Review Article

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Important features, applications and future perspective of zinc oxide based reduce graphene oxide nanocomposite

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Abstract: The Sustainable Development Goals (SDGs) play key role in propelling transformative changes, and highlighting burning issues to be addressed to achieve successful global developmental initiative. Zinc oxide (ZnO) and reduced graphene oxide (rGO) are helpful in achieving various SDGs. However, both materials have some limitations. To overcome the drawbacks of individual ZnO and rGO nanomaterials, ZnO/rGO nanocomposites are designed. This review aims to highlight issues being faced by ZnO and graphene oxide and their resolution through the development of ZnO/rGO nanocomposite. Various characterization techniques are discussed to explore physio-chemical properties. As the composite materials exhibit enhanced charge carrier separation, and extended lifetime of photoinduced charge carriers, making them a promising agent for improved photocatalytic degradation of pollutants, biosensors, and energy conversion and storage devices such as solar cells, and lithium batteries etc. Morphological relationship with various activities and potential applications in numerous fields, including controlling environmental pollution, biomedical, bio-sensing, energy etc., have also been discussed to explore their importance. Additionally, for further advancement to design next generation material several recommendations have been given. The knowledge gained from this review is the way for the development of next-generation nanomaterials, capable of addressing global challenges in energy and environmental sustainability etc.

Keywords: solar cell; lithium ion battery; water pollution; biomedicine; machine learning; 3D and 4D printing of nanomaterials

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1 Introduction

The Sustainable Development Goals (SDGs) agenda is presented for the better and sustainable future of all human beings, which covers the smooth time scale protocols from 2015 to 2030 by the United Nations.[1] The SDGs focus on addressing global challenges, such as environmental issues, poverty, inequality, climate change, and social injustice, etc. to provide equal access to sustainable life.[2] For effective sustainable life development, these goals focus that research and comprehensive understanding of material science are essential. Various stakeholders have endeavored to grasp and apply the SDGs in creating new initiatives that address each goal.[3,4] In last decades' nanomaterials with their unique properties have been renovated in various industries. They are being used in healthcare, water treatment, and sustainable resource management. Among them binary nanomaterials cover many area of SDGs as these material revealed promising applications in various field including environmental cleanup, electronics, gene therapy, targeted drug delivery[5] and agriculture (Zero hunger)[6,7] due to their high surface area, reasonable band gap energy, and chemical reactivity.[5,8]

Similarly, zinc-based reduce graphene (ZnO/rGO) nanocomposites which is binary nanomaterials play a crucial role in addressing several SDGs such as good health and well-being (SDG 3), clean water and sanitation (SDG 6), affordable and clean energy (SDG 7), industrial innovation and infrastructure (SDG 9), responsible consumption and production (SDG 12).[2,9] The ZnO/rGO nanocomposites exhibit high photocatalytic properties in which ZnO has a high surface area and rGO has high electron transport capabilities. This synergistic effect makes them effective for removing pollutants, including heavy metals and organic dyes, from waste water.[9] The ZnO/rGO have excellent applications in SDG 3 such as antimicrobial, antioxidant, cytotoxicity for good health and wellbeing.[10] Moreover, it has promising application in SDG 7 such as solar cells,[11] Super capacitors,[12] lithium and Sodium ion batteries.[13]

In this review, we discussed different issues of ZnO, GO and rGO nanomaterials. The important characterization techniques which explore various aspects of the ZnO/rGO nanocomposite, such as Fourier transform infrared (FTIR) spectroscopy, Raman spectroscopy, X-ray diffraction (XRD), UV-visible spectroscopy, scanning electron microscopy (SEM) and transmittance electron microscopy (TEM) are deliberated in detail. The role of ZnO/rGO nanocomposite in SDGs, their applications in various fields including controlling environmental pollution, role in solar cells and batteries, bio-sensing and biomedical applications etc. have been explored to highlight their importance and future implications. In the last part of the review, the morphological relationship with various activities and future recommendations has also been discussed.

2 Zinc oxide

The study of one-dimensional and two-dimensional materials has been an emerging field in nanoscience and nanotechnology. ZnO nanomaterials have attracted significant interest because of their remarkable properties in photonics, optics, and electronics. The synthesis of ZnO thin film nanomaterial has been a booming field since the 1960s due to its potential uses as catalysts, transducers, and sensors.[14] Its unique physical and chemical properties including electrical, mechanical, chemical and optical properties, the ZnO listed multifunctional material.[15,16] ZnO may be present in one, two and three dimensions and depicted different structures.[17–19] Many metal oxides with wide band gaps like TiO_2,[19] ZnO[20] and SnO_2[21] are usually used as photo-anodes for dye sensitized solar cells (DSSCs). Wide band gap, high optical transparency, and numerous morphologies such as nanorods,[22] nanoflowers,[20] nanotubes, and nanowires[23] make ZnO one of the most interesting materials to be used for photo-anodes in DSSCs. ZnO has been identified as a battery active material with high energy density of 650 Ahg^{-1}. Its good electrical conductivity (up to 230 cm^{-1}) is also greater than the oxides of other transition metals. Ridhuan et al. prepared ZnO electrode materials for solar cell by hydrothermal method which exhibit excellent physiochemical properties.[22] ZnO nanoparticles have been extensively investigated for their potential as antimicrobial agents, particularly in enhancing their effect against pathogenic bacteria and viruses. These particles possess unique physicochemical properties that significantly influence their interactions with microorganisms and exhibit their biological and toxicological responses.[23] ZnO NPs are becoming more widely used in agriculture due to their potential to boost crop production and improve plant nutrition. These nanoparticles serve as nano-fertilizers, aiding in better seed germination, stronger plant growth, and improved plant development. These nanoparticle also help in controlling plant diseases and contribute to healthier soil conditions due to their antimicrobial characteristics.[24] ZnO NPs exhibit excellent anticancer activity by producing reactive oxygen species (ROS), which damage the mitochondrial membrane of cancer cells. This disruption triggers a chain reaction involving caspase enzymes, causing cell death, or apoptosis, in the cancer cells.[25]

2.1 Limitations of zinc oxide nanomaterial

Despite potential applications in many fields, ZnO nanomaterial is facing some issues which limiting their uses in various field. For example, in case of heterogeneous catalysis hydrogen defect occur, causing a variation in the ratio of zinc to oxygen ratio. The presence of empty oxygen sites can lead to trap one or two electrons.[26,27] Furthermore, intrinsic defects present, such as donor and acceptor defects, and their energies lie in the range of ~0.05–2.8 eV.[28–30] The two most common forms of ionic defects are known to be oxygen vacancies and zinc interstitials as illustrated in Figure 1. Both

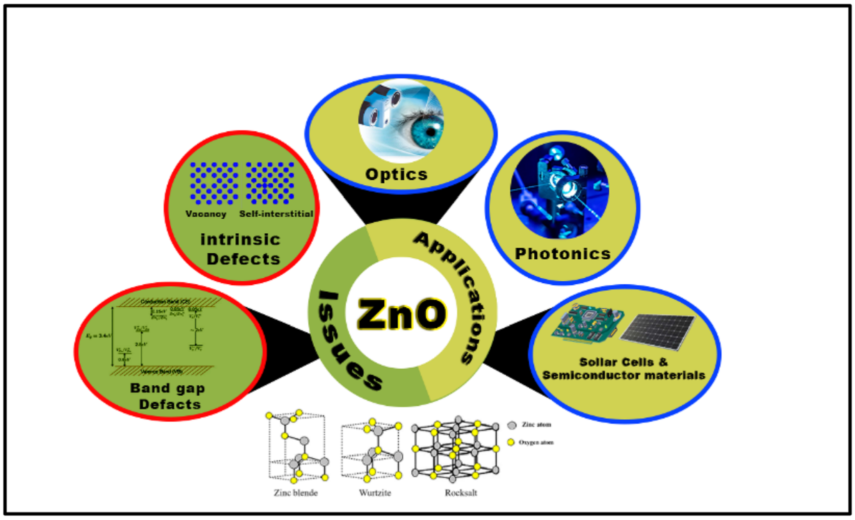

Figure 1: Applications and issues of ZnO nanoparticles.

defects donate two electrons and it is not easy to distinguish one from the other using electrical measurements.[29,30] Due to its broad band, such as 3.37 eV, high bond energy 60 meV, high thermal and mechanical properties that lie in-between ionic and covalent semiconductor material.[31,32] ZnO nanoparticles have potential applications as an anticancer agent, there are still several challenges that limit their broader application. Such as potential toxicity to healthy cells, not specific to targeted cancer cells, and difficulties in managing how the nanoparticles are released and behave inside the body[33,34] ZnO NPs can also be used in agricultural but the excessive use may cause phyto-toxicity, harming plant tissues, and disrupting essential physiological processes.[35,36]

3 Graphene oxide

Graphene is an allotrope of carbon arranged in a two-dimensional honeycomb lattice, exhibits excellent properties that make it a revolutionary material. Graphene has high electrical conductivity (about 1010 S/m),[34] and a remarkable carrier mobility (about 20 m^2/Vs).[37] Its thermal conductivity is up to 5300 W/mK,[38] alongside an intrinsic tensile strength of 130 GPa and a Young's modulus of 1,100 GPa, making it incredibly strong and stiff. Graphene exhibit high specific surface area (SSA) of 2,620 m^2/g and outstanding transparency, allowing ~97.7 % of light to pass through, further enhancing its potential for diverse applications.[39] Over the last decade, graphene oxide (GO) has attracted much attention from researchers due to its excellent mechanical, electrical, thermal, and optical properties. Apart from that GO can be prepared easily from raw material such as graphite which is cheap and readily available.[40] Based on structural point of view graphene and GO are almost similar but in GO the carbon plane is decorated heavily through oxygen containing functional groups, which makes the GO hydrophilic in nature.[41] GO nano-sheets are the most inspiring material for their potential electrode supporting materials in super-capacitors and other electrochemical energy storage devices. The inhibition of accumulation and restacking is of highest significance to enhance the use of graphene based nanomaterials in energy storage devices.[41,42]

GO can be converted to reduced graphene oxide (rGO) which lead to removal of oxygen containing functional groups and restore the sp^2 bonding network. This restoration of sp^2 bonding improves the electrical conductivity and hydrophobicity of rGO, making it a promising material in various applications compared to GO.[43] GO can be reduced through different methods such as micro-mechanical exfoliation and chemical vapor deposition etc. to rGO.[40] The cost-effectiveness and higher production capacity approach for developing good-sized rGO makes it widely usable instead of GO.[44] As rGO is mostly exposed to aggregation and agglomeration problems because of the Van der Waals interactions, result in a considerable loss of effective specific surface area and reduces capacitance. Therefore, much attention has been given on improving the specific surface area of rGO in view of the large-area applications in super capacitors.[44] The distinct properties of GO and rGO make them promising material for wide spread applications. In the term of applicability GO having rich oxygen containing surface functionality, highly suitable for various application including bio-sensing, drug delivery and waste water treatment to absorb various pollutants.[45] These oxygen containing functional groups increase hydrophilicity, making it highly dispersible in aqueous media. In contrast, rGO having partially restored sp^2 hybridization and graphene-like structure with fewer oxygen containing functional group, exhibits enhanced electrical conductivity.[46] This enactment in electrical conductance making rGO more favorable for use in energy storage devices such as super capacitors and batteries, sensors and photo- and electro-catalysis.[40,46] In general, the choice be subjected to the application, but rGO is frequently preferred in energy related field due to enhanced conductivity and stability.[47,48] GO is favored in biomedical applications such as bio-sensing and drug delivery because of its surface functionalities and biocompatibility.[49,50]

3.1 Limitations of graphene oxide and reduce graphene oxide nanomaterial

GO formed a two-dimensional stable crystal that exhibits unusual electronic properties like the anomalous quantum Hall effect and remarkably excellent mobility of charge carriers at room temperature.[36,40,51] Apart from this, the oxygen functional group, such as hydroxyl, epoxy (1,2-ether), carbonyl, and carboxylic acid, in the GO structure, makes it more reactive with other substances, as shown in Figure 2.[52,53] The sp^2 hybridization of carbon atom are **disrupted** due to the presence of oxygen atoms which alter to enhance the insulating properties of GO.[39,54,55] rGO also contains some issues including it can Stone-Wales defects (pentagon/heptagon bonded carbon atom network), in which carbon atoms form pentagon and heptagon shapes instead of hexagons. These defects significantly affect its physical and chemical properties of rGO.[56]

4 Zinc oxide based reduce graphene oxide nanocomposite

ZnO/rGO nanocomposites are basically formed to resolve the issues of ZnO and rGO. They are useful in many fields including environmental challenges, biological problems, energy storage and conversion issues etc.[24,25] as shown in Figure 3. They are widely used as an electrode material in batteries because of their characteristic two-dimensional nanostructure, exceptional electrical and thermal conductivity. They have extraordinary theoretical SSA, unique mechanical strength and decent electrochemical stability.[57] They exhibit a wide range of biological applications such as strong antimicrobial and antioxidant activities, as well as effective drug delivery potential. They are useful in bio-imaging for medical diagnostics and non-biological applications such as food packaging and sunscreen products.[58] It can also be used in the agriculture field by promoting plant growth, enhancing nutrient uptake, and serving as an effective antimicrobial agent to protect crops. Due to its high surface area and adsorption capacity, it is a valuable material for soil remediation, particularly in removing heavy metals and improving soil quality for sustainable farming applications.[59]

4.1 Characterization techniques and features

For confirmation of ZnO/rGO nanocomposites formation, different characterization techniques are used including

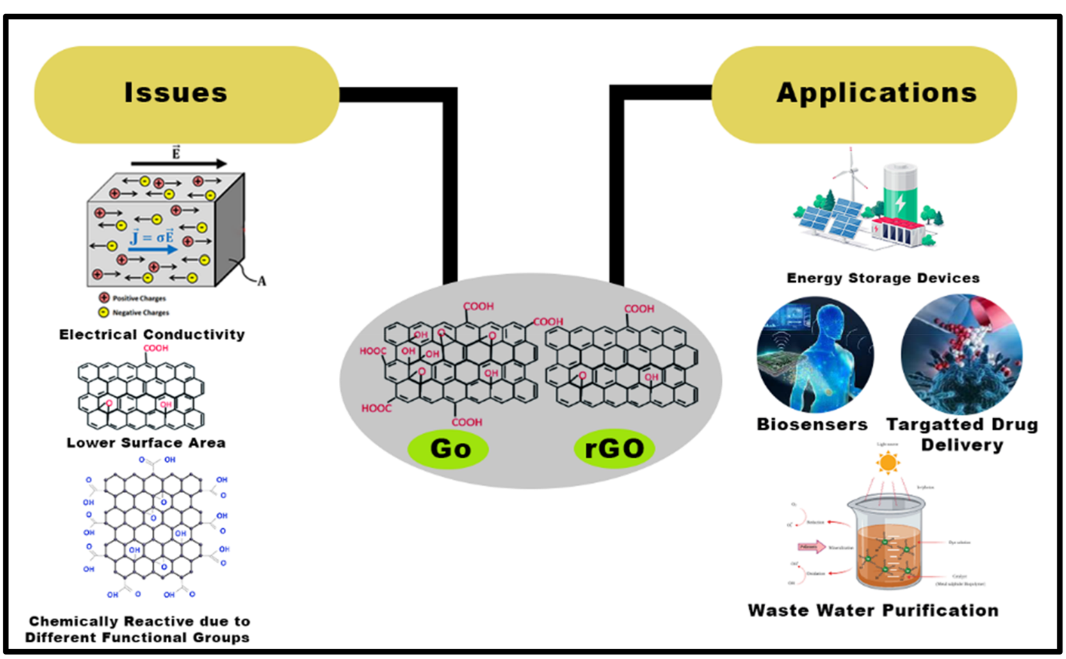

Figure 2: Applications and issues of GO and rGO.

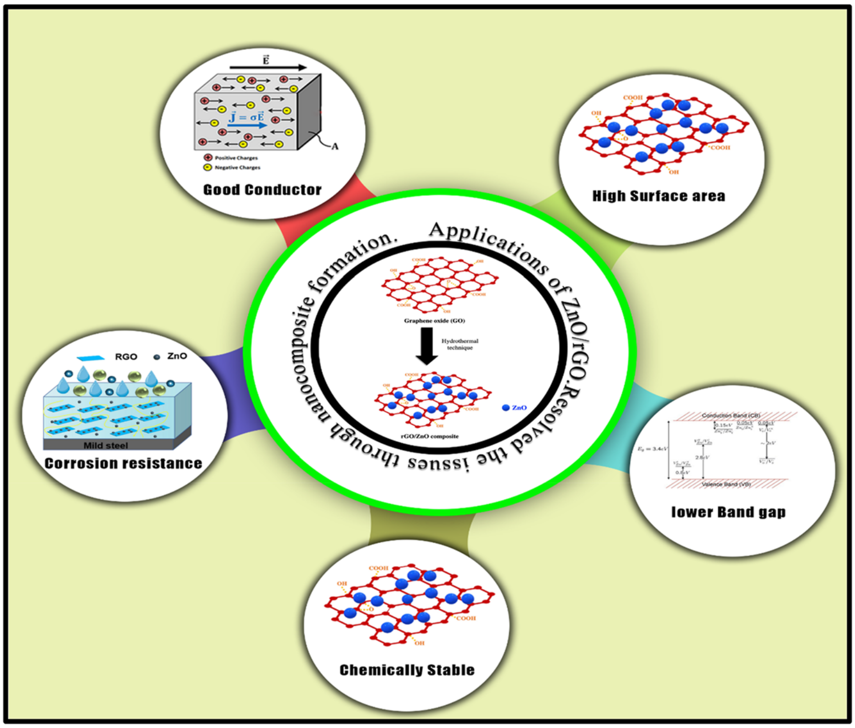

Figure 3: Applications ZnO/rGO nanocomposites.

XRD analysis, UV–visible spectroscopy, FTIR spectroscopy, Raman spectroscopy, SEM, TEM etc. Through these techniques various features of the nanocomposites can be explained. Some of the techniques which explore different features are explained in below:

4.1.1 XRD analysis

The XRD analysis is commonly used for the confirmation of the nanomaterials' crystalline nature and new phase formation.[60] Similarly, it is also used to confirm crystallinity and new phase formation of ZnO/rGO nanocomposite. For that purpose, ZnO, Graphite, GO, rGO and ZnO/rGO nanocomposite XRD diffractograms are obtained and then analyzed. The diffractograms of graphite, ZnO, GO, rGO and ZnO/rGO are presented in Table1, The presence of ZnO is confirmed by characteristic peaks corresponding to the hexagonal wurtzite structure of ZnO. These peaks appear at Bragg angles (2θ) of 31.7°, 34.4°, 36.2°, 47.60°, 56.6° and 62.9° corresponding to the (100), (002), (101), (102), (110) and (103) planes, respectively, which are characteristic of ZnO's hexagonal phase. Additionally, rGO's presence is confirmed by peaks at 24.5° corresponding to (002) plane of the hexagonal carbon structure. The presence of ZnO and rGO peaks in the diffractogram, revealing the successful formation of the nanocomposite. The degree of crystalline can be concluded from the intensity and sharpness of peaks. Commonly, ZnO crystallizes in two form: hexagonal wurtzite and cubic zinc-blende. Literature study have shown that ZnO mostly exist in wurtzite structure which is thermodynamically more stable. It also exists in zinc blende structure which is metastable and typically form in specific growth conditions such as in thin film or nanostructure. Moreover, according to Debye-Scherrer formula (Eq. (1)), the crystallite size of ZnO nanoparticles can be calculated from the most intense peaks.[60,61]

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (1)$$

Where D, K, λ, β and θ are the average crystallite size, constant (0.89), wavelength of the X-ray radiation (1.54 Å), full width at half maximum of peak and diffraction angle, respectively.

Zinc sulfide (ZnS), Cadmium Sulfide (CdS), Gallium nitride (GaN), cadmium telluride (CdTe), Zinc selenide (ZnSe) and gallium arsenide (GaAs) have similar structure with ZnO. ZnS, CdS, and GaN have wurtzite hexagonal structure while CdTe and GaAs show cubic zinc-blende structure.[62–64] Although, these NPs have similar structure with ZnO, but can be differentiated from each by various characterization techniques such as XRD, EDX, XPS and UV–visible spectroscopy. The XRD pattern of these NPs shows different characteristic diffraction peaks from each other. XPS and EDX techniques can be employed to confirm the elemental composition and oxidation states that help to determine the nature of given NPs. Additionally, UV–visible spectroscopy can also be used to differentiate these NPs

Table 1: Graphite, graphene, graphene oxide, reduced graphene oxide, zinc oxide and ZnO/rGO nanocomposite characteristics PXRD peaks.

Sample	XRD Peak at 2θ	Peak at corresponding plane	Crystal size (nm)
Graphite	26.7°	002	–
ZnO	31.7°	100	20.5
	34.4°	002	
	36.2°	101	
	47.60°	102	
	56.6°	110	
	62.9°	103	
	67.9°	200	
GO	10.26	001	–
	42.5	100	
rGO	24.5°	002	19.9
ZnO/rGO	26.5°	100	12.5
	31.7°	002	
	34.5°	101	
	36.3°	102	
	47.6°	110	
	56.7°	103	
	62.9°	200	
	68.2°	112	

because of their different band gap energies: 3.3, 3.6, 3.4, 2.4, 1.45, 3.10, and 1.42 eV for ZnO, ZnS, GaN, CdS, CdTe, ZnSe, and GaAs, respectively.[65–68]

4.1.2 UV–spectroscopy

The UV–spectra analysis is use for the determination of optical properties of semiconductor materials, the optical properties are exploit for different purposes such as solar cells devices fabrication as well as for calculation of band gap. Furthermore, this technique is used to find the purity of corresponding material.[69] Graphite, graphene, graphene oxide, reduced graphene oxide, zinc oxide and ZnO/rGO nanocomposite characteristics UV spectroscopic peaks are depicted in Table 2. In ZnO/rGO composite the peak observed at 303 nm which is because of $\pi \rightarrow \pi^*$ transition, this increase in absorption occur due to the contribution of rGO, due to which the surface charge increase, and this shifting of band leads to narrow band gap.[42,69,70] The band gaps of rGO and GO are 1.69 and 2.20 eV,[71] respectively. The low value of rGO is due to the reduction of some oxygen groups. The bandgap can thus be adjusted by managing the oxygen present in rGO. The energy bandgap value for pure ZnO is found to be 3.37 eV, and combined with GO, it increases to 3.63 eV. It has been observed that the excitation energy of ZnO nanoparticles increases with decrease of grain diameter according to the Kubo theory. The blue shift

Table 2: Graphite, graphene oxide, reduced graphene oxide, and rGO/ZnO nanocomposite characteristics, UV spectroscopic peaks.

Sample	Absorption bands (nm)	Transitions	References
Graphite	270	C=C	[74]
GO	230	C–C	
	300	C=C	
rGO	260	C–C	
rGO/ZnO	303	$\pi \rightarrow \pi^*$	[42,69,70]

of the ZnO bandgap in GO-ZnO can be attributed to the decrease in the size of ZnO nanoparticles. In contrast, rGO-ZnO exhibited a 3.25 eV band due to the increasing surface charge between ZnO and rGO. This led to the optical band gap shifting to a higher wavelength.[69,71]

4.1.3 FTIR spectroscopy

Infrared spectroscopy is a very important technique; it is use to characterize the vibration mode of molecules. These vibration modes are appearing due to different groups, elemental composition and bonding arrangement.[72] This technique is also used to analyze ZnO, GO, rGO, and ZnO/rGO their functional groups. The distinctive characteristics peaks of the analyzed material were observed at a specific point from where the nature and different functional groups were identified.[72–74] The ZnO, GO, rGO, and ZnO/rGO distinctive peaks are listed in Table 3.

4.1.4 Transmission electron microscopy and scanning electron microscopy

TEM is used for the determination of morphology, identifying structural defects, phase boundaries, and crystallographic information of nanomaterials.[75] Likewise, it is also used to determine the morphological characteristic of ZnO, rGO and ZnO/rGO nanocomposite.[76] The rGO have morphology like thin flat flakes which suggest that the rGO retained its graphene-like characteristics while also having some degree of structural deformation due to reduction process. This combination of flat and crumpled morphologies is typical for rGO and contributes to its unique properties and potential applications.[77] In ZnO/rGO nanocomposite, the spherical ZnO nanoparticles disperse uniformly in the smooth and flat nanosheet of the graphene oxide surface. Base on TEM analysis the dark region in the ZnO/rGO composite, the reduce graphene oxide surface act like a two-dimensional platform which indicates the uniform incorporation of ZnO nanoparticles on rGO surface.[75–77] SEM explains the surface morphology of

Table 3: Graphene oxide, reduced graphene oxide, zinc oxide reduce graphene oxide ZnO/rGO nanocomposite characteristics FTIR spectroscopic peaks.

Sample	Absorption bands (cm^{-1})	Vibrations	References
ZnO	3,407	O–H	[81]
	480	$Zn–O_{stretching\ and\ deformation}$	
	1,350, 1,600	–COOH stretching	
	2,830, 2,950	C–H	
	3,400	–OH	
GO	3,620–3,400	O–H	[75–81]
	2,963	$C–H_{(sym)}$	
	2,341	$C–H_{(Asym)}$	
	1,653, 1,426	$C{=}C_{(aromatic)}$	
	1,032–1,263	C–OH	
	600–914	C–O–C	
rGO	3,700–3,000	–OH	[81]
	1,650	cyclic hexagonal symmetry	
ZnO/rGO	480	Zn–O	[72,82]
	1,350, 1,600	$–COOH_{(stretching)}$	
	1,115	C–O	
	2,950, 2,850	C–H	

nanomaterials.[83] SEM confirmed ZnO attachment on the surface of rGO sheets as well as their morphology. SEM micrographs revealed that rGO sheets were twisted, folded and stromatolithic sheets with 10–25 µm height and 4–7 µm thickness.[84]

4.1.5 Raman spectroscopy

Raman spectroscopy is an important technique use to determine vibrational information, electronic and phonon properties as well as defects and impurities in a nanomaterial or compounds.[85] The Raman spectra analysis revealed, when carboxyl, hydroxyl, and carbonyl groups are removed from rGO during reduction it made up of partly healed carbon basal planes which exhibit two significant defect band (D-band) as well as graphite band (G-bands). The G-band in the sp^2 carbon atom is due to the first-order scattering of the E_{2g} mode in the hexagonal structure. The D-band is associated with the sp^3 hybridization of edge defects on the carbon basal plane.[86] The band of D-peak was observed at 1,360 cm^{-1} and G-peak at 1,581 cm^{-1}, though the D-peak observed at 1,360 cm^{-1} is caused by disorder and flaws in the hexagonal carbon structure. In chemically reduced rGO, the position of D-peak stays the same, while G-peak moves from 1,590 to 1,581 cm^{-1}, possibly as a result of rGO's self-healing properties, which allow it to rebuild its defective carbon atoms' hexagonal structure.[87] In case of ZnO/rGO the disorder and graphitic peaks observed at 1,355 and 1,581 cm^{-1}, whenever the ZnO is added to nanocomposite, both the D and G peaks shifted. Furthermore, when the intensity ratio of D to G-peaks is determined, less increasing of ZnO peaks is found, indicating that ZnO accumulated in rGO.[86,88] Table 4 displays the absorption bands of ZnO, GO, rGO and ZnO/rGO.

4.2 Applications

Many applications of GO, rGO and ZnO nanomaterials have been reported in the literature. However, these nanomaterials have also some limitation that pose problem in their exploration. The limitation of nanomaterials can be resolved through nanocomposite preparation, as a synergistic effect arising from both materials. To encounter the limitations, the ZnO/rGO nanocomposite are prepared which revealed better performance than the corresponding starting material. Some of the applications of the nanocomposite are discussed below:

4.2.1 Photovoltaic cells

Due to the immense increase in the world's population, there is always a great demand of energy because of the rapid consumption of fossil fuels (petroleum, natural gas, and coal).[89–92] Fossil fuels are not environmentally friendly; therefore, the development of green and clean renewable energy sources has become the main focus of recent research. Solar energy is an inexhaustible energy that has become one of the most economical and encouraging renewable energy. DSSCs and organic semiconductor solar cells are the third-generation solar cells after the initial silicon solar cells and semiconductor thin film solar cells.[11] DSSCs are a more effective and attractive alternative technology because of its low cost, easy manufacturing and

Table 4: Graphene oxide, reduced graphene oxide, zinc oxide rGO/ZnO nanocomposite Raman spectra analysis.

Sample	Absorption bands (cm^{-1})	Characteristic peak	References
ZnO	344	D-peak	[93]
	451	G-peak	
GO	1,344	D-peak	[89]
	1,587	G-peak	
rGO	1,360	D-peak	[85]
	1,590	G-peak	
ZnO/rGO	1,355	D-peak	[90]
	1,581	G-peak	

engineering as mentioned in Figure 4.[94,95] One of the most vital constituents of DSSC, the photoanode performs many functions, including supporting dye molecules, light absorption and radiation, and providing suitable energy levels for transferring charge carriers.[96]

ZnO/rGO nanomaterials have excellent photocatalytic applications because of their outstanding physical, chemical, and photo-electrochemical properties. The π-conjugation structure of rGO enhances the photocatalytic performance of ZnO by improving its ability to absorb light and separate charges, which allows excellent electron mobility and prevents the recombination of electron-hole pairs on the ZnO surface and increases its efficiency in capturing light, particularly in the visible spectrum.[88,97] This hybrid structure has significant applications in enhancing the power conversion efficiency of solar cells.[98] Sha Simiao et al. reported ZnO/rGO fabricated via one electrodeposition approach for DSSCs under different deposition potential and time, showing that DSSCs assembled with ZnO/rGO deposited at −1.1 V with 600 s achieved the highest photoelectric conversion efficiency of 1.07 %.[99] Many other researchers also obtained highest photo conversion efficiency via assembling DSSCs with ZnO/rGO as shown in Table 5.

4.2.2 Super-capacitors

Super-capacitors are the energy storage devices which are environmentally friendly. These have the ability to substitute the current traditional energy-related devices. Their greater power density, stability and performance compared to the existing energy-related devices made these suitable and superior storage energy devices.[100,101] Super-capacitors are widely used in the plug hybrid electric vehicles and backup power systems. Recently, transition metal oxides and phosphates are extensively used as potential electrode materials in supercapacitors and other energy-related applications.[102,103] ZnO/rGO nanocomposites have attracted significant attention in the development of supercapacitors due to their excellent electrochemical properties, offering unparalleled energy storage performance. ZnO high pseudo capacitance, coupled with rGO, exhibits exceptional electrical conductivity and surface area, resulting in the

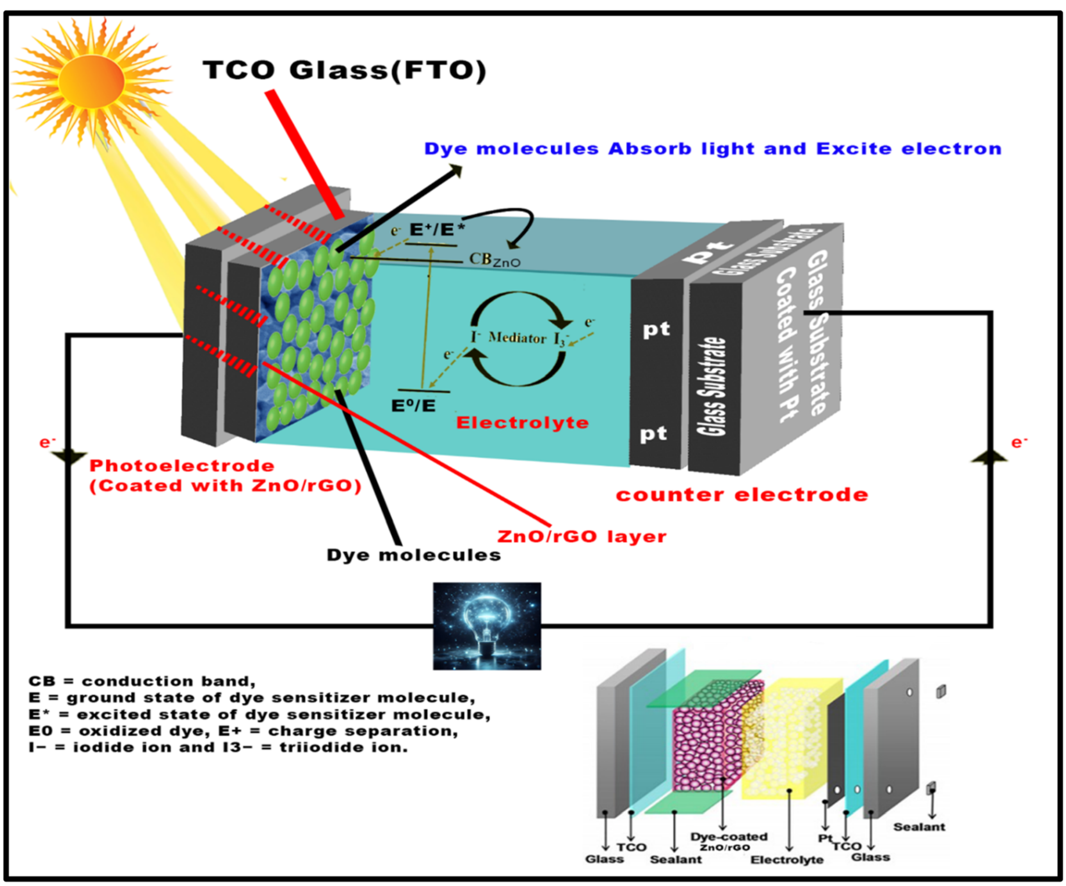

Figure 4: Mechanism and schematic diagram of photovoltaic cell.

Table 5: DSSCs assembled with the ZnO and ZnO/rGO photoanodes fabricated under different deposition potential, time and different concentrations of rGO.

Anode material	Deposition potential (V)	Deposition time (s)	Open circuit voltage (Voc) (mV)	Short-circuit current density (Jsc) (mA cm^{-2})	Fill factor FF	η (%)	References
ZnO (pure)	−1.0 V	600 s	346.730	2.330	0.560	0.460	[104]
ZnO/rGO	−1.0 V	600 s	359.110	2.860	0.570	0.590	
	−1.1 V	600 s	386.150	4.650	0.590	1.070	
	−1.2 V	600 s	374.780	3.750	0.580	0.820	
	−1.1 V	300 s	366.080	3.280	0.580	0.7040	
	−1.1 V	900 s	374.220	3.730	0.590	0.830	
	Percent of reduced graphene oxide (wt%)						
ZnO	0		0.48	3.43	0.49	1.08	[99]
ZnO/rGO1	0.1		0.64	2.87	0.51	1.25	[99]
ZnO/rGO2	0.5		0.63	2.50	0.70	1.47	[99]
ZnO/rGO3	1		0.64	3.02	0.60	1.55	[99]
ZnO/rGO4	5		0.30	1.27	0.28	0.14	[99]
ZnO nanoparticles	–		0.62	3.99	0.48	1.18	[105]
ZnO/Gr	–		0.43	1.93	0.13	0.44	[106]
ZnO/rGO	–		0.53	1.91	–	0.52	[107]

formation of hybrid electrodes with enhanced performance.[12] Haldorai Y et al. successfully synthesized ZnO/rGO composites using a green synthesis approach, indicating their potential as highly efficient electrode materials for supercapacitors. The composite exhibited an improved specific capacitance of 314 F g^{-1}, highlighting its excellent energy storage capability. The material exhibits excellent cycling stability, with a capacity retention of over 98 % after 1,000 charge-discharge cycles. This superior performance is attributed to the synergistic effects between ZnO and rGO, where ZnO provides more active sites for redox reactions, and rGO ensures efficient charge transport and structural stability.[108] P. Anandhi et al. successfully synthesized ZnO/rGO nanocomposite by the wet chemical precipitation method, maintaining 89 % of their initial capacitance after 5,000 charge-discharge cycles. Along with this impressive cyclic stability, the material also illustrates a high specific capacitance of 230 F/g, representing its strong potential for reliable and efficient energy storage in super capacitors.[109] Carbon nanotube composites offer high conductivity and flexibility,[110] while graphene-only electrodes provide large surface areas and double-layer capacitance behavior. However, ZnO/rGO hybrids combine the pseudo-capacitive properties of ZnO with the conductivity and structural support of rGO.[111] Table 6 Highlight enhanced specific capacitance of ZnO/rGO nanocomposite. The working principle of super capacitor is shown in Figure 5.

The small crystallite size has promising applications in energy storage, such as supercapacitors, which facilitate faster ion and electron transport, resulting in improved performance of devices like supercapacitors or batteries.[112] The smaller size of ZnO nanocrystalls provides a high surface area and more sites for charge accumulation and enhances contact with the electrolyte, due to which the capacitance or charge storage capability increases. Apart from that the nanoscale dimensions offer better structural stability during repeated charge–discharge cycles.[113] The presence of rGO as a conductive matrix ensures that the electrons can move rapidly through the composite, complementing the advantages of small ZnO particles.[114] Due to these properties the ZnO/rGO exhibit superior electrochemical performance, including higher specific capacity, better cycling stability, and enhanced rate capability.[115,116]

Table 6: Electrochemical performance of ZnO-based rGO composite electrodes.

Electrodes	Capacitance (F g^{-1})	Scan rate (mVs^{-1})/current density (A g^{-1})	References
ZnO/rGO	314	100 mVs^{-1}	[44]
ZnO/rGO	635	5 mVs^{-1}	[117]
ZnO/rGO	231.3	0.1 Ag^{-1}	[118]
ZnO-G	122	5 mVs^{-1}	[119]
ZnO/rGO	312	–	[120]
ZnO/rGO	719	5 mVs^{-1}	[121]
ZnO/rGO	275	5 mVs^{-1}	[122]
ZnO/CNT	411	20 mV s^{-1}	[123]

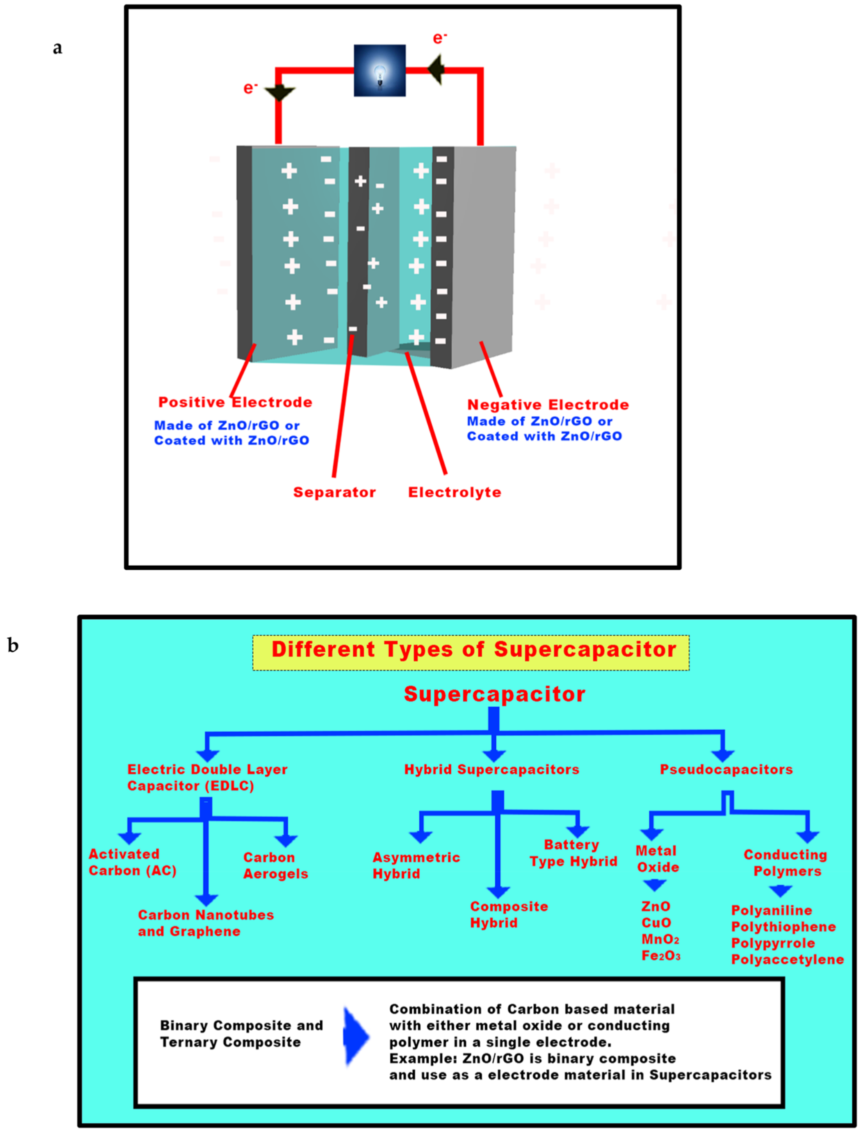

Figure 5: Supercapacitor, (a) mechanism and schematic diagram of ZnO/rGO super capacitor, (b) different types of super capacitors.

4.2.3 Lithium-ion batteries

Lithium-ion batteries (LIBs) gain significant attention to researchers for their high energy density, long cycling life, and efficiency, making them ideal for portable electronics and emerging high-energy applications like electric vehicles and industries etc.[124] However, improved performance has gripped efforts to develop low-cost and high-capacity materials such as transition metal oxides (TMOs). TMOs offer high theoretical capacities, their low conductivity and volume instability during cycling pose challenges.[125] These issues can be resolve by incorporating graphene-based materials, which enhance conductivity, provide additional lithium-ion storage sites, and stabilize volume changes. This approach has shown promise in improving the cycling life, rate capability, and overall sustainability of LIBs.[126] Researcher focusing on formation ZnO-based composite, obtained by integrating ZnO with carbonaceous material, particularly graphene[127] and rGO to enhance the application and performance of lithium ion batteries. By using such a material not only reduces volume fluctuation during charging and discharging, but also increases the electrical and ionic conductivities. Recently, ZnO/graphene and ZnO/rGO composites have provided high discharge capacity of 895 mA h g^{-1} after 110 cycles at 0.2 A g^{-1} and about 942 mA h g^{-1} after 400 cycles at 0.2 A g^{1} respectively.[127,128] Qi et al. prepared ZnO/GO nanocomposite as an anode material for LIBs by using facile-room temperature technique. The charging/discharging study of this material has shown that it exhibits high discharge capacity exceeding theoretical limit of ZnO (approximately 988 $mAhg^{-1}$) at first cycle. At second cycle the discharge capacity decrease to 1,000 $mAhg^{-1}$ and after 100 cycles the capacity decrease further to 550 mAhg^{-1}. A high initial columbic efficiency is reported for ZnO/GO, that could be accredit to electrolytes' decomposition and the formation of solid electrode interphase.[129] Wang et al. employed spraying dyeing method to prepare ZnO/rGO anode material for LIBs and achieved initial columbic efficiency of 69.8 %. Interestingly, ZnO/rGO maintained a high reversible specific discharge capacity of 793 mAhg^{-1} at 100 mAhg^{-1}, 608 mAhg^{-1} at 200 mAhg^{-1} and 327 mAhg^{-1} at 500 mAhg^{-1} after 200, 250 and 450 cycles, respectively.[130] The aforementioned studies show that both materials are promising candidates as anode materials for LIBs. However, superior long-term cycling stability and capacity retention make ZnO/rGO nanocomposite more suitable for practical application in LIBs as anode materials (Figure 6).

4.2.4 Environmental applications

ZnO/rGO composites have gained significant attention in environmental applications including high photocatalytic activity, adsorption capacity, and stability.[131] The photocatalytic process mainly consist of three steps: generation of electron-hole pair through photo excitation, separation of photo-generated charge carriers and lastly surface redox reactions, Figure 7.[132] In the photocatalysis, composite of graphene with ZnO could improve efficient charge separation and migration extended light absorption and photostability.[133] ZnO can generate reactive oxygen species (ROS) under light irradiation to degrade organic pollutants like dyes and pesticides.[134] Reduced graphene oxide (rGO) enhances this process by preventing electron-hole recombination, increasing photocatalytic efficiency, and extending light absorption into the visible range. The rGO also provides a high surface area and functional groups for adsorbing heavy metals such as lead and mercury, while ZnO adds additional active sites for binding.[135] rGO allows more efficient light absorption and interaction with target molecules, such as pollutants or water. The shorter diffusion paths for photogenerated electron–hole pairs minimize their recombination,[76,136] rGO, when coupled with ZnO nanoparticles enabling rapid charge transfer and improving the separation and mobility of charge carriers. This synergy between ZnO and rGO leads to enhanced light-driven catalytic activity, especially under UV or even extended into visible light due to possible quantum confinement effects at such small sizes.[135,136] Several studies have demonstrated the synergistic effect of rGO, enhancing the photocatalytic activity of ZnO under visible light. Zhao et al. studied that the rGO sheets accelerate charge transfer and improve the electron lifetime in ZnO, leading to an enhanced photocatalytic degradation of organic dyes under visible light irradiation.[84] Thuy et al. found that the combination of ZnO with rGO shifts the material absorption spectrum into the visible range, while simultaneously improving its charge separation and stability under repeated cycling. The presence of rGO not only boosts the material's photoactivity under visible light but also significantly improves its photostability.[137] Nguyen et al. reported the photocatalytic degradation of the dyes. MB and Rh–B under visible light irradiation by using ZnO/rGO nanocomposite with different ZnO morphologies: nanospheres, nanodisks and nanorods. Among them the higher photocatalytic degradation and adsorption of the dyes were found with ZnO/rGO composites.[138] ZnO/rGO can also use for removal of organic pollutant under visible light

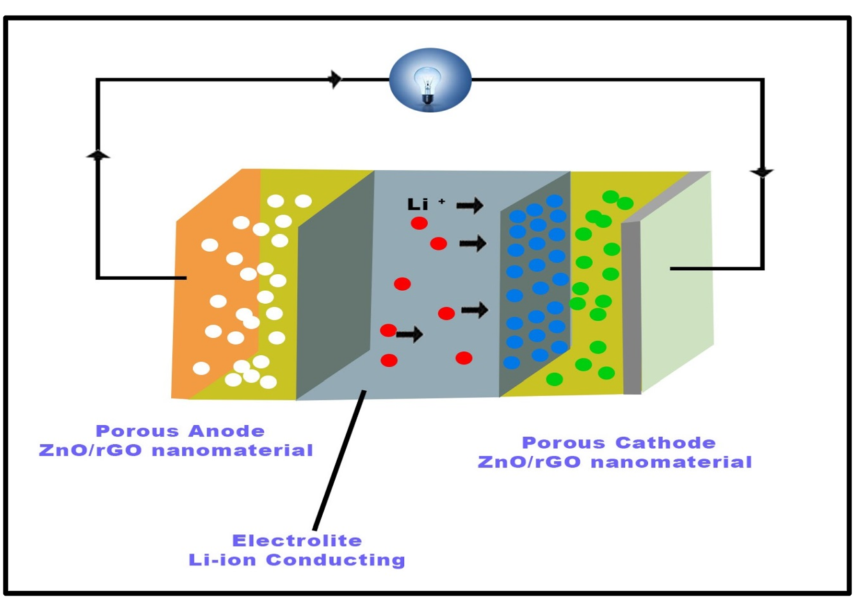

Figure 6: Mechanism and schematic diagram of lithium ion battery.

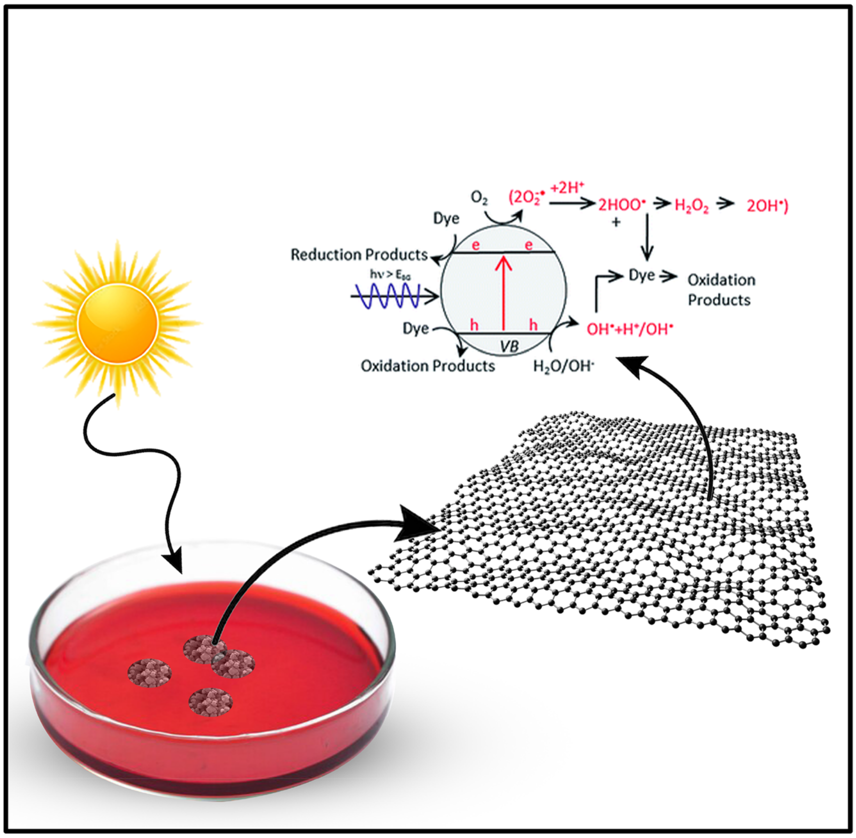

Figure 7: The proposed photodegradation of rganic contaminants in the presence of ZnO/rGO photocatalyst under UV light.

such as nonylphenol, roxarsone, ciprofloxacin etc. The synergistic effect between ZnO and rGO enhanced light absorption, charge separation, and the generation of reactive oxygen species, enabling efficient degradation of contaminants.[139] Figure 8, represent the schematic representation of photocatalytic process while photocatalytic comparison of ZnO and ZnO-based graphene nanosheet composites depicted in Table 7.

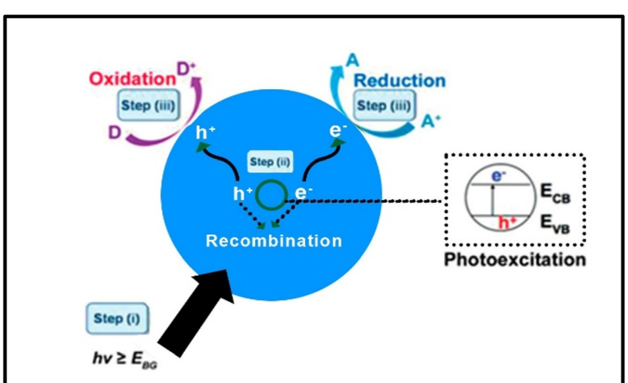

Figure 8: Schematic representation of the photocatalytic process.

4.2.5 Biological applications

The biological efficiency of ZnO, rGO and ZnO/rGO can be tuned by controlling their physiochemical properties such as particle size, morphology and the degree of reduction of GO. These physiochemical properties can be manipulated by various strategies such as method of synthesis, variation of precursor concentration, doping and surface modification. Literature study has shown that using cetyltrimethyl ammonium bromide as a template and changing $Zn(NO_3)_2$ concentration in sol-gel and surface-assisted synthesis leads to ZnO NPs with nanorod, nanoneedle and hierarchical structures with small particle size. These NPs have exhibited improved photocatalytic and antimicrobial activities due to large surface increasing contact with microbial cells area and improved light absorption capacity.[140,141] Thermal reduction of rGO at low temperature also improved the physiochemical properties of GO.[142] Similarly, the adjustment of rGO content in ZnO/rGO allow to optimized the composite's conductivity and interaction with biological entities thus enhancing its antimicrobial activities.[143] Surface modification with biomolecules either in green synthesis or in chemical synthesis methods reduces toxicity and improved interaction with biological system.[142,143] A study also has shown that doping with Cu also increase the biological activates of ZnO/rGO by improving its optoelectronic properties.[144]

4.2.5.1 Antibacterial activity

Recently, many researchers have been focusing on the design and development of the latest antibacterial agents that are not harmful to humans and are effective in action as well. Pathogenic bacteria revealed the emergence of antimicrobial resistance against traditional as well as commercial antibiotics. Due to this issue, the efficacy of antibiotics decreases continuously and limits their use. New antibacterial agents need to be designed to solve antimicrobial resistance. It has been observed that metal oxide nanoparticles have shown activity against gram-positive and gram-negative bacteria.[144,145] The transition metal oxides consisting of TiO_2, ZnO and CuO are the most important metal oxides with high antibacterial activity.[102] Among these, Zinc oxide nanoparticles have shown great antibacterial activity against a majority of the bacteria like *Escherichia coli* and *Staphylococcus aureus* as shown in Figure 9.[146] ZnO NPs damage bacterial cell membranes through radical generation by the electron-hole pair achieved from the photoexcitation, thus killing the bacteria by damaging the organic compounds in microorganisms.[147] Researcher produce ZnO-based hybrid nanocomposites through coupling with organic and inorganic substances.[148] Among them, GO[149] and reduced rGO[150] is promising materials to enhance the photocatalytic properties and thus increase the antibacterial activity of metal oxides.

Table 7: Photocatalytic comparison of ZnO and ZnO-based graphene nanosheet composites.

Pollutants	Catalysts	Band gap (eV)	Light source	Reaction time (min)	Removal efficiency (%)	References
Acridine-3,6-diamine	ZnO nanorods	–	Sunlight	25	79	153
	ZnO-graphene nanosheet	–	Sunlight	5	100	
Methylene blue	ZnO nanorods	–	Sunlight	30	78	
	ZnO-graphene nanosheet	–		5	100	
	ZnO nanorods	3.28		60	100	154
	ZnO/rGO	3.1		40		
	ZnO	3.12	Visible light	60	4.11	
	ZnO/rGO	2.94			93.78	
Methyl orange	GO	4.388	UV light	30	29	140
	ZnO/rGO	3.12			85	
Rhodamine B	GO	4.388			43	
	ZnO/rGO	3.12			90	
Ni(II)	ZnO/rGO	3.12	UV light	90	62	
Cr(IV)	ZnO/rGO	2.1	Visible light	120	84	141
Tetracyclin	ZnO/rGO	2.1	Visible light	120	64	

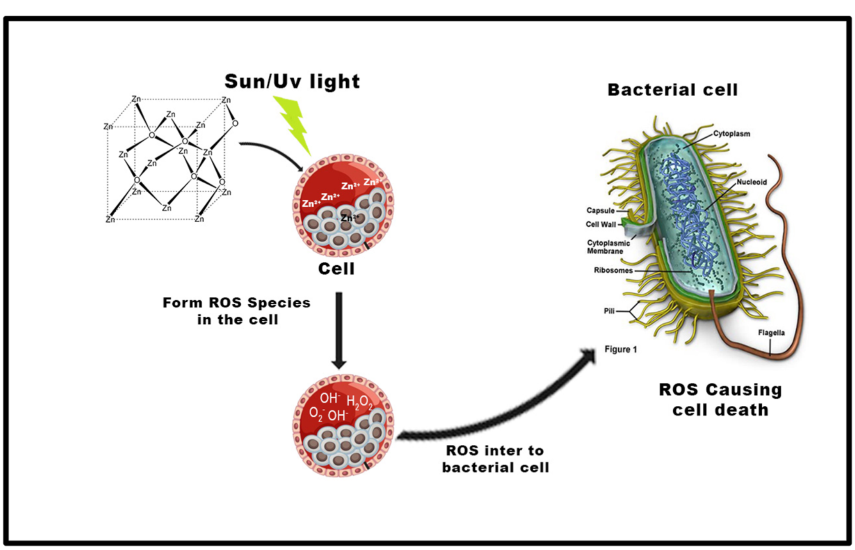

Figure 9: Mechanism and schematic diagram of anti-bacterial activity.

Beside dallying the recombination of electron-hole pairs, GO and rGO are less toxic than other carbon materials: fullerene, carbon nanotubes and graphene as well.[151] Additionally, the sharp edge of GO can damage cell membranes and kill the bacteria.[152] Raza et al.[143] report the synthesis of GO, rGO and ZnO NPs and their ZnO/GO and ZnO/rGO nanocomposite with 1, 5 and 10 wt% depicted as Zn-1GO, Zn-5GO, Zn-10GO, Zn-1rGO, Zn-5rGO, Zn-10rGO and compared their antibacterial activity against *E. coli* bacteria. The prepared ZnO, GO, rGO, Zn-1GO, Zn-5GO, Zn-10GO, Zn-1rGO, Zn-5rGO, and Zn-10rGO samples kill 84, 81, 73, 89, 92, 94, 85, 89, and 91 % of *E. coli* after 12 h at 37 °C, respectively. The enhance inhibition rate of nanocomposite is due the simultaneous activation of two antibacterial mechanisms, including dissolution of Zn^{2+} ions from ZnO and physical interaction between the sharp edges of GO or rGO sheet and *E. coli* following mechanism is followed.

Step 1; $ZnO + hv \leftrightarrow e- + h+$

Step 2; $H_2O + h+ \leftrightarrow \cdot OH + H^+$

Step 3; $e- + O_2 \leftrightarrow \cdot O-$

Step 4; $O- 2 + H^+ \leftrightarrow HO\cdot$

Step 5; $HO\bullet\ 2 + H^+ + e- \leftrightarrow H_2O_2$

The antibacterial properties of ZnO/rGO nanocomposites are enhanced by a multi-faceted mechanism that involves both the physical disruption of bacterial membranes and the generation of reactive oxygen species (ROS).[155] The sharp edges of rGO sheets are known to interact directly with bacterial cell membranes, causing mechanical damage through sharp edge-mediated membrane disruption. This results in an increased permeability of the membrane, allowing the leakage of essential intracellular components and finally leading to bacterial cell death.[155,156] ZnO nanoparticles in the composite generate ROS, such as hydroxyl radicals (•OH), superoxide anions ($O_2\bullet^-$), and hydrogen peroxide (H_2O_2), upon exposure to light or under oxidative conditions.[157,158] These radicals are highly toxic to bacteria as they damage cellular components, including lipids, proteins, and DNA, leading to oxidative stress and cause cell death. The synergistic interaction between rGO and ZnO enhances ROS generation through an efficient electron transfer mechanism which improves the ability of ZnO to generate ROS and thus more rapidly bacterial cell are disrupted.[159,160]

4.2.5.2 Cancer therapy

Due to large band gap, large excitation binding energy, and high piezoelectricity, ZnO exhibit a significant application in cancer therapy. The ZnO is considered as safe by Food and Drug Administration, when they are use against cancer cells they can't affect normal cell but they kill the cancer cells.[161–163] Due to the presence of large amount of oxygen containing functional groups and surface area of GO and rGo it is possible to synthesize their nanocomposite for excellent application in drug delivery such as in cancer therapy. For this purpose, ZnO/rGO nanocomposite were synthesized which have superior activity against cancer cells as well as minimum side effects[164–166] as shown in Figure 10. The anticancer efficacy was determined against cancer cells of human such as breast cancer (MCF7) and colorectal cancer (HCT116). ZnO/rGO nanocomposite show two folded higher anticancer efficacy against cancer cells as compared to ZnO nanoparticles. Apart from that the nanocomposite exhibit higher biocompatibility in normal cell (MCF10A and NCM460) as compare to ZnO Nps.[167]

4.2.5.3 Biosensors

Biosensor is analytical device which contain transducer system and bio-probe system which is used to detect and convert biological signals into electrical signals. Nanocomposite of Zno/rGO have improved carrier capacity and reduce charge recombination. ZnO/rGO exhibit improved electrical properties with minimum value of sheet resistance being 6.32 kΩ at 600 °C (Figure 11).[168] Due to thin sheet-like architecture, high surface area, electron transfer capability, biocompatibility, and bimolecular affinity, graphene oxide (GO) has been used most frequently in next-generation biosensors. To remove defects and enhance bio sensing properties of GO, the reduction of graphene oxide takes place or it can combine through other material.[168,169] The ZnO/rGO have excellent charge transfer capability, high electrical conductivity and abundant active sites to enhance sensing capabilities to detect neurological disorders, dopamine in human serum,[170] detection of glucose,[171] Urea[7] and protein.[172] The ZnO/rGO are used to detect organophosphorus pesticides residues through electrochemical sensors which is made or decoded with ZnO/rGO, in the environment, which is useful for the safety of environment as well for human being, the biosensors detect through electrochemical inhibition of acetylcholinesterase and immobilize to detect the organophosphorus pesticides.[173] Apart from that due to active surface sites, high surface area to volume ratios and high carrier mobility the ZnO/rGO have a significant application in electrochemical gas sensor devices. The synergistic effect of the nanocomposite enhanced their ability to sense and to bind quickly with macromolecules, which made them suitable for formation of gas sensor device.[174]

4.3 Effect of morphology of ZnO/rGO nanocomposite on their applications

The morphology of ZnO and rGO can manipulate their applications in different fields such as energy storage devices,[175] biological activities,[176] photo-catalysis, effect on

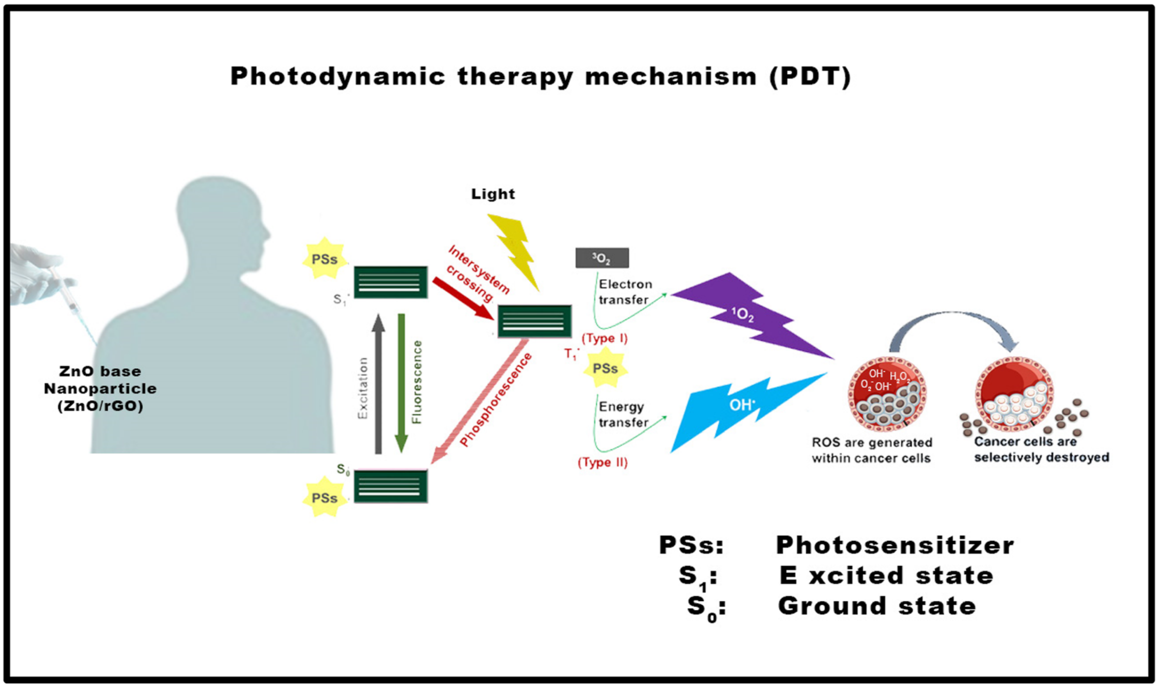

Figure 10: Mechanism and schematic diagram of cancer therapy.

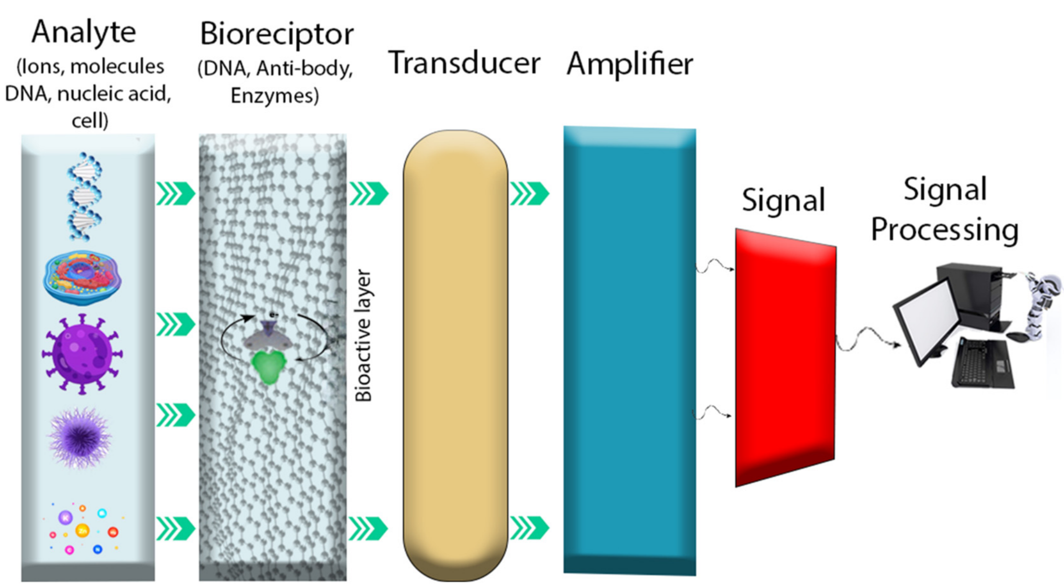

Figure 11: General schematic representation of biosensors.

adsorbents, photodetectors and electron transfer systems.[176–178] The ZnO/rGO nanowires exhibit high charge separation efficiency, and the recombination of electron-hole pair become lower, which is then more efficient to take part in radical reaction with dye molecule, due to which the photocatalytic properties of nanowires significantly increase.[179] ZnO/rGO nanorods exhibit improved mechanical strength and stability which have promising applications in electrode materials for super capacitor.[180] ZnO/rGO nanosheet have a wide range of applications in gas sensors,[181] anti-cancer drug delivery[182] and degradation of organic pollutants.[183] Zno/rGO nanoflowers have high surface area to volume ratio which lead excellent applications in catalysis, biomedical and bioremediation.[184] ZnO nanoparticles dispersed on rGO sheets, which enhances the surface area and electrical conductivity.[185]

4.4 Future perspectives of ZnO/rGO nanocomposites

For the Synthesis of ZnO/rGO nanocomposite ecofriendly and economically green synthesis should be followed. ZnO/rGO nanocomposites can be synthesized to minimize environmental impact and improve their scalability. One promising approach is the use of biological methods involving plant extracts, microorganisms, or enzymes to reduce and functionalize GO and ZnO. These methods not only avoid the use of toxic chemicals but also promote the formation of nanoparticles with well-controlled shapes and sizes.[185,186] It is needed to fabricate the nanocomposite in such method which not consume time and control the nanoparticle formation. For example, there are different methods which consume too much time such as hummers method,[187] chemical vapor deposition,[188] thermal decomposition,[189] mechanical exfoliation[41] and the wet chemical precipitation method which involves many oxidation/reduction steps which make the oxygen functional groups in graphene oxide sheet uncontrolled and further lead to defects in bio-sensing, catalysis, electrode materials and energy storage devices. Machine learning (ML) could be a game-changer in optimizing the synthesis of ZnO/rGO nanocomposites. By utilizing data-driven approaches, ML algorithms can help identify optimal synthesis conditions (e.g. temperature, time, precursor concentration) for maximizing photocatalytic efficiency, super capacitor performance, or antibacterial activity.[189,190] The integration of ZnO/rGO nanocomposites into 3D and 4D printing represents a highly innovative and promising avenue. 3D printing allows for precise customization of electrode structures, complex geometries, and multi-functional materials, which can enhance their performance in supercapacitors, batteries, and biosensors. The ability to print these composites into hierarchical structures can improve ionic conductivity, charge-discharge rates, and electrochemical stability.[191–193] One major challenge for ZnO/rGO nanocomposites is their long-term stability, especially in biological and chemical environments.[194] To prevent degradation and maintain their performance in applications like photocatalysis, biosensing, and energy storage, coating ZnO/rGO with additional protective layers could be explored. Protective coatings such as silica, polymers, or even other nanomaterials like TiO_2 or carbon nanotubes (CNTs) can shield the nanocomposite from environmental degradation, thereby improving its lifespan.

5 Conclusions

The combination of ZnO with rGO offers a valuable platform for developing advanced materials with excellent properties suitable for diverse applications. The synergistic effects of combining ZnO photocatalytic efficiency with rGO, conductivity and stability result in enhanced performance in various fields. Future research should focus on green and scalable synthesis techniques, the application of AI and machine learning for synthesis optimization, and their integration into 3D-printed multifunctional devices can be optimized composite structures, understanding charge transfer mechanisms, and exploring novel applications in various emerging fields. By addressing challenges such as material degradation and enhancing their multi-functionality, ZnO/rGO composites have the potential to revolutionize applications in energy storage, biosensing, catalysis, and environmental remediation. The continuous advancements in synthesis methods, functionalization, and device integration, ZnO/rGO nanocomposites are poised to become a key material in a wide array of cutting-edge technologies. Furthermore, their potential for industrial adoption in energy, environmental, and biomedical sectors could lead to the next generation of eco-friendly devices, smart sensors, and advanced energy storage solutions, which lead to innovation and improving the quality of life on a global scale.

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