Paper Fillers Innovations: From Design of Particles to Preparing Filler Composites

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The increase of filler content in paper is an effective way to reduce production costs and to promote the market competitiveness of paper mills. A shift from natural fillers to synthetic fillers has enabled improvements in the critical properties of paper. Meanwhile, innovations from single particles of filler to filler composites has made it possible to increase the filler content of paper. Among various filler innovations, the design of fiber/filler composites has aroused general attention from industry and academic researchers. However, concerns related to the cost and recyclability of composite fillers remain to be addressed.

Keywords: Mineral fillers; Paper strength; Nanocellulose; Filler Morphology; Filler composites

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Expectations of the Paper Industry Regarding Mineral Fillers

The use of mineral fillers in the paper industry has been practiced for many years. It is well recognized that paper brightness, opacity, smoothness, and ink receptivity are typically enhanced when mineral particles are incorporated into paper. More importantly, substitution of cellulosic fiber with cheaper mineral filler contributes to energy saving and cost reduction. The latter seems even more rewarding for a graphical paper mill, especially when the price of virgin pulp is increased. Therefore, it has been a recurring dream of papermakers to further increase of filler content in paper while meeting the requirements of runnability of paper machine and maintaining the quality of paper products.

Strength loss of paper resulting from the use of filler is the main barrier to increasing filler content in paper, since filler particles prevent the hydrogen bonding between cellulosic fiber. Thanks to the applications of strengthen additives and filler innovations, the filler content of printing and writing paper, as well as copy paper, has been doubled in the last few decades, *i.e.* from $10\% \sim 15\%$ to $25\% \sim 30\%$. Therefore, filler innovations are still hot topics from both the industrial and academic points of view.

Filler Innovations are Satisfying the Needs of the Industry

The inherent characteristics of natural filler, such as ground calcium carbonate, talc, and clay, endow paper products with high brightness, smoothness, and suitable paper strength. However, the relatively low specific surface area and high packing density that result from natural morphologies limits the improvement of other critical properties, such as paper bulk. The use of synthetic filler, such as precipitated calcium carbonate and calcium silicate, have enabled improved paper bulk, brightness, and printability (Hubbe and Gill 2016). It should be noted that the increased bulk of the paper before calendering

can allow the papermakers to achieve higher smoothness at a specified caliper. As the demand of increasing filler content, filler innovations have focused on enhancing the bonding between filler and cellulosic fiber, which results in an important opportunity, *i.e.* the innovation from single particle filler products to composites structure design of fillers. The composite innovations can be classified as follows:

- **Surface modification of filler**. Filler particles modified with biopolymers, such as starch, chitosan, can be prepared in the form of composites, which can potentially enhance the bonding between fiber and filler.
- **Filler preflocculation**. Filler particles can be preflocculated by polymers prior to blending with furnish. The preflocculated particles can be considered as composites. The formation of composites decreases the contact area between fiber and filler, which helps to increase fiber/fiber bonding.
- **Filler/fiber composites**. Filler/fiber composites have aroused general interest in recent years. These composites are formed by chemical or physical methods, such as *in situ* growth, co-flocculation, or co-grinding between filler and cellulosic material, such as fines, micro-fibrillated fiber, or cellulose nanofiber. On the one hand, the fiber component in the composite helps to enhance the bonding between fiber and filler. On the other hand, the filler particles aggregate during formation of composites, which also helps to alleviate the negative effect of filler on fiber-bonding.

Future of Innovations in Mineral Fillers: Opportunities and Challenges

Innovations in filler technology continue to make progress. The topic is addressed by the Agenda 2020 program (created by American Forest & Paper Association) and 2050 roadmap (developed by the Confederation of European Paper Industries). These initiatives recognize that the increase of filler content in paper is a key advancement in technology. In the future, developments involving chemical synthesis and nanomaterials may lead to composite fillers having stronger bondability with fiber. Meanwhile, the designability of the structure and function of filler composites is also a possible direction. However, there still exist many challenges. For example, although a high filler content of paper can be achieved by the use of composite filler, the cost can remain as an impediment to commercialization. In addition, aspects of the recycling of paper with composite filler may also need to be considered.

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