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AN OVERVIEW OF DENDRIMERS AS CONDUCTING NANOMATERIALS IN SUPER CAPACITORS

Pratyusha Satavalli[1, 2], Latha P.[2] and Mamatha Nakka[1]
[1]Department of Chemistry, VNR Vignana Jyothi Institute of Engineering and Technology, Hyderabad, India
[2]Department of Chemistry, Annamalai University, Tamil Nadu, India
E-Mail: spratyusha1@gmail.com

ABSTRACT

Dendrimers are synthetic polymers, which have become a most captivating research topic for scientists, owing to their unique structure and versatile properties. Hitherto, most of the research was done about how it influences the fields of medical and Industrial processes. However, a few research groups have focused on the applications of Dendrimers in the electrical field as a conducting material in Super Capacitors (SCs). The transfer of electron charge between the core and the surface units of dendrimers is unique for their energy reposition. Some studies reflect on the usage of Dendrimers as electrodes, scaffolds of electrode materials, and electrolytes. Herein, we acquaint a review of the various methods of utilising dendrimers in Super Capacitors.

Keywords: dendrimer, nanomaterial, graphene oxide, supercapacitor, energy storage.

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1. INTRODUCTION

The social and economic accomplishment of the nation which is brought up by the Industrial Revolution is still dependent on the depletion of fossil resources like coal, gas, and oil. This causes significant climatic changes due to the emission of greenhouse gases from various anthropogenic activities. The United Nations proposed a guideline (i.e., adaptation and mitigation) that resolves the imbalances in carbon catalog throughout the world [1-8]. As a measure of those adaptation and mitigation strategies, the restriction of fossil resource consumption and the evolution of renewable energy resources have been enormously followed [9-22]. These renewable energy resources generate electric power which needs an efficient device for electrochemical storage inclusive of batteries, electrochemical Supercapacitors (SCs), and fuel cells [23,24]. The most alluring nature of Supercapacitors when compared to Batteries is their rapid storage capability and improved cyclic stability. Latest innovations in SCs in the context of electrode materials and electrolytes have momentous power in bridging the space among batteries, fuel cells, and existing electrolytic-capacitor technology.

A Supercapacitor is a pulse current system wthatis meant for reaching the demands of high current and specific power (10000 W Kg^{-1}) for periods of less than 1 min. [1]. Hence Supercapacitors (SCs) can be used individually or along with any other energy storage device like batteries by employing them in hybrid vehicles, cranes, trains, and elevators for power efficiency and improved cyclic stability. Now, tremendous research is going on in Supercapacitor technology focusing on the improved electrical efficiency of electrode materials and electrolytes [27-30]. Divergent SCs are also useful to achieve the power demand for initiating and regaining braking energy within drive systems, which utilise power from a voltage cause [31, 32]. As SCs can even safeguard the backup power supplies against power disruptions, they are considered as the most feasible option for energy storage applications [33, 34].

The efficiency of SCs is dependent on various aspects including the type of electrolyte used and the electrochemical properties of the electrodes used [1]. Most carbonaceous materials like Graphene, carbon nanotubes, and activated carbon, with high surface area, serve as electrodes for Electric double layer supercapacitors (EDLCs) which can store charge through Non faradaic process at electrode –an electrolyte interface [35-37]. On the other side, metal oxides like MnO2 [38], RuO2 [39], Conducting polymers Polyaniline [40], Polypyrrole [41], and PEDOT [poly (3, 4-ethylenedioxythiophene)] [42] because of their delocalised pi electrons [43] and GO [Graphene oxide] [44] are good pseudo-capacitive materials which store charge through fast redox reactions at the surface. In contrary to the above two types, hybrid supercapacitors are of two types-One of them is capacitor type electrode SCs (e.g., redox/redox type such as EDLCs/redox type). The other one is battery-type electrode SCs [45, 46].

The working mechanism of the SCs was classified as shown in Figure-1.

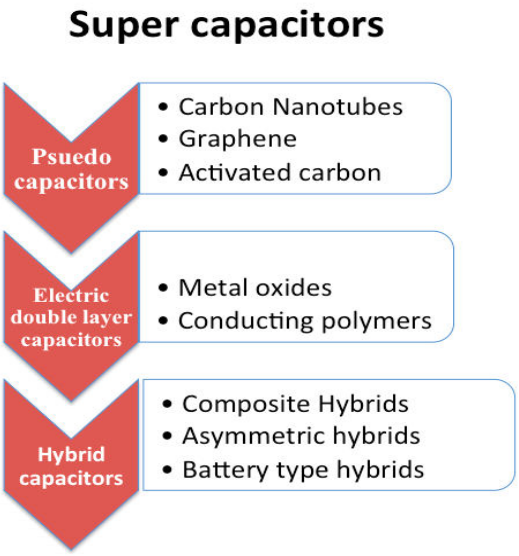


Figure-1. The working mechanism of SCs.

Various materials have been used as electrodes and electrolytic materials in SCs. Dendrimers are very useful contenders for both of the above-said purposes. These are fabricated three-dimensional hyper branched, spherical macromolecules which are marked by their highly branched 3D structure that imparts a high magnitude of surface functionality. They have three major parts-the Central Core moiety, branching units with variable surface groups, and cavities formed between the branched structures as shown in Figure-2.

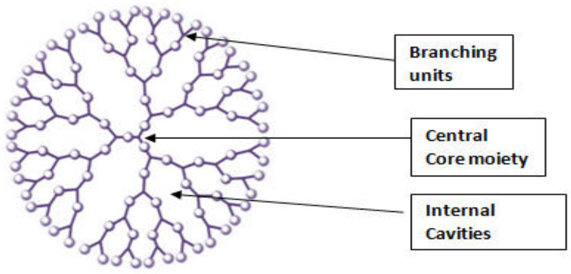


Figure-2. Structure of Dendrimers.

As their size and shape are accurately engineered, and are available at variable sizes/generations with their surface functional groups and cavities, dendrimers have a unique and surplus number of applications in a plethora of fields as shown in Figure-3.

1.1 Medical Applications

Dendrimers are used in targeted drug delivery where they act as a medium for carrying the drug to a specific cell or tissue of the body. According to the studies of Patri et.al targeted delivery of methotrexate is done by using PAMAM generation 5 dendrimer [47]. Dendrimers can be even used as gene transfer agents in gene therapy. Svenson et.al reported that PolyAmidoAmine (PAMAM) is used as a vehicle in gene therapy [48]. Magdalena *et al* reported that dendrimers also act as efficient MRI contrast agents [49]. To reveal that microRNA irregularities can result in genetic disorders Fengye *et al* proposed a PAMAM-MWCNT hybrid, which acts as an electrochemical biodetector for microRNA [50]

1.2 Industrial applications

Hawker. *et al* demonstrated that the electron-rich dendrimer synthesized from p(methoxycarbony1) benzyl and 3,5-dihydroxybenzyl alcohol acts as a good solubilizing agent comparable to Sodium dodecyl sulfate with recyclable solubilization property [51]. Dendrimer-operated magnetic nanoparticles, bound to the Fe-salen complex acts as an active and selective catalyst in oxidising various sulfides and sulfoxides according to the studies of Niakan *et al* [52]. According to the work of Dodangeh *et al* PAMAM dendrimers of variable generations were covalently attached to Curcumin, which is an efficient flouroscent chemos ensor. Especially they were reported as efficient metal cation chemosensors under a p^H range of 2-12 [53]. PAMAM terminated with bipyridyl derivative is an efficient non enzymatic sensor of H_2O_2 [54].

1.3 Electronic applications

According to the studies of Cho. *et al* Dendrimers play a vital role in modelling of Organic thin film transistors [55]. Dendrimers were manipulated into diverse forms of electrodes like Polyaniline nanowire arrays grown on dendrimer functionalised MWNT[56], Carbon nanotube/dendrimer composite [57], Dendrimer functionalised magnetic nanoparticles [58], Nickel hydroxide nanoplatelets grown through dendrimer over graphene sheets [59] and PAMAM [60] was used as electrolyte in Super Capacitors.

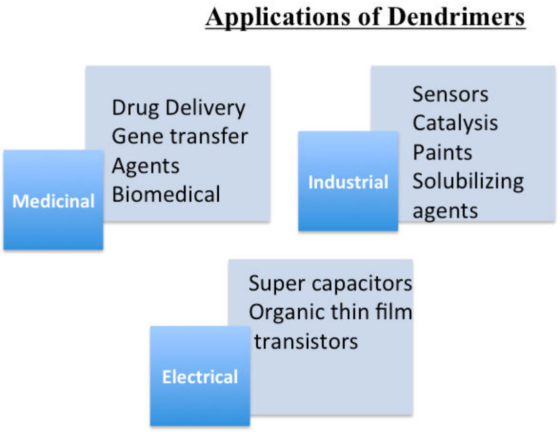


Figure-3. Applications of Dendrimers in various fields.

This review focuses on the utilisation of Dendrimers in various forms as electrodes and electrolyte materials in SCs, their efficiency in terms of capacitance, cyclic stability and energy storage mechanisms.

2. MATERIALS AND METHODS

2.1 Aligned Poly Aniline Nanowire Arrays Grown on Dendrimer Functionalised MWNT as Electrode Material

Jin *et al*., synthesized Poly aniline nanowire arrays grown on PAMAM dendrimer [56]. Initially, the COOH and COCl groups were introduced to MWNT by treating it with a mixture of nitric, sulphuric acids, and $SOCl_2$. Then covalently linking poly-amino dendrimer PAMAM on Multi-Walled Nanotube results in various reaction sites undergoing in-situ polymerisation with (Poly aniline) leading to ordered Poly aniline nanowire arrays as shown in Figure-4.

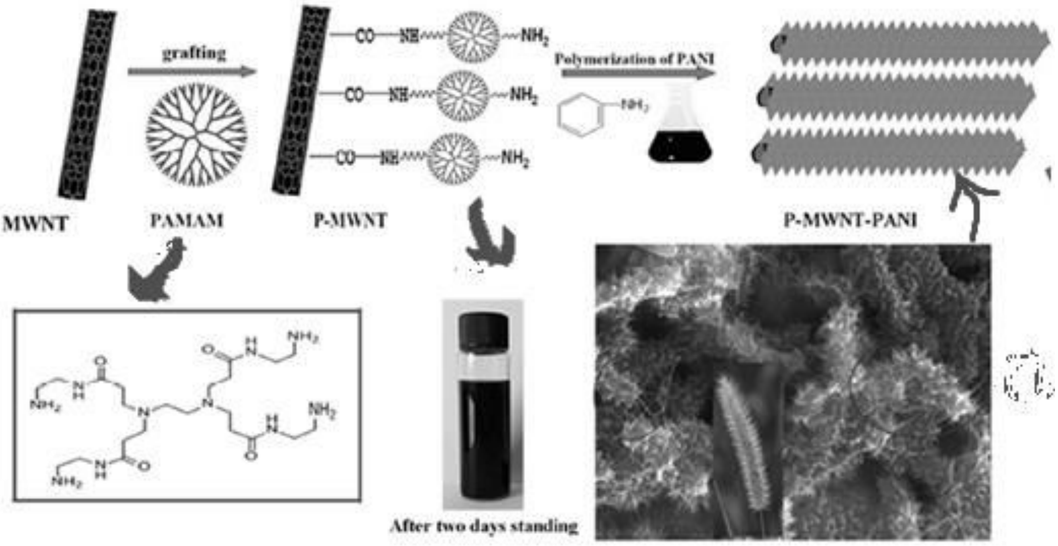


Figure-4. Framework for the synthesis of P-MWNT-PANI [56].

2.2 Hybrid of Carbon Nanotube/Dendrimer as Electrode

Paulo *et al*., prepared a hybrid of carbon nanotube / dendrimer. Commercially available MWCNTs were functionalised with amine and carboxylic groups then they are sonicated to form MWCNT with NH_2 and MWCNT with COOH , to this a mass of glycodendrimer is added and dissolved [57]. These dispersions of MWCNT with COOH/4-Gl and MWCNT with NH2/4-Gl were coated onto aluminium charge accumulators to form a single bilayer. An electrode was synthesized by depositing eight such bilayers and ionic electrolyte, EMITFSI (1-ethyl-3methyl imidazolium bis (triflouro methyl sulfonyl) imide was used in which the glass fiber membrane was soaked and embedded in between the CNT/dendrimer electrodes. They prepared 4 samples with 0, 10, 20, and 50 wt% by modifying the mass of dendrimer relative to the total mass of carbon as represented in Figure-5.

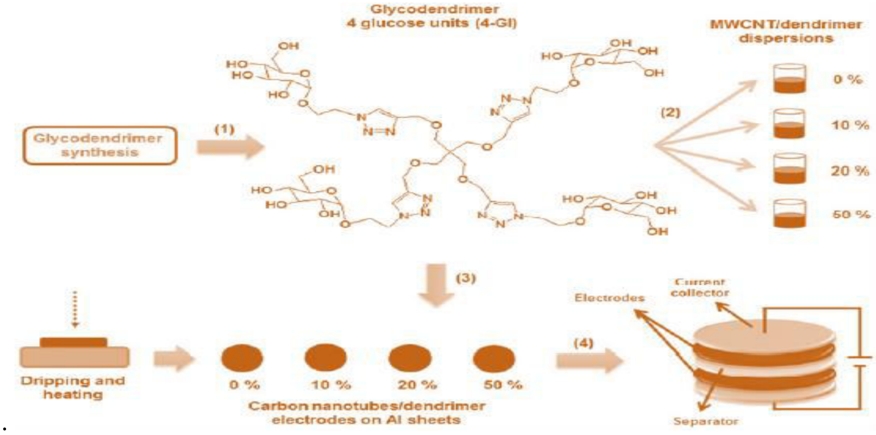


Figure-5. Schematic representation of the synthesis of supercapacitors [57].

2.3 Dendrimer Functionalised Magnetic Nanoparticles as Electrodes

Chandra *et al* synthesized Fe_3O_4@D-NH2 nanoparticles. A reaction mixture of Dieethylene triamine dissolved in methanol is mixed with methylacrylate and agitated for 5 days at 25^0c in a static atmosphere [58]. This results in a generation-1 dendrimer with 5 methoxy teminal moieties. Dried methanol and ethylene diamine were added to this and stirred which results in generation-1 dendrimer with 5 amino terminal moieties. This is then purified and the same procedure is repeated to synthesize generation-2 dendrimer with 10 amino end moieties. This formed redox active NH2-PAMAM dendrimer is then used to stabilize and functionalize Fe_3O_4 to synthesize the so-called Fe_3O_4@D-NH2 nanoparticles as shown in Figure-6.

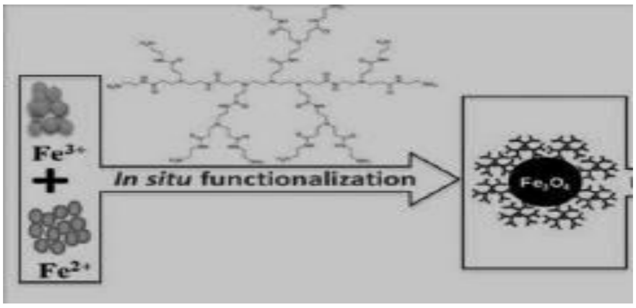


Figure-6. Graphical representation of the preparation of Fe_3O_4@D-NH2 nanoparticles [58].

2.4 $Ni(OH)_2$ Nanoplatelets through Dendrimer Assisted Growth on Graphene Sheets

Naveen *et al.* synthesized dendrimer oriented $Ni(OH)_2$ nanoplatelets developed vertically on graphene sheets (NDG) as electrode materials for supercapacitors. On dissolving $NiCl_2 \cdot 6H_2O$ in ethanol, PAMAM is mixed into the solution with riveting leading to the complexation of Ni^{2+} with DENs. In a water/ethanol mixture, GO synthesized from synthetic Graphite was dispersed and added to the reaction mixture (complexation of Ni^{2+} with DENs) under vigorous stirring. Hydrothermal treatment was done to the dark brown precipitates formed and then it is dried and filtered. In the same manner, various compositions of dendrimers to $Ni(OH)_2$ ratios were used to synthesize different $Ni(OH)_2$/DEN/GO (NDG) ternary composites like NG(without dendrimer), NDG-0.2(10 mg of dendrimer), NDG-1(50 mg of dendrimer), NDG-2(100 mg of dendrimer) [59].

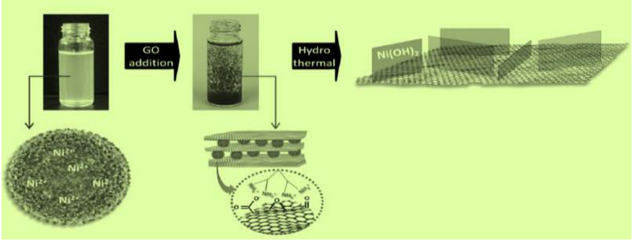


Figure-7. Schematic representation of the preparation of NDG ternary Composites [59].

2.5 Dendrimer as Electrolyte

Lin. *et al.* utilized PAMAM dendrimer as electrolyte in an Electric Double Layer Capacitors [60]. All essential characteristics required for an electrolyte can be fulfilled by PAMAM such as the deformation of surface groups to expose to more charges and also avoiding salt formation leading to effective charge density. PAMAM G-0.5 with carboxyl end groups have a negative charge.

3. RESULTS AND DISCUSSIONS

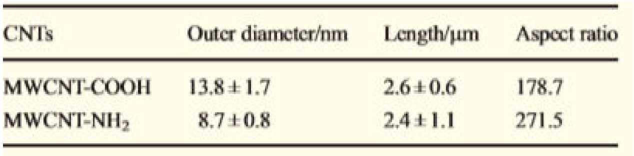
3.1 Jin *et al.* elucidated the structure of the samples they synthesized through FT-IR. It shows 2 peaks indicating the C-N and C=O bonds representing the functionality of PAMAM on MWNT. Then this MWNT and PANI chemical reciprocity was exhibited by a red curve in FT-IR and Raman Spectrum analysis. The ordered growth of PANI nano fibers on MWNT was analysed by XRD studies, BET-specific surface area, and SEM images [56]

The Cyclic Voltammeter (CV) and Galvanostatic charge-discharge (GCD) studies elucidate the electrochemical behaviour of these Composites. Cyclic Voltammeter studies tell that area and electric current density of P-MWNT-PANI composite are remarkably more compared to PANI, PANI-MWNT.GCD curves of the composite resulted in an improved capacitance of $540Fg^{-1}$ at $1Ag^{-1}$ of current.

Integration of P-MWNT directs the growth of ordered nanostructure of PANI, enhancing the rate proficiency of P- Multi-Walled NanoTube/PolyANIline but reducing the electrolyte's and active material's ion diffusion length. The Electrochemical Impedance Spectroscopy analysis is also in line with the outcomes of CV and GCD studies representing the enhanced surface area which is useful for good electrolytic ion diffusion to the composite surface. The obtained composite material has high cyclic strength compared to PANI and PANI-MWNT. The graph represented 85% of the original capacitance was maintained after 2000 charge – discharge series at 2 Ag^{-1} of current.

3.2 Paulo *et al*. Employed SEM, TEM studies to analyse the structure of the MWCNT-NH2, MWCNT-COOH, and the electrodes developed. TEM and other studies showed that the materials are of high purity. Fig-9 represents the Scanning Electron Microscope picture of one bilayer electrodes with 0, 10, 20, and 50 wt% of 4-GI dendrimer/MWCNT. Among the 4 only 20 and 50 wt% dispersions resulted in good electrode coverage, confirming its efficient capacitance.

Table-1. Physical properties of oxygenated and amino functionalised MWCNTs [57].



CNTs	Outer diameter/nm	Length/μm	Aspect ratio
MWCNT-COOH	13.8 ± 1.7	2.6 ± 0.6	178.7
$MWCNT-NH_2$	8.7 ± 0.8	2.4 ± 1.1	271.5

Electrochemical characteristics were analysed by Cyclic Volammeter and Galvanostatic charge-discharge studies. They exhibited a capacitance of 9.5, 13.1, 57, and 45.5 F g−1 for 0, 10, 20, and 50 wt% of the 4-Gl dendrimer, respectively at 10 mV s^{-1} of scan rate as given.

3.3 XRD and FT-IR were used for the structural characterisation of Fe_3O_4@D-NH2 nanoparticles by Chandra *et al* [58]. The FT-IR findings show N-H, O-H, Fe-O, and CO stretching vibrations. The stability of nanoparticles with dendrimer was also confirmed by magnetisation values [22]. TGA and TEM studies were also done for structural analysis. Surface properties like specific surface area and porosity were calculated from the N2 adsorption/desorption

Cyclic Voltammeter was used to assess the electrochemical behaviour of the sample material in five distinct electrolytes like Na_2SO_4, KCl, KCl-PBS (phosphate buffer), tetra butyl ammonium tetra fluoro borate (with 5mL of methyl alcohol) (TBATFB) and propylene carbonate.70-120 Fg^{-1} range of best capacitance values were recorded based on the material load in different electrolytes.

GCD measurements show the retention of capacitance after 500 cycles for the magnetic nanoparticles decreases in this order in these electrolytes Na_2SO_4, KCl, KCl-PBS, Propylene Carbonate, and TBATFB.

3.4 The ternary composite's structural confirmation by Naveen et.al was done through XRD, XPS, FESEM, and HRTEM, etc. techniques [59]. The reports of XRD reflect the growth of crystalline $Ni(OH)_2$ and the effect of Dendrimers in the growth of microstructures. The XPS studies were used to calculate the corresponding quantities of $Ni(OH)_2$ to dendrimers. FESEM studies reported the impact of dendrimers on microstructural morphology. All the above studies represented that the composite, with no dendrimers has accumulated $Ni(OH)_2$ nanoparticles on the surface of Graphene, in contrast, NDG-0.2 composite exhibited the start of ordered development of $Ni(OH)_2$ and the NDG-1 composite has clear vertically developed $Ni(OH)_2$ nanoplatelets on Graphene layers as a result of the amount of the dendrimers involved. The NDG-2 ternary composite had an irregular growth of nanoparticles due to the high amounts of dendrimer used.

The Cyclic voltammogram studies of NG, NDG-0.2, NDG-1, and NDG-2 reflect specific capacities of 496, 560, 1226, and 1013 Cg^{-1} respectively. It depicts that dendrimer-assisted $Ni(OH)_2$ nanoplatelets growth perpendicular to Graphene, in the form of NDG-1 influenced the effective increase of capacitance compared to NG, NDG-0.2, NDG-2.

Galvanostatic Charge discharge studies were in good agreement with the above studies exhibiting a high capacity and long cycle life by NDG-1. The high capacity retention of NDG was observed even at 5000 cycles which is a consequence of its microstructural features. Even in asymmetric full cells the NDG-1 when combined with RGO, exhibited more energy and power densities.

3.5 NMR and EPR (Electron Paramagnetic Resonance) studies of G-5 and G-2 PAMAM performed by Lin *et al*. reported that PAMAM is an unstable electrolyte [60]. The NMR studies showed that after 24 hours of solvolysis, the amine groups began to detach and almost disappeared at 48 hours. GPC (Gel Permeation Chromatography) studies of PAMAM were conducted in DMAc and observed low molecular weights compared to theoretical values. The variation can be due to the use of high temperatures during the run and the mobile phase used.

4. CONCLUSIONS

The review highlights the utilisation of Dendrimers in Super Capacitors as conducting materials in versatile approaches. The reports displayed that Dendrimers were engineered into various forms of electrodes such as they become a foundation for building the electrode materials, as an electrolyte in various mechanisms of Super Capacitors with good efficiency. However, the amount of research done in the path of idealization of dendrimers as conducting materials in Super Capacitors is very narrow. Our effort of compiling the respective methodologies should drive the attention of researchers in the concerned fields to achieve the goal.

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