

POLYVINYL BUTYRAL (PVB), VERSETILE TEMPLATE FOR DESIGNING NANOCOMPOSITE/COMPOSITE MATERIALS: A REVIEW

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Abstract

In current article, the versatile behaviour of Polyvinylbutyral (PVB) and its ability to form composite materials with various inorganic species is reported. PVB has hydrophilic vinyl alcohol groups and hydrophobic vinyl butyral groups. These groups behave as promoters of polymer adhesive and binders for organic moieties. The composite materials of PVB have been synthesized via physical as well as chemical both protocols. PVB is used as a constituent part in the formation of composite, induces a specific property in a resulting one which are utilized by various ways since it has stronger in binding ability, sharper optical clarity and able for providing flexibility and toughness in the formed composite. Various sophisticated instrumentation techniques eg FTIR, XRD, FESEM, TEM etc. are reported for characterizations of samples. The composite materials have excellent film formation properties, and can be potential candidate for photoelectric as well as photovoltaic applications. The inorganic conducting species which do not have film formation ability can be useful by composite formation along with PVB. The green protocols for synthesis of composites may also useful for biological applications.

Key Words: Composite Material, Hydrophilic, Hydrophobic, Nanofibre, Antireflection coating.

INTRODUCTION

The composite materials are always remaining embedded in humankind since the beginning of human civilization. A great deal of composites is available around us and is also synthesized in laboratory, since these are being used for the preparation of significant articles. Any material, formed by combining two or more materials in order to accomplish some booster properties in resulting one, than the sum of the individual components is specified as composite. A vast verity of composites is available around us as found to be present in natural resources and also synthesized in the laboratory by chemists for daily use material. The nomenclature of the materials based on its component size, induced while initiating the composites, resolves to be calling them as micro or nano. Micro composites may simply be thrash out as composite materials since these are anciently came in commencement, parading miscellaneous examples like a conjugated polymer poly(1,4-phenylenevinylene) (PPV), was synthesized inside the channels of the mesoporous silica MCM-4 to know the electrooptical properties of conjugated polymers present in encapsulated form[1] and a composite gel is produced by composing (PNIPAAm and clay platelets) by ice-template assembly (freeze-casting) and UV-initiated cryopolymerization, found to be thermo responsive and having marvelous mechanical potential [2]. Composite material is formed just to get the best performance/property of any component, unattainable in individual form since in composite form it shows different characteristics from its previous form and typically produces a very advantageous picture regarding to different aspects e.g. composite materials are employed as material used for electromagnetic radiation protection [3], as anode material to enhance electrochemical properties in lithium ion battery [4,5], to offer a good deal of fire retardency when made by the combining with polypropylene with sepiolite and with organo-sepiolite [6]. In bio-molecular field, a composite made up of DNA and the cyclodextrin-immobilized poly (allylamine) (PCD) shows properties of both the doublestranded DNA, like intercalation and cyclodextrin, such as encapsulation of organic molecule in intramolecular cavity [7].

As composites are unificated by the combination of two or more constituent materials, when any phase of such materials induces particles in the nano range then termed as nano composites. Nano range is considered to be less than100 nm. The properties of nanocomposite materials improve tremendously and diverge from the properties of constituent materials. There is an increment of stability in nanocomposites due to being smaller in size (surface/volume ratio). Same factor operates behind their mechanical strength, being smaller in size, in nanoparticles more surface area is available to interact with other substance producing relatively influencer mechanical strength [8,9].

With respect to applications for various fields, lots of nanocomposites synthesized, by using different methods, have been brought up such as two-dimensional molybdenum disulfide (MoS_2) and silver nanoparticles (AgNPs) as are reported to have admirable graphene properties [10]. Graphene/SnO₂ (GS) nanocomposites are being synthesized in an acidic medium and have been used for the determination of electrochemical properties of dopamine, ascorbic acid, and uric acids [11], Polyluminol and graphene oxide nanosheets are synthesized electrochemically to show supercapative and pseudocapative behavior [12]. Lots of reports are



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available in literature to synthesize the nanocomposites using different metals or constituent materials. There is a wide area of nanocomposites, used to satisfy different required aspects like synthesis of copper-modified ordering mesoporous carbon-silica nanocomposites (MCSs) is made to take place which is used to enhance adsorption of dibenzothiophene (DBT) selectively [13]. Nanocomposites of CeO2.CuO.ZnO oxides have been synthesized which exhibited Photoluminescence, photocatalytic and antibacterial activities [14] while nanocomposites of Fe_3O_4 onto ZnO/AgCl are synthesized at low temperature, which are magnetically separable and show greater photocatalytic activities under visible-light irradiation [15]. TiO₂-SiO₂ thin films nanocomposites with different compositions (up to 100 mol % TiO₂) were deposited on silicon by using sol-gel dip coating method [16]. In a same way nanocomposites consist of chiral polyamide-imide (PAI) and modified zirconium nanoparticles (ZrO₂NPs) with polyvinyl alcohol are synthesized using green method [17]. Silver nanoparticles were synthesized in situ in chitosan hydrogel with the help of sodium tripolyphosphate which shown great potential to be used as drug delivery [18], in the same way silk-gold nanocomposite is synthesized which is used to prepare the nerve conduits for functional peripheral nerve regeneration [19] and carbon-gold nano composite (CGN) is used as immune probes for the formation of electrochemical immune sensor to be used in determination of cancer biomarkers [20]. There is a series of number of nanocomposites made up of TiO_2 as one of the constituent applying in various fields on the basis of their applications, it applies in the photocatalytic degradation of organic pollutants in water like chlorobenzene, phenol methyleneblue dyes etc [21-23], for the removal of radioactive metal ions like cesium (Cs⁺), cobalt (Co²⁺) and europium (Eu³⁺) from aqueous solution [24], to increase potential of lithium ion battery [25] to betters up the performances of biosensors, photo catalysts and other optoelectronic devices [26].

Polyvinyl butyral (PVB), a resin, being stronger in binding, sharper in optical clarity, and being able for providing flexibility and toughness, opens a way of being used as a constituent part of a composite while solving an issue of applications in numerous fields since composites and nanocomposites of PVB are being used to fulfill many important aspects. PVB has vast applications as laminated safety glass, paint, adhesive, primers binders, etc. This random copolymer is completely amorphous in nature and is being synthesized from polyvinyl alcohol and butyraldehyde in acidic medium (Figure-1). The random structure of PVB results a glassy polymer with no apparent crystallinity except at very high alcohol content. PVB found to be polyhydroxy and Polyacetal, since it contains predominant proportion of (Major) butyral, hydroxyl and (very less) acetyl groups [27] (Figure-2). Among the groups present in PVB moiety, vinyl alcohol group is hydrophilic while vinyl butyral group is hydrophobic in nature.



Polyvinyl alcohol

Polyvinyl butyral

Figure 1: Synthesis of polyvinyl butyral from polyvinyl alcohol and butyraldehyde [31].

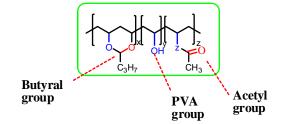


Figure 2: Structure of polyvinyl butyral represents different groups.

These groups behave as promoters of polymer adhesive and binders for organic photoconductors in electrographic process. The non-polar, polar components of PVB can interact preferably with other macromolecules and it may therefore be compatible with both hydrophilic and hydrophobic components. The percentage alteration of any monomer component can manipulate hydrophilicity or hydrophobicity of polymer [28]. Therefore, it is important to understand the intermolecular interaction of vinyl alcohol and vinyl butyral moieties. It may also facilitate the use of PVB as a copolymer with potential miscibility with polymers having widely dissimilar groups. Polyvinyl butyral is served as a material of interlayer in laminated automotive safety glass to prevent glass fragments from being spread out on sudden, damaging and breaking and to make safe from heat or light, resulting of UV radiations or solar energy by diminishing their effects since exhibits a micro porous structure and hence may be used in ultra



filtration membrane too. PVB is soluble in polar solvents which enlarge its ionic conductivity resulting prevention in its vulnerability to splash caused by non polar substances [29,30].

While examining optical and electrochemical properties of PVB, it is very stable on these criteria however mechanical properties may be enhanced by the induction of nanofillers, providing it a mechanical support [31]. The FT-IR spectrum of polyvinyl butyral is shown in *Figure 3*, Representing characteristic peak for OH group, shown at 3442 cm⁻¹, which is little broader peak, deviating from its normal value ~3600 cm⁻¹ to show the presence of intermolecular hydrogen bonding. The peak for carbonyl group appears at 1639 cm⁻¹.

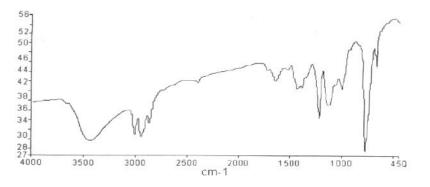


Figure 3: FT-IR Spectrum of Polyvinylbutyral (PVB).

PVB INDUCED COMPOSITE MATERIALS

PVB is cheaper pitted polymeric material consummates a wide range of industrial applications. Numerous materials, necessitating for different types of applications, involve the use of PVB. It provides a wide range of applications described as -

In the Preparation of antireflection coating

To assemble antireflection coating PVB is used along with tetraethyl orthosilicate (TEOS), a silica precursor. The reaction is made to take place through sol-gel path, where hydrolysis and condensation of TEOS is catalyzed by basic condition, provided by ammonia while ethanol is used as solvent. Since PVB is a sticky material for glass and plastic with magnificent pellucidity, may be applied as a modifier while preparing high transmittance organic-inorganic hybrid antireflecting coating.

The properties of antireflecting coating were characterized with the help of different spectroscopic techniques, i.e. FT-IR absorption spectroscopy, transmission electron microscopy (TEM), programmable rheometer, particle size analyzer UV-VIS spectrometry, ellipsometry, atomic force microscope (AFM) and contact angle measurement. The modification of silica with varying amount of PVB also revamps its properties. The transmittance property of a traditionally pure silica coating lessens in moisture since it exhibits easy water absorption property from surroundings. Induction of PVB to silica introduces hydrophobic groups into silica sol. It is observed that modification of silica with 4% PVB greatly enhances its moisture resistance property. On modification of silica with PVB, the particles of the modified sol grow faster which may attributes to the formation of the PVB bridges among silica particles to provide denser coating due to higher viscosity.

Figure 3 shows TEM images of normal and PVB modified sol for two weeks aging time indicating that PVB modified silica consisting larger particle size which will lead to more scattering of light on the surface of anti reflecting coating since larger particle size provides rougher surface. This anti reflecting coating is crack free before and after heat treatment till160 °C and is highly transparent [32]. PVB modified silica consists of larger particle size which will also lead to more scattering of light on the surface of antireflecting coating since larger particle size provides rougher surface. This anti reflecting coating since larger particle size which will also lead to more scattering of light on the surface of antireflecting coating since larger particle size provides rougher surface. This anti reflecting coating is crack free before and after heat treatment at 160°C and is highly transparent [32].



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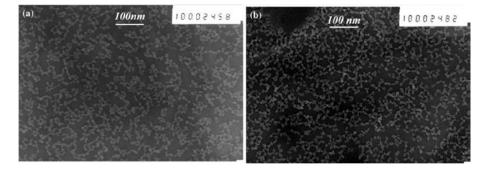


Figure 4: shows TEM images of normal and PVB modified sol after aging.

Mesopores in Zeolites

Aluminosilicate zeolites are materials used as catalyst in different processes and as adsorbent in ion exchange technique based industries. In zeolites channel and cavity range between 30-150 nm, providing furnishing in its working behavior but since there is a limitation of pore size in zeolites, which results undesired moderate diffusion rate of reactant and product. This shortcoming might be removed by shortening the pore size. Definitely there would be few obstacles while achieving the right way like in the duration of its synthesis, its colloidal nature and consumption of larger amount of template make this work harder. To overcome these problems PVB is used to generate mesopores in the ZSM-5 zeolites in both inter and intracrystal manner.

PVB is a material possessing hydroxyl groups in its structure to provide sufficient affinity towards silica and to allow PVB gel to disperse easily in silica surface. Synthesis of mesoporous ZSM-5 is made to take place through three different routes, by direct hydrothermal treatment of PVB/silica composite using sol-gel path way, by crystallizing PVB hydrothermally, impregnated Aerosil-200, and by the re-crystallization of zeolite precursor in the presence PVB; for the clarity, the zeolite samples formed using above these methods are denoted as sample-A, sample-B, and sample-C while the conventional ZSM-5 synthesized without PVB is denoted as Con-ZSM.

It is shown in *Figure-5* sketches, the XRD patterns of ZSM-5 samples which are synthesized by different methods. These all samples possess the characteristic diffraction peaks occurred at 2 of 7.9° , 8.8° , 23.1° , 23.9° , and 24.4° . It is seen that the three samples synthesized in the presence of PVB also exhibit good crystallinity; hence the presence of PVB gel in the synthesis system does not produce negative effect on the formation of ZSM-5 zeolite. However, the XRD pattern of Sample-C little differs with slight broadening in its pattern showing that sample-C is polycrystalline with the aggregate nano size units. TEM micrograph is represented in *Figure-6 (a-d)* which informs that conventional ZSM-5 synthesized without PVB does not possess mesopores in it since there is no clear bright spot showing its presence.

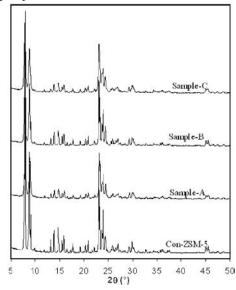


Figure 5: XRD pattern of conventional and mesoporous ZSM-5.



The TEM of Sample-A indicates that there are lots of mesopores in a single crystal appearing as bright parts shown on the surface of ZSM-5 crystal. Sample-B mesopores are smaller, distinctly different from Sample-A structure; the Sample-C is polycrystalline with an aggregate of nanosized particles [33].

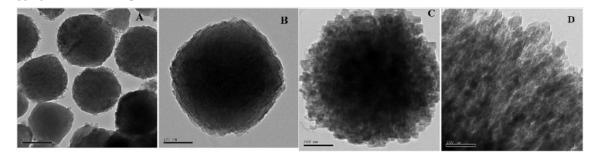


Figure 6: (A-B) TEM image of regular ZSM-5 synthesized by conventional method, (C-D) picture of mesoporous ZSM-5 (Sample-A) synthesized by hydrothermal treatment of PVB/silica composite.

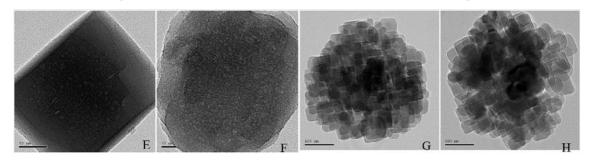


Figure 6:(E-F) TEM picture of mesoporous ZSM-5 (Sample-B) synthesized by direct hydrothermal treatment of PVB impregnated Aerosil-200, and (G-H) TEM picture of mesoporous ZSM-5 (Sample-C) synthesized by re-crystallization of preformed ZSM-5 in the presence of PVB [33].

Preparation of polymeric electrolyte

PVB is found to be a suitable polymeric material in the formation of solid electrolyte which is a component of electrochromic cells. The electrochromic material based devices operates the optical properties like optical transmission, absorption, etc. and these devices are being used in advanced applications like smart-window, electrochromic mirror and electrochromic display devices. There are two types of electrolytes, found to take place while the electrochromic cells are incepted that are liquid electrolyte and solid electrolyte. Since liquid electrolyte based cells often face the issue of dissolution of electrochromic film, which results degradation of device, contrary solid electrolyte based cells give better performance hence has become a topic of emphasis. Recently polymer solid electrolytes, based on lithium salts are prominence path in the manufacturing of the electrochromic devices. The basic function of the polymeric material is to solvate salt to form polymer electrolyte which is done by the functioning of the ether group present in host polymer. Now a days, the ethylene oxide group based polymers are excessively used to fulfill this purpose; however while using this path, we must reduce the crystallinity of polymer in order to achieve high conductivity at room temperature.

PVB, completely amorphous in nature, is used as a desired material in electrolyte since the resulting PVB complexes are also amorphous and hence resulting electrolyte remain independent of non-trivial problem of crystallization of complexes with polymer like problem faced in case of polyethylene oxide (PEO). *Figure 7* shows typical X-Ray diffraction pattern for pure PVB samples, PVB, LiI in 1:0.15 ratio and PVB, LiI in 1:0.2 ratio which are represented as sample 0, sample 3 and sample 5 respectively.

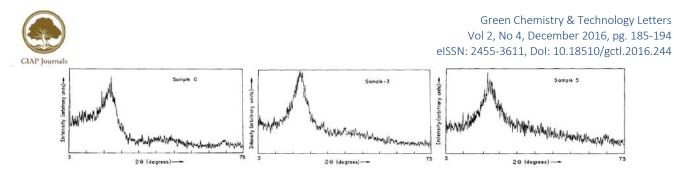


Figure 7:X-ray diffraction pattern for different weight ratio PVB: LiI= 1:0 (sample-0), 1:0.15(sample-3) and 1: 0.2 (sample-5).

In the XRD pattern of pure PVB, there is no crystalline peak which reveals PVB as completely amorphous in nature; while the XRD pattern of sample 3 and sample 4 show presence of crystalline character. With the addition of LiI, the amorphicity of the sample reduces and the conductivity too. The reason behind the decrease in conductivity is suggested to be increasing in viscosity, since the viscosity produces the decrease in the mobility of ions.

An electrochromic device consisting of PVB based solid polymeric electrolyte, showed reasonably good switching behavior indicating that PVB based solid polymeric electrolytes are cheap, robust, electrochemically stable ion conductors and can be used in electrochemical devices commercially [30].

Synthesis of mullite nanofibre

Mullites are also termed as porcelainites which are rare silicate minerals. These are formed by the combination of alumina and silica and are resistant of phase change at high temperature. Ceramic fibers are material with properties of thermal shock damage resistance, chemical stability, flexibility and dielectric properties which facilitate their use as reinforcement material.

A number of methods have been introduced for the synthesis of such fibers, like sol-gel method, melt-spinning method, solution spinning method and electrospinning method. Among these methods electrospinning method for the preparation of nanofibre is a point of attraction since last few decades because of its versatility and cost-effectiveness for the fabrication of one dimensional nano or micro fibers. Electrospinning fibers have a characteristics of production of ultrafine fibers hence are able to fulfill a wide range of applications related to medical point of view such as wound dressings, tissue scaffolds, drug delivery, filtration as well as shielding clothing for the military applications.

Mullite nanofibre are synthesized by the addition of varying amount of Polyvinyl butyral (PVB) in the prespinning solution in order to find out the most appropriate viscosity of the solution at which the nanofibre with smaller size, smoothness and most uniform appearance are obtained by electrospinning process.

The surface morphology of the nanofibre with different PVB content was investigated through scanning electron microscopy (SEM). *Figure 8 (a-g)* shows the effect of different PVB amount on the morphology of the nanofibre. *Figure a* and *b* indicate that when PVB amount is less than 4.0 wt% then completely and partially beads are obtained while on increasing its amount, ranging from 4.0 to 6.0 wt%, as shown in the *figure (c) to (e)*, continuous nanofibre with normal cylindrical morphology are obtained. On further increase in PVB amount to 8 wt%, this led to the impression of wide and flat ribbon structured morphology of the nanofibre.

It is observed that the viscosity of the PVB prespinning solution played an important role on the morphology of the mullite nanofibre. The incessant electrospun nanofibres with proper features are achieved while taking PVB amount at 4 to 6 wt% [34].



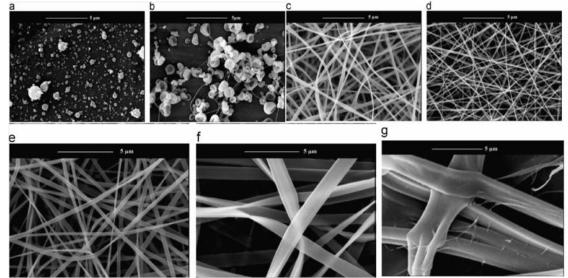


Figure 8: Morphologies of electrospun fibers with (a) 0 wt%, (b) 2 wt%, (c) 4 wt%, (d) 5 wt4%, (e) 6 wt%, (f) 7 wt%, (g) 8 wt% of PVB.

Synthesis of Alumina fiber

Due to their high strength, thermal stability and other properties, alumina fibers are used as reinforcement of metals or ceramics since it forms a continuous sequence while arranged to form a fibers. Alumina fibers are used in the production of number of house hold products due to its high temperature insulating property.

The Alumina fiber was prepared by the synthesis of polyvinyl butyral-AlNO₃ composite sol. The synthesis of composite sol takes place by a sol gel process in aqueous medium resulting alumina fiber with smooth surface and uniform diameter, in the whole process ethanol is used as solvent. The alumina fibers have been characterized by X-ray diffraction (XRD), FTIR, SEM, DTG/DSC etc.

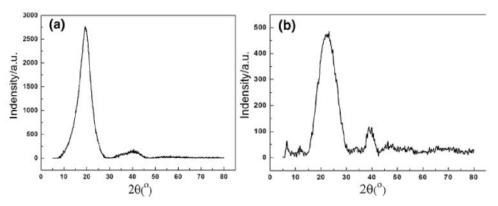
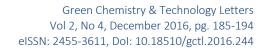


Figure: 9 Representing XRD pattern for (a) PVB and (b) AN-PVB [35].

In *figure 9 (a)* and *(b)* XRD pattern of PVB and PVB-AlNO₃ is represented. The XRD pattern of PVB-AN suggested the successful reaction between PVB and AN, here there is a presence of a new peak at $2 = 6^{\circ}$ in contrast to the XRD pattern obtained for PVB. Moreover, in PVB-AN pattern there is a absence of reflection peaks of AN which may attributes to dehydration of composite gel by the evaluation of NO₂ gas to give Al₂O₃ Which can be represented by the following equation.

$$4AINO_3 \longrightarrow 2Al_2O_3 + 12NO_2 + 3O_2$$

The FT-IR spectra of PVB, AN and PVB-AN also show the peaks at 2493 cm⁻¹. The FTIR spectrum of PVB-AlNO₃ arisen due to OH vibration in COOH group indicating that there is some reaction taking place [35].





Polyvinyl butyral is cheaper in cost and tough polymeric material which is served to satisfy a wide range of industrial applications [29], like it has outstanding adhesive properties with many materials such as glass, metal, plastics and wood. The PVB is predominantly used in safety glass laminates, specifically in automotive and architectural glass [36], comprises a protective interlayer to bond two panels of glass. The bonding process takes place under heat and pressure, making the PVB layer optically clear while binding the two panes of glass concurrently. Once sealed together, the glass "sandwich" behaves as a single unit and appears in the same as normal glass [37]. Polyvinyl butyral resins are also used in a range of applications including technical ceramic binders, inks, dye transfer ribbon inks, paints and coatings, binders for reflective sheet and binders for magnetic media. PVB is seemed to be suitable as a matrix to form nanofibre with Polyaniline (PANI) by electrospinning (ES) in the construction of surface acoustic wave humidity sensor [38]. Using PVB, photovoltaic thin film solar modules are also being manufactured. PVB may be availed to construct the photovoltaic circuit on a glass sheet. Solid state reference electrode has been assembled with the use of polymeric membrane of polyvinyl butyral (PVB), Ag/AgCl and NaCl for decentralized chemical measurements.

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