Nanoparticle-based Antimicrobial Paper as Spread-breaker for Coronavirus

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Paper, Packaging and Tissue industries are already indoctrinated with the antibacterial and antimicrobial concepts, producing special packaging materials to preserve food and fruits and antibacterial tissue papers. These are mostly based on antimicrobial organic compounds and inorganic materials. In the meantime, lots of advancements have taken place in nanotechnology with production of a number of nano materials which have antibacterial properties. In fact, the antimicrobial efficiency of some of the nanoparticles is many times more than that of traditional antimicrobial additives.

For combatting the sub-microscopic bacteria and viruses, antimicrobial paper incorporating nanoparticles, can be very effective. It is particularly appropriate at the present juncture to use such papers and products to arrest spread and kill the deadly coronavirus. The key technologies available now in use for production of antimicrobial paper and products, are reviewed, along with test results in reducing the percentage of virus on the antimicrobial paper surface and thus spread of the virus.

Finally, the impact of the coronavirus epidemic on the paper industry has been forecast, and techniques for formulating the nanoparticle-based antimicrobial composite and its addition during paper manufacturing are discussed.

Introduction

We have witnessed the horror of the coronavirus epidemic since the beginning of this year, causing deaths of almost half a million and infection of several millions of people throughout the world. Governments, hospitals, testing laboratories and security staff members, doctors and scientists are all playing their roles to arrest infections due to this epidemic. Some industries are also working hard to meet the rapid production and supply of testing and personal protective equipment to combat the spread of the virus. In this challenging time, we are all looking to do everything we can to reduce the spread of infection, to protect ourselves and others, and this includes broadening the availability of antimicrobial papers to all sectors.

Writing and printing, packaging, tissue and even newsprint sectors of the paper industry, have roles to play at the present juncture as all these papers are used by society in everyday life: in schools, colleges, offices, hospitals, transport, travel, restaurants, grocery and other goods packaging, face and nose wiping tissue papers and newspapers. The coronavirus may contaminate these papers and spread infection to others. Therefore, all eyes are now on the paper industry to supply paper, packaging and tissue papers, having antimicrobial properties so that bacteria and viruses, when they come into contact with the treated paper, are neutralised and unable to spread.

Paper is such a common commodity and used in such large quantity that developing manufacturing facilities for thousands of tonnes of antibacterial paper per day, can be a really Herculean task. However, in view of this deadly disease, the paper industry will have to pull on the socks to prepare itself in a more challenging way. On the other hand, the industries, who will be able to show positive results on antimicrobial properties, will benefit more than ever before and will create history.

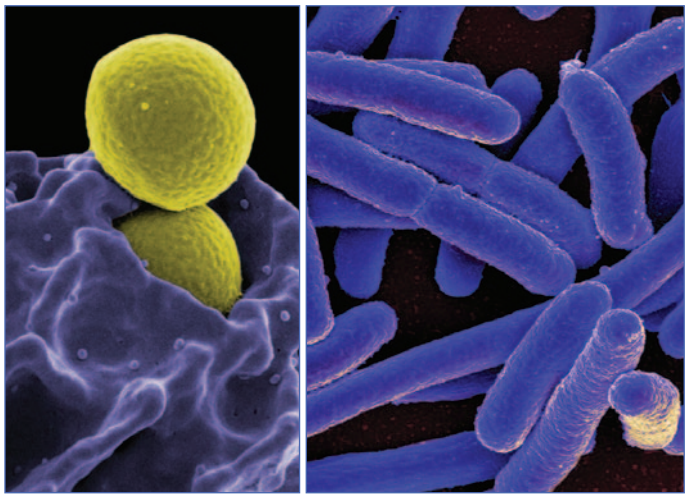

Figure 1. Photomicrographs of bacteria: (a) spherical *Staphylococcus aureus* (MRSA), (b) rod-like *Escherichia coli* (both courtesy of NIAID).

Definitions and Concepts

The term bacteria was devised in the 19th century by the German botanist Ferdinand Cohn, and it is based on the Greek word *bakterion* meaning a small rod. Bacteria are single-celled microorganisms that multiply by simple division, having round, rod-like, spiral or lamentous single-celled (*Figure 1*) or non-cellular bodies often gathered into colonies (5).

To survive and reproduce, bacteria need energy and this comes in many forms: the most common involves oxidation of organic compounds, but others include photosynthesis, oxidation of inorganic compounds (such as ammonia, nitrite and some metal cations) which is a mode unique to bacteria, and anaerobic respiration, where electron-acceptors include such anions as nitrate and sulphate. As the chemical reactions necessary for production of energy are performed, the bacteria expel waste products, which, for example, in the case of glucose oxidation results in production of ethanol, or for autotrophic bacterial reduction of nitrate results in liberation of nitrogen gas.

The vast majority of bacteria are considered either harmless or positively beneficial to humans, but some are pathogenic and a few cause infectious diseases in humans such as meningitis, typhus and bacterial pneumonia. In other cases it is the chemical expelled that explains why bacteria can be problematic; for example, *Clostridium botulinum* produces botulinum toxin, the most potent toxin known to mankind, while *Streptococcus mutans*, a component of dental plaque, metabolises sugars and produces acid which attacks tooth enamel.

Worthy of special attention are Methicillin-resistant *Staphylococcus aureus* (MRSA) bacteria (or super bugs) which are of major concern (6). The excessive use of antibacterial household cleaners and antibiotics is said to help super bugs survive, because if bacteria are exposed to repeated doses of these products they will mutate genetically, in the course

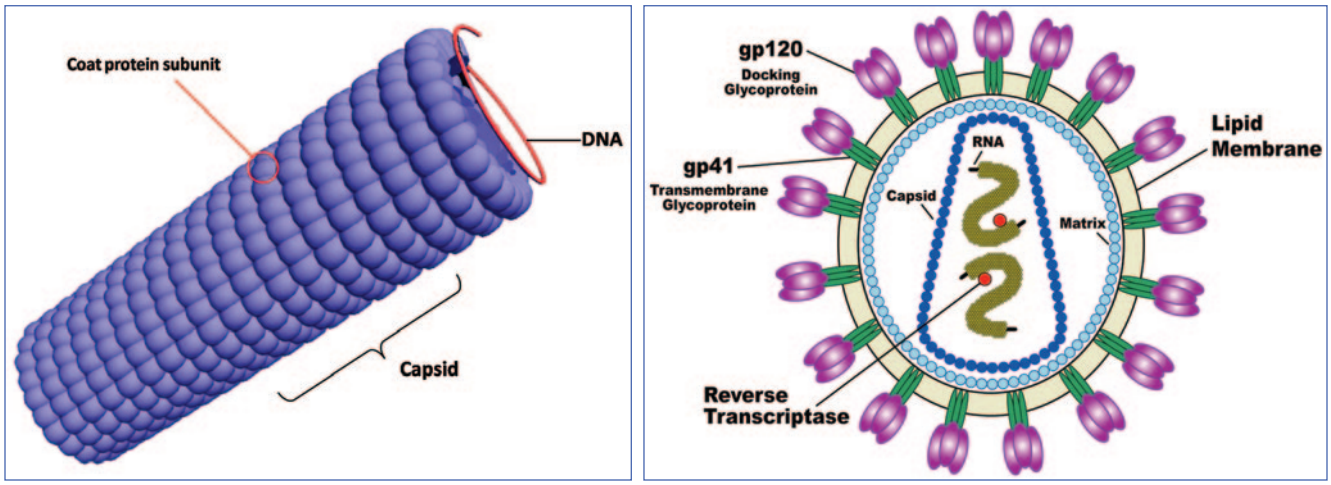

Figure 2. (a) Graphic of a helical virus protected with a capsid or protein shell (courtesy Arionfx); (b) Graphic of a virus protected by a lipid membrane (courtesy NIAID).

of which they acquire resistance.

Turning now to viruses, these are not living entities but are infectious agents that have developed the ability to enter a host and replicate within its cells. A virus consists of genetic material (DNA or RNA strands) packed within a protein shell (or capsid) (*Figure 2a*); in some cases, an external layer of lipids may also be present (*Figure 2b*). They are sub-micron in size, so smaller than bacteria, and in general there are three main morphological types: helical, icosahedral, and complex.

Antimicrobial agents are chemicals used to kill or inhibit the growth of micro-organisms whether they are bacteria, viruses or fungi (7). They can be bactericidal, killing the organism concerned, or bacteriostatic, inhibiting the growth of the organisms concerned thereby giving the immune system of the host time to act.

SARS-CoV1, SARS-CoV-2 and Surfaces

Before delving into the depth of antibacterial paper and products, where they serve as spread-arresters, it is worthwhile understanding the difference between SARS-CoV1 (the virus responsible for the SARS outbreak in 2002/3) and SARS-CoV2 (also known as 2019-nCoV), which is the virus responsible for the current COVID-19 outbreak (see *Figure 3*).

Scientists at NIH (2) compared how the environment affects SARS-CoV-2 and SARS-CoV-1. SARS-CoV-1, like its successor, emerged from China, and infected more than 8,000 people in 2002/3; it was eradicated by intensive contact tracing and case isolation measures and no cases have been detected since 2004. SARS-CoV-1 is the human coronavirus most closely related to SARS-CoV-2. In the stability study, the two viruses behaved similarly, which unfortunately fails to explain why COVID-19 has become a much larger outbreak.

The NIH study (2) attempted to mimic virus being deposited from an infected person onto everyday surfaces in a household or hospital setting, such as through coughing or touching objects. The scientists then investigated how long the virus remained infectious on these surfaces (1-3) and found that viable virus was still detected:

- in aerosols for up to 3 hours,
- up to 4 hours on copper,
- up to 24 hours on cardboard, and
- up to two to three days on plastic and stainless steel.

These times will vary under real-world conditions, depending on factors including temperature, humidity, ventilation, and the amount of virus deposited. Nevertheless, the results provide key

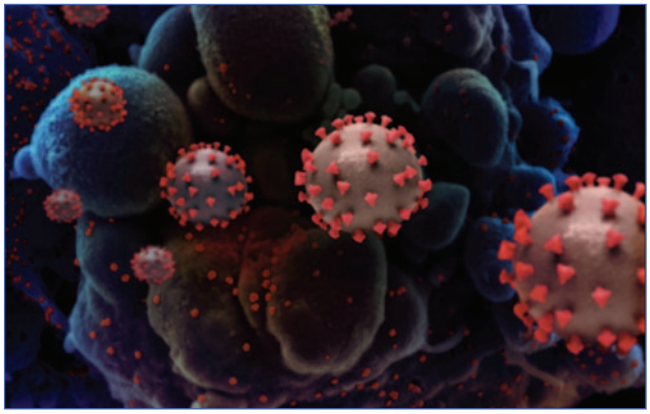

Figure 3: SEM Image of Novel Coronavirus SARS-CoV-2 (Courtesy NIAID).

information about the stability of SARS-CoV-2, which causes COVID-19 disease, and suggests that people may acquire the virus through the air and after touching contaminated objects. So the findings emphasise the importance of hand washing and disinfecting frequently touched surfaces to protect against infection.

The scientists highlighted additional observations from their study (2):

(a) If the viability of the two coronaviruses is similar, why is SARS-CoV-2 resulting in more cases? Emerging evidence suggests that people infected with SARS-CoV-2 might be spreading virus without recognising, or prior to recognising, symptoms. This would make disease control measures that were effective against SARS-CoV-1 less effective against its successor.

(b) In contrast to SARS-CoV-1, most secondary cases of virus transmission of SARS-CoV-2 appear to be occurring in community settings rather than healthcare settings.

So these findings show that SARS-CoV-2 is actually quite similar to SARS-CoV-1 in terms of stability in the environment. This means we can learn from our experiences with SARS in 2002-2004 to gain insights into infection control, especially in healthcare settings. On the other hand, it indicates that the major differences in the epidemiology of these viruses probably arise from other factors, especially the ability of SARS-CoV-2 to be transmitted by people not exhibiting clear symptoms. These results will inform future epidemiologic investigations that will be necessary to understand spread of this virus person to person.

Antimicrobial Paper

Like all other industries and organisations in the world, the pulp and paper industries also never anticipated that coronavirus could become a world calamity, keeping all activities at a standstill and causing thousands of deaths. While developments by the paper industry on reducing the hospital-induced diseases such as MRSA have been on-going for the last few decades, suddenly a new challenge has appeared and all focus is now on inventing new products to arrest the spread of coronavirus.

Based on the duration of antibacterial effects and the type of agent used in production of paper, it may be classified into two categories (see *Figure 4*):

A) Antibacterial paper with long lasting antibacterial property, and

B) Antibacterial paper with short duration of antibacterial property.

Obviously, for combatting coronavirus, the first category is much needed, while the second category may be used for common purposes, notably as hand, face and nose wipes. Indeed, the recent developments on Antibacterial grades involve mostly (A) types of papers, based on incorporation of nanoparticles having antimicrobial properties (7). *Figure 5* summarises the various antimicrobial spread breakers.

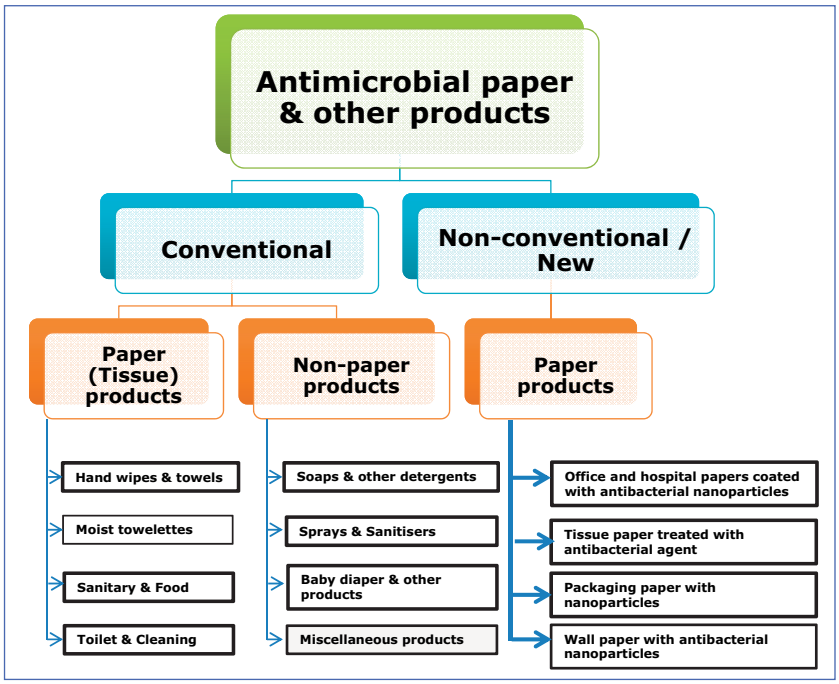

Figure 5. Antibacterial paper and products, which can act as spread breakers for coronavirus.

Bacteria and virus are in the micron and submicron sizes and in order to react effectively with these micro-organisms, harbouring help of nanotechnology, which deals in particle sizes even smaller than these micro-organisms, can serve the objective effectively. Moreover, because of the high surface area and antibacterial properties, the nanoparticles can act as killers for the viruses and the bacteria (7).

The efficacy of Ag-nanoparticles as antibacterial agents, is well-known and many products based on Ag-nanoparticles have been produced including additive for antimicrobial paper (5, 7-12).

Considering the ongoing coronavirus outbreak, apart from staying at home, people need other items to halt the spread of this respiratory disease. Various nanotechnology products are available (13) to equip people for combating COVID-19. Even though nanoparticle- or nanofibre-improved respiratory masks and gloves are required to be employed outside to limit the number of people exposed to potential risks, soaps, sanitisers, disinfectants, shampoos, and detergents, composed of antiviral and antibacterial nanomaterials, can be used inside to fight against this pandemic. Graphene, nanodiamond, polymer nanofibres (e.g., polyacrylonitrile), and such nanoparticles as silver, titanium dioxide, and copper oxide are commonly incorporated into these categories of products so as to contribute to their proficiency.

The overwhelming demand for nano-enabled or nano-enhanced gowns, aprons, scrubs, and protective gear has set off a race among health centres. Other much-needed goods such as air filtration systems containing high-efficiency particulate air (HEPA) filters play a pivotal role in capturing bacteria, mould spores, and viruses to prevent airborne contamination in hospitals. Such systems, activated by UV light on the basis of nanostructured absorbers, are certified to filter the majority of airborne infectious agents down to 3nm.

A new development is the first-ever molecular photo catalyst air purifier works which is based on a chemical reaction between the nanoparticles and the pollutants under the presence of UV light. Moreover, state-of-the-art systems based on electric filtering remove ultra-fine particles down to 0.3nm, using an electric shock. This ventilation system does not contain filters that can get clogged, therefore the purification efficiency stays high at all times (12).

It is felt that nanotechnology is under-utilised in a time of crisis like now (14). It is exigent to review the various research studies undertaken for contributions, likely to be made

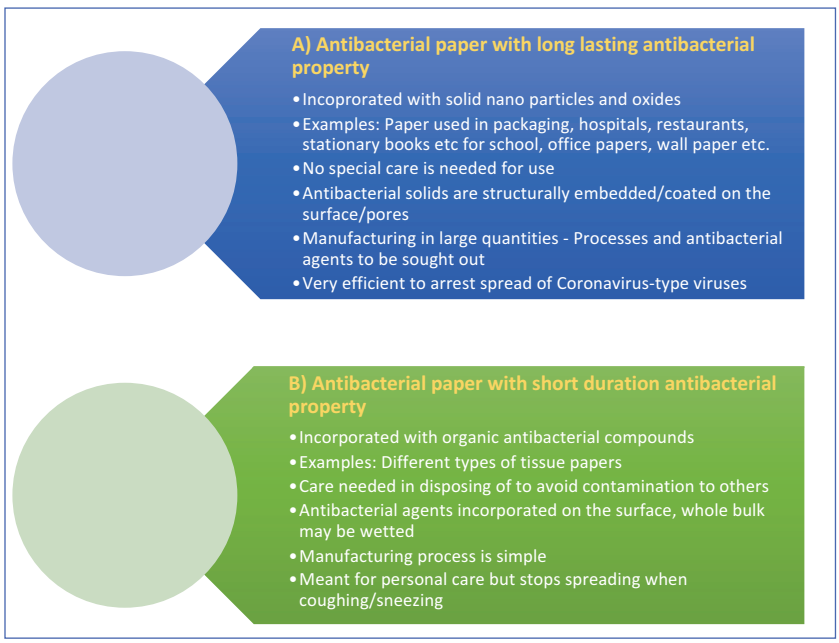

Figure 4: Graphic demarcating the two types of prominent antibacterial paper (7).

by nanotechnology not only to stop spreading of the virus but also in diagnostics and treatment.

i) Addmaster – antibacterial and antiviral treatment

Addmaster (15) in UK is now the major producer and supplier of antibacterial additives for paper as well as textiles, paint and coatings. It is engaged in developing antibacterial additives and testing against MRSA in hospitals and other healthcare settings. In addition it has gained wide experience and expertise on antiviral products and is now tackling the coronavirus crisis. Numerous tests on coronaviruses have been made by this Company, particularly the feline strain as it is similar to COVID-19. [See Page 26]

Their Biomaster antimicrobial additives provide long-lasting and effective product protection against harmful bacteria, biofilm, fungi and mould for the lifetime of the end product without affecting its structural and physical qualities. This is particularly important in hygiene critical situations, where every step possible should be taken to reduce cross-contamination, especially as studies have shown that bacteria such as MRSA can survive on paper for up to 38 weeks.

ii) Antibacterial Printing Technology

Galloways Printers Ltd (16) based in Cheshire is also using Biomaster additives in their print formulations aimed in particular at:

- Food service industry for order books and menu cards,
- Education for books and stationery,
- Travel and leisure sector, for in-flight safety cards,
- Health care, for patient files.

The company is also looking to protect workstations with protective paper coverings of this antibacterial paper so that staff can hot desk safely.

iii) James Cropper Products

The PaperGard grade produced by James Cropper (17) is treated with Biomaster anti-microbial silver ion technology. It is aimed at a wide range of applications, from premium boxes for luxury brands to greetings cards and envelopes. [See Page 27]

iv) American Eagle Products

American Eagle Paper Mills (18) has introduced a new antimicrobial paper line; the Eagle Armour suite of paper products has been designed to reduce the transmission of communicable diseases. Again these have been treated with Biomaster silver ion technology. What is special about Eagle Armour antimicrobial paper is that it inhibits the growth of bacteria without sacrificing paper performance or appearance, producing paper with 92% brightness from recycled pulp.

v) Nekoosa Coated Products

Nekoosa Coated Products is another American company using Biomaster additives, this time in what the company claims to be the world's first antimicrobial carbonless paper. According to their website, this technology will not affect the paper's properties such as dry toner/laser compatibility or carbonless image permanence. This product is aimed at any high traffic environment such as hospitals, classrooms, exam rooms, offices, clinics and day-care centres which use carbonless forms (19).

vi) Antimicrobial Tissue Paper using Nanocellulose

At NC State's College of Natural Resources (20), researchers have developed a new tool that can help prevent the spread of COVID-19 and other infectious diseases: antimicrobial tissue paper. It is based upon a hydrophobic spray-coating of chitosan (Ch) and cellulose nanocrystals (CNCs) composite. Hand-sheets were prepared, spray-coated with Ch, CNC, and their composite coating (ChCNC), and tested for antimicrobial activity against Gram-negative bacteria *Escherichia coli*. The highest bactericidal activity was observed with ChCNC-coated tissue paper, inhibiting up to 98% microbial growth. Plasma treatment further improved the antimicrobial activity of the coatings.

vii) Cascades Disposable Paper Towels

Cascades took a different approach for their Antibacterial paper towels, launched in 2012, which contained micro-encapsulated benzalkonium chloride coated onto the surface. As the user dries their hands, the capsules break, transferring the active agent onto the user's skin where it kills bacteria (21).

viii) Sofidel Disposable Paper Towels

Sofidel (22), one of the world's leading manufacturers of tissue paper for hygienic and domestic use, continues to innovate and diversify its solutions for personal and household hygiene proposing the new Nicky Defend, a disposable tissue paper towel with antibacterial lotion designed for the Consumer market and specifically developed to guarantee proper hygiene**.**

Generally used in public spaces and workplaces (Away-from-Home), disposable paper towels are now making their entrance into homes, offering a soft, strong and absorbent alternative to fabric towels for drying the hands. Disposable antibacterial towels are designed to retain the germs and bacteria present on wet hands and prevent them from multiplying, and the special interfolded dispensing system allows the towels to be taken out one at a time, protecting the product against external contamination.

ix) Antiviral Facial Tissues

An antiviral facial tissue (23) is treated with a solution to deactivate cold and flu viruses in cough, sneeze, or nasal discharge to prevent spreading of the virus. While these tissues could help with this, there are several real-world-use factors that affect how well they work.

Antiviral three-ply facial tissue has a moisture-activated middle layer, which is treated with an anti-viral formula consisting of citric acid and sodium lauryl sulphate (a surfactant found in many soap and cleansing products). According to the manufacturer, when cough or sneeze residue hits the middle layer, the tissue begins working immediately, killing nearly all cold and flu viruses it captures.

The packaging for Kleenex Anti-Viral tissues notes which germs it has been tested against: it claims that it inactivates 99.9% of rhinoviruses type 1A and 2, influenza A and influenza B, and respiratory syncytial virus (RSV) within 15 minutes. However, the Kleenex Anti-Viral tissues are not proven to protect against coronaviruses, which cause 10% to 30% of viral upper respiratory infections.

In the patent application made in 1986, Kimberly-Clark explained that the antiviral components needed to be placed in a middle layer of the three-ply tissue because they could be irritating if they came in contact with the skin. The tissues reached the market in 2003, and the packaging was updated in 2009 to note that the tissues would inactivate HIN1 influenza (swine flu).

Antiviral tissues do not deactivate viruses in or on our body, so they do not shorten the course of illness. In theory, they might reduce the chance that the virus is spread to someone else who might come in contact with the discarded tissue. Colds and influenza are spread in two main ways. First, by droplets spread through the air when one coughs or sneeze, which can travel as far as six feet. Covering our cough or sneeze helps prevent this

spread, but it will not eliminate it completely. Using a tissue will reduce the spread of germs whether it is an antiviral tissue or not.

Viruses are also spread by droplets settling on surfaces or virus being transferred to surfaces from our hands that are contaminated by our own respiratory secretions. A cold virus can live outside of the body or on a hard surface for three or more hours.

If one uses the tissue and cannot dispose of it immediately, it is possible that the antiviral tissue would deactivate the virus, so the tissue would be less infective to others who would come in contact with it. However, it may be noted that these tissues do not deactivate the viruses on our hands or face, as the sides that touch our skin are not treated with antiviral formula. One still needs to wash the hands well after using the tissue to avoid spreading germs.

Whether or not someone could catch a virus from coming into contact with our used tissue also depends on whether enough time has elapsed since it was used (since they do not work immediately) and the amount of discharge (too much may overwhelm the antiviral agents in the sheet).

Demand for Antiviral Packaging to increase

The coronavirus pandemic continues to rewrite the nature of global society in 2020, and with it the attitudes and behaviours of consumers now and for the foreseeable future. Anxiety around the avoidance of germs and other contaminants is driving demand for safer products with higher integrity, and so innovation that provides added assurance can help remove barriers to consumption (24).

Novel coronavirus provides a turbocharged boost to consumer anxiety, but also the potential for innovation, associated with the Sterilised Society trend. Certain categories and packaging types stand to benefit from a wider concern; UHT products in heat-treated aseptic packaging can tout their credentials as part of a specialised and sterile production process and final product. Furthermore, research is ongoing into antibacterial and antiviral polymers and biopolymers for packaging – materials that are enhanced with active drug elements that are efficient and exhibit low toxicity. Demand for such materials in day-to-day consumer products may rise significantly post-COVID-19, as consumers are likely to maintain concerns and habits learnt during this difficult period.

Antibacterial Personal Care Wipes market

Antibacterial personal wipes are used for cleaning and disinfecting purposes and often come folded and wrapped for convenience. They are made of tissue, paper, or nonwoven and contain antibacterial ingredients which are effective at killing 99.9% of harmful germs (25). Most antibacterial personal wipes offer protection from a broad range of bacteria and micro-organisms.

Leading companies are focused on offering gentle Antibacterial Personal Care Wipes that biodegrade in 28 days and are made of 100% renewable plant fibres to ensure that no waste is generated that might pollute the environment. Therefore, the development of new and innovative biodegradable Antibacterial Personal Care Wipes is projected to provide a huge growth opportunity for the key players operating in the market during the forecast period.

Based on product types, the antibacterial personal wipes market is segmented into sanitising, skincare, and wound cleaning. The sanitising segment held the largest share of the market in 2019 and is estimated to register the highest CAGR in the market during the forecast period; at a CAGR of 9.3% from 2019 to 2027.

Manufacturing Paper containing Nanoparticle Additives

One of the likely problems, to be faced in the manufacturing of antibacterial paper in the paper mill is how to incorporate the nanoparticles onto the paper or board. There could be many techniques for manufacturing of the antibacterial paper. Antibacterial nanoparticles such as Ag (silver), which may be available in ionic form (Ag^{+}) or reduced metallic form (Ag^{o}); these being quite costly, one cannot afford to add anywhere and any amount in the manufacturing process.

(i) During stock preparation

It cannot be added at the stage of stock preparation because one cannot ensure that the trace amount of additive will adhere to the paper surface.

(ii) During surface sizing

At the stage of size press, one may add but it has to be experimented thoroughly; the other safe stage where it can be added, is during coating. So, the first thing to be sorted out is at which stage it is to be added. A metered size press could serve the purpose appropriately.

(iii) During coating

Addition of the nanoparticle as such alone, cannot result in a uniform and well-coated product so that the resulting papers acts as antibacterial product for whole life period or long period. Metallic ions having antibacterial and antifungal capabilities, such as silver, copper and zinc, are impregnated in a mineral or blended with a carrier to form the composite for application as a coating.

(a) **With regular coating pigments**: metal ions can be embedded in some mineral or coating pigment regularly used in coating, namely calcium carbonate (precipitated or ground), titania, kaolinite clay or often talc mineral.

(b) **With other clay minerals**: extensive studies were carried out to exchange the metallic ions in the interlayer spaces of clay minerals such as vermiculite (26) and montmorillonite (27). Attempts may be made to incorporate the silver ions with the regular pigments used in the paper coating, such as kaolinite clay, calcium carbonate and titania (28).

Inorganic antibacterial material is usually in the form of a composite. This is considered as highly safe (non-volatile) and heat-resistant compared to organic materials. Appropriate release of antibacterial metallic ions from the composite can effectively inhibit the birth and growth of harmful microbes. Silver has a broad spectrum of antibacterial activity, while exhibiting low toxicity towards mammalian cells (29).

By impregnating silver or copper ions into the nanometre openings between the layers of vermiculite, it is possible to render the composite with antibacterial functions if the metal ions can be released gradually from the composite.

The minimum concentration of silver ions to inhibit *Escherichia coli* is as low as 0.78μg/ml. Silver-loaded inorganic antibacterial materials, such as Ag-zeolite, Ag-glass, Ag-zirconium phosphate, Ag-phosphorite and Ag-TiO_2, have been developed. These products generally show high antibacterial activity and low toxicity. However, the high reactivity of silver was found to be a problem. Examples include the reduction of silver ions to elemental silver, causing the loss of antibacterial ability, and the change to dark colour after ultraviolet irradiation or heating of the composite.

(c) **With kaolin**: Kaolin has some antibacterial properties and is used to absorb toxins and bacteria like the other clays. New multifunctional materials possessing antibacterial and antifungal properties have been derived from clay

minerals and other inorganic materials by using cation exchange reaction and surface modification (30).

Conclusion

There exists a lot of confusion about the Antimicrobial products meant for different micro-organisms to reduce proliferation and spread of diseases caused by bacteria and viruses. While there are some products and papers developed for MRSA and even SARS CoV-1, there has only been a beginning made to combat the present coronavirus (SARS CoV-2).

The antibacterial papers can be divided into two categories in terms of the antibacterial additive used; the Ag-nanoparticle based antibacterial additive and the tissue paper, where mostly organic compounds are added.

The Ag-ion based paper has been tested to be working very well, reducing the Feline coronavirus to >95%. Three paper companies (one in UK and two in USA) have already started producing antimicrobial papers at industrial scale for use in hospitals, packaging, school stationary, restaurants etc.

The other category is the antibacterial tissues, where the antibacterial additives, which are mostly organic compounds, are applied on the surface. While these tissue papers can serve to reduce bacteria and virus at the time of sneezing or coughing, these cannot reduce the spread of present coronavirus infection; rather caution should be taken in disposing of the used tissues.

Efforts for application of nanotechnology in producing appropriate antibacterial papers, should be intensified as many metallic and oxide nano materials are available. These have been tried extensively for producing multitude of materials (6) for use in all domains and it is high time to intensify research to incorporate them for production of efficient antibacterial products. It is proposed that the antibacterial nanoparticles should be incorporated into the commonly used pigments for coating or in some clay minerals such vermiculite and montmorillonite and the composite product to be added at the stage of surface sizing or coating.

The demand for antibacterial paper and products is likely to increase in future because of the impact of coronavirus inculcating use of safe and hygienic products with antimicrobial properties.

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Disclaimer

Much information was collected from the websites or was received from the organisations noted. The author takes no responsibility for the claims made.

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