

# Developments in nano-additives for paper industry

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**Abstract** The economic benefits of the paper industry made it one of the most important industrial sectors in the world. This review will focus on the recent development of nanotechnology, in the context of additives used in the paper industry. Nanotechnology is used to alter the production process based on the changes towards resource-based and industrial knowledge, prompting the changes in a similar direction, with much greater emphasis on stability. Nanofiber, nanofiller, nanocomposites, and nanoscale chemicals used in paper applications forms the crux of the work. The application and production of nano-additives to the paper industry have attracted the attention of scientists and researchers, due to the resulting profound improvement to the properties of the produced papers, encompassing mechanical, printability, glossiness, and gas barrier properties. The purported benefits of nano-additives are numerous, among them high surface area, strength, and low weight, high stiffness, and maintained sustainability. However, the integration of nanotechnology in the paper industry could not be up-scaled or commercialized, due to a few challenges, which are increased cost, lack of compatibility among materials, and knowledge gap.

**Keywords** Nanoparticle · Surface area · Adsorption · Wettability

## An overview: paper industries

The global production of paper and paperboard is now comfortably in excess of 400 million tons, with the slow-down in the USA and Europe more than offset by corresponding growth in Asia [1]. The Asian production is in excess of 40 %, while ~30 % comes from Europe and ~25 % from North America [2, 3]. This total volume is only expected to rise significantly, in real (not inflated) terms, in 2014, as the Asian market starts to make up for the decrease in the American and European markets [4].

The primary stage of the manufacturing process of paper is related to material preparation, pulp manufacturing, pulp bleaching, paper manufacturing, and fibers recycling [5]. The refined, and if necessary, bleached wood pulp, is prepared as a thin slurry, together with a number of additives, especially fillers, to create the correct mix for the wanted paper quality [2, 4] (Fig. 1).

## Manufacturing: paper industries

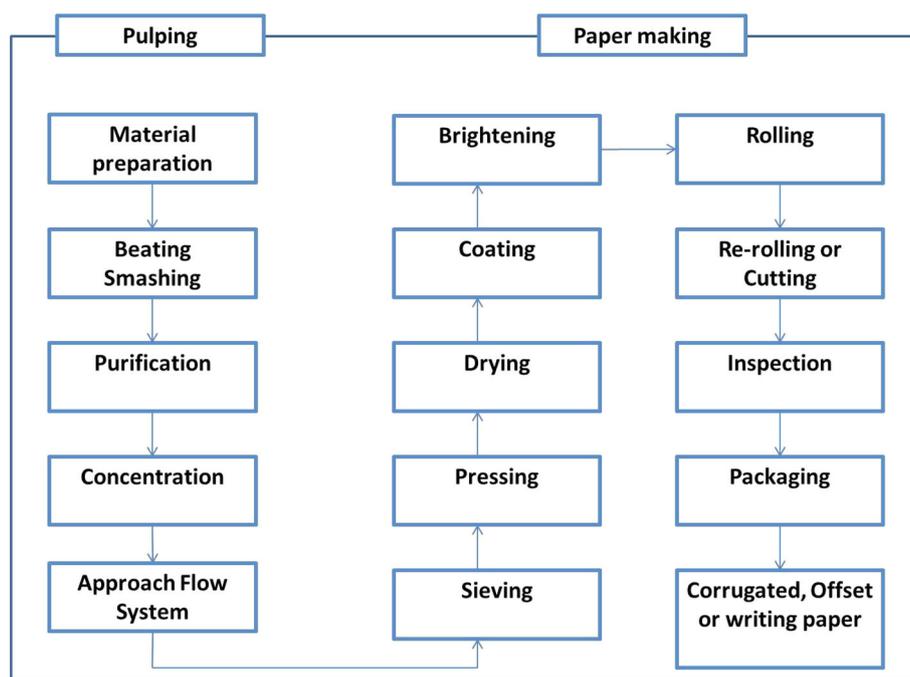
The paper manufacturing industry converts different materials into variant board products and paper via mechanical and chemical process known as pulping [3]. With advances in the paper industry, more attention has been paid on producing high-quality paper in a cost effective manner [6]. The final properties of the paper, such as its quality, weight, surface gloss, smoothness, and reduced ink absorber, are of paramount importance [7–9]. The incorporation of nanostructured and low levels of organic into the paper formulation is expected to enhance the properties of the produced paper.

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**Fig. 1** The general flow system of papermaking



### Current nanotechnology

Nanotechnology is of great importance in almost all modern-day industries, targeting high-quality and efficient market potential [10–12]. The large interest in the nano-scale range is due to the fact that nano-additives possess enhanced properties compared to its bulk counterparts [11]. Current nanotechnology is leading the development and global economic growth. Nano-scale techniques are also prominent in multi-disciplinary fields, providing glimpses towards exciting new capabilities for materials, devices, and systems [12, 13]. Nano-additives with unique advantages are expected to remodel the facets of controlled technology. The main purpose of nanotechnology, in this context, is to control and manipulate materials to obtain special functionalities. Nanotechnology-based developments provide incremental and evolutionary changes [14], and are a new tool that is recently added to a wide range of applications, with pulp and papers among them.

### Nanotechnology: paper industries

The unique advantages and properties of the paper can be improved via nano-additives. The advances of nanotechnology resulted in the development of multiple nano-additives, which is the byproduct of extensive fundamental studies associated with advances in nanotechnology. The size of nano-additives is significant, due to a variety of factors [13–15]. Light scattering attribute are affected by nano-additives, gloss and opacity, the calendaring steps,

the rate of drying, and paper penetrance. The nanosize particles have a propensity to aggregate [16]. In reality, due to the insufficient specific surface, it is difficult to obtain larger diameter nano-additives [17, 18]. For example, due to the bigger surface area that competes for sizing, furnishes containing tiny particles nano-additive are more resistant towards the internal sizing agent.

### Nano bleaching agents: paper industries

The pulp and paper industry decolorize or whiten papers with a bleaching agent. Chromophores generally contain groups of colored atoms [19–21], which are capable of absorbing visible light, characteristic wavelengths, and reflect or transmit part of the light [20]. These absorbing groups of chromophores are destroyed by oxidation/reduction of the bleaching agents [22–24].

### Nano calcium silicate particle

Nano calcium silicate are examined as fillers in 45–55 g m<sup>-2</sup> newsprint, where its opacity increased [25–27] by about 3.7 points for the 45 g m<sup>-2</sup> newsprint at 2 wt% loading, while for the 55 g m<sup>-2</sup> newsprint, it was about 1.3 points [26]. This basically proves that nano calcium silicate is capable of increasing the newsprint's sheet's opacity [27]. The excellent light scattering surface afforded by the nano-structured framework is desirable, resulting in enhanced opacity [28–30]. Both these factors will result in a significant improvement to print quality.

The nano phase calcium silicate platelets are stacked to form an open network structure, which enables it to quickly absorb printing ink, preventing the forwarding image from being visible on the reverse side of the paper [31]. Overall, nano calcium silicate results in higher quality printed image, localization of ink dots, and prevent undesirable spreading across the paper [29, 32].

### Nanosizing agents: paper industries

Protecting the filler or glaze by applying sizing or size is a practical technique of incorporation in pulp and paper industry [33–35]. The paper and textile manufacturing industry utilized sizing to alter the absorption and wear characteristics of the resulting material; it is the phase used for oil-based surface provision for gilding (in this context), recognized as a mordant [36–38], and is used for some art techniques by painters and artists to prepare papers and textile surfaces [39, 40].

### Nano silica/hybrid

Properly selecting nano sizing is crucial towards the transmission of functional confidants, including anti-fungal and UV protection. This is achieved mostly through the presence of nano silica via dewatering, which is the act of lowering the water transmission rate, while retaining water retention and strength [41]. Nano silica usually provides paper with the high abatement and anti-bacterial activity, which usually results in high degradation [42]. The introduction of nano silica usually results in the reduction of print-through and the improvement of optical properties [43]. The addition of 1.8 wt% of nano silica reduced print-through by about 30 % and increased the opacity and brightness by about 1.7 points each, collectively representing a significant improvement [44]. Furthermore, nano silica reduced print-through by 40 % at a 2.2 wt% addition for yellow directory papers, which is rather significant. Some studies compared coated and uncoated nano silica, and confirmed that coated nano silica results in good print quality, optical density, color gamut, water permanence, color performance, and dimensional stability as opposed to uncoated ones [45–47] (Fig. 2). Nano silica can be prepared by the sol–gel technique, which involves simultaneous hydrolysis and condensation reactions. A sol of sodium silicate, silicon alkoxide, or halide gets converted to a polymeric network of gel. Partial hydrolysis of orthosilicate takes place when it is mixed with water and ethanol.

Nanostructured silica, with an open network structure and regulated by regulated precipitation from geothermal water, has been synthesized and used bleaching agent. It

resulted in a print through reduction of 30 % for newsprint and 40 % for full-scale paper mill manufacturing [48].

In advanced, nanostructured silica with an open network structure by regulated precipitation from geothermal water has synthesized and used bleaching agent. It found that print through reduction of 30 % for newsprint and 40 % for attained for full-scale paper mill manufacturing [48].

### Nano titanium oxide/hybrid

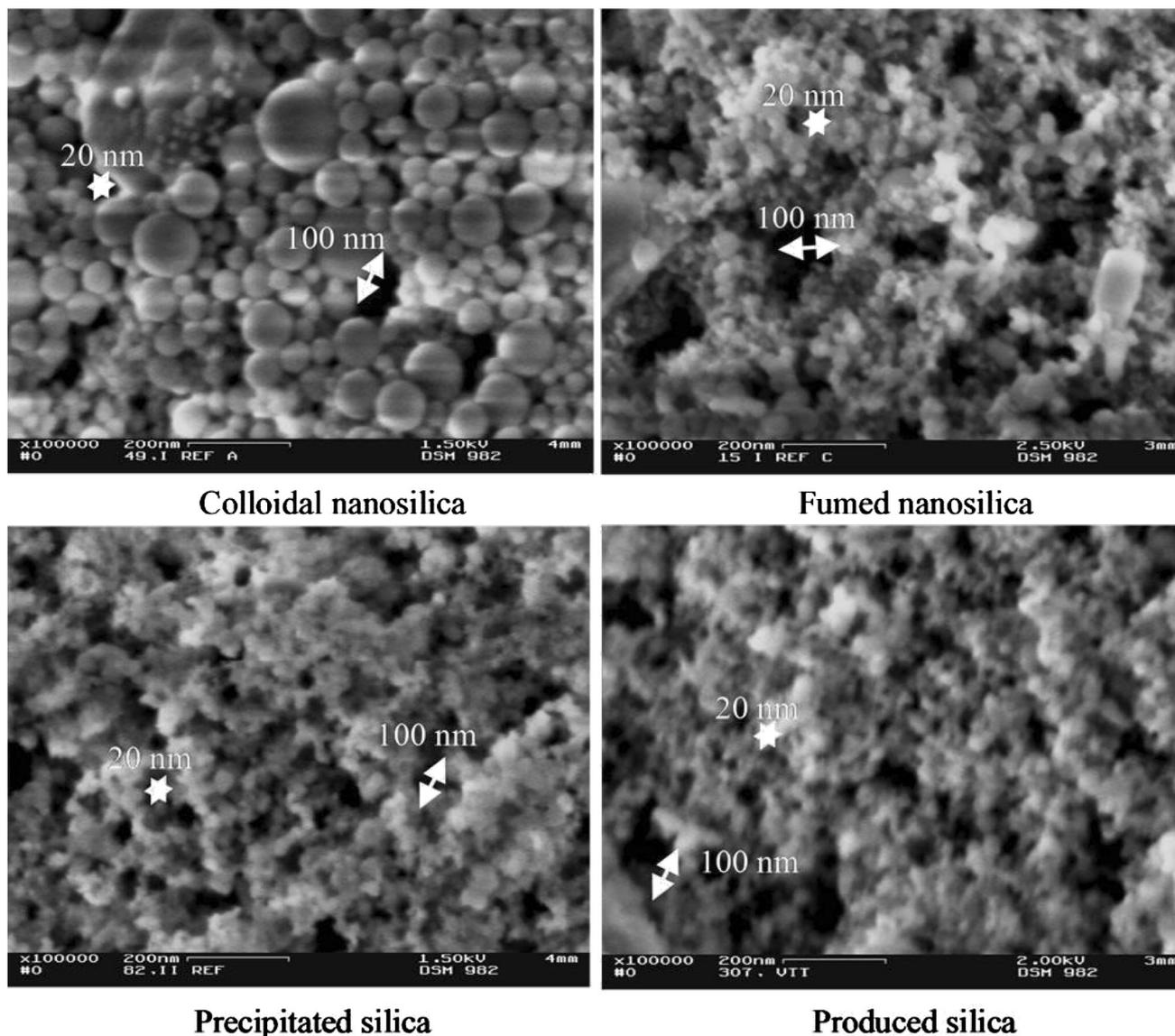
TiO<sub>2</sub> imparts deodorizing and antimicrobial to the paper, on top of improving its opacity, refractive index, and whiteness [49]. However, some disadvantages, such as cost and dependence on UV, restrict its usage to printing papers on a commercial scale [49–51]. As an alternative, TiO<sub>2</sub> hybridized with other metal oxides, such as ZnO, demonstrated excellent chemical and optical properties, which makes it viable for a broad range of antimicrobial and UV applications of paper [52]. Coating the surface of the paper with TiO<sub>2</sub>/ZnO gave way to superb anti-fungal and UV properties, which are essential towards the enhancement of its life [53].

### Nano wet/dry strength agents: paper industries

Tensile properties of the paper both in wet and dry phases can be improved by crosslinking the cellulose fibres based on its covalent bonds during wet/dry strength chemicals, which do not break upon wetting [54–57]. The variety of the wet strength chemicals are involved with process improvements, such as wet strength resins or neutral sizing agents [58–60].

### Nanocellulose

Paper products made of nanocellulose have been widely used as packaging materials, owing to their promising attributes such as biodegradability and sustainability with low environmental impacts. Generally, cellulose is a semicrystalline linear polysaccharide compound of  $\beta$ -1,4 linked glucopyranose units, with its polymer chains combined with hydrogen bonds forming bundles of fibrils consisting of ordered excellent crystallinity and disordered amorphous domains [61, 62]. The crystalline domains can be isolated in the nano-scale, with highly ordered and regular rod-like nanocrystals, after the removal of the amorphous domains via acid hydrolysis, also known as cellulose nanofibers, crystalline nanocellulose, or nanowhiskers [62–69] (Table 1). Furthermore, nanocellulose can be obtained from cheap and abundant renewable natural resources such as wood, plants, vegetables, and other agricultural residues and processing waste [65].



**Fig. 2** SEM image of coatings containing silica pigment on the fine paper surface. Magnification of 100,000 [45]

**Table 1** Nanocellulose derivatives and their dimensions

Nanocellulose derivatives	Diameter (nm)	Length (nm)	Aspect ratio (L/d)	References
Microcrystalline cellulose	>1000	>1000	1	[62]
Micro fibrillated cellulose	10–40	>1000	100–150	[63]
Microfibril	2–10	>1000	>1000	[64]
Cellulose whisker	2–20	100–600	10–100	[65]
TEMPO-oxidized nanocellulose	3–4	>1000	1000	[66, 67]
Carboxymethyl nanocellulose	5–15	>1000	200–100	[68, 69]

However, the hydrophilic properties of nanocellulose created a low H<sub>2</sub>O vapor barrier of paper-based packaging material [66]. Consequently, it limits the requirement for high barrier or extended applications in packaging [67]. This encourages the advent of research works on chemical

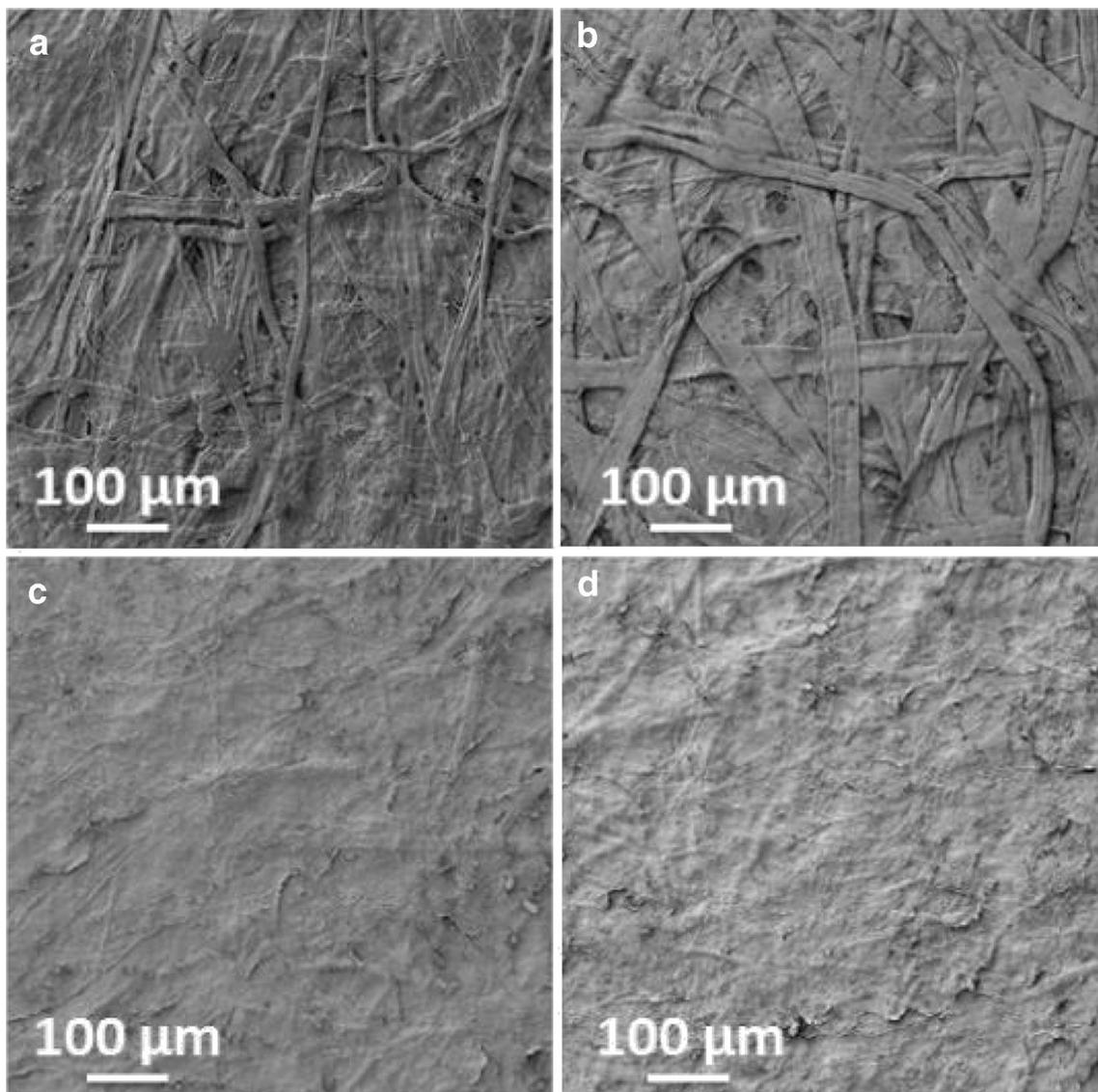
modification of nanocellulose, intended to improve its hydrophobic properties. Over the past few years, some works have successfully realized an eye-catching cellulosic nano paper, with remarkably high toughness using wood nanofibril [68]. Increasing tensile index yields a

significantly stronger paper, mostly via the addition of nanocellulose. Sheet drainage is also improved, although it comes out as slightly impaired. To overcome this limitation, cationic polymers, such as polyacrylamide, have been used as a fixative for the retention of nanocellulose, as well as for better suspension drainability [69, 70]. Compared with micro-sized cellulose, nanocellulose are more effective as an additive for the paper industry, due to the interactions between the nanosized elements, which is connected by hydrogen bonds, form a percolated network when the nanocellulose is dispersed in the pulp's slurry [66–85].

Therefore, it was determined that the nanocellulose paper was a network composed of inter-wined nanofibrils

(Fig. 3), with an aspect ratio exceeding 100, and with a tensile index of 214 MPa, which is quite close to that of industrial steel (250 MPa) [69]. The orientation of nanofibrils cellulose is assumed to be incorporated into the inter-fibrillar region and form lateral interlinks between fibrils via  $H_2$  bonding.

However, there are still several challenges associated with this approach, such as the efficiency of grafting and tailored barrier properties of the paper being produced. Therefore, increasing the hydrophobic properties of paper products and improving vapor barrier properties by lowering its water vapor transmission rate is fast gaining some guarded interest [70]. Many methods have been applied for nanocellulose surface modification, including acetylation,



**Fig. 3** SEM micrographs of the four paper samples: **a** standard tracing paper. **b** laboratory paper sample. **c** microcrystalline cellulose paper and **d** Nanocellulose paper [85]

silylation, grafting, and the use of coupling agent [71, 72]. For example, some research works focus on the development of carboxylated nanocellulose, based on TEMPO (2,2,6,6-tetramethylpiperidine 1-oxyl) mediated oxidation [73–75]. Through this modification, a nanocellulose, with a promising thermal and mechanical properties, is obtained [74, 76]. The produced paper possessed extremely high tensile strength and tearing resistance in the machine direction, as well as air permeability.

### **Nano pigment: paper industries**

Pigment is an abundant component in the paper industry, and naturally, the most important factor affecting the final properties of the paper [77, 78]. Paper pigment agents generally consist of inorganic compounds, such as kaolin, calcium carbonate, binder, soluble co-binder, dispersants, water as carrier, and other additives [79, 80]. The use of nano pigments with an appropriate binder in the coating formulation will help fill the micro-voids and offer better barrier properties [81, 82].

### **Nano kaoline**

Nano kaoline is one of the most widely used pigments in the paper industry, and it is used in a large range of coating applications. Nano kaoline or kaolinite, with the chemical formula of  $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ , is the principle nano mineral in China clay [82]. Structurally, it is a dioctahedral 1-to-1 layered  $\text{Al}_2\text{O}_3$  to  $\text{Si}_2\text{O}_5$ , and consists of two basic units, including an octahedral sheet comprising of closely packed  $\text{O}_2$  and OH where Al, Fe, and Mg atoms are arranged in the octahedral coordination [83]. The second structural unit is the  $\text{SiO}_2$  tetrahedral layer, where the silicon atom in the center is equidistant from four  $\text{O}_2$ , or possible OH. The inherent impurities in nano kaolinite include titaniferous minerals, iron oxides, and mica, each giving a different color, from yellow to dark brown on produced paper [84–86]. It has processed for paper industry applications that demand very high brightness, low yellowness, and other physical and optical properties [85].

Various modification methods applied to cover the disadvantages of natural nano kaoline included centrifuge or sedimentation process, acid activation, chemical bleaching, thermal treatment or calcination [83, 85–87]. Surface modification of nano kaoline about the intercalation process involved the insertion of low molecular weight organic reagents included guest molecules (urea, formamide, potassium acetate and dimethylsulfoxide) between layers consisting of two-dimensional arrangements of tetrahedral and octahedral sheets [88]. Indeed, intercalation not only enhances the possibilities of kaoline exfoliation as nanostructured mineral, but also modifies the nano kaoline

surface via complexation of  $\text{H}_2$  bonding [89]. The formation of intercalates happens when the organic material located and stacked between kaolinite layers. Chemical bleaching is a standard beneficial technique used to enhance the brightness of nano kaoline for high value applications [86]. The process discolors the nano kaoline and allows the small proportion of total iron removal in solution. In some of the studies, nano kaolin modified with polymer material resulting black liquor after alkaline pulping. The modification carried out in order to change its nature [90, 91]. The results suggest enhance of the mechanical attributes and the water absorption of that manufactured nano kaoline through polymer [92]. Thus, the prepared paper can use for the specific use that needs greater absorption and higher dewatering time [93].

### **Nano ZnO/hybrid**

Nano ZnO is a material with potential pigment suitable for paper coating to expose brightness and greater printing properties. Nano ZnO provides paper coating pigment a high covering power apart from anti-fungal and UV protecting properties due to its extremely small size [94]. In some work, ZnO/starch composites were prepared by a simple and novel wet chemical method using zinc nitrate and NaOH as precursors and soluble starch as stabilizing agents [95].

### **Nano carbonate**

The use of nano calcium carbonate is important when opacity is needed at a low-basis weight; they are invaluable in packaging grades where low permeability is combined with opacity to protect food from light [96–99]. The presence of nano calcium carbonate, however, affects fiber to fiber contact and reduces paper strength [100–103]. Other properties are improved rendering the paper useful for special purpose [104–106]. For example, the fine proportions of nano calcium carbonate and nano kaolin used for the coating of paper surface [107].

### **Nano retention agents: paper industries**

Improvements in the retention of a functional chemical in a substrate are achievable by the retention agent which is a chemical process [108]. Eventuate is that totally less chemicals are used to get the same effect of the functional chemical and less chemicals goes to waste [109, 110].

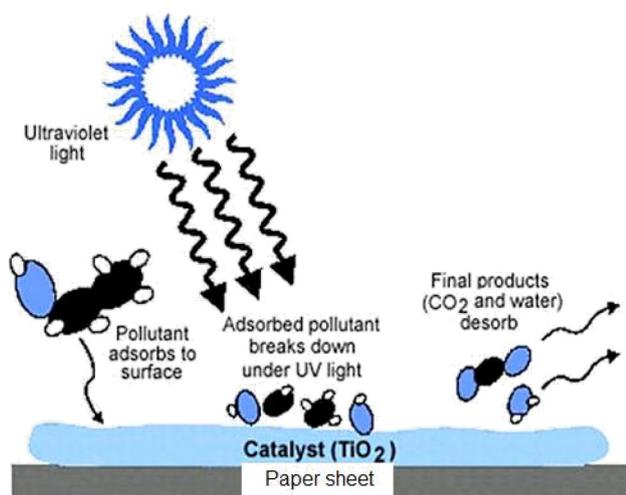
### **Nano zeolite**

Nano zeolite filler materials used for improving properties of asphalt and paper functioning when mixing with pulp

[111]. Meanwhile, the addition of nano zeolite also assists in removing gas emissions within the scope of high quality paper. This is due to their crystal structure that composed of millions of channels and cavities with a very wide surface area [112, 113]. Furthermore, natural nano zeolite has a significant tendency to attract moisture and can return the water absorbed without any change in the crystal structure and moisture absorption properties [114]. Therefore, activated nano zeolites are extensively used as desiccants in the paper industries due to the said characteristics. Compared to synthetic zeolites, however, natural zeolite provides significant advantages of cost effectiveness, abundant resources and easier accessibility towards paper industries [111–114].

### Nano TiO<sub>2</sub>

Nano TiO<sub>2</sub> is possible used as retention agent for supporting materials on paper sheet. There are two possible mechanisms on nano TiO<sub>2</sub> as a retention agent, where a nano TiO<sub>2</sub> suspension is coated on the sheet surface and wet end addition, where nano TiO<sub>2</sub> is deposited onto individual fibre before sheet formation, leading to a bulk distribution of the TiO<sub>2</sub> loading [115–120] (Fig. 4). For example, the surface properties of nano TiO<sub>2</sub> with hexadecanoic acid demonstrated that the wetting and dispersion of the mixture improved greatly. The paper with nano TiO<sub>2</sub> had higher dynamic elastic modulus than the unfilled ones [116]. This encourages the development a modern method of advance paper with super hydrophobic surface prepared with the addition of modified nano TiO<sub>2</sub> to cellulosic fibers [115, 118]. Superb speed homogenizer was used to disperse Nano TiO<sub>2</sub>, and the surface modification was done using coupling agent, 3-(trimethoxysilyl) propyl



**Fig. 4** TiO<sub>2</sub> paper sheet in photocatalysis system [120]

methacrylate [116]. The results show that the increasing hydrophobicity of water contact angles for modified paper from 126.5° to 154.2°.

A binder can be used in the latter approach not only to increase the TiO<sub>2</sub> loading and dispersion in the paper, but also to protect the fiber against radiation. Thus, the produced paper displays high stability when exposed to solar like light [119]. For example, some studies carried out on sludge recovered from paper ash containing nano TiO<sub>2</sub> that obtained via acid treatment with that the crystal structure of TiO<sub>2</sub> of the anatase and crystal size was 10–20 nm [120]. The stability and degradability activity of nano TiO<sub>2</sub> in the paper sludge then evaluated by gases acetaldehyde removal. It found that the obtained paper capable of completely decomposing acetaldehyde to CO<sub>2</sub> under solar and UV light [121].

### Nano mineral filler: paper industries

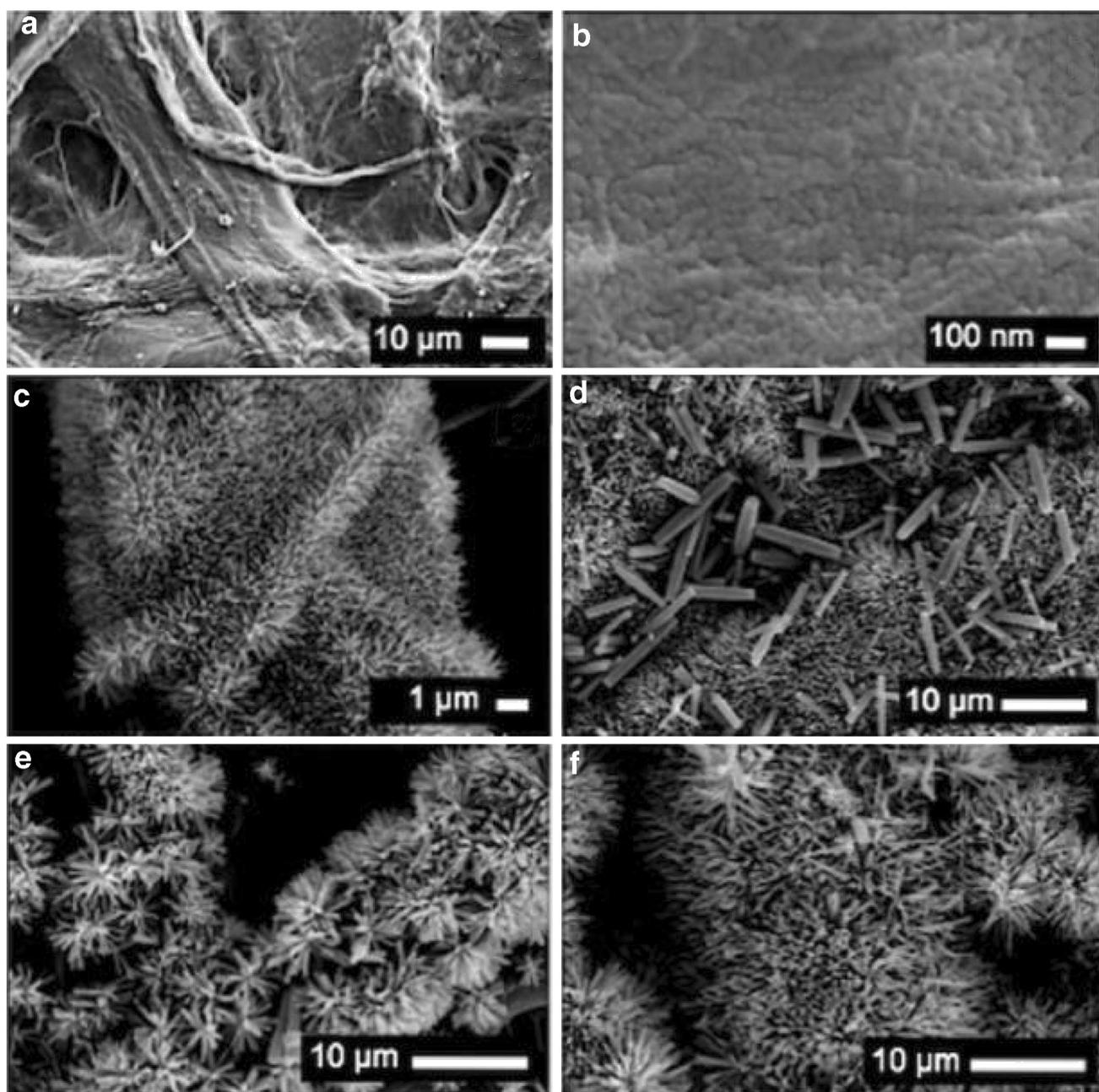
Due to the hydrophilic, fibrous, organic and highly porous nature, paper simply subjected to ultraviolet degradation, microbial attack and high water vapor transmission rate [122, 123]. In addition, the sizing agents included starch that applied to the surface of the paper make the condition still worse [124–126]. To overcome these disadvantages, novel nano mineral fillers being attracted as paper coating to meet the improved quality standards of excellence-grade papers [127].

### Nano clay

Low gas permeability is one of the most interesting property enticements from nano clay as an additive in paper industry, particularly for the nylon based paper [128]. It claimed to supply a 100-fold lessening of O<sub>2</sub> permeability and provide a CO<sub>2</sub> barrier giving improved the total shelf life of the paper [129]. Conventionally, nano clay used as barrier layers in multilayer polyethylene terephthalate bottles, meat and cheese containers. Nano clay also used as replacement for ethylene glycol copolymer in food packaging and high density polyethylene containers [128, 130]. Therefore, once nano clay applied in paper industry, it can be act as waterproof barrier [131]. It offered a more suitable alternative to conventional polyethylene and acrylic acid that are commonly used and has a high potential application in paper beverage technology [132].

### Nano calcium carbonate

Some studies examined the light scattering of precipitated calcium carbonate filler coated with novel silicate and zinc sulfide nanoparticle [27]. The precipitated calcium carbonate showed an enhancement in the light scattering from a pigment coating of paper using nano-additives [28, 29].



**Fig. 5** SEM micrographs showing **a** untreated handsheet paper; **b** after seeding with ZnO nanoparticles; **c** ZnO nanorods grown on paper at a concentration of 10 mM for 10 h; **d** ZnO nanorods grown

on paper at a concentration of 10 mM for 20 h; **e** ZnO nanorods grown on paper at a concentration of 20 mM for 10 h; and **f** ZnO nanorods grown on paper at a concentration of 20 mM for 20 h [137]

#### Nano TiO<sub>2</sub>/hybrid

Some studies reported on the photocatalytic degradation of xylene with nano TiO<sub>2</sub> hybrid with beta-cyclodextrin coated paper [116]. Paper coated with a combination of nano TiO<sub>2</sub> and beta-cyclodextrin had a better effect on the degradation than paper coated only with nano-TiO<sub>2</sub>, which the hybrid system has a significant synergistic effect on the degradation of xylene [120].

#### Nano superconductor agents: paper industries

Superconductor agent is an element or compound that will conduct electricity without resistance below a certain temperature towards produced paper [133–135].

#### Nano ZnO

ZnO nano additives can also use on a paper surface without the use of binders. ZnO nano additives can coat onto paper,

which achieved first by ammonia and heat treatment of zinc oxide and cellulose, followed by ultrasound treatment [53, 136]. Paper products coated with zinc oxide nano additives exhibit antibacterial activity against *E. coli*. This is showing that, with the addition of nano ZnO, an excellent antifungal and UV protecting properties of paper consequently produced (Fig. 5). Meanwhile, demonstrated that the brightness, whiteness, smoothness, print density, print uniformity, picking velocity and oil absorbency of the paper significantly improve with addition on certain composition of nano ZnO [137].

### Nano dispersion agents: paper industries

Dispersing agent or dispersant or plasticizer or a super-plasticizer is either a non-surface active polymer or a surface-active substance added to a pulp suspension, to improve the separation of cellulose particles and to prevent settling or clumping [138–140].

### Nano polymer

Many of nano polymers, especially those with high charge densities can be used as additives

for papermaking and they should be used in dilute solutions, to facilitate their dispersion throughout the stock, which in consequence avoid high turbulence [141]. In general, nanopolymer is a polymer-based material with a complex architecture, high density and durability. Nanopolymer reinforced some components of plasma conduits in emergency repairs or hazardous conditions. A wide variety of water-soluble nano polymers used for papermaking and categorized as naturally occurring polymers included nano polysaccharides/protein, modified naturally occurring polymer and synthetic polymers [142].

Nano polyacrylic acid or nano polyphosphates often used, with anchoring cations, to reduce the zeta potential of some fillers [143]. This helps filler dispersion, but may interfere with their aggregation during drainage [144, 145]. Neutral synthetic nano polymer retention aids included nano polyacrylamide, nano polyvinyl alcohol and nano polyethylene oxide [146]. They are generally inexpensive nano polymer, easily formulated for high molecular weight about several millions g/mol and added to the dry strength [143]. In other studies, polymeric nano xylan may use as the strength-increasing paper additives. In contrast to dissolving pulp production, a high polymeric nano xylan concentration is desirable for paper grade pulp to increase the pulp yield and strength properties [141]. The subsequent addition of isolated polymeric nano xylan during the paper making process may improve mechanical properties. Because the pulping and bleaching processes introduce COOH groups, the cellulosic fibers carry a slightly

negative charge [142]. These negative charges cause electrostatic repulsion between single anionic fibers and reduce the possible inter fiber linkages [146]. The absorption of polymeric nano xylan as cationic additives into the anionic pulp surface decreases the electrostatic repulsion effect and therefore enhances the inter-fiber linkage and mechanical properties of the produced paper [147]. Cationic side groups lead to higher additive retention; however, a number of cationic groups are not directly proportional to the mechanical properties of the paper [148]. For example, a slight cationization of polymeric nano xylan to the degree of substitution of 0.1 causes increase in tensile and tear strength of paper up to 60 %. Polymeric nano xylan exhibited as a good O<sub>2</sub> barrier, suggesting possible applications in food packaging [149]. Polymeric nano xylan may easily extract from agricultural residues than wood because woods contain more lignin and the xylan integrated strongly into the lignified tissue [150].

### Certain challenges associated with nano additives

Due to their lower particle size nano-additives need special attention so that they could be retained in the fiber matrix during the papermaking process [60, 107]. Papermakers can select an appropriate nano-additives and the process for the development or enhancement of desired paper properties or making a new product.

### Conclusion and future prospective

Nano-additives can add new functionalities to paper industries. The main parameters of nano-additives are particle size and size distribution, aspect ratio, stabilization and surface modifications. Being lower in particle size, nano-additives could provide better performance than traditional additives such as adsorption, which can benefit from a high surface area. With respect to this, they may confer certain unique attributes and functionalities of paper, which is also one of the major concerns of papermakers. Incorporation of nano-additives into paper slurry could also have such attributes as high smoothness, good appearance or other previously unimagined functions. The advantages combined with the use of nano-additives expected to persuade worldwide papermakers to attract to this potentially promising research area and to make every effort to create positive breakthroughs.

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### Compliance with ethical standards

**Conflict of interest** The authors declare that there is no conflict of interests regarding the publication of this paper.

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