# FEDERAL UNIVERSITY OF TECHNOLOGY MINNA

# GENERATIONAL MATERIALS MODIFICATION TOWARDS A LOW COST AND EFFICIENT CONVERSION OF LIGHT INTO ELECTRICITY: MY CONTRIBUTIONS

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*B.Sc (ABU), MTech (FUTMINNA), PhD (ABU)*
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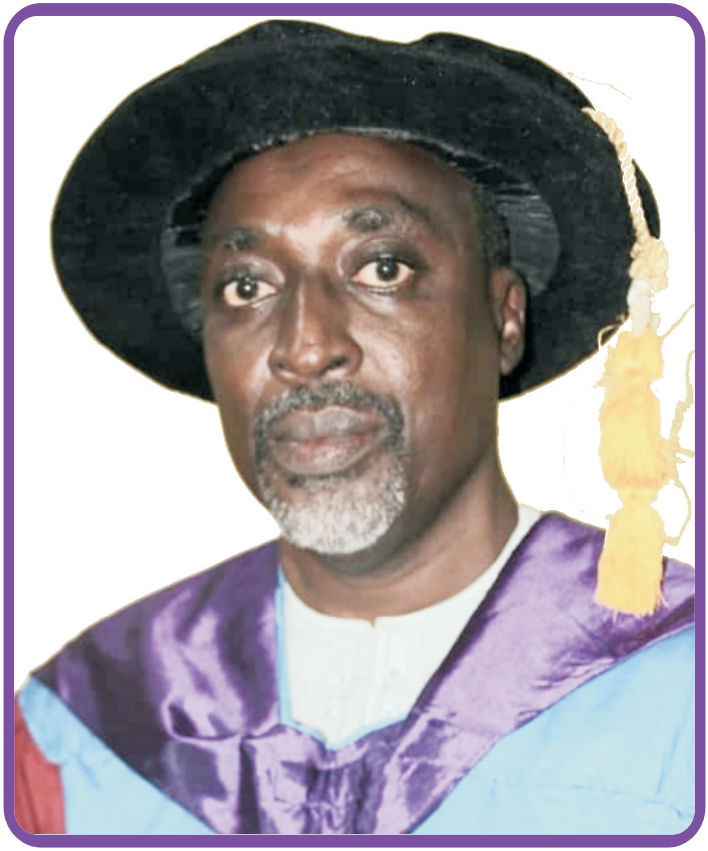

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***Professor of Physics***

**ALHAMDULILLAH**

PRAISE BE TO ALLAH THE POWER AND THE GLORY

**May peace and blessings of Allah be on the effulgent light that fills the Universe and surrounds all places of existence.**

**QUOTES IN MARBLE**

**"Allah is the Light of the heavens and the earth**. The similitude of His light is like a niche within which is a lamp, the lamp is within glass, the glass as if it were a brilliant [white] star, lit from a blessed olive tree, neither of the east nor of the west, whose oil would almost glow even if untouched by fire. Light upon light! **Allah guides to His light whom He wills**. And Allah sets forth parables for mankind, and **Allah is Knowing of all things"**

The Quran 24:35

## 1.0 INTRODUCTION

### 1.1 My Discipline

I am first and foremost a Physicist which is the science of nature that deals with the laws and properties of matter and the forces that act upon them. It is the most fundamental scientific discipline. My reading Physics for my first degree was by divine providence as I had applied for and was admitted to read Mathematics at Ahmadu Bello University, Zaria.

I am secondly a Solid State Physicist, that branch of condensed matter physics that specialises in the study of physics of solids and attempts to understand how the behaviour of atoms and molecules gives solids their observed properties. It forms a theoretical base of materials science.

By research orientation, my interest is in Materials Science (materials synthesis and characterisation) which is the study of properties of solid materials and how their properties are determined by materials composition and structure. Most of my academic research is in the synthesis and characterisation of materials for low cost and efficient conversion of light into electricity.

### 1.2 Interest in Light

My interest in light started at a very young age, about seven years of age. As children we used to go play hide-and-seek at *Katamba* or *Zaure* of the palace of one of the ruling houses of Bida – the Usman Zaki ruling house. The frontage of *Katamba* of the palace is shown in Plate 1:


Plate 1: Etsu Usman Zaki palace, Bida

The *Katamba* has a very large space inside and there only one light bulb is at the entrance and non-inside the *Katamba*, so the light from the bulb streams only straight along the aisle (walk way) and the inside of the *Katamba* to the left and right recesses

was pitch dark, which makes it possible for someone in the recess of the Katamba to see anyone walking into the Katamba without the person seeing him. As a young boy of about 7 years this was a mystery to me and baffled a young mind, that had no knowledge then of principles and characteristics of light. This heralded my first interest in light and then of courses the verse of the Quran I quoted above, **"Allah is the Light of the heavens and the earth...."**

### 1.3 Light

Generally, *"Light is that which causes the sensation of vision",* that was the first definition I got from my first teacher in physics Dr. N. A. Oyedum in my form three. Through the sense of sight, light is a primary tool for perceiving the world and communicating with it.

In a more technical term, Light is defined as the electromagnetic radiation with wavelengths between 380 (blue light) and 750 nm (red light) which is visible or detectable to the human eye. Electromagnetic radiation extends from γ rays and X-rays through to short radio waves and to the long radio waves shown in Fig. 1

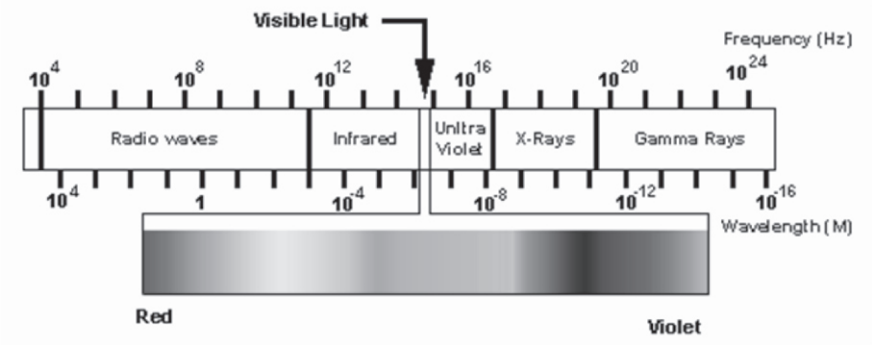



Fig. 1: Electromagnetic spectrum (Anonymous, 2006)

This is often referred to as 'the electromagnetic spectrum'. An alternative physical (quantum mechanical) description of light is to consider radiation as being emitted as discrete parcels of energy, called photons, which have dual nature – that of a particle and a wave expressed in Physics by the de Broglie relation

$$P = h/\lambda$$

The L.H.S is the particle side and R.H.S give the wave-like behaviour.

**2.0 Solar Energy Resource**

World energy needs is predicted to reach 30 TW of energy resources by the year 2050 to maintain economic growth (Almosni, Delamarre, Jehl, Suchet, Cojocaru., *et al*., 2018). To meet this demand significant fraction of alternative energy resources must come from renewable energies given their abundance and environment friendliness. A sustainable global energy system requires a transition away from energy sources with high greenhouse emissions.

Scientists believe that the sun is the best candidate that can offer a fully developed sustainable solution for the possible energy crisis. Therefore, the direct solar energy conversion through solar cells can be considered as a mainstream renewable energy resource once their manufacturing cost is decreased to an affordable level comparable with other available energy resources.

The amount of solar energy reaching the surface of the earth every hour is greater than the amount of energy used by the earth's population over an entire year. Thus effective utilization of this vast resource could solve energy demand of the world. This energy is renewable and sustainable. Solar energy

utilization is thus steadily growing alternative form of reliable energy technology today because of its abundance and it is less harmful to the environment than other methods of energy production such as coal, fossil fuels and nuclear.

Abundant solar energy in addition to maintaining life on the planet can be utilized based on anthropogenic processes in three basic forms: Electricity from the direct conversion of solar energy using semiconductor materials (solar photovoltaics, PV); electricity from captured thermal energy (concentrated solar power, CSP), and heat from the sun (solar thermal). Solar thermal utilises the direct use of heat energy from the sun for various application such as (a) Solar water heating (b) solar heating of buildings (c) solar distillation (d) solar pumping (e) solar drying of agricultural and animal products (f) solar furnaces (g), while solar photovoltaic application is the direct conversion of solar energy into electricity at the atomic level. Our interest is in the latter. Also, in the context of depleting of oil deposits and increasing pressure from global warming, solar cells have gained a lot of interest emerging as a credible alternative to fossil fuels due to the abundance of solar energy. Hopes for pervasive and use of this abundant and environmentally benign energy is dependent on its ease of availability which is a function of reduction in its cost to achieve grid parity (when use of alternative energies, like solar costs less than, or equal to, the price of using power from conventional sources i.e fossil fuels) with enhanced efficiency.

## 2.1 Materials Research in Solar Energy Conversion

Materials science and research has gone a long way since the discovery of the photovoltaic effect, in making solar cells more efficient and attempts to achieve cheaper photovoltaics.

Increasing interest and use of solar energy is predicated on fundamental materials research, dedicated to reaching highest performances and industrial developments, turning laboratory results into commercial systems. The challenges in conversion of solar energy into electricity are materials availability and processability to achieve low cost. Extremely low cost could be achieved if highly scalable and low-cost processes could be used to produce high-quality materials.

Materials study, synthesis and characterisation (basic research) holds promise for improvement of existing PV materials, the successful identification of new materials and achievement of low-cost processing procedure systems (required technological development) and turning laboratory results into commercial systems (market application). This is summarised in Figure 2

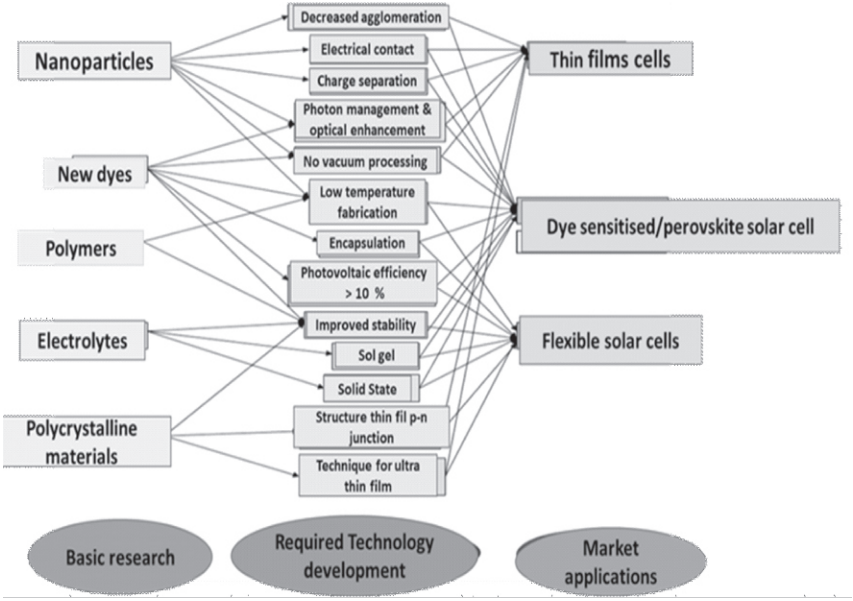



Fig. 2: Basic materials research underway with the technology

developments required to achieve the desired applications (Asim *et al.*, 2012)

My major contribution to the advancement of Physics, particularly Solid State Physics and specifically materials science is in the simple and low cost synthesis procedures, characterization and optimization of various solar cell materials components with an overall view of achieving cost reduction and increasing efficiency

## 2.2 Photovoltaics

Photovoltaics use semiconductor materials to generate electricity from solar energy. A semiconductor is a solid, mostly crystalline, material such as silicon, selenium or germanium. Semiconductors have electrical conductivities greater than insulators but lower than metals which are good conductors. They are insulators at low temperatures, however at high temperatures or when excited by sunlight, they conduct electrons. They have a lower band gap compared to that of insulators as shown in band gap diagram of conductor insular and semiconductor in Fig. 3.

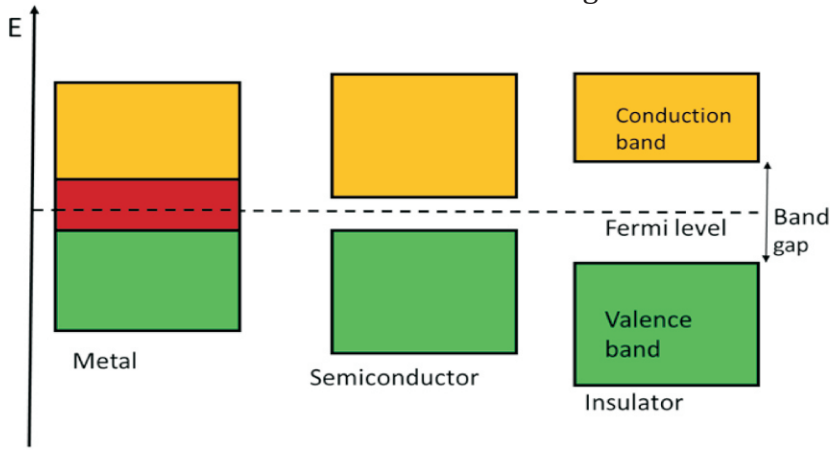



Fig. 3:Energy band diagram of conductor, semiconductor and insulator

The most commonly used semiconductor element is silicon belonging to column IV of the periodic table which has four valence electrons and bond to four other atoms of the same element to have a fully satisfied valence of eight electrons. A number of compounds can be semiconductors having a valence of eight electrons, such as Gallium arsenide (GaAs), and cadmium telluride (CdTe), are examples of III–V, and II–VI semiconducting compounds.

## 2.3 How do Photovoltaics Work?

First and second generation solar cells are based on P-N junction, called P-N junction solar cell. The principle of operation of a p-n-junction solar cell is illustrated in Figure 4. When the light falls on the device, the light photons of certain wavelengths are absorbed by the semiconducting material (excites electrons which move from the valence to the conduction band) and electrical charge carriers, electrons and holes, are generated. These carriers diffuse to the junction where a strong electric field exists, the electrons and holes are separated by this field and produce an electric current in the external circuit, this current, called photo-current, depends on the incident photon intensity and the nature of semiconductors that constitute the junction device.

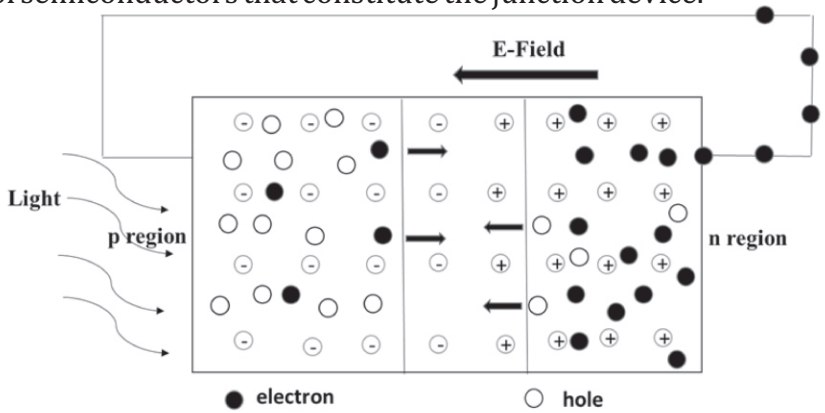



Figure 4 P-N junction solar cell under light illumination

## 3.0 Generations of Solar Cells

Solar cells are devices which convert sunlight directly into electricity. They are classified as first generation, second generation and third generation. They are usually made of semiconducting materials.

## 3.1 First Generation

Generally, first generations solar cell is based on Silicon (Si) solar cell. It consists of large-area, high quality single-crystal, single layer p-n junction diode devices. The fabrication of such cells involve high energy and labour inputs making these very costly. For instance, to get high purity Silicon, we start with quartzite gravel or crushed quartz, which is reduced in coke to give metallurgical grade Silicon. The metallurgical grade Silicon is then converted to $SiHCl_3$. This is then reduced in Hydrogen to yield semiconductor grade polycrystalline Silicon. Obtained Polycrystalline Silicon is then converted to high grade crystalline silicon by Czochralski growth scheme or floating zone scheme. The high grade Silicon is then cut into wafers and ready to use. These processes make the fabrication process of the silicon based solar cell complex and quite costly.

## 3.2 Second Generation

The drive for the reduction in cost of solar cells lead to the second generation solar cells. Second generation solar cells are thin film based solar cells. They are also P-N junction based solar cells. To get required films, techniques such as vapour deposition and electroplating are advantageous, aqueous solution techniques were developed to reduce production costs.

Thin (solid) films are solid materials of less than 1000 nm grown on another material called substrate, as shown in Figure 5.


Fig. 5: Thin solid film on a substrate

Thin films belong to low dimensional systems (LDS), whose properties can be very different from the bulk specimen in quite unexpected way. Though bulk materials can be replicated in thin film, the intriguing aspects of thin films are the many alloys, compounds and systems of unique composition that can be fabricated in thin film form. They have wide applications in microelectronics, optoelectronics, corrosion and wear application magnetics sensors, etc.

Thin films can be fabricated in various ways. The techniques can be divided to physical and chemical methods. In physical methods the film material is moved from a target source with some form of energy to the substrate. These techniques such as thermal evaporation, sputtering, electron beam evaporation are widely used in one-component films, like metal films, though configurations for multiple component depositions do exist. Chemical film fabrication methods involve chemical reactions and the precursors are mostly components undergoing reaction at the substrate surface or in the vicinity of the substrate. These methods include liquid phase epitaxy, vapor phase epitaxy, chemical bath deposition, successive ionic layer deposition among others (Smith, 1995).

Among the different thin film materials synthesis methods, aqueous solution techniques possess certain advantages. These techniques are cost effective and are technically undemanding to the experimentalist. Furthermore, relatively mild conditions are

used. The deposition is carried out at or close to room temperature and under normal pressure.

My interest was in liquid phase methods, such methods include Chemical Bath Deposition (CBD), Solvothermal and Hydrothermal Methods, Successive Ionic Layer Adsorption and Reaction, (SILAR) and sol-gel methods. The motivation for use of such methods is to develop reproducible and relatively simple route(s) for applications keeping in mind that low capital cost is important for successful commercialization, for instance thin film photovoltaic devices.

### 3.2.1 CdS/CdTe thin film solar cell and challenges

Thin film photovoltaics have two very important layers that determine to large extent the performance of the photovoltaic or the solar cell. These layers are the window layer and the absorber layer. Fig. 6 shows a CdS/CdTe solar cell

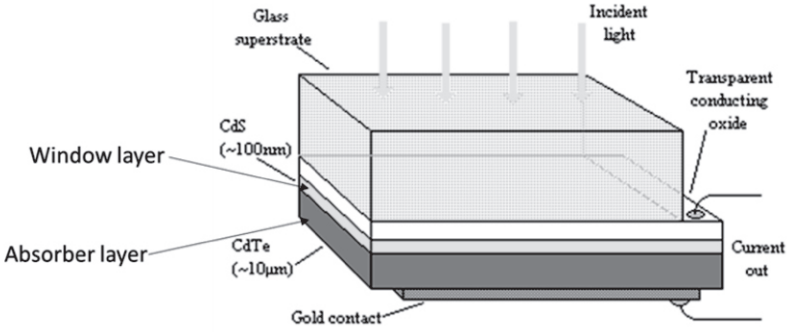



Fig. 6: Schematic of CdS/CdTe solar cell with a CdS film as window (not to scale) and CdTe as absorber (Carlsson, 2001)

The window material must be thin enough and have a wide enough band gap (**2.8 eV or more**) to let all available light through the interface (heterojunction) to the absorber layer. Though Thin-film cadmium telluride (CdTe) is regarded as one

leading material for the development of cost-effective photovoltaics (PV), however, the presence of toxic heavy metal Cd raises concern, thus search to alternative less toxic materials. Also CdS/CdTe based solar cell CdS window material has a band gap of 2.4 eV, which is less than the required gap of 2.8 eV. This leads to high window absorption loss with about 36% of the incident radiation lost to photon energy > 2.4eV and a subsequent lower efficiency of the solar cell. The use of ZnS film with a Band gap of 3.6 eV would have been appropriate as window material, however, it is highly resistive and hard to dope and could significantly increase the cells series resistance.

### 3.2.1.1 My Contribution

In this regard my contribution was in attempting to combine CdS and ZnS to increase the bandgap of resulting compound, exploiting the merits of CdS and ZnS to obtain a more effective window material. This was done through the co-deposition of CdS and ZnS using chemical bath deposition, with the aim of tailoring the ban gap of resulting $Cd_{1-x}Zn_x$ from 2.42 eV (CdS) to 3.6 eV (ZnS) by compositional control.

The relative ratio of Cd, Zn was varied with a solution composition parameter y = 0.3, 0.5 and 0.7, where

$$y = \frac{[ZnCl_2]}{[ZnCl_2] + [CdCl_2]}$$

with a bath temperature between 80 and 85 $^0$C. The film had a thickness of between 70 and 200 nm and a resistivity of ~$10^2$ to 7 x $10^3$ Ω cm as the y values increased.

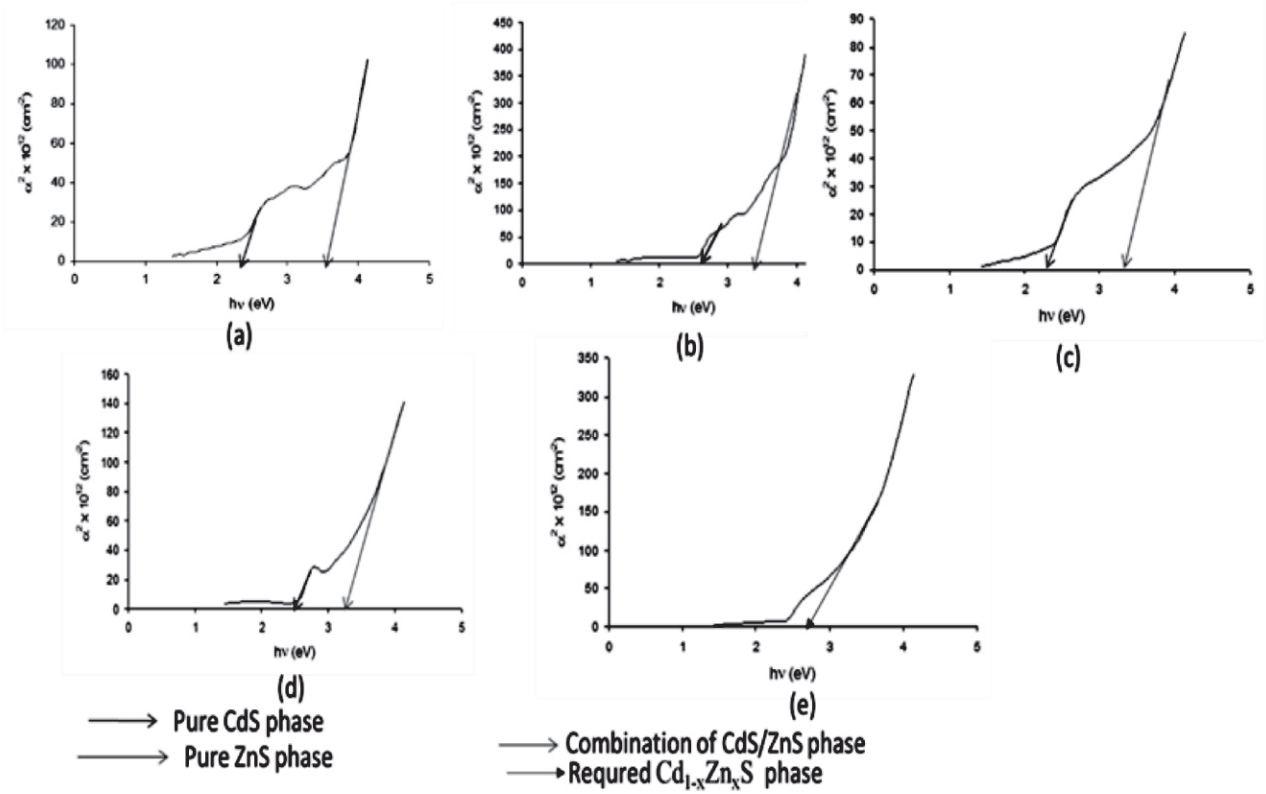



Fig. 7: Band gap plots of synthesised $Cd_{1-x}Zn_xS$ films (Isah *et al.,* 2008)

Fig. 7 shows the gradual evolution of the two phase of CdS and ZnS (a), (b) and (c) to a single phase CdZnS at the optimised deposition parameters. At the optimized deposition parameter, a pure $Cd_{1-x}Zn_xS$ phase, with x = 0.19 ($Cd_{0.81}Zn_{0.19}S$) and an improved band gap from 2.42 eV pure CdS to 2.7 eV of the synthesized $Cd_{0.81}Zn_{0.19}S$ was obtained (Isah *et al.,* 2008 and Isah, 2013).

### 3.2.2 Copper Indium Gallium Sulphide(Selenide) $CuIn_xGa_{1-x}S(Se)_2$ (CIGS) thin film solar and challenges

Copper Indium Gallium Sulphide(Selenide) $CuIn_xGa_{1-x}S(Se)_2$ (CIGS) is another thin film solar cell that has found commercial production. However, limitation in supplies for indium and tellurium, and the wide fluctuation in prices of indium and tellurium present a big challenge in achieving low cost solar cell, Thus the development of a most promising material, Copper Zinc Tin Sulphide (CZTS), made of earth abundant materials as replacement for CIGS.

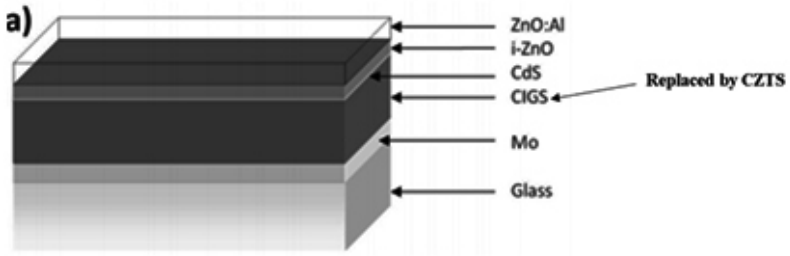



Fig. 8: CIGS based Solar cell (Ezealigo *et al.,* 2017)

CZTS is derived from the CIGS structure by the isoelectronic substitution of two In or Ga, atoms respectively by one Zn and one Sn atom. CZTS has the potential for availability, reproducibility in industrial scale, costs and environmental safety.

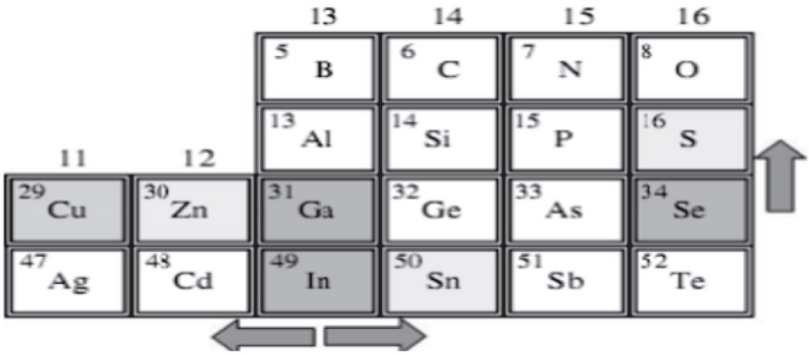



Fig. 9: A part of periodic table showing the logic of changing from CuIn(Ga)$Se_2$ (CIGS) and $Cu_2ZnSnS_4$ (CZTS) .

Theoretical calculations have shown that conversion efficiency as high as 32% is possible for CZTS solar cell (Ito and Nakazawa, 1988). Also calculated minimum raw materials cost concluded that the cost of raw material for CZTS PV technology is much lower than that of the existing PV technologies (Wadia *et al.,* 2009).

In the synthesis of CZTS, there is a competition between phase pure CZTS growth and the growth of binary [$Cu_2S$, ZnS, SnS, and $SnS_2$] and ternary phases [$Cu_2SnS_3$] counterparts as the material itself possessing complexity of this quaternary complex due to having three metal cation and one anion.

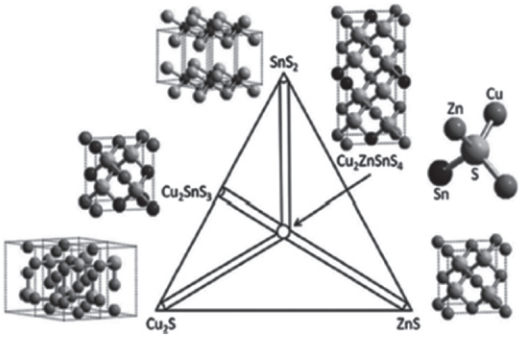


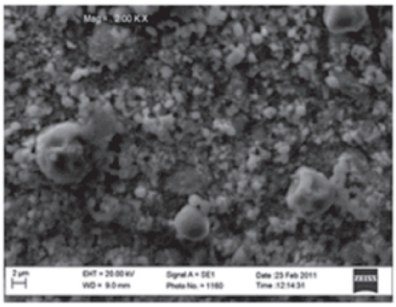

Fig. 10: The pseudoternary phase diagram to form the CZTS phase and other secondary phases along with their crystal structures. (Du *et al.,* 2014)

The CZTS phase formation is more complex and narrow in phase stability, as seen in (Irani *et al.,* 2016) Fig. 10, thus getting pure CZTS phase can be challenging

### 3.2.2.1 My Contributions

**(a) Sulfurisation of thermally deposited metallic precursors**

In this regard, Copper Zinc Tin Sulphide (CZTS) thin films were fabricated by two stages, sequential thermal evaporation of metallic precursors and subsequent sulfurisation. The resulting CZTS was investigated for their morphological, structural and optical properties. The temperature of substrates used was set at 150 °C throughout deposition. Sulfurisations were done for 3 hours at 550 °C with ramping rate of 10 °C/min. The composition ratios, the structural morphological, optical and electrical properties of CZTS thin films were investigated. Analysis of the X-ray diffraction films reveals the presence of tetragonal structure and cubic structure. The morphology of the CZTS thin film as obtained from SEM analysis reveals that the particles are nearly spherical in shape and distribution for the sulfurised sample.



(a)

| Sample | Cu/(Zn+Sn) | S/metal | Band gap (eV) |
|---|---|---|---|
| Zn_Sn_Cu_100 | 0.44 | 0.83 | 1.49 |
| Zn_Sn_Cu_150 | 0.92 | 0.83 | 1.51 |
| Zn_Sn_Cu_200 | 0.98 | 0.91 | 1.50 |
| Standard | 1.0 | 1.0 | ~1.50 |

(b)

Fig. 11: (a) Scanning Electron Micrograph (SEM) surface image and (b) metallic ratio and band gap (Isah *et al.,* 2013b)

The films show an increase in grain size with increase in Cu/(Zn + Sn) ratio, which indicates an enhancement of the grain growth under Cu – rich conditions. The optical band-gap of the CZTS samples has been estimated to be between 1.49 and 1.51 eV, appropriate for high conversion efficiency for solar-cell absorber (Isah *et al.,* 2013b).

***(b) Hydrothermal Synthesis of CZTS***

Apart from thermal evaporation electron beam evaporation is also used. The challenge however in use of such deposition system is the requirement for sophisticated system and high vacuum which eventually adds to cost.

We thus considered a one-pot chemical synthesis of CZTS, by hydrothermal method. It is a non-vacuum method that does not require any expensive precursors or equipment. It uses water as a solvent for the precursor chemicals with temperature raised above 100 °C in a sealed reaction container (an autoclave). The autoclave, normally consisting of a stainless steel shell with a Teflon liner as shown in Fig. 12

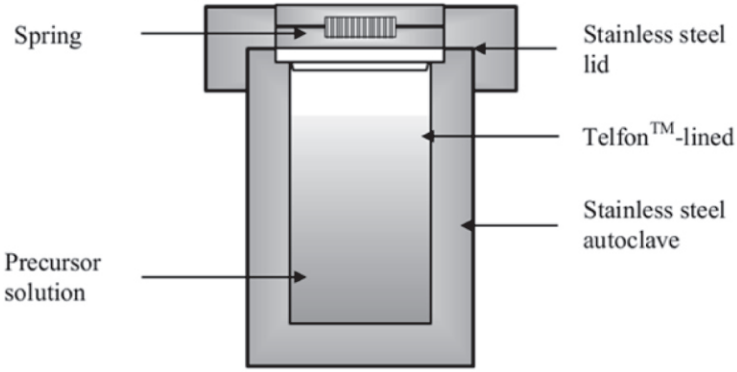
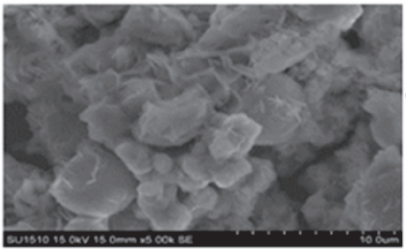
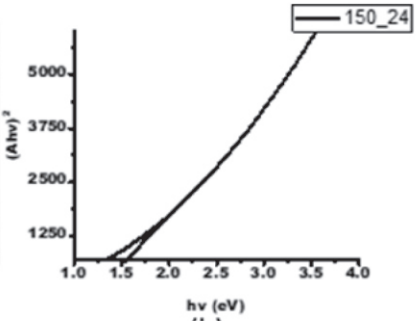



Fig. 12: A schematic diagram of hydrothermal synthesis setup (Habib, *et al.,* 2012)

Our contribution was optimising the hydrothermal deposition parameters of CZTS. The parameters considered were temperature, reaction duration (Mohammed *et al.,* 2019) and surfactant. In doing so different surfactants of Polyacrylic Acid (PAA), Polyvinylpyrrolidon (PVP), Polyethylene glycol (PEG) were used, and reaction temperatures of 150 °C, 190 °C, 230 °C and 270 °C and reaction times of (in steps of 40 o) and reaction durations of 12 h, 24 h, 36 h and 48 h were applied. The surfactant PAA, with reaction temperature of 150 °C and duration time of 24 h gave Cu: Zn: Sn: S atom ratios of ($Cu_{1.96}ZnSn_{1.14}S_{4.14}$) very close to stoichiometric ($Cu_2ZnSnS_4$) that had non-existing ZnS, SnS, $Cu_3SnS_3$, $Cu_3SnS_4$, $Sn_2S_3$ secondary and ternary phases.



(a)




(b)

| Sample | Cu/Zn/Sn/S |
|---|---|
| 150_24 | 1.96:1.00:1.14:4.14 |
| Ideal stoichiometry | 2.00:1.00:1.00:4.00 |

(c)

Fig. 13: (a) SEM image (b) band gap plot of the CZTS (c) elemental ratio (Mohammed *et al.,* 2019)

The scanning electron microscope showed flower-like particles which are composed of several nanospheres. The obtained band gap 1.5 eV was obtained, which is ideal for device application as absorber material in thin film solar cell

### 3.3 Third Generation

Third generation solar cells also referred to as 'emerging concepts' were introduced to solve the high costs of production in first generation, enhancing poor electrical performance of second generation while maintaining very low production costs. This new generation include dye-sensitised, Quantum dot and perovskite solar cells, organic photovoltaic solar cell (OPV) technology. They generally have not only low cost production but have potential for high efficiency. Our contributions were in DSSC and Perovskite solar cells.

#### 3.3.1 Dye Sensitised Solar cells

Dye sensitized solar cells (DSSCs) are low-cost alternative to photovoltaic silicon and thin film cells on the basis of materials and process costs. This cell is extremely promising because it is made of low-cost materials and does not need elaborate apparatus to manufacture. Even though its conversion efficiency is less than the best thin-film cells, its price-performance ratio is envisaged to be high enough to allow them to compete with fossil fuel electrical generation.

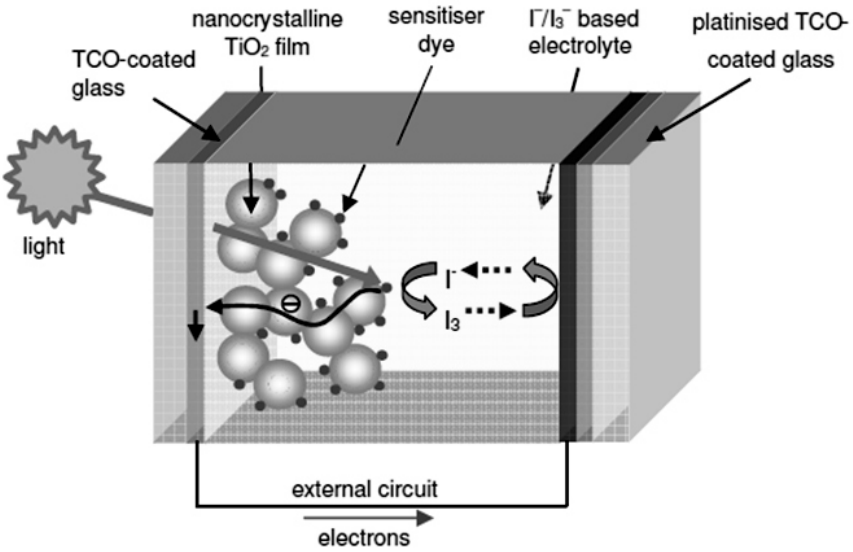



Fig. 14. Schematic of a liquid electrolyte dye-sensitised solar cell. Source: (Gratzel, 2003)

At the heart of the system is a ***mesoporous oxide layer*** composed of nanometer-sized particles usually $TiO_2$ (anatase). Attached to the surface of the mesoporous oxide is a monolayer of the charge transfer dye called ***photosensitiser dye.*** A photon is absorbed by the dye D (Photoexcitation) and excites an electron in the dye,

$$hv + D \rightarrow D^*$$

the photo-exited excited dye then injected into the conduction band of the mesoporous oxide semiconductor, the dye becomes ionised ($D^+$) and electron transport through the metal oxide film to the TCO-coated glass working electrode;

$$D^* + TiO_2 \rightarrow D^+ + e_{cb}^-(TiO_2)$$

The original state of the dye is then restored by electron donation from an **electrolyte**, such as the iodide/triiodide couple.

$$2D^+ + 3I^- \rightarrow I_3^- + 2D$$

The iodide is regenerated in turn by the reduction of triiodide at the **platinised counterelectrode** as follows,

$$I_3^- + 2e^- \rightarrow 3I^-$$

The circuit being completed via electron migration out of the device from the $TiO_2$ conduction band through the external load. (Grätzel, 2003).

### 3.3.1.1 My contributions

Our contribution in this regard are in use of natural dyes as **Photosensitisers** and **plasmonic** effects and synthesis of and alternative counter electrode possible cost reduction and enhancing the efficiency of solar cell

### A. Dye photosensitiser

The performance of the Dye sensitized solar cell depends much on the dye used as sensitizer, the absorption spectrum of the dye and the anchorage of the dye to the surface of $TiO_2$ photoelectrode. Basic Requirements for a dye are: (i) they should have panchromatic utilization (whole range of visible light) (ii) have high incident photon-to-current conversion efficiency (IPCE) (iii) Strong chemical bonding on semiconductor oxide surface (iv) Low cost. Synthetic ***Organometallic Dyes*** used mostly in DSSC though have the first three of requirements are rather expensive

Generally, **synthetic** transition metal coordination compounds (ruthenium polypyridyl complexes) are used as the effective sensitisers because of their intense charge-transfer absorption in the whole visible range (Qin, 2010) and highly efficient metal-to-ligand charge transfer. However:

(i) ruthenium polypyridyl complexes contain a heavy metal, which is undesirable from the point of view of the environment

(ii) the process of synthesizing the complexes is complicated and costly. Alternatively, natural dyes can be used as sensitisers with an acceptable efficiency.

The advantages of natural dyes over commercial synthetic dye are their availability and low cost, easy extraction and environmentally benign. These systems, being biocompatible/biodegradable are potential sensitizers, capable of delivering environmental friendly systems.

There are four classifications of natural plant pigments: chlorophyll, carotenoid, anthocyanin, and betalain, thus possible candidates used as photosensitisers in DSSCs. Our contributions were in:

### *(a) Chlorophyll and anthocyanin natural dye*

We looked at chlorophyll from pawpaw leaves and anthocyanin from flame tree flower and mixture of the two on the efficiency of Dye sensitized solar cell, from the ethanolic extract of chlorophyll from pawpaw leaves and anthocyanin extract from flame tree flower. The results show low efficiencies of 0.2 % for both chlorophyll and anthocyanin extract, however the cocktail of both gave improved photoelectric efficiency of 0.27 % (Kimpa et al., 2012), as shown in Table1.

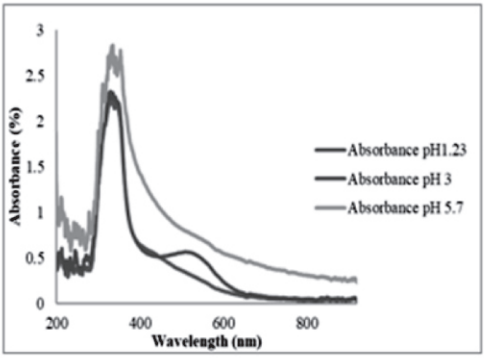
Table 1. Obtained photoelectrical parameters

| Dye | $I_{SC}$ ($mAcm^{-2}$) | $V_{OC}$ (mV) | FF | $\eta$% |
|---|---|---|---|---|
| Ruthenium | 7.4920 | 0.639993 | 0.4972 | 1.185 |
| Flame tree flower | 0.6680 | 0.503934 | 0.5880 | 0.200 |
| Dye cocktail | 0.74418 | 0.517958 | 0.6900 | 0.270 |
| Pawpaw leaf | 0.6490 | 0.503938 | 0.6053 | 0.200 |

(Kimpa *et al.,* 2012)

### *(a) Betalain natural dye*

Betalain is another interesting class of natural pigments, whose purified extracts from commercial sources have been subjected to photoelectrochemical study. Betalains consisting of the yellow betaxanthins and red–violet betacyanins absorb visible radiation over the range 476–600 nm. Betalains are characterized by high molar extinction coefficients in the visible region and pH-dependent redox properties. They are relatively stable over pH range 3–7. Acidic conditions seem to favour betalain-sensitized photo-electrodes with high optical densities capable of complete absorption in the visible range of 400–600 nm. Thus water extract of betalain pigment derived from red bougainvillea glabra flower used as sensitiser exhibited a pH dependence of the photoelectric permformance of fabricated DSSC




(a)

| Extract sources | $I_{SC}$(mAcm$^{-2}$) | $V_{OC}$ (V) | Efficiency (η%) |
|---|---|---|---|
| $pH_{1.23}$ | 1.13 | 0.43 | 0.21 |
| $pH_{3.0}$ | 3.72 | 0.59 | 0.98 |
| $pH_{5.7}$ | 2.27 | 0.61 | 0.57 |

(b)

Fig. 15. (a) UV-Vis absorbance an (b) obtained efficiencies (Isah *et al.,* 2015)

Dye extract at a pH of 3 displays broad absorption peak in the 480–560 nm range resulting from $\pi - \pi^*$ transitions due to the

mixed contributions of the yellow–orange betaxanthins (480 nm) and of the red–purple betacyanin (540 nm). At a pH of 5.7, the extract has a highest but flattened absorption spectrum in the long range wavelength indicating a suppressed absorption peak at this pH. The dye extracts at pH 1.23 and 3.0 have about the same absorption intensity in short wavelength range.

The DSSC at 1.13 pH had the lowest efficiency due to betanin degradation in very strong acidic environment with resultant inefficient light harvesting by the dye. At high pH betanin is poorly absorbed onto titania due to its inhibition by indicaxanthin competing with betanin for adsorption sites, thus the lower efficiency at pH 5.7 compared to 3.0 which had considerably higher efficiency (Isah *et al.,* 2015).

### *(c) Dye co-sensitisation*

We also exploited dye co-sensitisation, using a combination of two natural dye, anthocyanin and betalain. The result show a synergic photon absorption effect and thus higher efficiency as compared to when each dye was used alone (Isah, Sadik and Jolayemi, 2017).

## B. Modified Photoelectrode (Counter electrode)

### (a) Surface Plasmons and plasmonic enhanced Counter electrode

Noble metal nanoparticles (NPs) have widely been applied in various research fields, including optical sensors, catalysts, and surface-enhanced Raman scattering, due to their characteristic optical and electrical properties, not available with bulk material (Jing, Zhang, and Wang, 2013). Surface plasmons are the collective oscillation of electrons that propagate through a metal/dielectric interface. At the metal/dielectric interface, the electrons are excited by the electromagnetic field (photon or

light), and the interactions between surface electrons and electromagnetic field constitute the surface plasmon. When the incident photon wave-vector matches that of the surface plasmon, localized surface plasmon resonance (LSPR) results, leading to strong photon or light absorption.

On the basis of the LSPR effect, plasmonic nanostructures have been increasingly exploited to enhance the performance of photovoltaic devices, for which the mechanism has mainly been described to have a strong light scattering effect and local field enhancement. Recently, many efforts have been devoted to increasing the light harvesting efficiency by introducing plasmonic structures into various types of solar cells.
We synthesised plasmonic silver nanoparticles Ag NPs using Successive Ionic Layer Adsorption and Reaction- a wet chemical synthesis method similar but different from Chemical Bath Deposition (CBD).

SILAR-method for the deposition of thin films is based on sequential reactions of solvated ionic components on the solution-solid interface. Cation $[KL_p]^{n+}$ and anion $[AL'_q]^{m-}$ are reacting forming solid compound $K_mA_n$. The method requires that the solubility constant of the compound formed by the precursors K and A should be low enough. The overall reaction can be presented as follows:

$$m[KL_p]^{n+} + n[AL'_q]^{m-} \longleftrightarrow K_mA_n\downarrow + mpL + nqL'$$

SILAR is generally a four step procedure (cycle):

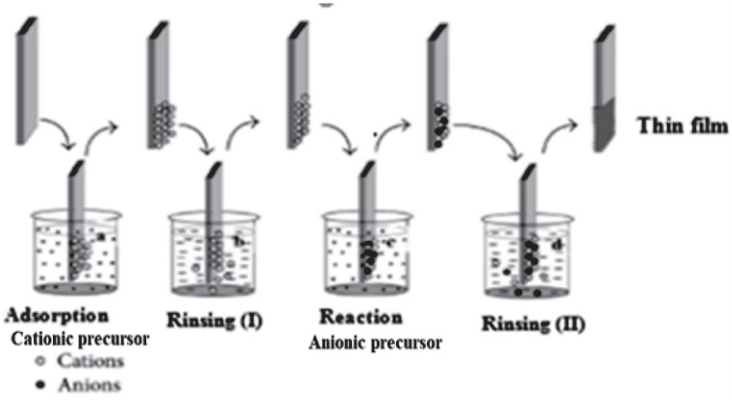



Fig. 16: SILAR processes (Pawar *et al.,* 2011)

**(I) Adsorption**:

In the first step of SILAR process, solvated cations $[KL_p]^{n+}$ present in the precursor solution are adsorbed on the surface of the substrate forming a double layer of inner (positively charged of the cation) and outer (negatively charged from counter ions of the cation)

**(ii) Rinsing (I):**

In this step excess unadsorbed precursor is rinsed away from the diffusion layer by deionized water resulting in saturated electrical double layer.

**(iii) Reaction:**

In the reaction phase the adsorbed saturated electric double layer is introduced into the solvated anionic $[AL'_q]^m$ precursor. This results in the reaction of the adsorbed saturated electric double layer with the anionic precursor to form the solid material $K_mA_n$

**(iv) Rinsing (II):**

The last step rinses the counter ions of both types of precursors as well as the reaction by-products out of the system

It offers simple, inexpensive and time saving procedures, which can be carried out at room temperature with no restrictions on substrate material, dimensions or its surface profile and the thickness of the film or nanoparticle can be easily controlled. We used Successive ionic layer adsorption and reaction (SILAR) method for the intercalation of Ag nanoparticles into the pores of mesoporous $TiO_2$ photoelectrodes of betalain-sensitized DSSCs. A unique, novel and first time report of synthesis of AgNps on $TiO_2$ by SILAR (Isah et al:, 2016) using silver diamine complex $[Ag(NH_3)_2]^+(aq)$ as the cationic precursor. The ionic equation of the complex ion is given as:

$$[Ag(NH_3)_2]^+(aq) \leftrightarrow Ag^+ + 2NH_3$$

Tin (II) chloride dissolve in in HCl is used as the anionic precursor

$$SnCl_2(aq) \leftrightarrow Sn^{2+} + 2Cl^-$$

which reduces $Ag^+$ thus

$$2Ag^+ + Sn^{2+}(aq) \rightarrow 2Ag(s) + Sn^{4+}$$

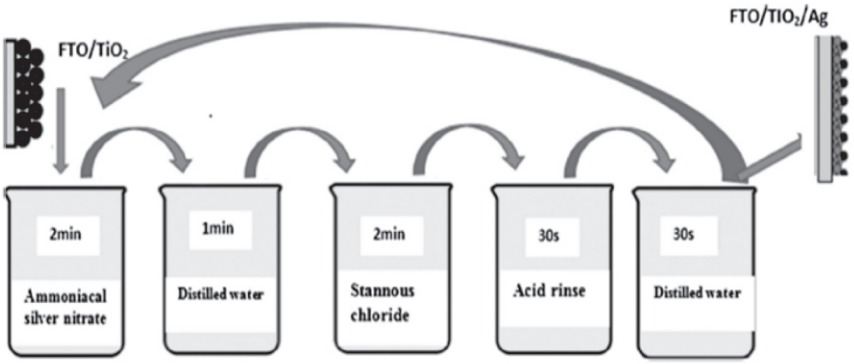
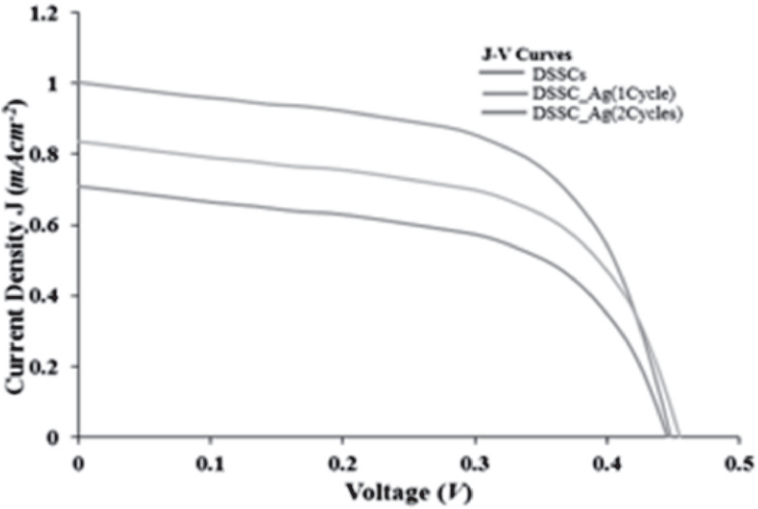



Fig. 17: $TiO_2$/Ag nanocomposite synthesis procedure (Isah *et al.,* 2016)

The fabricated DSSC solar cells from the AgNps plasmonic photoelectrode showed a 50 % increase in the solar cell efficiency for a one SILAR cycle and a 22% enhancement of efficiency as shown in Fig. 18.




(a)

| Photoanode | $J_{sc}$ ($mAcm^{-2}$) | $V_{oc}$ (V) | η (%) | % Increase in η |
|---|---|---|---|---|
| $TiO_2$ (bare) | 0.70 | 0.45 | 0.18 | 0 |
| $TiO_2$_Ag(1cycle) | 1.01 | 0.45 | 0.27 | 50 |
| $TiO_2$_Ag(2cycles) | 0.83 | 0.45 | 0.22 | 22 |

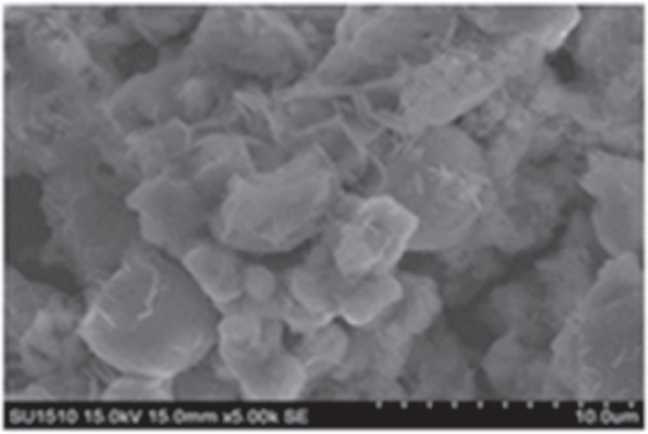
(b)

Fig. 18: (a) I-V characteristics and (b) table of obtained photoelectric parameters (Isah *et al.,* 2016)

**(a) CZTS as counter electrode**

The nanosphere-like shapes of the CZTS (shown in Fig. 18a) synthesised using hydrothermal synthesis were nanoporous

(from SEM micrograph) suggesting good candidates for use as counter electrode for DSSC.




(a)

| Sample | $J_{sc}$ (mA/cm$^{-2}$) | $V_{oc}$ (V) | Efficiency (%) |
|---|---|---|---|
| PEG based CZTS | 1.9 | 0.475 | 0.902 |
| PVP based CZTS | 3.8 | 0.475 | 1.490 |
| PAA based CZTS | 9.5 | 0.475 | 3.200 |

(b)

Fig. 19: (a) SEM image and (b) efficiencies of the CZTS electrode based DSSC (Mohammed, 2019)

The obtained CZTS were thus used as counter electrodes for DSSC with very good results, the PAA surfactant based CZTS does however showed better promise than those of PEG and PVP (Mohammed, 2019). The DSSC fabricated using PAA surfactant based CZTS had the highest efficiency

### 3.3.2 Perovskite solar cell

Even though continuous research efforts have contributed to the significant advances in the DSSC performance, leakage of liquid electrolyte posed some challenges inducing difficult issues for commercialization. This challenge is solved through substituting a solid hole transport material (HTM) for the liquid electrolyte without changing the basic concept of dye-sensitized solar cells to yield solid state dye sensitized solar cell (ssDSSC). However, to achieve high power conversion efficiency (PCE) from solid-state sensitized solar cells required new light absorber with an absorption coefficient greater than 0.5 x $10^4$ $cm^{-1}$. Perovskite-structured organometallic halides tend to possess such properties with ability to absorb light within a thin layer of about 2 μm. Thus the development of completely new solar cell architecture called Perovskite Solar cells (PSC).

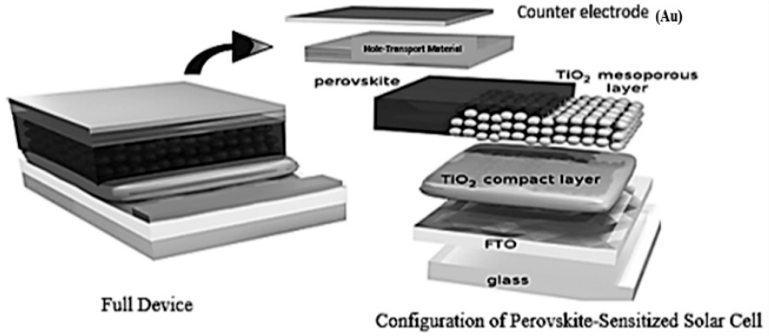



Fig. 20: Schematic Configuration of Perovskite-Sensitized Solar Cell (Juarez-Perez *et al.,* 2014)

PSCs have of late attracted significant attention due to the large diversity of low-cost processes existing to produce them, the versatility of structure and materials that can be used, its excellent absorption and high carrier diffusion length, and the fact that its power conversion efficiency (PCE) have dramatically improved to over 20% in a relatively short period, with prospects of exceeding the theoretical maximum efficiency for solar cells.

### 3.3.2.1 Contribution

We tapped into these frenzied efforts at looking at the various components of PSC with intent of cost reduction and improving on stability and PCE. We investigated the prospect of replacing expensive gold/silver with less expensive nanocomposite of multi-walled carbon nanotubes and platinum nanoparticles as counter electrode required for the regeneration of hole transport material (HTM).

The idea of using Pt-MWCNT nanocomposite is leveraged not only on the large surface area presented by MWCNT but its excellent electrical conductivity, thermal stability, its special tubular porous structure as well as the good electrical property of Pt.

Small amount of platinum nanoparticles was impregnated into carbon nanotubes, so as to have more space for light absorption on the cells and increase the efficiency of the cells. The Pt nanoparticles were evenly distributed on the outer surface of the carbon nanotubes as seen in Fig. 21.

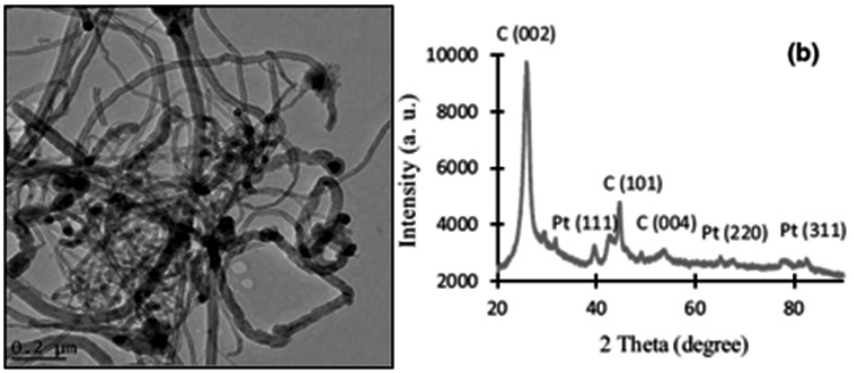



Fig. 21: (a) Pt nanoparticles evenly distributed on surface of the Carbon tubes (b) XRD results showing C and Pt peaks (Ibrahim *et al.,* 2020)

The electrical conductivity of the developed nano composite had conductivity between that of Pt and CNT (Ibrahim *et al.,* 2020). The results showed that the prepared nanocomposites will be a good electrode material in solar cell applications.

**Other contributions**

Among my other research work related to materials for solar energy application are the use of SILAR in deposition and studies of the optical properties of CdS (Isah, 2008a; Isah, 2008b), studies on $CuAlS_2$ thin films synthesis by thermal evaporation (Moreh *et al.,* 2013) and studies of thermally oxidized metal oxides (Isah *et al.,* 2013) and (Isah *et al.,* 2016).

## 4.0 CONCLUSION

The world is moving towards more of renewable energy and the country Nigeria cannot afford to be left behind. The United States for instance is already mooting the idea of having 40 % of its energy demand from solar energy by the year 2035. Other more advanced countries have similar targets.

The conversion of light into electricity in form of photovoltaics especially with abundant solar resource has the potential to supply much of the country's electricity demand apart from addressing the negative impact on environment from use of fossils fuels energy generation. Also with significant room for cost reductions, from new and cheaper materials, low cost materials synthesis procedure and improvement in conventional materials, solar technology can increase its contributions to the reliability and resilience of the power grid. Thus basic fundamental research plays a great role in achieving the main objectives of pervasive photovoltaic usage. In short, materials science (physics) plays a critical role in achieving the goals of low cost, efficient and renewable sustainable energy source and in understanding device fundamentals based on materials research.

Generally, materials scientists, (materials physicists, materials chemist and materials engineers) are still significantly low. Thus, the need to have more materials scientist to solve our technological challenges is paramount. Though in general Government does spend huge amount of resources in sending scientists abroad for training, however, failure to provide adequately paid employment (low salaries) and facilities when they return home, have led to challenges of proper re-assimilation on return home. This challenge has led to many overseas trained graduates remaining in the developed countries of training to the detriment of their home country. The long term solution is to improve status and facilities in the country. With provision of improved facilities training can be carried out in the country, rather than in developed countries, curbing the challenge of re-assimilation on return home.

Due to competing demands for fund from government, it is suggested that specialized facilities and equipment be concentrated in a few sites across the country (research centres or facilities centre) for not only efficient utilisation but also proper maintenance by specially trained hands. Maintenance is a real problem, as is the case in laboratories in developing countries with much unserviceable equipment, and no one available to carry out even minor repairs. Thus, the imperative for such specialised research centres, with trained requisite manpower. This can be more effective, as the centres should be large enough to have their own workshop and repair facilities. Such centres should also have a broad technological focus which serves to promote a common interest among individuals from different scientific and engineering disciplines so as to engender collaborative multi-field research efforts. This will create an effective meeting ground for professionals of different disciplines for joint research programmes to be initiated and good research results obtained through cross breeding of ideas.

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**Peace and blessings of Allah's be on the most luminous lightening that heralds the profitable rain-clouds which fills all intervening seas and receptacles.**

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# Brief Profile of the Inaugural Lecturer

Prof. Kasim Uthman Isah was born in Bida, Niger State on 11th Sepatember 1965, to the family of Alhaji Usman Isah (Usman Maidariya), a pioneer educationist in Northern Nigeria and Hajiya Zainab.

He attended Jinadu Anglican School, Falomo, Lagos and the Saint John's Primary School, Bida for his primary education. He went to Government Secondary School, Minna between 1977 and 1982. He attended the Ahmadu Bello University for his BSc (Hons) in Physics and PhD in Physics (Solid State Physics). His Master's degree was at the Federal University of Technology, Minna.

He joined the Federal University of Technology on the 7th of Januarry1991 as a Graduate Assistant, after a two-year stint at the College of Education, Minna as an Assistant Lecturer. He rose through the University system to become a Professor of Physics (Solid State Physics) in 2016.

He has held various positions of responsibility such as Departmental Examination Officer, 2000-2002; Sub-Dean, 2009-2010; Head of Department of Physics, 2010-2014; Acting Dean School of Physical Sciences, 2018-1019; member of University Governing Council, 2013-2017 and presently the Director, Anti-corruption and Transparency Unit of the University. He has also served as member/chairman of numerous committees.

In the course of his academic activities he has supervised 7 PhD students both within and outside Federal University of Technology Minna and many Masters students.

He has served severally as PhD external examiner to the University of Lagos; Federal University of Agriculture, Makurd; Nigerian Defence Academic, Kaduna in Nigeria; the University of Pretoria, and the University of Witwatersrand, Johannesburg, both in South Africa.

He has also served as Associate Professor and Professorial Assessor to Kano State University of Science and Technology, Wudil, Kano State; Ahmadu Bello University, Zaria; Federal University, Lokoja; Usmanu Danfodio University, Sokoto; Gombe State University, Gombe; and Federal University, Gusau.

Prof. Isah has participated and presented research papers in several conferences and workshops in Nigeria, Tunisia, Zambia and Rwanda and has served as member of editorial board of a number of Journals in Nigeria and outside Nigeria, aside from having successfully organised two International Conferences for the School of Physical Sciences, Federal University of Technology, Minna. He recently won a multimillion naira research grant from the Petroleum Technology Development Fund, to further our research on perovskite and plasmonic enhanced solar cell.

He is a member of the Nigerian Institute of Physics (NIP) and the Materials Research Society (MRS).

He is happily married and the marriage is blessed with four children.