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A review on zinc oxide composites for energy storage applications: solar cells, batteries, and supercapacitors

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ABSTRACT

Zinc oxide (ZnO) is used for various purposes because of its special physico-chemical properties, including large band gap, high binding energy of exciton, nontoxicity, high chemical and thermal stability, large piezoelectric constants, and wurtzite crystal structure with various and widespread applications in electronics, optoelectronics, biochemical sensing, biomedical, and energy-saving systems. This review mainly aimed to present the recent improvement in ZnO-based composite materials with utilization in energy storage systems with a specific focus on lithium-ion batteries, dye-sensitized solar cells, and supercapacitors. The first part of this paper looks at the structure and properties of ZnO and then describes some of the most common synthesizing methods of ZnO composites, including electrochemical, chemical, solvo/hydrothermal, and physical deposition methods. Finally, the recent advancement of ZnO-based composite materials applied in energy storage systems was discussed. ©2021 JCC Research Group.

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1. Introduction

Energy consumption is rising steadily due to industry and agriculture development as a result of population and economic growth. Accordingly, the natural resources of energy are becoming increasingly rare, and the nonrenewable resources of energy will eventually run out in the future. However, these energy resources are extensively applied in industries utilizing energy storage technologies. A promising solution to the world's energy crisis is to replace potent, sustainable, and low-cost

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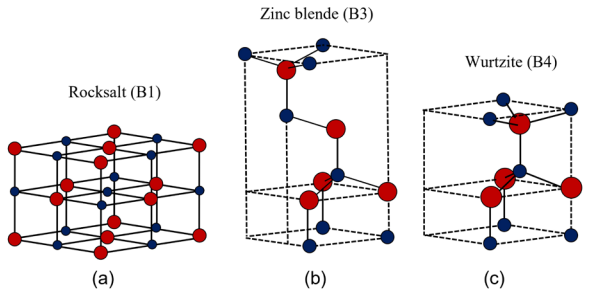

Fig. 1. Schematic image of different crystal structures of ZnO: (a) cubic rock salt (B1), (b) cubic zinc blende (B3), and (c) hexagonal wurtzite (B4). Red and dark blue spheres are Zn and O atoms, respectively.

renewable resources for energy. An effective energy storage management is required to address discrepancies among productivity, the quality of power, environmental restriction, and expenses. Due to this reason, many research reports have been conducted on the energy storage field to develop and present effective energy storage systems critical in the development of renewable energy [1, 2].

ZnO is a semiconductor in group II-V with a wurtzite crystal structure, large band gap of about ~3.7 eV, a high exciton binding energy equal to 60 meV, and piezoelectric characteristics [3]. ZnO has many functional applications regarding its excellent properties, including great chemical stability and photo stability, excellent electrochemical coupling coefficient, and a wide spectrum of wavelength absorption. ZnO has low toxicity, biodegradable, biocompatible, therefore known as a material of choice for biomedical and pro-ecological uses [4, 5]. ZnO is to metal oxides like TiO_2, CuO, and CeO_2 in several cases, due to its relatively low cost, with no surface water, and easy synthesis procedure [6]. It is s an n-type semiconducting material with high chemically and thermally stability and a broad range of applications, including photocatalysis, energy-saving systems, piezoelectric, optoelectronic, electrochemistry, biosensors, and biomedical in a variety of morphologies and structures [7-9]. ZnO is high stable thermally and mechanically at room temperature, which makes it desirable to be utilized in electronics, laser technology, and optoelectronics. ZnO has applications as a photocatalyst in hydrogen formation, energy generator, converter, and sensor due to its pyro- and piezoelectric characteristics. Also, ZnO is a key compound in the ceramic industry due to its piezoelectric constant, rigidity, and hardness. ZnO has antibacterial properties in darkness at a pH range of 7-8. In addition, ZnO nanoparticles (NPs) display visible fluorescence emission, especially when they are synthesized by sol-gel, while the hydrothermal synthesis method makes them suitable as UV emitters [10-13].

A huge number of various methods were presented to synthesize the well surface structured ZnO composite materials, including electrochemical, chemical, solvo/hydrothermal, and physical deposition processes. ZnO-based composites have several benefits, including low price, eco-friendly, acceptable electrochemical reversible, excellent specific capacitance, great power and energy density, convenient cycling stability, and facile synthesis methods, presenting the potential of candidates for electrochemical supercapacitors. The ZnO/carbon composite materials have decent cycling stability because of the synergistic effect of the two-layer carbon capacitance and faradaic ZnO capacitance. The materials consist of carbon, including reduced graphene, graphene oxide, carbon nanotubes, and carbon aerogel, have been applied as electrode materials. Therefore, the composite materials fabricated of carbon material and ZnO might result in a supercapacitor having improved capacitance property [14]. Plenty of studies have been done to develop composites made up of ZnO with polymer matrices, including poly(methyl methacrylate) (ZnO/PMMA), poly (methacrylic acid) (ZnO/PMAA), polystyrene (PS/ZnO), and poly(ethylene glycol) methyl ether) ZnO (PEGME) composite to create novel groups of polymers with combined characteristics of the organic polymeric materials and inorganic particles. The synthesized ZnO/polymer composites have exceptional electrical, thermal, and optical characteristics, which empower them to be applied in extended industry sectors [4, 15, 16].

Recently ZnO composite materials have been drawn tremendous interest owing to their possible applications in energy storage devices, including supercapacitors, solar cells, and Li-ion batteries. ZnO nanostructures have been known as suitable material as photo anode to reached high performance in solar cells. Photo anode made of the nanocrystalline shape of a dye-sensitized semiconductor is the key part in the transformation of solar to electrical energies. ZnO has been thought of as an excellent candidate for the anode in Li-ion batteries, LIBs, due to the great hypothetical capacity of 978 mAh/g, cost-effective, great diffusion coefficient, abundance, and low toxicity. ZnO can be combined with different lithium storage compounds to provide great and steady capacities. Supercapacitors are extensively applied due to their great power density, rapid charging, elevated lifetime, convenient temperature properties, saving energy, and environmental friendliness [1, 7, 17-19].

In this review, the recent development and researches in ZnO-based composites used in energy storage applications have been presented and discussed. The particular emphasis has been given to ZnO-based composite materials applied in the energy-saving system, including LIBs, supercapacitor, and dye-sensitized solar cells. At first, a brief introduction about the structure, characteristics, and various synthesis methods is presented. Then, different applications of ZnO-based composite materials used in energy storage systems will be discussed in detail.

2. Structure and properties of ZnO

2.1. Atomic and crystal structure

Zinc oxide (ZnO) is an inorganic compound semiconductor, which is insoluble in water and soluble in bases and dilute acids accompanied with elevated melting point (1975 ºC) at which it decomposes. The iconicity of ZnO lies at the border between ionic and covalent semiconductors [20]. The different crystal structures of ZnO are rock salt (or Rochelle salt) (B1), zinc blende (B3), and hexagonal wurtzite (B4), as presented in Fig. 1. The structure of wurtzite is thermodynamically stable at ambient temperature and pressure. The stable structure of B3 is formed by growing on cubic substrates, and the B1 could be achieved at quite elevated pressures [20].

The noncentral symmetric structure and the polar surfaces are two key properties of the wurtzite structure. ZnO wurtzite crystalline structure has the lattice parameters *a* and *c* as 3.2495 Å and 5.2062 Å, respectively [21]. The wurtzite structure could be defined as alternating planes of O^{2-} and Zn^{2+} tetrahedrons stacked along the c axis. Oppositely charged ions result in positively charged (0001) Zn and negatively charged (0001) O polar surfaces, result in surface energy divergence, normal dipole moments, and spontaneous polarization along the c axis, as well as a deviation in surface energy. Some characteristics of ZnO, including spontaneous polarization and piezoelectricity, are due to this polarity which in turn is the main parameter in defect generation, etching, and crystal growth [22].

2.2. Intrinsic aspects

ZnO semiconductor has a wide band gap (E_G=3.437 eV at 2K) and high exaction binding energy (E_b=60 meV), which make it suitable for various applying in varistors, piezoelectric transducers, and transparent conductive films. The polycrystalline materials are the only required

material for several of these applications; although, the recent improvement in the fabrication of large-area single crystals makes it possible to produce UV and blue light emitters as well as high temperature and high power transistors. The lack of asymmetry center in the wurtzite, as well as a great electromechanical coupling, translates into strong piezoelectric and piraleptric properties [23]. The benefits accompanied by a large band gap are comprised of high power operation, low noise generation, high breakdown voltage, and the capability to withstand large electric fields. ZnO has a high excitation binding energy of 60 meV. It is a paramount oxide with near-ultraviolet emission and transparent conductivity. Due to its non-central symmetry, ZnO presents piezoelectric behavior, which is the main feature for the manufacture of electromechanical coupled transducers and sensors [24, 25].

Generally, the electrical transport characteristics of ZnO are related because of the wurtzite anisotropy in the crystal structure. Electrical ZnO is a transparent, conductive semiconductor presenting impurity- or defect-dominated n-type conductivity at room temperature. This type of conductivity is a result of the large band gap and the presence of impurities and defects having donor ionization energies. The n-type behavior and the specific nonamphoteric and donor-like property are innate to ZnO. For ZnO to be a technological success as a material for UV optoelectronic devices, a crucial problem is the ability to reliably produce n-type and p-type ZnO materials. N-type doping is easily achieved. However, like most wide-band-gap semiconductors, ZnO exhibits asymmetric facility of dopability through one way (n-type) but not the other way (p-type). In theory, it is possible to obtain some grade of p-type doping in ZnO; however, these structures are likely to be metastable, evidently achievable experimentally, and complicated to be precisely refabricated because of the extended alteration in ZnO microstructures, alongside the seemingly paramount role of defects at high density of interfaces and surfaces between grain boundaries [23].

The high breakdown strength and high saturation velocity are two important characteristics of ZnO, which turns it to be attractive in electronic devices. Also, ZnO exhibits high radiation hardness compared to alternative semiconductor materials, including GaN, CdS, GaAs, and Si, resulting in increasing ZnO utility in space applications. The laser action of optically pumped ZnO in the UV region in low and high temperatures has been shown [23].

3. Synthesis techniques of ZnO composites

A large variety of ZnO composite materials could be synthesized and prepared with ZnO via different routes such as electrochemical, precipitation process, solvothermal, hydrothermal, microwave, emulsion, microemulsion, sol-gel process, precipitation from microemulsion, mechanochemical, physical/chemical deposition processes, biological (green synthesis), and microfluidic reactor-based methods [4, 26]. By using these methods, it is possible to obtain products with different shapes, sizes, and spatial structures. Some of these approaches are discussed in detail in the next sections.

3.1. Electrochemical method

The electrochemical synthesis route has been employed for a large range of metallic, semiconducting, and oxide nanoparticles (NPs). The method has been used to directly form metal NP films, such as CuO, CdS, and ZnO NPs on graphite or H-terminated silicon surfaces [27]. The electrochemical method is cheap with a high production yield. They also offer the option of both batch and continuous procedures [27]. Electrochemical synthesis techniques provide a simple and feasible substitute to expensive routes, including thermal evaporation or laser ablation. Furthermore, it is possible to control the growth of metals, oxides, compound semiconductor nanowires, and conducting polymers by utilizing the electrochemical method [28]. The accurate process control comes from their electrical nature, which permits to control of the aspect ratios, lengths, and the compositions of the synthesized materials by changing the different parameters of the electrochemical synthesis procedure, including charge flow, current density, solvent polarity, the distance between electrodes, as well as temperature are critical for controlling particle size selectivity of the electrochemical synthesis process [27].

A two-layer ZnO film was synthesized by electrochemical synthesis to be utilized as a photo anode in dye-sensitized solar cells (DSSC) [29]. The synthesis method was a one-step potentiostatic electrodeposition and pyrolysis method on a conducting fluorine-doped tin oxide (FTO) glass. The film consisted of an under a layer of porous ZnO NPs and an over a layer of nanosheets of ZnO (ZnONS/NP). The synthesized composite exhibited improved self-established light scattering property compared to the thin film of ZnO NPs. A general method comprised of two sequential electrochemical steps was presented to synthesize three-dimensional (3D) graphene (ERGO)– ZnO composites [30]. At first, the 3D ERGO matrix was synthesized through the electrochemical reduction of a concentrated graphene oxide dispersion. Then, ZnO was coated onto the prepared matrix by electrochemical deposition to yield a graphene-based 3D porous composite, which was suitable to be used as electrodes in the electrochemical apparatus.

Recently, Daideche et al. applied the electrochemical polymerization method to synthesize polyaniline/ZnO composite films (PANI/ZnO) on transparent indium-doped tin oxide (ITO) glass substrate via using the cyclic voltammetry technique. The synthesis was performed in the sulfuric acid medium using aniline monomer precursor and LiClO4 salt at room temperature. The results showed that adding ZnO doping in the polyaniline polymer matrix result in improved electrical conductivity of the composite [31]. Xu et al. showed a two-step electrochemical deposition method to synthesize different hierarchical ZnO nanostructures, such as diverse morphologies of ZnO crystals, ZnO rod arrays on sheets, ZnO needles on needles, and ZnO rods on rods [32]. The hierarchical nanostructures fabricated by this method were uniform and pure, which could be a candidate to be used as photoanodes in DSSCs.

Polyoxometalates (POMs)-assisted electrochemical method has been used to fabricate uniform ZnO hollow spheres at room temperature with high photocatalytic activity [33]. The results showed that it is possible to synthesize controllable spherical ZnO particles by this simple, one-step electrochemical method [33]. This method has also been used to synthesize spindle-like ZnO with small NPs and porous structures in ionic liquid [34]. The electrochemical deposition was mentioned as a routine method used to fabricate high-quality 1D ZnO films used in perovskite solar cells, but the obtained morphologies often differ, caused variable power conversion efficiencies (PCEs) [35, 36]. ZnO structured films fabricated by this method were used to serve as electron transport layers in solid-state solar cells. Well-conducted ZnO layers were deposited in a chloride medium and developed with nanostructures ranging from arrays of nanowires to form a dense, well-covering layer. The nanostructured oxide layers showed higher power conversion efficiencies compared to the dense ones [35, 37].

3.2. Chemical methods

Chemical methods are the most frequently used method to synthesize ZnO composites in the laboratory and on industrial scales [26]. There are various chemical syntheses routes, including mechanochemical, emulsion, microemulsion, sol-gel, and chemical vapor deposition. These methods are usually categorized based on the source of energy or the size selection [27].

The mechanochemical process is a simple and cheap method to fab-

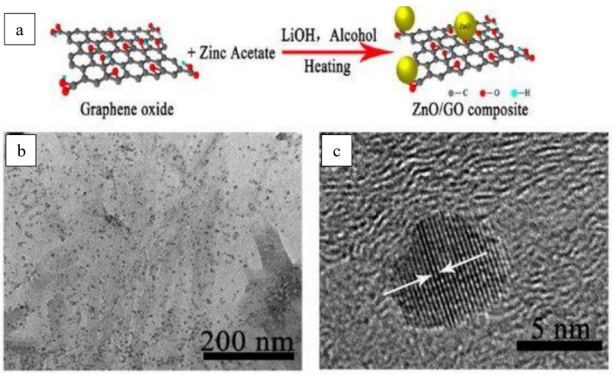

Fig. 2. The process of preparing GO-ZnO nanocomposites by solvothermal method (a), TEM micrograph illustrating dispersion of ZnO nanoparticles on graphene sheet (b), and HRTEM image presenting the lattice fringes of ZnO nanoparticle (c). Reprinted from [58].

ricate ZnO NPs on a large scale [38, 39]. This method includes high-energy dry milling to start a reaction by ball–powder affects ball mill at low temperature. The mechanochemical method has several advantages, such as the small size of particles, the restricted tendency for agglomeration, and the homogeneous structure and morphology of crystalline. To synthesize ZnO NPs by this method, anhydrous $ZnCl_2$ and Na_2CO_3 are used as starting materials and NaCl as a reaction medium and separator of ZnO NPs [4, 40]. Several composites such as fluorapatite-zinc oxide (FAp-ZnO) composite nanopowder [41], Mn-doped ZnO nanoparticles [42], silver doped ZnO NPs [43], and carbon nanotubes (CNT)/ZnO hybrid materials [44] have been synthesized by the mechanochemical process.

Micron- and submicron-sized emulsions synthesis methods are usually referred to as microemulsions. The microemulsion is a separate, clear phase with low viscosity, which is thermodynamically stable. The microemulsions method is applied to start polymerization on a large scale via hard controlling of monomers, radical initiators, and the dispensation of co-surfactants between two phases of aqueous and organic [27]. ZnO–$ZnWO_4$ nanocomposite was synthesized by microemulsion method with surfactant assistant [45]. The particles size was decreased by increasing the level of $ZnWO_4$ up to 30 wt%. The addition of 30 wt% $ZnWO_4$ caused a deviation in the value of the energy band gap from 3.2 to 3.16 eV. The prepared composite had the highest activity with 96% photodegradation efficiency. In another study, both hydrothermal and microemulsion synthesis methods were applied to prepare zinc oxide/activated carbon nanocomposites with controlled particle size and even distribution [46-48]. A microemulsion of an oleic acid/n-Butanol/sodium hydroxide solution was utilized as a precursor for preparing zinc oxide/activated carbon composites.

The sol-gel method is another chemical synthesis method that permits solid materials from colloidal suspensions. Sol-gel contains a solid matrix (gel) which is filled with a solution of solid particles or chemical reagents (sol). The sol-gel route includes consecutive chemical reactions, including hydrolysis, condensation, polymerization, and the development and aging of particles [49]. The sol-gel method is attracting a lot of interest because it is simple, cost-effective, reliable, and repeatable, with quite mild conditions of synthesis. It is possible to modify the surface of zinc oxide with certain organic compounds by the sol-gel method to fabricate composite materials with modified properties and a broad range of uses [4].

Many reports investigated the synthesis of ZnO composite materials in the literature. A well-crystallized material of $ZnWO_4$-(ZnO) composite was prepared by sol-gel synthesis method from aqueous solutions of Zn^{2+} and WO_4^{2-} at pH = 7 or pH = 10 without the addition of any surfactant [50]. Different structures and morphologies of ZnO/SnO_2 nanocomposites have been prepared by sol-gel route followed by annealing the precipitate at 600 and 750 °C [51]. The prepared composites are potentially used in solar cells, gas sensors, photocatalysts, and optoelectronics. Porous zinc-copper composite aerogels made of nanosized and microsized particles were synthesized by the epoxide addition sol-gel approach [52]. The synthesized ZnO-CuO aerogels are of particular interest because of their specific catalytic and sensing properties. V_2O_5-ZnO nanoflakes composites were prepared by employing a simple sol-gel method [53]. The study revealed that anchoring V_2O_5 in the ZnO composite resulted in improvement of the photocatalytic performance of the composite under irradiation of visible light. Han et al. studied a simple sol-gel method for the synthesis of ZnO-coated activated carbon aerogel with high surface area and well-crystallized ZnO NPs [54].

3.3. Solvo/hydrothermal method

Solvothermal is a heterogeneous synthesis method in which precursors are dissolved in a solvent under high pressure and temperature in a closed reactor. The reactor (referred to as autoclave) is resistant to the effects of corrosive solvents at high temperatures and pressure. Two important factors of chemical (nature of solvents, precursors, and mixing methodology) and thermodynamic (temperature and pressure) can affect the characteristics in the solvothermal method, which could be checked by designing special reactors [20].

Sandwich-like GO/ZnO nanocomposites were prepared by solvothermal synthesis method in ethylene glycol medium by graphene and zinc acetylacetonate oxide as precursors. The previously formed ZnO nanoparticles had a diameter of near 5 nm and were deposited uniformly and densely on both sides of graphene sheets to make a sandwich structure. The synthesized ZnO-coated graphene nanocomposites can enhance the sensing and photocatalysis behavior of ZnO [55]. A facile solvothermal method was applied to prepare graphene-ZnO nanocomposites (G-ZnO) in which the crystalline ZnO nanoparticles (30–70 nm) were homogeneously deposited on the graphene sheets. The synthesized G-ZnO nanocomposites had a convenient capacitive behavior (122.4 F/g) relative to GO (2.13 F/g) and rGO (102.5 F/g) as a good candidate of electrode material applicable for high-performance supercapacitor [56]. An electrolyte-gated transistor was fabricated using a channel layer gallium–indium-zinc-oxide NPs by solvothermal method [57]. Fig. 2 presents the process of preparing GO-ZnO nanocomposites by the solvothermal method as well as the TEM and HRTEM images of ZnO nanoparticles (c)

The hydrothermal process is a kind of solvothermal process in which water is used as the solvent. Hydrothermal synthesis processing may include three key steps: hydrolysis and hydrogen bonding, polymerization, and precipitation. To control the size in this method, surface chelating agents are used. The two main advantages of the hydrothermal synthesis method are the cost-effectiveness and reduced environmental effect. The hydrothermal system contains precursor solution, water pumping tube, a reactor with proper precursor delivery and product extraction, a cooling device, filtering, and a container for the final product [20, 59].

Hydrothermal route accompanied with Pluronic P123 triblock copolymer was applied to produce zinc oxide/graphene (ZnO–rGO) nano-composites via the in-situ reduction of graphene oxide (GO) and zinc acetate $(CH3COO)_2Zn{\cdot}2H_2O$). The prepared ZnO–rGO nanocomposite samples showed improved hydrogen sulfide sensing behavior compared to ZnO [60]. Carboxylated graphene–ZnO (G-COOZn) composites were synthesized as coating ZnO NPs on graphene sheets using a single-step thermal process by $Zn(NO_3)_2$. G-COOH sheets and carboxylated graphene (G-COOH). The electrochemical characteristics of the prepared G-COOZn supercapacitor revealed its electrochemical capacitance function in the range of 0–1 V and a special capacitance of ~238 F g^{-1} at 50 mA cm^{-2} of current density. This approach provided a facile and direct method to deposit ZnO NPs on graphene sheets and

might be used for preparing alternative types of hybrids based on GO sheets for technological uses [61]. A photocatalytic Pd-ZnO-EG nanocomposite composed of palladium (Pd), ZnO, and exfoliated graphite (EG) was synthesized in a one-step hydrothermal method with higher photocatalytic activity and better removal efficiency of Acid Orange 7, widely used as an organic pollutant model, compared with zinc oxide and Pd-ZnO [62]. A simple hydrothermal route at a temperature of 100 °C was applied to synthesize the composites of ZnO–graphene, which showed a strong and wide absorption peak in the visible spectrum as well as an intense light absorption in the UV region which make this composite a suitable photocatalyst under visible and UV light [63].

A combination of two synthesis methods of hydrothermal procedure and electrospinning was applied to prepare hybrid composites in which ZnO in the form of nanoflakes was wrapped with carbon nanofibers (CNFs) and the fabricated composites with application in SCs as a potent electrode material with higher specific capacitance (260 F/g) compared to pristine ZnO NFs (118 F/g). The synthesized ZnO/CNFs composite also showed good capacity retention (73.33%). The results showed great potential applications of ZnO/CNFs composite to improve energy storage devices with high energy and power densities [64].

3.4. Physical deposition method

The physical deposition synthesis process involves the deposition of the supplied material onto the substrate by physical approaches. Some of the physical deposition methods include evaporation, sputtering and spin casting. The physical evaporation method involves a vacuum chamber where the substrate and a source of the deposited material are located there. The vacuum is needed for easy evaporation of the source material molecules in the chamber and deposit on all surfaces. Then, the source material is heated to be boiled and evaporated. In the sputtering method, energetic ions generated in a glow discharge bombard the "target," causing the removal of its atoms or molecules and then deposited over the substrate. This technique can be performed at a lower temperature compared to the evaporation one. Finally, in spin casting, spraying or spinning is used for deposition; therefore, the material to be deposited shall be dissolved in a solvent to make a liquid phase. The solvent has been evaporated during a baking process, and a thin film of the source material coats on the substrate. The spin casting method is mainly beneficial in polymer layers production [65].

Manthina et al. synthesized a new ZnO nanorods composite structure covered by TiO_2 NPs by the layer-by-layer electrostatic deposition process. The novel zinc oxide-TiO_2 composite showed better transportation of electron transport in company with nanorods as well as a surface made by NPs, which make this composite a good candidate in the collection of charge as well as light. Also, the DSSCs which contained the novel composite films showed improved values of current values compared to the titanium oxide films [66].

A nanostructured composite of ZnO-carbon black (ZnO-CB) was synthesized by atomic layer deposition (ALD) as the anode material used in lithium-ion batteries. By applying the ALD method, extra oxygen vacancies are created in the zinc oxide structure as well as stronger bindings among the zinc oxide and surfaces of carbon result in improved the electrical connection through discharging and charging process to have a stable solid electrolyte interphase (SEI) layer [67]. ALD method was used to deposit single and multi-layers of TiO_2 and ZnO on hydroxyl functionalized MWCNTs [68]. CNTs were homogeneously coated with layers of TiO_2, ZnO as well as the mixture of them. Composites that were fabricated by the coating of the combination layer showed less catalytic activity because of increasing the average densities and therefore decreasing the surface areas as compared with CNTs covered by one oxide layer [68].

Different lengths of ZnO nanowires (1-4 μm) were deposited by a simple solution-phase deposition process onto ITO glass substrates and employed as photoanodes in solar cells. Increasing the length of zinc oxide nanowire structure results in more dye molecules adoption by ZnO nanostructure and improved the photovoltaic performance of DSSCs [69]. The sputtering physical deposition technique was applied to synthesis aluminum-doped Zinc oxide (ZnO: Al or AZO) and used as transparent conductive oxide films in thin-film photovoltaic cells. The distinctive process development appropriate for large area deposition was presented for this specific application, with the key compromise between optical properties (transparency and light scattering) and electrical characteristics (conductivity) [70].

Physical vapor deposition (PVD) associated with dip-coating was applied to synthesis ZnO/multi-walled carbon nanotube (MWCNT) doubled coating on Mg-0.8Ca-3Zn alloy [71]. The fabricated ZnO/MWCNT doubled coating composite presented greater compressive strength compared to the bare Mg alloy and one-layer coated ZnO after soaking in simulated body fluid (SBF) solution. The deposition synthesis method, which was used to fabricate the composite of ZnO (the inner layer) and MWCNTs (the outer layer) on the surface of the bare Mg alloy sample, was a method to increase the corrosion resistance of the composite [71].

4. Application of ZnO composites for energy storage devices

To face the energy challenge and to present an unpolluted environment, the advent of sustainable technologies and energy resources and is essential. Resources of renewable energy, particularly solar energy, is one of the suitable candidates for eco-friendly energy sources to decrease or replace the world energy demands on fossil fuels.

4.1. Application of ZnO composites for solar cells

Solar energy, as an attractive source of free and endless energy, is one of the potential candidates for nonrenewable energy resources. There are some types of technologies based on solar energy, such as solar heating, solar photovoltaics, and solar thermal electricity. The photovoltaics (PV) technique, which converts solar energy to electrical energy, is abundant, inexhaustible, and clean [19]. The application of new and complex materials in advanced solar cell technologies, namely dye-sensitized, perovskite, multi-junction, and quantum dots, opens the door to provide effective and constant energy conversion. No one of the established solar cell technologies have nearly reached the hypothetical energy conversion due to energy bandgap and losses of thermalization and transmission, which are closely associated with the characteristics of the active compounds such as defects. Also, the stability of the active compounds is effective on the lifespan or permanence of solar cells technologies. Ideal characteristics of charge transport materials (CTMs) applied in solar cells include proper energy levels related to the high efficiency of solar light absorption, acceptable conductivity, great carrier mobility, and effective excited charge carrier extraction [72].

Zinc oxide composite materials with outstanding properties, including wide bandgap, outstanding electron mobility, high electron affinity, good stability, and great conductivity, have been recognized as ideal materials in the solar cell. In addition, ZnO can be coupled with materials with smaller energy gaps in order to absorb light in the visible region, for example, semiconductors with a narrow band gap, organic polymeric materials, and dye sensitizers [73].

Perovskite material has the chemical structure of ABX_3. In this structure, "A" and "B" are two different sizes of cations, and X should be an anion. Perovskite solar cell (PSC) was first presented as a sensitizer for photovoltaics by Kojima et al. in 2009 and has received extensive

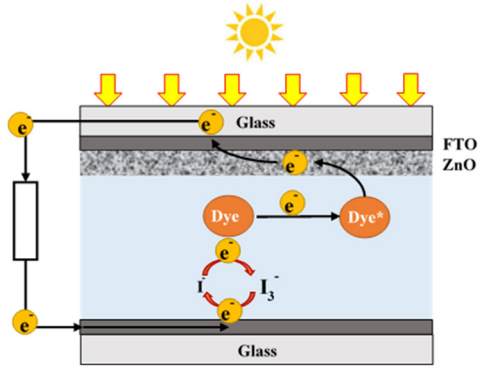

Fig. 3. Schematic overview of a typical DSSC contained ZnO.

attention as a light absorption layer [74]. Its solar to electric power conversion efficiency (PCE) was 3.8 % which was improved to more than 20% PCE in the past few years [75, 76]. PSCs are photoelectrochemical systems with a structure including several layers of electron transport material (ETM, e.g., TiO_2, ZnO), transparent conductive substrate, a hole transport layer (HTL), perovskite light absorption, and an Au- or Ag-back electrode [77]. It is important to design a proper ETM layer with high electron mobility to reach high photovoltaic efficiency. ZnO composites are extensively used as an ETM layer for the extraction and transportation of photogenerated electrons. Also, they can use as photogenerated hole-blocking to defeat the charge recombination in perovskite films. Regarding the key role of ZnO composites in the photovoltaic performance of PSCs as the ETM layer, it is important to control their specification, especially the morphology, trap states, energy level alignment, and interfacial characteristics [73].

The first dye-sensitized solar cell (DSSC) based on titanium dioxide (TiO_2) was fabricated in 1991 by Grätzel et al. [78]. DSSC has arisen as a favorable photovoltaic device because of the good PCE, cost-effectiveness, low toxicity, more than 10% efficiency of energy conversion, and proper long-lasting stability [18]. A DSSC typically contains three side-by-side thin layers of a mesoporous TiO_2 film, a dye sensitive to sunlight coated on TiO_2 film (e.g., ruthenium bipyridyl derivative), and an organic liquid electrolyte, necessarily having triiodide and iodide ions acting as the redox couple. The sandwich of these three layers is placed between two conducting glasses, one of the coated a thin TiO_2 buffer layer and the other one covered by a platinum layer. ZnO has regarded as the best substitute for TiO_2 in DSSCs due to the same electron affinities, similar band gap energies, and the fact that ZnO has a much higher diffusion current than TiO_2 with high electron mobility and stability against photo corrosion. Moreover, several designs of photoanodes can be fabricated by various morphologies of ZnO for DSSCs, perhaps more than any other metal oxides. The crystalline structure of ZnO is conductive to anisotropic growth, which is different from the crystalline structure of TiO_2, which makes it the main choice for DSSCs with photoanodes containing nanosheets, nanowires, or nanorods [79-81]. ZnO should attain higher efficiency to be used in DSSCs. In this regard, the DSSCs contained nanostructural zinc oxide showed significantly improved efficiency in comparison to thin films of zinc oxide because of the large surface area, improved light collecting, and abilities to light scattering [82]. A schematic overview of a usual DSSC is presented in Fig 3.

1D nanostructure ZnO composites with different morphologies, such as nanorods, nanotubes, nanowires, and nanofibers, have been widely investigated to be applied in DSSC, and extensive studies have been performed about it in the literature compared to the other morphologies. Among 1D ZnO nanostructures, nanotubes assume to be the better candidate to use in DSSCs due to their higher surface-to-volume ratio and the porous structure relative to ZnO nanowires and nanorods. The hollow and oriented structure of nanotubes may provide both the increased porosity and the large surface area for adsorption of photosensitizer dye, result in effective light absorption, which can enhance the efficacy of DSSCs. Also, ZnO nanowire structures have been advised as proper candidates for photoelectrode arrangements due to their appropriate length, which can deliver a straight route to the rapid movement of the photoelectrons to the conducting surface of the photoanode. In addition, porous ZnO nanofibrous is the other morphology suitable to be utilized as photoanode in DSSCs, which were less studied [19]. The comparison of the various structure of ZnO composites for DSSCs application and their photovoltaic characteristics are summarized in Table 1.

4.2. Application of ZnO composites for batteries

The improvement and advancement in batteries as an effective storage device has been taken centuries, from the first battery, invented in 1800 by Alessandro Volta [92], to the lithium-ion batteries introduced by Sony in 1991 [93]. LIBs, one of the most appropriate choices for energy storage devices, are superior to the other batteries regarding the number of cycles [94]. The key parts of a Li-ion cell are shown in Fig. 4, representing a positive electrode (cathode), a negative electrode (anode), and a Li-ion conductive electrolyte in between. The operating

Table 1.
Photovoltaic performances of different composites of ZnO in DSSCs application

Composite	Synthesis method	J_{sc} (mA/cm²)	V_{oc} (V)	FF (%)	PCE (%)	Ref
Li doped ZnO	MW-Hydrothermal	4.18	0.67	44	1.23	[83]
ZnONWAs on ZnO-TiO_2	Hydrothermal	3.51	0.656	38	0.91	[84]
ZnONNAs on ZnO-TiO_2	Hydrothermal	5.70	0.617	42	1.47	[84]
ZnO/MWCNT	Doctor blade	12.82	0.67	63	5.39	[85]
Ag-doped ZnO	Sol-gel	9.09		39.76	2.02	[86]
Ga co-doped ZnO	Sol-gel	4.94	0.65	75	2.43	[87]
Cu doped ZnO	Co-precipitation	0.81	0.47	63	0.89	[88]
Mn-doped ZnO	Hydrothermal	11.5	0.63	58	4.20	[89]
ZnO-TiO_2 NF	Electrospinning	12.03	0.79	66	6.33	[90]
ZnO+g-C_3N_4 modified TNTs	Solvothermal	14.68	0.69	61.2	2.45	[91]

Jsc= Circuit current density, FF= fill factor, Voc=Open circuit voltage
MW = Microwave-assisted
NWAs= Nanowire arrays
NNAs= Nanoneedle arrays
NF= Nanofiber
TNT= TiO_2 nanotube

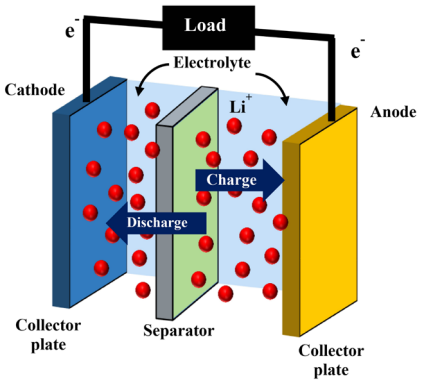

Fig. 4. Schematic of a Li-ion cell.

approach of a Li cell is built on the movement of Li-ions between the two electrodes, able to store Li. $LiCoO_2$ cathode, carbon anode, as well as LiPF6 electrolytes have been used in the first Li-based batteries that had a capacity of about 140 mAh and 3.7 V and an efficacy of 50 % that was quite low [94].

LIB is still the main power and energy source for plenty of electrical devices because of the great power density and energy density. The restrictions caused by anodes with carbon-based strictures have been controlled by using different alloys, and metal oxide compounds like transition metal oxides (TMOs), including $MnxO_x$, ZnO, and TiO_2, because of their high hypothetical capacity. Particularly ZnO has been regarded as a promising anode due to its great hypothetical capacity of 978 mAh/g, low price, low toxicity, high availability, and great lithium-ion diffusion coefficient. Although, the actual use of ZnO is hindered by the quick capacity decay. Through the electrochemical process of discharging and charging, a large volume of expansion occurs, which causes loss of the disconnection of electricity between the anode powders. To overcome the capacity fading and increase the long cycle life of batteries, several strategies have been applied, such as numerous surface morphologies, nanostructures (one, two, and three dimensional), adding conductive mediators, and doping with diverse elements [7].

The synthesis of binder-free electrodes made of zinc oxide nanosheets coated on graphite was reported as an anode electrode with great capacity and stability to be applied in the lithium-ion battery. The novel electrode composite was prepared via hydrothermal route followed by thermal evaporation procedure. The composites with a thicker graphite layer showed an excellent capacity of about 600 mAh/g and the lowest capacity fading. It is related to the particular micro size porous arrangement with proper interrelated nanoparticles (less than 15 nm) that provides a huge space for accepting the changes in volume following the lithium insertion/deinsertion procedure. In addition, graphite is important for stabilizing the structure and shape through discharging/charging procedure upon electrochemical cycling, leading to improve the specific capacity and increase the reversible capacity of zinc oxide nanosheets [95].

A hierarchically organized self-assembled ZnO composite was synthesized with the assistant of graphene oxide to improve the electrical conductivity of ZnO. The results revealed the excellent reversible capacity, great durability, as well as improved rate performance of the synthesized composite. The enhanced performance of the novel composite was due to the unique self-assembled form, which exhibited great specific capacity, great electrical conductivity, reduced lithium-ion diffusion pathway, firm SEI layer [96]. Feng et al. fabricated various ZnO-graphene hybrid composites via different amounts of graphene by hydrothermal route for lithium-ion battery. The composites, which contained 71.45 wt. % of graphene showed great reversible specific capacity and perfect rate function. In addition, it had a significant reversible capacity of 240 mAh g^{-1} at the low cut-off voltage (0.01-1.0 V). This outstanding outcome was related to the addition of graphene to the composites, which diminishes the amount of expansion through the electrochemical cycling procedure [97].

The single-nozzle electrospinning synthesis method was applied to fabricate the core/shell structure of zinc oxide/carbon nanofiber composites by the various amount of carbon nanofibers (CNFs). The composite made of zinc oxide/CNF-0.5 showed well cycling stability and enhanced capacity. The CNFs improved the electrical conductivity as well as speed up the kinetic of the electrochemistry process. In addition, CNFs could inhibit the volume expansion of zinc oxide through the charging/discharging procedure [98]. Xiao et al. synthesized a novel composite containing ZnONPs compacted in a three-dimensional hierarchically carbon frame via a hydrothermal method. The new ZnO@CF composite showed improved specific capacity, extended lifetime as well

Table 2.

Electrochemical performance comparison of ZnO-based composites for lithium-ion batteries

Composite	Synthesis methods	Reversible capacity mAh/g	Cyclic number	Current density mA/g	Ref.
SnO_2-ZnO-C	hydrothermal and balling	1192.2 723.6	300	200 5000	[103]
ZnO QDs@porous carbon	Carbonization	990 357		100 2000	[104]
ZnO/Co_3O_4/C	Self-sacrificial template.	1316 1101	100 25	100 1000	[105]
ZnO/rGO	Spray drying and heating	793 608	200 250	100 200	[106]
CF@ pore-ZnO	ZnO nanoparticles coated onto CF	510 395	300 1000	100 2000	[107]
CNT@ZnO	liquid-phase	709.2	50	100	[108]
ZnO/3DOM-mC	Dual-templating/ *in situ* solution growth	973.3	100	1396.6	[100]
ZnO/CNFs-PA	Electrospinning	702		200	[109]
Peapod-like ZnO@C	Hydrothermal and calcination	565.1 246.6	200	200 4000	[110]
Carbon coated ZnO microspheres	Hydrothermal	527	100	100	[111]
ZnO-C nanosheets	Hydrothermal / CVD	851 804	50 200	100 1000	[112]

as enhanced rate performance. In conclusion, the compacted three-dimensional carbon frame could arise active electronic interactions by providing electron paths. In addition, the firm and resilience of novel carbon structures are significantly involved in diminishing the stress as well as volume changes in electrode compounds through the lithium insertion/deinsertion process [99].

Zinc oxide NPs encapsulated in 3D macro-/mesoporous carbon composites (ZnO/3DOM-mC) were synthesized to use as anode materials in LIBs. The prepared composites showed perfect electrochemical performance due to their special hierarchically porous morphology. Moreover, the specific mesoporous structure of the prepared composites permitted a larger space to accept volume changes through the charging/discharging process. The 3D carbon-based structure increased the electrode power [100]. A simple route-extended Strober method was applied to synthesize core-shell zinc oxide/carbon nanosphere composites, and the core-shell structure improved the specific capacity of composites due to the reduction of lithium-ion diffusion path as well as inhibition of the pulverization initiated from huge changes in volume through the lithium insertion/deinsertion procedure caused capacity fading decrease [101].

Nanostructured zinc oxide loaded/N-doped carbon foam (N-CF) composites were synthesized via a vapor-assisted two-step thermal method. The fabricated composite showed stable discharge capacities as well as excellent rate capabilities. The improved electrochemical characteristics of prepared composites were originated from a specific CF matrix which enhanced the close interaction between CF matrix and ZnONPs. In addition, these specific structures inhibited the growth of ZnONPs through the prolonged cycling time [102]. The comparison of the various structure of ZnO composites prepared by different methods for Li-on batteries application and their characteristics are summarized in Table 2.

4.3. Application of ZnO composites for supercapacitors

Supercapacitors (SCs) have been introduced as a novel category of energy storage devices with distinguished importance due to their quick charge-discharge rates, longer lifespan, and excellent power capability. The applications of SCs include backup energy sources, hybrid electric vehicle-based green transportation, and any other emergency power supply systems. SCs are divided into two groups based on the mechanism of energy storage: electric double-layer capacitors (EDLCs) and pseudo capacitors. EDLC is fabricated as a product of a double layer on the surface of an electrode. But, the pseudo capacitor keeps charge on the surface of the material and in bulk individually by a double layer mechanism and faradaic reaction. Therefore, the pseudo capacitor stores the charge 10-100 times more than EDLC [10]. EDLC capacitors collect the electronic and ionic charges among an electrode and electrolyte border and carbon-based materials. Usually, mesoporous carbon materials (activated carbon) are used as electrode materials in EDLCs and, recently, carbon nanotubes and graphene-like materials. In contrast, pseudo capacitors store charges through redox reactions at the surfaces of the electrode material, which is generally made of metal oxide and conducting polymers. EDLCs own lower energy density and higher power density compared to the pseudo capacitors. The EDLC can provide long time cycle life, but pseudo capacitors have higher specific capacitances. Therefore, the integration of the behavior of pseudo capacitors and EDLC, which is built on the metal oxide and carbon composite electrodes, could open new horizons to long cycle life and great specific capacitance of SCs [113].

The choice of appropriate electrode materials is crucial due to their effect on the performance of the SCs. Over the last years, several transition metal oxides, including RuO_2, Co_3O_4, ZnO, MnO_2, MgO, Fe_2O_3, and NiO, have been studied as a possible electrode material for SC. ZnO can be thought of as a potential candidate for SC electrodes because of its good electrochemical activity; although, the low conductivity and cyclic stability are the major problems for its application in SCs [64]. To solve this issue and increase the specific capacitances, hybrid composite electrodes have been fabricated by loading composite materials to ZnO electrode materials. The practical improvement is the result of the synergetic effect of separate ones in the composites through the particle size reduction, specific surface area increase, and higher porosity and raising the number of active sites. ZnO-based composite materials have many benefits, including low cost, environmentally friendly, good electrochemical reversibility, excellent special capacity, superior energy and power densities, proper cycling stability, and facile synthesis, which make them a suitable candidate for electrochemical supercapacitors [7].

Reduced graphene oxide (rGO)/ZnO nanocomposite prepared by ultrasonic-assisted solution method has been studied as an electrode material for supercapacitor application. The electrochemical studies showed the increased specific capacitance (312 Fg^{-1}) of the composite compared to the pure ZnO (200 Fg^{-1}) with improved cycling stability up to 1000 cycles and confirmed its ability as a proper electrode material for supercapacitors [114]. The graphene-zinc oxide thin film was synthesized by the ultrasonic spray pyrolysis process, which attained the maximum specific capacitance (635 F/g) [115]. In another report, rGO-zinc oxide nanocomposites were synthesized by hydrothermal process and applied in SCs, which achieved a maximum value for the capacitance of 719.2 F/g [116]. The three-dimensional graphene (3DG) framework has gained

Table 3.
Various ZnO composites for SCs application

Composite	Synthetic method	SC (F/g)	Scan rate (mV/s)	Current density (A/g)	Electrolyte	Ref.
rGO/ZnO	Ultrasonic spray pyrolysis	635	5	-	1 M KOH	[115]
RGO/ZnO	One step using $scCO_2$	314	100	10	2 M KOH	[125]
ZnO@rGO	Ex-situ wet chemistry	949	-	1	2 M KOH	[126]
ZnO/N–OHPC	Dual-templates with dicyandiamide	720	-	1	6 M KOH	[127]
ZnONPs/graphene	Electrospraying	89	-	1	1 M Na_2SO_4	[128]
ZnO nanocones	Chemical etching	378.5	20		1 M KOH	[129]
3D MnO_2 nanowire/ZnO nanorod	Low temperature solution	747	2			[130]
CC/ ZnO@C@NiO CSNAs	Hydrothermal	677		1.43	PVA/KOH	[131]
ZnO/Co_3O_4	Hydrothermal/ reflux/ annealing	1135	5	1		[132]
Mo:ZnO@NF//AC@NF	Hydrothermal	2296	5	1	3 M KOH	[133]

ZnO/N–OHPC: ZnO/nitrogen-doped hierarchically porous carbon
CC/ ZnO@C@NiO CSNAs: carbon cloth/ ZnO@C@NiO core-shell nanorods arrays

a lot of attention because of quick electrical charge transmission and the great possible solutions for the previously mentioned difficulties. 3DG/ZnO nanorods were fabricated by the hydrothermal method, which could achieve a maximum value for specific capacitance (554 F/g). 3DG/Zinc oxide composites were electrochemically more stable compared to the other ZnO-graphene composites [117].

Several pieces of research were carried out on the development of zinc oxide/carbon-based composites as electrodes to use in SCs. An electrode material fabricated of ZnO/ carbon nanotubes composite can achieve a specific capacitance (SC) of 324 F/g [118]. Kalpana and co-workers fabricated an electrode of ZnO/carbon aerogel and attained the maximum SC of 500 F/g [119]. In another study, ZnO/activated carbon composites were prepared by the sol-gel route and could achieve SC=160 F/g and 500 cycles stability [120]. An electrode fabricated of ZnO/carbon composites synthesized by the green chemistry approach could achieve SC=820 F/g. Additionally, an electrode composite made of ZnO/carbon retained 92% of capacitance retention up to 400 cycles with maximum SC of 92 F/g [121]. Xiao and co-workers could fabricate nanocomposites of core-shell carbon sphere (CS)@zinc oxide by a simple method of low-temperature water-bath to achieve SC= 630 F/g at the current density of 2 A/g [122].

The composite materials consisting of ZnO/metal oxide of MnO_2 and NiO have been studied to be used as an electrode in SCs with low price, the large hypothetical capacity of 1370 and 2573 Fg^{-1}, respectively [7]. The composites of ZnO-MnO_2 nano cables were synthesized by the method of high-temperature annealing/hydrogenation and could achieve the capacitance of 138.7 mF/cm^2 [123]. The electrode composites fabricated of core-shell nanofiber structure of zinc oxide-MnO_2 could achieve the value of SC= 907 Fg^{-1}. Pang and co-workers could synthesize zinc oxide–NiO porous nanostructure and achieved a maximum of SC=649.0 $Fg^{-1}$with and cycle stability of 400 cycles [124]. The comparison of the various structure of ZnO composites synthesized by various methods for SCs application and their characteristics are summarized in Table 3.

5. Conclusions and future insights

Extensive studies have been conducted on the principal features of zinc oxide-based composite materials regarding synthesis, development, modification, as well as their application in energy storage systems. In this review, a summative view of zinc oxide-based composite materials was presented with specific consideration on the applications in solar cells, batteries, and SCs. ZnO-based composite materials demonstrated outstanding energy storage capabilities as electrode materials used in SCs as well as lithium-ion batteries. Zinc oxide is usually supposed an active compound to be used in batteries with the predominant energy density of 650 A h/g. The high electrical conductivity of zinc oxide is about 230 S/cm which is more than any other oxide material. Also, ZnO-based composite materials could be used as photocatalysts to degrade the molecules of dyes in the visible light region. The morphology, particle size, the orientation of crystals as well as oxygen defects are some of the parameters which affect the photo degradation as well as the longtime constancy of zinc oxide composite materials. There are several approaches and strategies to increase the photocatalytic characteristics of zinc oxide, including fabrication of nanostructured arrays, integration to other materials such as noble metals, as well as doping and insertion of carbon compounds.

Extensive development has been identified on zinc oxide-based composites with various structural designs as well as synthesis procedures. Similarly, more researches could be made to fabricate nanocomposites with diverse compositions and examine different characterization techniques regarding morphological, structural, and different applications in energy storage devices. The low cost and facile syntheses procedures of ZnO-based composite materials make it feasible and capable for future applications. Although a huge number of studies and researches were conducted to synthesize and development of ZnO-based composite materials for various energy storage applications, further studies should be considered on how to enhance long-term durability, production on a large scale, and price. Therefore, more studies should be projected to explore the adjustable characteristics of zinc oxide-based composites in the area of energy storage systems.

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Conflict of interest

The authors declare that there is no conflict of interest.

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