

A Few Prospective Compositions for the Chalcogenide Photovoltaics

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Abstract

The intensive effort has made to design new amorphous chalcogenide alloy for the photovoltaic as well as other optoelectronics applications. This work describes a short over view on photovoltaic materials, the relevance of inorganic photovoltaic materials, the development history of Cu (InGa) Se (CIGS) photovoltaics, prospects of chalcogenide photovoltaics. Along with the successful synthesis of $\text{Cu}_{20}(\text{In}_{14}\text{Ga}_9)\text{Se}_{45}\text{Te}_{12}$ (CIGST-1) and $\text{Cu}_{25}(\text{In}_{16}\text{Ga}_9)\text{Se}_{40}\text{Te}_{10}$ (CIGST-2) chalcopyrite materials in amorphous form. Structural analysis of the bulk materials was performed from the X-ray diffraction (XRD), Field Emission Scanning Microscope (FSEM), Differential Thermal Analyzer (DTA) and micro Raman analysis. While, the presence of elemental concentrations has been confirmed from the Energy Dispersive Spectroscopy (EDS). Subsequently, thermally evaporated thin films surface morphology and roughness parameter have been analyzed by using the Atomic Force Microscopy (AFM). The 100nm thin films, current-voltage (I-V) and resistance-voltage (R-V) characteristics at room temperature and in the temperature range upto 200°C, under applied voltage range upto 40 V have also been discussed. Outcomes of the structural analysis demonstrates bulk materials have an overall amorphous structure, and their thermally evaporated deposited thin films have low roughness. Structural analysis also reveal bulk CIGST-2 composition has single phase amorphous structure. This material thin film has a smooth surface morphology with a lower and higher values of I and R at room temperature. While, in the temperature range up to 200°C it has a higher and lower I and R respectively. The physical variation in these materials could be explained with the help of the chemical bond theory of solids.

Keywords

CIGST Chalcopyrite Materials; FSEM; Raman Spectroscopy; AFM; I-V Characteristics

Introduction

Photovoltaic Energy

Energy is integral part of every activity of mankind. Fossil fuels like oil, coal, and natural gas, have been the major energy source of power in modern society

for many years. While, the limited availability of fossil fuel sources, the unfavorable environmental effect caused by burning fossil fuels, and the concern about the energy security arising from the heavy dependence on imported fossil fuels, world demand rising to energy, have made finding sufficient supplies of clean energy urgent and of utmost importance in the next half century. One of the most promising and hugely under-utilized, alternative energy sources is solar energy, which is clean, renewable, safe, ubiquitous and abundant, covering 0.1% of the land on Earth with 10% efficient solar conversion systems would be sufficient to power the world. Photovoltaic (PV) cells are also commonly known as solar cells which directly convert sunlight in electricity to involve one of the major methods to capture and convert solar energy. At present solar generated electricity provided a very small portion of the world's electricity, nearly less than 0.1% even though its recent growth of 35–40% per annum owing to economical factor; solar electricity is about five to ten times more expensive than electricity generated from fossil fuels (especially coal).

The high costs of PV module manufacturing and installation are two dominant parameters for solar electricity. First-generation of PV modules mainly based on crystalline silicon wafers and a substantial portion of module manufacturing cost approximately \$300/m²; which depends on the cost of materials. Several modules already made in high volumes and have a little potential for further cost reduction. Therefore investigators proceed toward to second-generation thin film PV technology based on materials such as amorphous Si, CdTe, and Cu(In,Ga)Se₂ those offer advantages of lower material costs till now. To achieve more efficient PV cells "third-generation" comes in picture with ultrahigh efficiencies and/or ultra-low costs. Investigators believe this generation proposed modules would be significantly reduce the cost-to-efficiency ratio, which make solar electricity competitive against or even cheaper than fossil fuel

generated electricity.

PV Materials

Traditionally, PV materials are the inorganic semiconductors which can form suitable junctions with other materials. Several semiconducting materials have been shown a PV effect, but only a few of them have sufficient commercial interest due to satisfying essential constraints to minimising the thickness and their enough availability. Ideally, absorber material must have an efficient high absorption coefficient ($10^5/\text{cm}$) of terrestrial light and direct energy bandgap semiconductor with a bandgap of 1.5 eV, high quantum efficiency of excited carriers, long diffusion length and low recombination velocity. Material should have ability to form a good electronic junction (homo/hetero/Schottky) with suitably compatible materials. The high optical absorption defines the optimum thickness of an absorber in a solar cell because order of a film thickness inverse of the optical absorption coefficient. To minimize the processing and manufacturing difficulty the elemental materials should be chosen as simplest as possible.

However, it is mirror fact no suitable semiconducting material available with direct optical energy bandgap close to 1.5 eV. Most widely investigated silicon has an indirect bandgap with a gap 1.1 eV; by means there is no an ideal material for effective solar absorption. Si wafers have to be at least 50 mm thick in the optical enhancement technique to improve the effective absorption. In order to this boron-doped diamond-like carbon and fullerene films have also been identified as potential materials for photovoltaic applications with a promising results. A much wider choice of materials also available with two-component alloy/compound films. Metastable alloy film a-Si:H has been discovered somewhat serendipitously with large tailorable bandgap, easy dopability and high optical absorption coefficient. Indeed, in the 1980s, a-Si:H based thin film solar cell technology challenged the supremacy of crystalline Si cells. Besides metastable a-Si:H alloy, other two-component (binary) GaAs, CdTe, Cu₂S, Cu₂O, InP, Zn₃P₂ etc, materials are attractive for the thin-film solar cells. Of course, GaAs, InP and their counterpart alloys/ or compounds are ideal for photovoltaic applications but too expensive for large-scale commercial production. With increasing number of components, the number of possible materials are increasing in the geometrical proportion: like, alloys and compounds of I–III–VI ternaries and related quaternaries can form a host of suitable photovoltaic

materials. Such materials phase diagram is quite complex in bulk form. Therefore, it must be emphasized the increasing number of components with low cost of materials. In the form of microcrystalline thin films, the phase diagram is fuzzy and more complicated. Indeed, it is sufficiently relaxed to allow the formation and coexistence of all possible crystal structures which distributed heterogeneously.

The thin film of a material creates ab initio by the random nucleation and growth processes of individually condensed/ reacting atomic/ ionic/ molecular species on a substrate. The structural, chemical, metallurgical and physical properties of such materials strongly depends on a large number of deposition parameters and thickness. Advantage of thin-film it may encompass a considerable thickness range with varying from a few nanometers to tens of micrometers: might be best defined in terms of the birth processes. Thin films of the material can be produced by a number of existing methods with the geometrical control. Ideally an efficient solar cell material thin film should have following properties;

- Absorption coefficient must have high enough: it require less thickness and consume less material.
- The materials used for photovoltaics must be available in abundance.
- The material should non be toxic; so that, it could not influenced the health from any hazardous effects..
- The material must be stable; it should not degrade with time.
- Materials must have tendency to deposit from the variety of physical, chemical, electrochemical, plasma based and hybrid techniques.
- Microstructure of the film from one extreme of amorphous/nanocrystalline to highly oriented and/or epitaxial growth should be controlled by the deposition parameters and substrate or used technique.
- Material should provide a wide choice of shapes, sizes, areas and substrates.
- Owing to relaxed solubility condition and a relaxed phase diagram, the doping or alloying must be in the compatible manner (but in many cases incompatible materials are also useful).
- Surface and grain boundaries could passivate

from suitable materials.

- Material must have tendency to form different types of feasible electronic junctions, like, single and tandem junctions.
- Surface and interface of the materials should modify to get an interlayer diffusion barrier and surface electric field.
- Surfaces should also modify to achieve desired optical reflectance/transmission characteristics and optical trapping effects.
- Despite of conservation of energy and materials, thin-film process should be in general eco-friendly.
- Photovoltaics material used for space applications should have high energy particle radiation to create the defects in material structure for degrade their quality and functioning. The material must have high radiation resistance.

History of CIGS Photovoltaic Materials or Solar Cells

First report on $\text{Cu}(\text{In,Ga})\text{Se}_2$ (CIGS) photovoltaic material or solar cell was became in 1975, when Bell laboratory scientists had achieved 12% solar energy conversion efficiency where a layer of CdS evaporated onto a CuInSe_2 single crystal. After this investigation the intensive development has been made in CuInSe_2 thin films. In the University of Maine the thin films were deposited by dual source evaporation of CuInSe_2 & Se; the small area devices were built in the substrate configuration with an efficiency of 5%. In 1980s commercial companies were started developments of CIGS solar cells. At the commercial stage two different preparation concepts for CIGS were adopted. First co-evaporation process whereas, Cu, In, and Se were deposited from the separate evaporation sources, the efficiency was demonstrated more than 10%. Second was deposition–reaction process using Cu and In metallic precursors with H_2Se as the reacting chalcogen source. Outcomes of these two CIGS solar cells have demonstrated that the deposition–reaction technique better device fabrication technology than the co-evaporation approach. The cause was interpreted as; during the reaction step the absorber layer doped with Na. In the year 1995 Na-containing glasses had became standard and both technology and investigators demonstrated the co-evaporation and deposition–reaction, almost with similar efficiency (~14%). But deposition–reaction technology had attracted much attention due to its easy handling and

versatile industrial uses.

Further investigation inters into more complicated evaporation processes, to achieve three stage co-evaporated solar cells. First three stage co-evaporated solar cells was invented in 1994 by the National Renewable Energy Laboratories (NREL), with an efficiency of 20%. Around 2000 an effort was made to improve the co-evaporation technology; for this production and a research group had designed line-evaporation source and able to deposit on a substrate. In 2008, several laboratories around the world had experience to prepare a CIGS solar cell with efficiency more than 19%. However submodules of $\text{Cu}(\text{In,Ga})(\text{S,Se})_2$ formed by the deposition–reaction process was repeated with efficiency around 16%. Hence both materials are still in competition and may pursued parallel.

Prospects of Chalcogenide Photovoltaics

It is predicted that chalcogenide photovoltaics can have substantially contribute to future energy supply in some terawatts capacity. At present, chalcogenide module production is in the gigawatts/year range which mainly driven from CdTe module producer. Investigators assessed the future cost reduction of potentials thin film photovoltaics. Photovoltaic technologies CdTe and $\text{Cu}(\text{In,Ga})(\text{S,Se})_2$ have nearly same cost perspective for large scale production. A chalcogenide thin film module contains, depending on the chalcogenide film thickness, some grams per square meter of metals and chalcogens. The materials indium and tellurium are considered most critical in terms of availability. In order to estimate the maximum installation possible based on each material, two different approaches can be used: (i) use of published material reserves and (ii) extrapolation of current annual material extraction. The indium exploitation with Cu ore (although also contained in zinc, lead, tin, and tungsten ores) is getting much attention. Therefore one can conclude from the current investigations the chalcogenide thin film solar cells based on CIGS can substantially add to the world power supply.

The objective of this work, to present a few chalcogenide compositions; which can be use as photovoltaic materials as well as other optoelectronics applications (like electrical switching). Vast number of past reports on CIGS material demonstrates; this composition alloy in general has a crystalline structure and it's difficult to form amorphous structure. Cause,

complex metallic, semi metallic, non-metallic compositions, therefore this composition (although a few reports on its amorphous form) is designated as chalcopyrite material. To achieve a high performance chalcogenide photovoltaic/ optoelectronics use; investigator has successfully synthesized the $\text{Cu}_{20}(\text{In}_{14}\text{Ga}_9)\text{Se}_{45}\text{Te}_{12}$ (CIGST-1) and $\text{Cu}_{25}(\text{In}_{16}\text{Ga}_9)\text{Se}_{40}\text{Te}_{10}$ (CIGST-2) compositions alloys in the amorphous (or semiconducting) form. By introducing the an additional element Te in CIGS matrix. It is expected the incorporation of the chalcogenide metallic element Te in CIGS matrix would increase the phase transition temperature with higher order structural hindrance. To create a larger number of charge sits at the micro level in the alloy. In order to describe these two materials; demonstrate the amorphous structure, higher order of phase transition temperatures, Raman spectroscopic band for the bulk materials. Subsequently, 100 nm thin films AFM surface morphology, three dimensional view and roughness parameter and I-V and R-V characteristics at room temperature as well as temperature dependence variation under applied voltage range upto 40V are also discussed.

Experimental Details

Bulk samples were prepared by the melt quenched technique. High purity elements Cu, In, Ga, Se and Te (99.999% Aldrich) have been taken in the appropriate compositional ratio 25:16:9: 40:10 and 20:14:9:45:12. Properly weighed materials kept into clean quartz ampoules were evacuated and sealed under at the vacuum of 10^{-5} torr. The materials sealed ampoules were put into electric furnace and heated up to 1200°C with a slow heating rate and maintained the furnace temperature for 36 hours. To ensure the homogeneity of molten materials ampoules were continuously rotated with the help of an electric motor in last 12h heating process. After getting the desire melting time ampoules were frequently quenched in NaOH containing ice water. Prepared ingots of the materials were collected by breaking the ampoules.

Materials thin films were deposited on a glass substrate ($1.5\text{ cm} \times 5\text{ cm}$) by employing the thermal evaporation method. To ensure the homogenous surface of the thin film maintained the high vacuum (order of 10^{-6} Torr) under normal atmospheric pressure, throughout the material deposition process. After achieved the desired thin film thickness the deposition system power was turn off. Further,

deposited thin film on glass substrate was left as it is in a high vacuum chamber for the normal cooling. The deposited and cooled thin film on a glass substrate (two substrates for each composition) was removed from the sample holder.

Characterizations

The amorphous structure of the prepared materials was confirmed by X-ray Diffractometer (BRUKER D8 ADVANCE, $\text{CuK}\alpha$, $\lambda = 1.54 \text{ \AA}$). To characterise the surface morphology of the materials ULTRA 55, Field Emission Scanning Electron Microscope (Karl Zeiss) with EDS attachment equipment was used. While, to describe the amorphous to crystalline phase transformation the Differential Thermal Analyzer (DTA) equipment were used. Further the LabRAM HR Raman system having 514.5 nm wave length Argon LASER with a CCD detector in backscattering geometry range of 50 to 300 cm^{-1} , equipment was used for the spectroscopy measurement.

Surface morphology, three dimensional view and roughness parameter of the materials thin films was performed from the Atomic Force Microscope (AFM), Agilent 5500, in a surface contact mode. Subsequently, I-V and R-V characteristics of the thin films at room temperature as well as in temperature range upto 200°C , in the variable voltage range upto 40 V were performed under two probe surface contact mode from the PM5 with Thermal Chuck, Agilent Device Analyzer B1500A equipment.

Results

Bulk Material Analysis

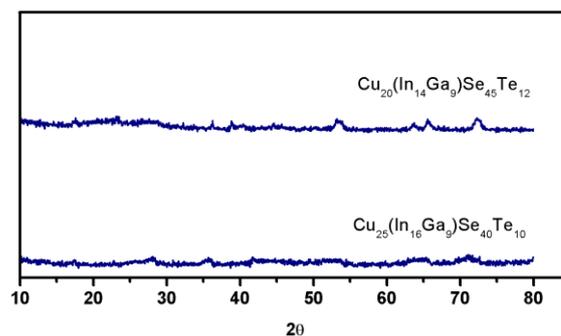


FIG. 1 XRD PATTERNS OF $\text{Cu}_{25}(\text{In}_{16}\text{Ga}_9)\text{Se}_{40}\text{Te}_{10}$ AND $\text{Cu}_{20}(\text{In}_{14}\text{Ga}_9)\text{Se}_{45}\text{Te}_{12}$ ALLOYS

To confirm the amorphous nature the XRD patterns of $\text{Cu}_{20}(\text{In}_{14}\text{Ga}_9)\text{Se}_{45}\text{Te}_{12}$ (CIGST-1) and $\text{Cu}_{25}(\text{In}_{16}\text{Ga}_9)\text{Se}_{40}\text{Te}_{10}$ (CIGST-2) bulk alloys are exhibited in Fig.1. XRD pattern of the CIGST-2 alloy is exhibited a good amorphous nature; while, the CIGST-1 composition

demonstrates a few very low intensity broad peaks. Such very low intensity peaks in amorphous materials are arise due to occurrence of fine clusters in homogenous amorphous structure. Therefore, this result reveals the elemental solid phase solubility in CIGST-1 composition inferior than CIGST-2 composition alloy. This result also consist to several past reports on CIGS alloys; in which it is demonstrated a chemical threshold can be exist for the Cu concentration in between 23-24%. Hence, the outcome demonstrates the overall amorphous nature of these bulk materials.

Subsequently, to further confirm the amorphous structure also analyzed the surface morphology of the CIGST-1 and CIGST-2 alloys by the high resolution field emission scanning microscope technique (FSEM). Obtain microscopic surface morphology of the CIGST-1 composition alloy at 100 nm and 20 nm is given in Fig. 2 (a, b) and energy dispersive spectroscopy (EDS) result is exhibited in Fig. 2 (c).

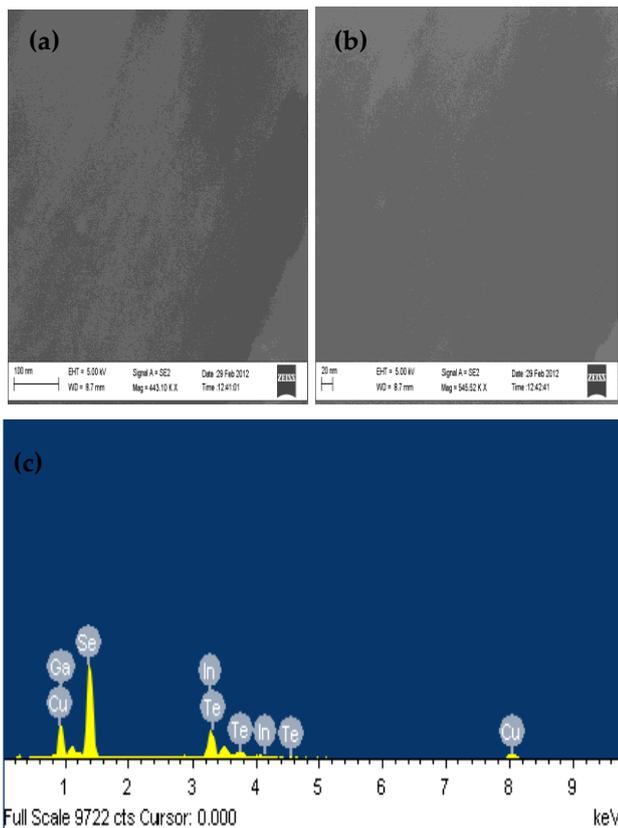


FIG. 2 (A, B, C) FIELD EMISSION SCANNING MICROSCOPE SURFACE MORPHOLOGY AND EDS RESULT FOR $Cu_{20}(In_{14}Ga_9)Se_{45}Te_{12}$ ALLOY

Outcome of the surface morphological analysis demonstrate; there is no specific growth at the nano scale, accept a dark spot in 100 nm scale, this may cause the existence of very fine clusters within the complex structure (this also appeared in XRD pattern

of the alloy). Hence FSEM result is also providing experimental evidence regarding the formation of amorphous structure of the alloy. The EDS spectroscopy result could provides the evidence of the elemental presence in the alloy in desired amounts ($\pm 2-3$).

The homogenous surface morphology of CIGST-2 alloy at 100 nm and 20 nm scale is exhibited in Fig 3 (a, b) and corresponding EDS pattern is shown in Fig 3 (c). This composition alloy surface appears quite homogenous than CIGST-1; by means this composition elemental diffusion rate higher than CIGST-1. Further this composition EDS measurement has also shown the presence of elemental alloy in appropriate amounts ($\pm 2-3$).

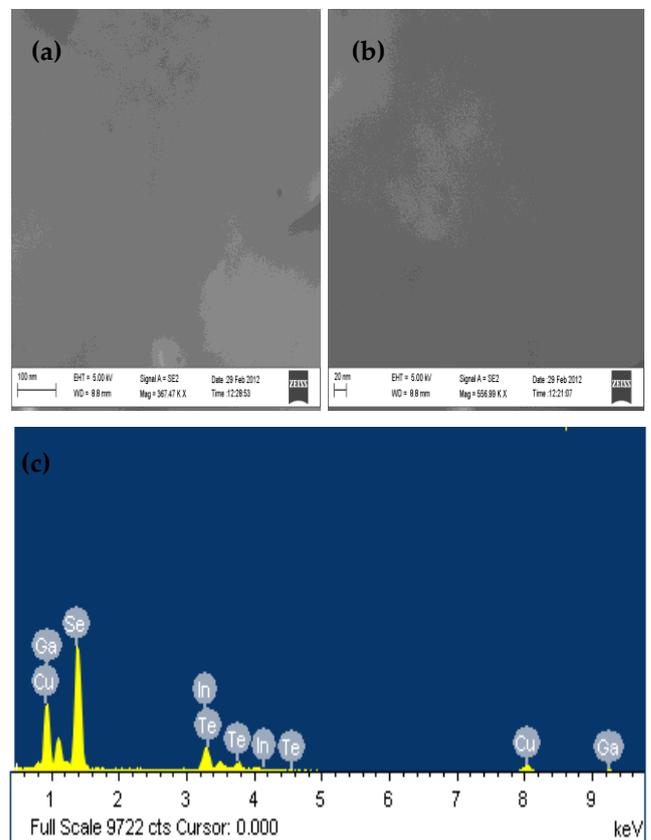


FIG. 3 (A, B, C) FIELD EMISSION SCANNING MICROSCOPE SURFACE MORPHOLOGY AND EDS RESULT FOR $Cu_{25}(In_{16}Ga_9)Se_{40}Te_{10}$ ALLOY

The elemental concentration tailoring in the CIGST-1 and CIGST-2 optoelectronic materials were done to achieve the higher order phase transition temperature; aim was to verify the sustainability of materials under the rapid thermal shock. Owing to truth, the low temperature phase transformation is one of the major problem in chalcogenide photovoltaic materials. To verify this property in these materials performed the DTA analysis (here DTA technique preferred over to

DSC due to its rapid thermal process). Obtained DTA single outcome with respect to temperature and reduce mass temperature for the CIGST-1 is exhibited in Fig. 4 (a, b), while, the CIGST-2 composition alloy DTA outcome is given in 5 (a, b). DTA single of CIGST-1 composition has reveal this material amorphous to crystalline phase transformation begin from ~345°C. While, CIGST-2 composition signal shows the rather higher value (~380° C).

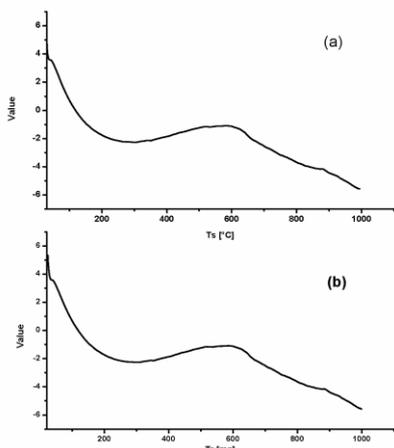


FIG. 4 (A, B) DIFFERENTIAL THERMAL ANALYZER SIGNALS FOR $Cu_{20}(In_{14}Ga_9)Se_{45}Te_{12}$ ALLOY

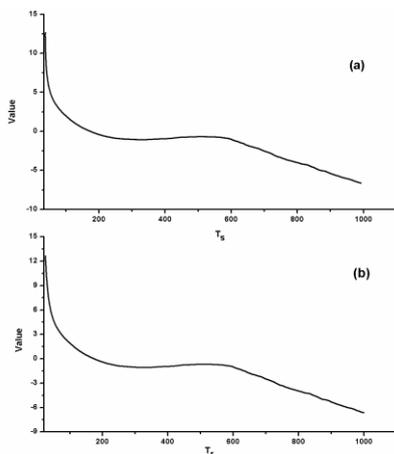


FIG. 5 (A, B) DIFFERENTIAL THERMAL ANALYZER SIGNALS FOR $Cu_{25}(In_{16}Ga_9)Se_{40}Te_{10}$ ALLOY

Thus the DTA result demonstrates these materials have high thermal sustainability. This is surprising in such materials; cause, metallic chalcogenide alloys have been generally exhibited the amorphous to crystalline phase transition temperature around 100-200°C. But this finding demonstrating compositions CIGST-1 and CIGST-2 has quite higher value of phase transition as compared to normally reported amorphous chalcogenide alloys. This may be due to single phase bond formation in the complex alloys; in which strong metallic nature Cu, semi-metallic In, metallic chalcogen element Te and non-metal Se and

Ga are present.

To confirm the single phase/or kind of phase formation in the alloys also performed the micro Raman measurement in the wave number range 0-500 cm^{-1} . The recorded spectra for the CIGST-1 and CIGST-2 is exhibited in Fig.6. It is well defined, Raman spectra can also provide the microstructural correlation for the materials. In chalcopyrites Raman modes are grouped into low, central and high energy frequency bands. Most of the Raman modes consist a very low scattering cross section and overlap on each other; means difficult to resolve even at low temperature. Raman spectra A1 symmetry mode, corresponding to the antiphase vibration of the anions in the lattice, it would usually the most intense band in the spectra of chalcopyrite compounds.

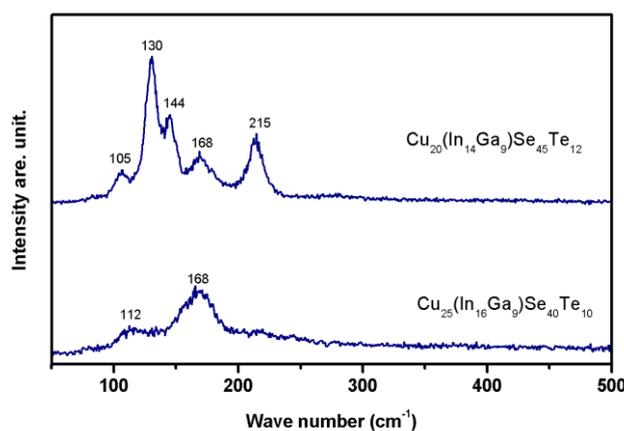


FIG. 6 RAMAN SPECTRA OF $Cu_{25}(In_{16}Ga_9)Se_{40}Te_{10}$ AND $Cu_{20}(In_{14}Ga_9)Se_{45}Te_{12}$ ALLOYS

The most intense characteristic spectra in CIGS chalcopyrite is reported at 175 cm^{-1} and corresponding band energy in between 173–216 cm^{-1} . Here in CIGST-1 we have obtained two distinct Raman band at 168 cm^{-1} and 215 cm^{-1} , with a lower limit band shift. This lower Raman shift might be arise due to the addition of the chalcogen element concentration in CIGS matrix. The existence of low frequency phonon bands in such materials have also been demonstrated, it can be lies in between 80-154 cm^{-1} . In CIGST-1 spectra these low frequency phonon bands appear at 105 cm^{-1} , 130 cm^{-1} and 144 cm^{-1} . By the investigators the distinct low frequency peaks in the raman spectra have been related to very fine crystallinity or clustering within the system. However, the CIGST-2 Raman spectra profile has shown a broad band A1 symmetry mode at 168 cm^{-1} with accompanied a small shoulder of the low frequency phonon band at 112 cm^{-1} . The appearance of the single broad Raman band in CIGST-2 alloy demonstrates; it has ordered amorphous structure

than CIGST-1 composition. The finding of the Raman measurement is also consisting with XRD, FSEM and DTA outcomes for these bulk materials.

Thin Film Analysis of the Materials

This study emphasizes toward to possibility of photovoltaic property in these materials, therefore, it is worth to describe the thin film properties of the CIGST-1 and CIGST-2 materials. Atomic Force Microscopy surface morphology of thermally evaporated, 100 nm thin film of the CIGST-1 composition is given Fig7 (a, b, c). Here Fig 7 (a) represents the surface morphology of thin film; the corresponding three dimensional view of surface cross sectional area 5µm×5µm is given in Fig 7 (b).

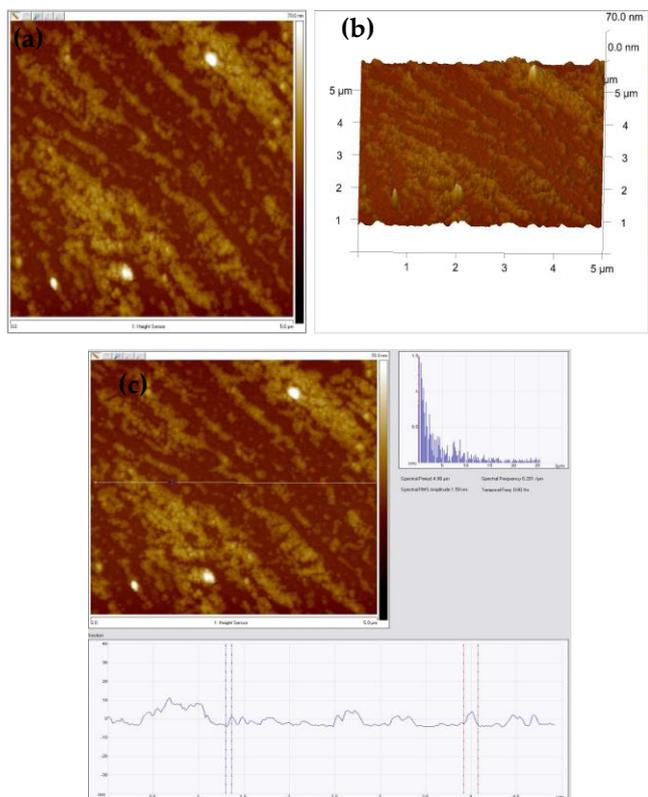


FIG. 7 (A, B, C) AFM SURFACE MORPHOLOGY, THREE DIMENSIONAL VIEW AND ROUGHNESS PARAMETER PROFILE FOR CU₂₀(IN₁₄GA₉)SE₄₅TE₁₂ THIN FILM

Appearance of bright and dark regions in surface morphology demonstrates the existence of clusters within an amorphous homogenous phase. The bright region in the outcome may appear due to partial solid solubility of heavy element (Cu) with other relatively lighter elements of the CIGST-1 alloy. 3D view of the thin film surface morphology provides the evidence absence of sharp crystallization in homogenous morphology by showing a few island order of less than 15 nm. Therefore, it can be considered normal amorphous phase alloy. Fig 7 (c) represents the thin

film surface roughness profile; which has analyzed as per outlined in microstructure: the corresponding pattern is given below in the figure. Therefore, outcome demonstrates CIGST-1 composition thin film surface not a completely smooth; it has on average roughness order of 5-10 nm. Cause, inhomogeneous phase mixing in the amorphous phase of the alloy. Further, Fig 8 (a, b, c) represents the AFM surface morphological parameters of the CIGST-2 composition thin film. Thin film surface of the CIGST-2 appears (see Fig 8 (a)) quite homogenous and nearly free of cluster formation within the alloy intrinsic structure.

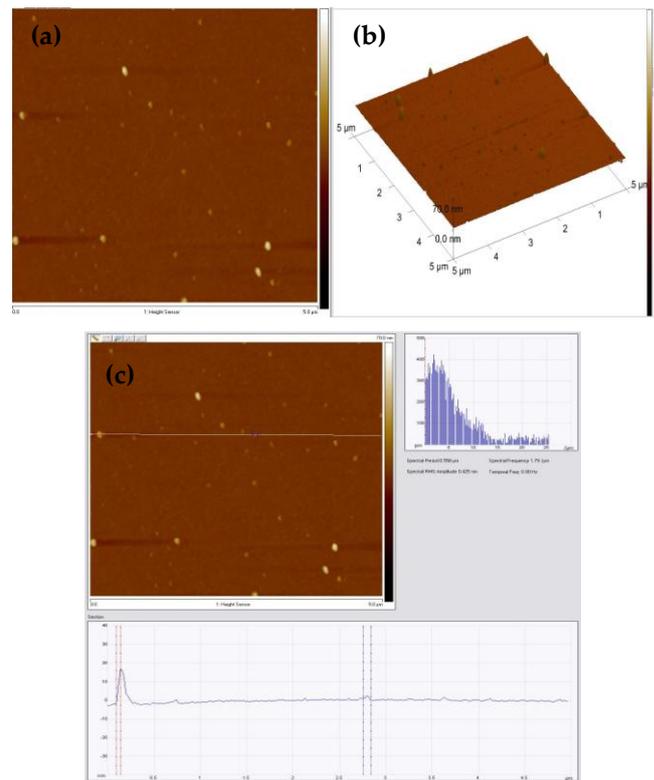


FIG. 8 (A, B, C) AFM SURFACE MORPHOLOGY, THREE DIMENSIONAL VIEW AND ROUGHNESS PARAMETER PROFILE FOR CU₂₅(IN₁₆GA₉)SE₄₀TE₁₀ THIN FILM

3D view (see Fig 8 (b)) of this alloy thin film is also showing a smooth surface in a cross sectional area 5µm×5µm; in which it has exhibited a few very small sizes (order of 0.55 nm) islands. Fig 8 (c) represents thin film surface roughness profile for CIGST-2 composition. Roughness parameter of the thin film is obtain by drawing a horizontal straight line across on the surface micro-morphology by selecting two distinct regions (one smaller point lies on straight within smooth surface and other larger particle lies on the line) on the straight line (see Fig 8 (c)) and taken the difference between two points on the surface. Accordingly analysis has been done, the obtained outcome has built a roughness profile pattern. Roughness profile of CIGST-2 thin film reveal, it has

quite a smooth surface with a very low surface roughness parameter value. Thus AFM surface morphology analysis outcome of the CIGST-2 thin film demonstrates it has good surface property than CIGST-1 thin film. The appeared difference in thin films surface morphology of the CIGST-1 and CIGST-2 might be due to change in complex intrinsic structure cause tailoring the amounts of alloying elements.

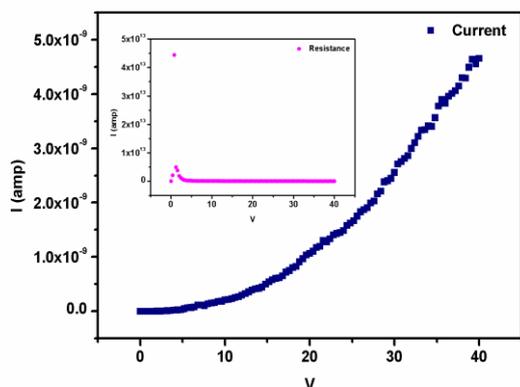


FIG. 9 I-V AND R-V CHARACTERISTICS FOR $\text{Cu}_{20}(\text{In}_{14}\text{Ga}_9)\text{Se}_{45}\text{Te}_{12}$ THIN FILM AT ROOM TEMPERATURE

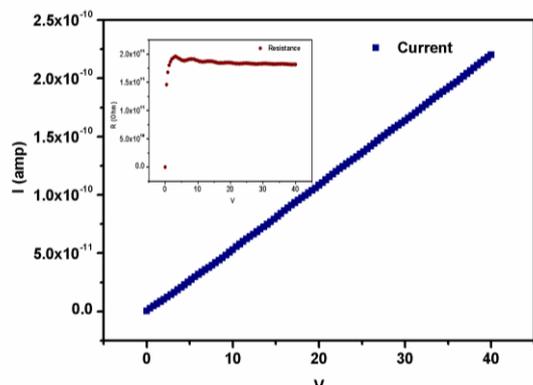


FIG. 10 I-V AND R-V CHARACTERISTICS OF $\text{Cu}_{25}(\text{In}_{16}\text{Ga}_9)\text{Se}_{40}\text{Te}_{10}$ THIN FILM AT ROOM TEMPERATURE

In order to extend the thin films study in terms of the prospective photovoltaic uses, performed the electrical I-V and I-R measurements for the CIGST-1 and CIGST-2 composition alloys; under applied voltage range upto 40 V, at room temperature as well as in temperature range upto 200°C. Fig 9 exhibits the current variation with the applied voltage range at room temperature and inset profile demonstrates the corresponding resistance for the CIGST-1 composition thin film. Result reveal CIGST-1 material has no current growth in the applied voltage range upto 5 V, the corresponding initial resistance average order is $10^{13} \Omega$. With increasing voltage more than five volt current has follow the increasing trend and above the 15 voltage system exhibits a sharp increase in current upto 40 volt and reaches maximum order of 4.5×10^{-9}

amp. The corresponding resistance is also showing a sharp decline near to applied voltage 5V, afterward a very low constant resistance is obtained. High resistance and negligible current growth at low voltage $< 5\text{V}$ demonstrates the non-conducting behavior of the CIGST-1 material; while, the increasing current with increasing voltage reveal the semiconducting nature of the deposited thin film material.

CIGST-2 alloy thin film I-V and R-V (see in inset) characteristics in the above said applied voltage range at room temperature is exhibited in Fig.10. CIGST-2 thin film has shown the electrical conduction even at below the 5 V and it increases continuously up to 40 V and archive the current growth order of $\sim 2.25 \times 10^{-10}$ amp. This indicates CIGST-2 thin film has more metallic character than CIGST-1; this also consists with the DTA findings.

The increase in metallic behaviour in CIGST-2 thin might be due to presence higher amounts of Cu and In; while the amorphous character remains preserved owing to amounts of Se and Te in the configuration. Inset profile of CIGST-2 thin film shows the initial resistance order of $1.5 \times 10^{11} \Omega$, afterward it is nearly constant ($\sim 1.85 \times 10^{11} \Omega$) throughout the applied voltage range. Thus the CIGST-2 current conduction ability at low voltage makes them superior for the photovoltaic application. Besides its low current conduction ability in voltage range 5-40 V at room temperature in comparison to CIGST-1. Thus the outcome reveals CIGST-1 composition could be useful for the optoelectronics applications.

In order to define these alloys as photovoltaic materials it is valuable to describe the temperature dependence current conduction behaviour. Because in real application temperature variation also affects the photovoltaic module performance. The temperature dependence current growth of the CIGST-1 thin film is exhibited in Fig. 11; in the temperature range 30-200°C. This measurement was performed at 10°C successive temperature interval under applied voltage range upto 40V for the each temperature data point. Inset profile in Fig.11 is exhibited a clear view of temperature dependence current growth from room temperature to 100°C in the applied voltage range; for the each temperature data point. Obtain outcome demonstrates the maximum current at 200°C is order of 4.8×10^{-6} amp for the CIGST-1 thin film. Subsequently it also reveal CIGST-1 system thin film has uneven current growth upto 170°C (upto 100°C is exhibited in inset profile). The uneven current growth with temperature can be

directly relate to the existence of fine clusters within the homogeneous structure of material. Due to involvement of rising temperature and increasing applied voltage, it is quite possible clusters bonds broken and creates the sudden increase in charge carriers at a specific temperature. Along with this outcome demonstrates CIGST-1 thin film current increases rapidly in between the temperature range 180-200°C. Cause the excess supplied heat energy creates large number charge of carriers.

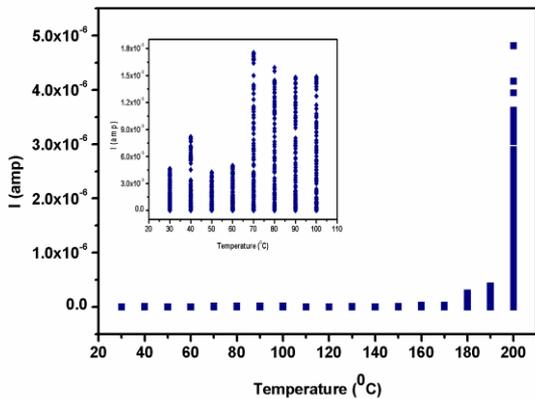


Fig. 11. TEMPERATURE DEPENDENCE CURRENT VARIATION OF $Cu_{20}(In_{14}Ga_9)Se_{45}Te_{12}$ THIN FILM UNDER APPLIED VOLTAGE RANGE UPTO 40V

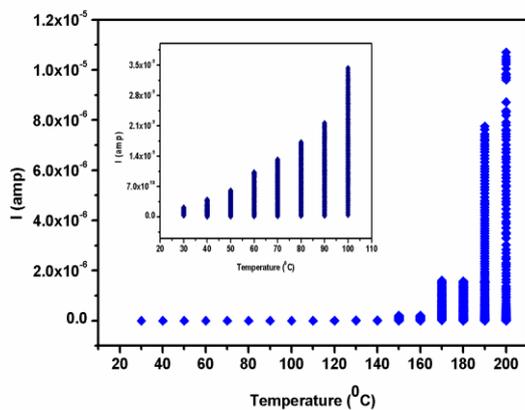


FIG. 12 TEMPERATURE DEPENDENCE CURRENT VARIATION OF $Cu_{25}(In_{16}Ga_9)Se_{40}Te_{10}$ THIN FILM UNDER APPLIED VOLTAGE RANGE UPTO 40V

CIGST-2 thin film temperature dependence current growth in range 30-200°C, in the applied voltage range, at each data point is shown in Fig.12. This alloy thin film has an ordered increasing current profile at each temperature data point (see inset profile in Fig.12). Obtained outcome demonstrates, the current growth of this thin film is order of 1.1×10^{-5} amp at 200°C. This systematic (free to abrupt change in current conduction) increase in current with increasing temperature under applied voltage range demonstrate;

CIGST-2 thin film has a homogenous amorphous structure, in which, the current conduction governs through charge carries of defect states or lone pair electrons. This temperature dependence current growth CIGST-2 also has one noticeable point from the view point of technological applications; the room temperature I-V characteristic of CIGST-2 thin film has exhibited inferior current growth value compare CIGST-1 thin film; while, the temperature dependence CIGST-2 thin film has exhibited superior current growth than CIGST-1 thin film. This indicates; CIGST-2 alloy thin film has a large number deep localised defects states or loosely bonded hydrogen like bonds. While; CIGST-1 alloy thin film has a greater number of Van-der Waal like weak bonds in the alloy complex structural geometry. Here Fig. 13 represents the temperature dependence resistance variation for the CIGST-1 and CIGST-2 thin films under applied voltage range for each data point. Initial resistance of CIGST-1 thin is higher and it is abruptly decreased with increasing temperature. On the other hand, the CIGST-2 thin film resistance initially quite lower than CIGST-1, and it gradually decreases with increasing temperature. Specifically the temperature range 30-60°C is important for photovoltaic application; in this range CIGST-2 thin film has lower order resistance and a consistent decline: in comparison to CIGST-1 thin film.

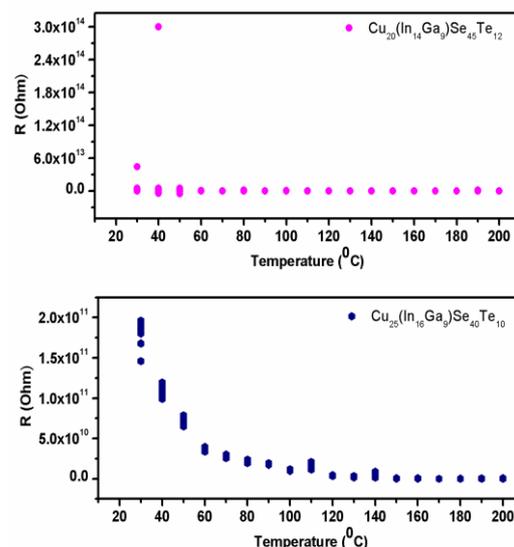


FIG. 13 TEMPERATURE DEPENDENCE RESISTANCE VARIATIONS OF $Cu_{25}(In_{16}Ga_9)Se_{40}Te_{10}$ AND $Cu_{20}(In_{14}Ga_9)Se_{45}Te_{12}$ THIN FILMS UNDER UNDER APPLIED VOLTAGE RANGE UPTO 40V

Discussion

The physical variation in bulk as well as in thin films

of CIGST-1 and CIGST-2 could be interpreted as; the atomic size, density and alloying concentrations may play an important role in physical properties variation of bulk and thin films of the alloys. The element Cu (atomic radius 1.45 Å and density 8920 kg m⁻³) has been taken owing to its high melting point, metallic nature and high order UV/Visible light absorption ability. With the prime objective it can enhance the thermal as well as mechanical stability of the material. Second alloying element In (atomic radius 1.56 Å and density 7310 kg m⁻³) responsible to work as a recombination blocker within the complex CIGST-1 and CIGST-2 alloys with the increasing atomic mismatch ratio in configuration. Ga (atomic radius 1.36 Å and density 5904 kg m⁻³) has been used for the band tailoring of the alloys. However, Se (atomic radius 1.03 Å and density 4819 kg m⁻³) is the base element to make active layer photovoltaic cell material. This element responsible to form long chain as well as ring within the complex structure, consequently it provides the large number of defect states in the complex configuration. The additional chalcogen element Te has been incorporated as cost of Se (in past reported CIGS) with the aim it would increase the thermal stability, defect states and decrease the Se rings within the intrinsic complex configurations of the alloys.

In this subsequent, it is possible to cubic close-packed Cu, tetragonal In, orthorhombic Ga, monoclinic Se and trigonal Te structurally uneven alloying can form; Cu-Cu, In-In, Ga-Ga, Se-Se, Te-Te, Cu-In, Cu-Ga, Cu-Se, Cu-Te, In-Ga, In-Se, In-Te, Ga-Se, Ga-Te, Se-Te, Cu-In-Ga, Cu-Se-Te, Cu-Se-In, Cu-Se-Ga, Cu-In-Te, Cu-In-Te, Cu-In-Ga-Se, Cu-In-Ga-Te, Cu-Ga-Se-Te, In-Ga-Se-Te and Cu-In-Ga-Se-Te, homopolar, heteropolar binary, ternary, quaternary and penternary bonds, with the distinct energies. Such a complex bond formation is creates a large number of unsaturated bonds in sudden frozen alloys configurations. These unsaturated bonds, defects, correlated traps could produced all types of molecular bonds and defects within the complex intrinsic structure, therefore, systems creates a large number of charge carrier centres for electrical conduction. Since, CIGST-1 composition has lower concentrations of Cu & In and higher concentrations of combined chalcogenide chalcogen elements Se & Te in comparison to CIGST-2 composition alloy. Owing to higher concentrations of the chalcogen elements CIGST-1 system frozen state has a strong tendency to form chain and ring very quickly in amorphous structure. But the presence of

strong chain broker metallic element Cu and bond modifier element In with fixed amount of Ga concentrations could restrict the homogeneous structure formation. Here, possibly heavy elements Cu and In could not completely dissolve into the Se and Te chains. Therefore, CIGST-1 alloy has formed very fine clusters within the preserved homogenous amorphous structure. Which has appeared in XRD, Raman spectroscopy and in the AFM surface morphology of thin film. Due this consequence no current growth or non-conducting behavior has appeared at low voltage; at higher voltage it has followed the increasing current trend at room temperature, as well as in the temperature dependence I-V profile. By showing the uneven increasing current trend below the ~170°C in the applied voltage range for the each data point.

CIGST-2 alloy contains higher concentrations of heavy elements Cu & In as cost concentrations reduction of the Se & Te. The increase and decrease in concentrations of Cu & In and Se & Te in the CIGST-2 have affected the chemical equilibrium of the alloy, as a consequence, perhaps rate of solid state reaction for chalcogen elements Se & Te is would have slow down and sufficient time scale available to dissolve Cu & In in complex intrinsic structure before the growth of the homogenous amorphous phase under the frozen condition. The increase in metallasity and slow solid state reaction in the CIGST-2 alloy also has appeared in DTA signal. The AFM surface morphology and less order of roughness parameter for the CIGST-2 thin film is also an evidence about the homogeneous phase mixing within the amorphous configuration. At room temperature current growth in CIGST-2 thin film at low voltages has appeared due to presence of sufficient amount of metallic element Cu in complex alloy configuration. The increasing current trend with increasing temperature in this thin film; owing to the production of the deep localized [47-51] correlated traps or defects within a homogenous structure.

Conclusions

In brief, author has presented a short over view on photovoltaic materials, the relevance of inorganic photovoltaic materials, the development history of the Cu(InGa)Se (CIGS) photovoltaics, prospects of chalcogenide photovoltaics. Along with also discussed the successful synthesis of two new [Cu₂₀(In₁₄Ga₉)Se₄₅Te₁₂ (CIGST-1) and Cu₂₅(In₁₆Ga₉)Se₄₀Te₁₀ (CIGST-2)] compositions for the photovoltaic applications.

Experimental findings revealed the CIGST-1 bulk and thin film has an amorphous structure accompanied with very fine clusters in intrinsic complex configuration; it has none conducting nature at low voltages. While, with increasing temperature it has shown the increasing order but in fluctuating manner upto a certain temperature range under specific applied voltage range (up to 40V), for the each data point, afterward it followed the regular trend. This might be due to the existence of two non-homogenous amorphous phase mixing in the complex homogenous intrinsic structure of the alloy.

On the other hand, CIGST-2 alloy bulk and thin film has exhibited the single amorphous phase with a high order phase transition temperature and metalliasity. Experimental findings revealed, this composition thin film has a smooth surface morphology with a low order roughness parameter. Subsequently, electrical measurement outcome of this thin film demonstrates; it have conducting nature with a low value of the resistance at the room temperature under the low applied voltages. The presence of sufficient amount heavy metallic element as well as the homogenous phase mixing in complex alloy configuration has produce a large number structural traps and defects in the system. Therefore this composition can be considered a promising candidate for the photovoltaic application.

Thus, the obtained experimental evidences on the CIGST-1 and CIGST-2 bulk and thin films demonstrates: CIGST-1 alloy would be useful for optoelectronics (like electrical switching etc) applications, however, CIGST-2 is a suitable composition for photovoltaic application.

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