reflection spectra. The analysis of data gives value of several optical parameters including refractive index, extinction coefficient and energy band gap. It has been observed that the nanostructured glasses show both direct and indirect band gap. The literature suggest that size reduction brings dramatic change in type of energy band gap. The band gap can be direct or indirect depending on nanostructurization of the sample. Moreover, the values of optical constants such as refractive index and extinction coefficient are also found to enhance significantly on size reduction. Some studied show increase while some report decrease in energy band gap on nanostructure formation in sample. The increase in band gap was due to decrease in disorderness and defects in the system while decrease is attributed to increased band tails and defects in system. The electrical properties of the nanostructured glasses in literature were studied using I-V characteristics at different temperatures. The studies show that two types of behaviour are being observed in these glasses. The literature shows that conduction in nanostructured glasses takes place via thermally assisted tunnelling and
variable range hopping of charge carriers corresponding to higher and lower temperature ranges respectively. Moreover, it was reported that nanostructure formation augments the electrical conductivity by order of $10^4$ as compared to its bulk counterparts. Mechanical properties of nanostructured chalcogenide have been given importance due to enhancement in these properties. It was reported that microstructural features like grain size and shape, their distribution, pores and their distribution, other flaws/defects and their distribution, surface condition, impurity level, second phases/dopants, stress, duration of its application and temperature affect the mechanical properties of nanocrystalline materials. Further, an appropriate fraction of nanocrystalline grains in nanoglass can result in good strength, hardness and ductility, which is useful for the synthesis of novel nanograinned glass/crystalline composites with high strength, high hardness and superior ductility. The literature study on thermal properties of nanostructured glasses shows that grain size, grain boundary, temperature and presence of defects affect the thermal properties of nanostructured glasses. Further, literature reports reduced thermal conductivity in nanocrystalline coatings arises from the enhanced phonon scattering due to the presence of numerous closely spaced grain boundaries. Also, the combined effect of increased boundary phonon scattering and scattering from defects, contributes to the further reduction of thermal conductivity.

The study of various properties of nanostructured glasses shows that these materials are tunable according to application. They can show direct or indirect band gap on size reduction. The electrical properties also can be improved for opto-electronic applications. The amorphous structure can be ordered as per requirement to get the desired mechanical properties. The structural arrangement, grain size, defects etc, will overall rule the properties of these nanostructured glasses and makes them suitable candidate for many scientific and industrial applications. Beside this, nanoglasses have some limitations. Nanoglass production require vast developed economic methods for producing large quantities of nanoglasses with well-controlled microstructures. The microstructures depend on several factors as discussed above in paper, so controlling these factors and ensuring the applicability is big challenge. In coming future, nanoglasses seem to open the way to a new class of non-crystalline materials with tunable atomic and electronic structures and, hence, new properties (in comparison to glasses produced by quenching the melt) are expected. More focus must be given to thermal and thermoelectric properties in view of their applications as these properties are scantily been studied in context of nanoglasses. Study of properties and development of new nanoglasses may permit the development of technologies for better future of mankind.

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Disclosure statement

The authors declare no conflict of interests.

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