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REVIEW

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Cellulose-based hydrogels for personal care products

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Name of cellulose is referred to a type of natural carbohydrate with many hydroxyl groups and high water absorption capacity. Cellulose is the most abundant natural polymer, found as the main constituent of plants (plant cellulose). Some bacteria such as *Acetobacter xylinum* are also able to synthesize bacterial cellulose. Cellulose-based hydrogels are superabsorbent materials, which make 3D networks. Chemical bonds or other cohesive forces such as hydrogen bonding or ionic interactions connect the cellulose chains together. Hydrogels can swell and absorb water and other aqueous fluids in their 3D networks, but they are insoluble in them. Nowadays, an increasing demand emerges for biodegradable materials and products made from renewable resources such as cellulose. The excellent biocompatibility of cellulose has prompted the large use of cellulose-based personal care products. Cellulose hydrogel is used for these products as the thickener and stabilizing agents or as moisturizing agent to improve the skin feel of the product. Hygienic cellulosic absorbent products such as diapers, panty liners, tampons, paper towels, and tissue papers are used as personal care products. These products are available in different absorbency ratings from junior to ultra-absorbency. Using cellulose-based hydrogel, superabsorbent products are made. In this review, applications of cellulose-based hydrogels in personal care products were reviewed.

KEYWORDS

cellulose, diaper, hydrogel, personal care products, tampon

1 | INTRODUCTION

Superabsorbent polymers (SAPs) are primarily used as an absorbent for water and aqueous solutions for diapers, adult incontinence products, feminine hygiene products, and similar applications. Undoubtedly, in these applications, superabsorbent materials will replace traditional absorbent materials such as cloth, cotton, paper wadding, and cellulose fiber, which suffer from some limitations such as low efficiency, low recyclability, and leakage.

Commercial production of SAPs began in Japan in 1978, for use in feminine napkins. This early superabsorbent was a cross-linked starch-g-polyacrylate. Polyacrylic acids eventually are replaced previous superabsorbents, and they are the primary polymer used for SAPs today. In 1980, European countries further developed the SAP for use in baby diapers. This first diaper using this technology used only a small amount of polymer, approximately 1 to 2 g. In 1983, a thinner diaper using 4 to 5 g of polymer and less fluff was marketed in Japan.[1]

Absorbent hygiene products (AHPs) have made an important contribution to the quality of life and skin health of millions of people.

The category of AHPs consists of the following 3 groups of AHPs with benefits such as the softness, smoothness, leakage prevention, strength, and protection provided by nonwoven fabrics:

- Baby diapers
- Feminine hygiene products
- Adult incontinence products

Since in the mentioned applications good skin care has a crucially important role, so using biocompatible and biodegradable materials without any negative effect on human skin and environment are preferred.

2 | DIFFERENT TYPES OF PERSONAL CARE PRODUCTS

Baby diapers, sanitary pads, panty lines, and adult incontinence products typically consist of a top layer (nonwoven or perforated film), an absorbent core (fluff pulp and/or superabsorbers), a back sheet (plastic film, nonwoven), and a fastening system (tape, belt, or Velcro). A tampon typically consists of a cover stock, an absorbent core, and a string. Some types of tampons also include an applicator. Each type of these products can include different products such as the following items in Table 1.[2]

The average diaper is based on 4 primary components. The top layer that is in direct contact with the body is made from a permeable polypropylene nonwoven. The next component is a layer that temporarily stores the urine and distributes it onwards to the absorbent layer. These components are both made from fluff pulp and SAP. The outer layer has to hold the diaper together and prevent leakage. It may contain small microholes to allow air to pass through. This layer is made from polyethylene (PE). In addition, a diaper contains a fastening system such as tape, plus elastic and dyes and print (Figure 1).[3]

In terms of weight, the fluff also makes up the largest proportion of a sanitary towel (approx. 66% of the total weight). Cotton is the largest constituent material by weight in the tampons (approx. 90% if the applicator is excluded). Cotton buds and cotton wadding are products that use the same materials as tampons. On the other hand, fluff (approx. 78%) and SAP (approx. 19%) are the main components of breast pads (Figures 2 and 3).[3,4]

Other disposable products, such as disposable bed linen, mattress covers and draw sheets, surgical gowns, and diaper liners, contain many of the same materials as diapers, incontinence care products, and sanitary towels.

Mattress covers, for example, may be made from several layers of cellulose, nonwoven fabric, and a plastic coating. They may also contain fluff pulp to increase absorption. Some products state that they have seams that are strong enough that the patients can be lifted and that they have edges treated with paraffin to ensure against leakage. The plastic film, which stops moisture from permeating through the products, may be PE, for example, or a bio-based plastic film. The products may be laminated with plastic glued in place (water-based dispersion adhesive or hot-melt adhesive), or the plastic film may be directly extruded onto the product. Use of nonwoven products has risen in recent years, particularly in the health sector, in place of cotton/polyester products. There are various ranges of disposable bed linen on the market that are made from nonwoven fabric with polypropylene fibers.[4]

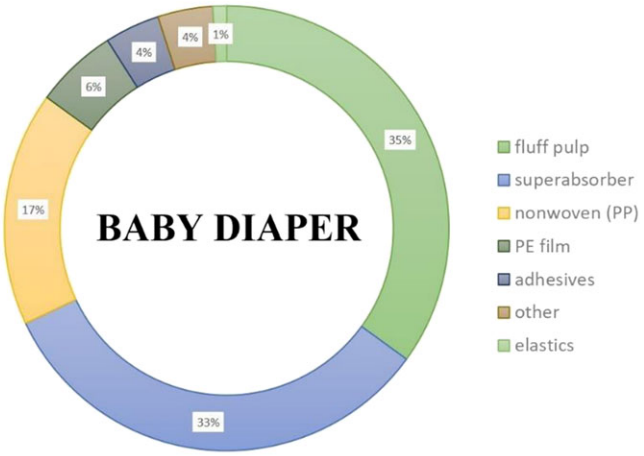

FIGURE 1 Average baby diaper composition.[3] PE, polyethylene; PP, polypropylene

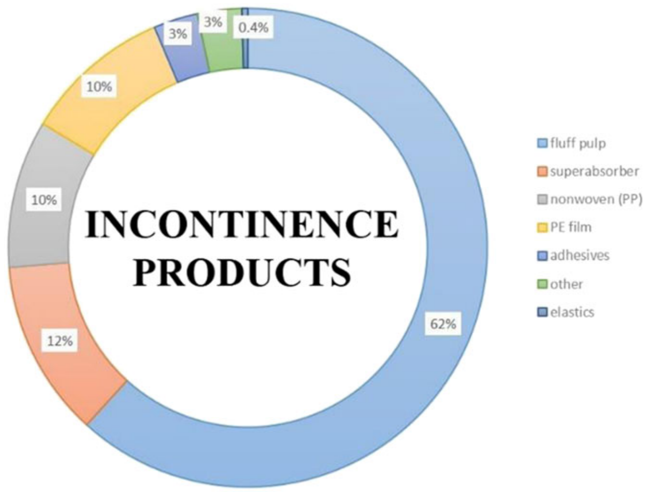

FIGURE 2 Average incontinence products composition.[3] PE, polyethylene; PP, polypropylene

2.1 | Feminine hygiene products

Sanitary napkin was the first disposable article of commercial significance and introduced in 1921. The first menstrual tampon was introduced by Tampax Corporation in 1933. The original Tampax brand tampon was made of cotton fiber wrapped in a nonwoven rayon coversheet. Today, the absorbent medium used in most modern tampons is cellulose fluff.

Another development in absorbent technology occurred in the 1960s with the advent of superabsorbents. Superabsorbent diapers were introduced in Japan in 1979, but have only become widely used in the West since 1984.[5]

House et al claimed that 26% of the world's female population is in their reproduction stage.[6] The female hygiene products that exist

TABLE 1 Classification of absorbent hygienic products

Product Groups	Products
Feminine sanitary protection	Sanitary towels, sanitary napkins, pant liners, panty shields, tampons, breast pads
Baby diapers	Baby diapers, pant diapers, training pants, swimming pants
Adult incontinence products	All-in-one products: Contains both the absorbent core and the outer shell with fastening (tapes, hook & loop, belts), insert pads (needs additional product for fastening), pants/briefs, liner pads, mesh briefs supports, bed protection, under pads

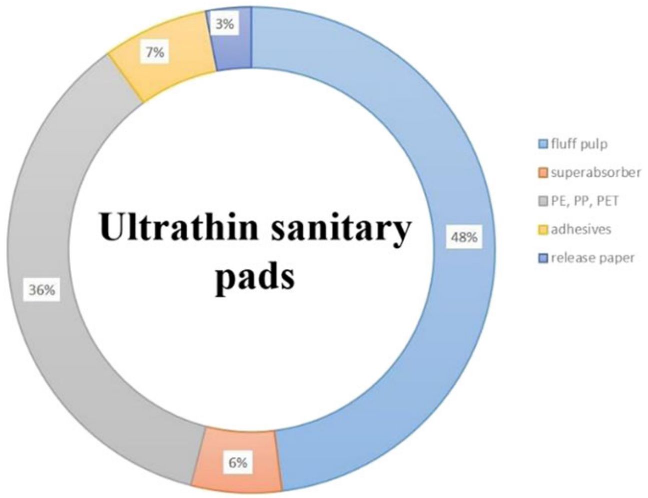

FIGURE 3 Average ultrathin sanitary pads composition.[3] PE, polyethylene; PET, polyethylene terephthalate; PP, polypropylene

today can be categorized into main groups, external and internal products, and these are divided into smaller groups.

- Disposable napkins, reusable napkins, and, finally, cloths and rags are in the group of external hygiene products for women.
- The internally used methods are different types of protections that are inserted into the vagina. They either absorb the blood or hold the blood inside the body for the user to take out when full. The category is divided into 2 under groups: tampons and menstrual sponges.[7]

The feminine hygiene products market includes different products such as sanitary pads, tampons, panty liners, and shields. Sanitary pads are the most commonly used feminine hygiene products and are expected to grow at notable rate owing to increasing demand from developing markets. These types of products are manufactured from a wide range of synthetic or natural raw materials and used by women to maintain their personal hygiene.

The most important factors affecting the demand for feminine hygiene products are the rising level of consumer education, increase awareness about alternative hygiene products, and modernization in retail format. So the feminine hygiene products market is expected to garner $42.7 billion by 2022.[8]

2.2 | Baby diapers

Before 1936, diapers were mainly either cotton-based ribbed toweling or cotton muslin-type material, both of which, with laundering, could be reused. In the late 1930s, early forms of tissue-based disposable under pads and diaper inserts were introduced in Sweden. Between 1936 and 1942, rubber pants were used to hold the cellulose pad in place. In 1950, the company introduced a new version of the product on the basis of bleached cellulose wadding with a knitted mesh outer layer, which could be inserted into a holding pocket in a rubber pant. Further developments in the mid-1980 owed much the availability of improved SAPs and better methods to add the polymer to the pulp core.[4]

The global baby diaper market covers different types of diapers provided by the manufacturers within the baby diapers. In fact, the different types of baby diapers manufactured around the globe are cloth diapers, disposable diapers, training nappy, swim pants, and biodegradable diapers. Depending on the type of absorbent materials used in manufacturing, disposable diapers are also divided into ultra-absorbent disposable diapers, regular disposable diapers, superabsorbent diapers, and biodegradable disposable diapers.

Nowadays, frequent use of diapers to maintain hygiene and prevent rashes in babies' skin, increasing birth rate, rapid urbanization, and continuously improving economic conditions in developing countries are the main factors, which drive the growth of baby diapers market. Global baby diapers market is expected to garner $59.4 billion by 2020. It seems that increase in disposable income and rising health care expenditures have driven the demand for baby diapers.[9]

2.3 | Adult incontinence products

Absorbent products specifically designed for adult incontinence are the newest category of hygiene products. Their use in Europe began in the late 1960s. The design is based on the technology developed for baby diapers and feminine hygiene protection. The key performance requirements such as protection from leakage, comfort, discretion, and skin dryness are similar in kind but differ in degree according to the severity of incontinence.[3]

There are different types of incontinence such as stress, urge, and mixed urinary incontinence as well as fecal incontinence. The condition affects people of all ages including young children and teenagers, women especially after childbirth, and men often with prostatic problems. People with neurological disorders and older people, particularly those with mobility and cognitive impairment, are vulnerable to incontinence problems.

The factors causing skin health problems in the incontinence pad area are the same for adults with incontinence as for babies and infants and are mentioned as below:

- Elderly skin is more fragile and vulnerable to the injury being thinner, less stretchable, and less resilient; as a result, it has poorer healing properties.
- Urinary incontinence is not always accompanied by fecal incontinence, so the mixing of fecal matter and enzymes with urine in adults is not commonplace as it is with babies.

Products that are highly absorbent and that reduce the effects of occlusion by the use of "breathable" materials are likely to reduce skin overhydration and will have consequent skin health benefits.[10]

There is a driving growth in the market for adult incontinence products such as adult diapers, under pads, light incontinence pads, absorbent underwear, and other products due to the rapid aging populations and the rising awareness about the hygiene and the convenience of these products. So the personal hygiene manufacturers are continuing to deliver on unique new products to satisfy a range of consumers who have a range of different needs.

According to recent data, adult incontinence is the fastest growing category in retail tissue and hygiene globally, with $7.2 billion in sales in 2015. Retail volume for these products is expected to grow at a compound annual growth rate of 7% through 2020, with the number of units sold projected to hit 25.8 billion in 2020.[11]

3 | THE COMPONENTS OF AHPs

The AHPs are designed for absorbing body fluids such as urine or menstrual fluid and feces especially in baby diapers and some incontinence products.

The fluid must be readily accepted, distributed, and absorbed by the structure of these hygienic products. Layered constructions and skin-friendly materials are selected for prolonged use.

There are 4 principal functional layers in these products, each of which is engineered to optimize overall product performance.

- Top sheet or facing: The layer next to the user's skin must be capable of allowing fluid to pass readily through to the next layer. It is important that fluid is not retained within the structure of this layer so that the amount of time that moisture is in contact with the skin is kept to the minimum. The softness of feel is a critical attribute for skin contact material.
- Acquisition and distribution layer (ADL): Fluid passes through the top sheet into the ADL where it is temporarily stored while capillary action causes it to spread over a larger area. This facilitates maximum utilization of the absorbent core structure. As most SAPs can take a few moments to fully absorb fluid, the ADL plays an important role in managing fluid during this critical stage. Any absorbent hygiene product with a high superabsorbent and low fluff pulp content, such as a baby diaper, will place high demands on ADL performance. Products with high fluff pulp or low SAP levels can often function without a separate ADL because of the good capillary properties of the fluff fibers.
- Absorbent core: The fluid storage layer typically relies on an appropriate blend of fiberized fluff pulp and SAP to absorb and retain urine or menstrual fluids. Total capacity can be engineered to a level appropriate for any individual product application. Twin cores can also be used to manage high fluid levels in heavy incontinence products. Many modern "ultrathin" feminine hygiene products use an engineered air-laid substrate as an absorbent layer. This is generally a precombined structure of multiple layers, containing both SAP and fluff pulp and other suitable capillary fibers. The resultant product is extremely thin and discreet in use. The absorbent core in modern tampons is made of the cellulose-based absorbent material, of either rayon (viscose) or cotton, or a mixture of both.
- Back sheet or outer cover: The function of the back sheet is to provide a fluid impervious barrier so that moisture is contained within the structure of the absorbent hygiene product. Many types of films are suited to this application, most commonly used is low gauge PE. Many products are subsequently enhanced by the addition of soft nonwoven covers to the PE, which can be colored or printed on to suit market needs. It is also generally accepted that a breathable film or nonwoven layer is of benefit in maintaining good skin condition, particularly in baby diapers and panty liners. Polyethylene can be made breathable film at extrusion stage by creating micropores in the substrate, and fibers in the nonwoven layer can be treated to be hydrophobic. These 2 processes allow air to pass through the film while maintaining an effective fluid barrier.[3]

4 | SUPERABSORBENT MATERIALS IN PERSONAL CARE PRODUCTS

Superabsorbent polymers are hydrophilic materials that are able to absorb physically and retain large amounts of water or aqueous solution.[12] As SAPs, superporous hydrogels (SPHs) are formed by covalently cross-linked hydrophilic polymers.

The first generation of SPHs was made from highly hydrophilic acrylamide (AM), salts of acrylic acid (AA), and sulfopropyl acrylate. Superporous hydrogel composites are the second generation of SPHs. For making SPH composites, a matrix swelling or a composite agent is used. A composite agent is a cross-linked water-absorbent hydrophilic polymer that can absorb the solution of monomer, cross-linker, initiator, and remaining components of the SPH synthesis.

The third generation of SPHs is presented by hybrid SPHs produced by adding a hybrid agent to the SPH previously made. Hybrid agents are natural or synthetic water-soluble or dispensable polymers capable of chemical or physical cross-linking. It is possible to produce an interpenetrating polymeric network with this method (Figure 4).

For example, AM-based SPH is synthesized in the presence of sodium alginate and after that cross-linking occurs between alginate chains and calcium ions forming a hybrid SPH.[13]

Superporous hydrogels can also retain this liquid under a certain pressure and therefore are suitable for use in sanitary products such as diapers and sanitary towels. Today, the greatest volume of SAPs comprises full synthetic or of petrochemical origin, and they are mostly produced from the acrylic monomers (Figure 5), most frequently AA, its salts, and AM.[12,14]

The global SAP market is expected to reach USD 7.96 billion by 2020, registering a compound annual growth rate of 7.6% between 2015 and 2020. This growth is fueled by the high demand from Asia-Pacific, the Middle East and Africa, and Latin America and the ageing population in developed countries and the thinner diapers. The market for diaper is the largest application for SAPs and hence driving the demand for the polymer in the recent years.[15]

Synthetic SAP-based products include a polyacrylamide copolymer, sodium polyacrylate, polyvinyl alcohol copolymers, and ethylene maleic anhydride copolymer. Polyacrylamide copolymer is likely to witness fastest growth rate owing to properties such as high water retaining and absorption.[16]

While most sanitary products such as diapers, sanitary towels, and incontinence care products are becoming thinner and lighter due to use of SAPs, there may be obstacles to use synthetic SAPs.[12] Volatile AA price and supply deficit are likely to challenge industry participant growth. The raw material deficit in the market is likely to increase manufacturing costs and bring down profit margins. The world's decision for environmental protection potentially supports the ideas of

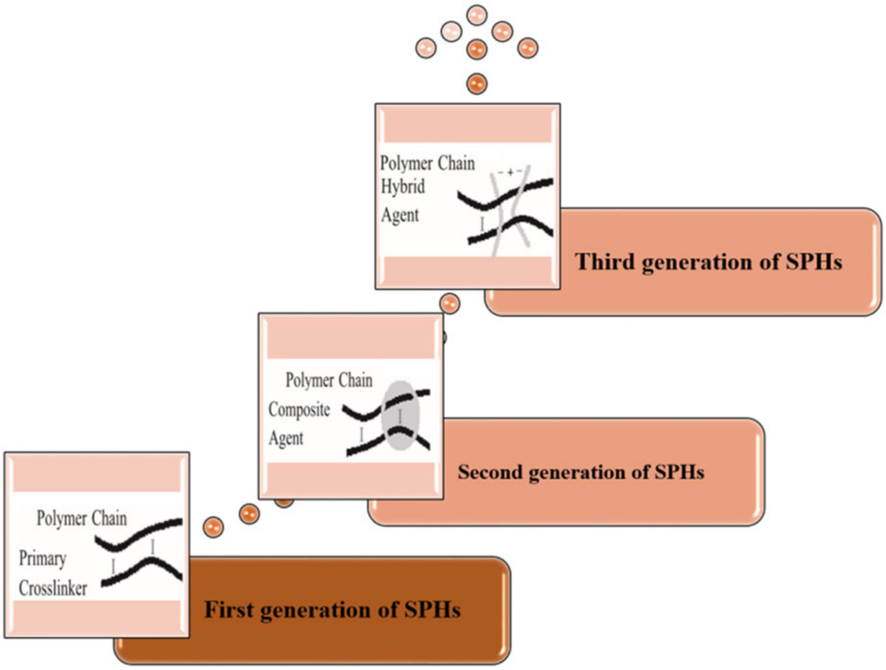

FIGURE 4 Structure of various superporous hydrogels (SPHs) generations

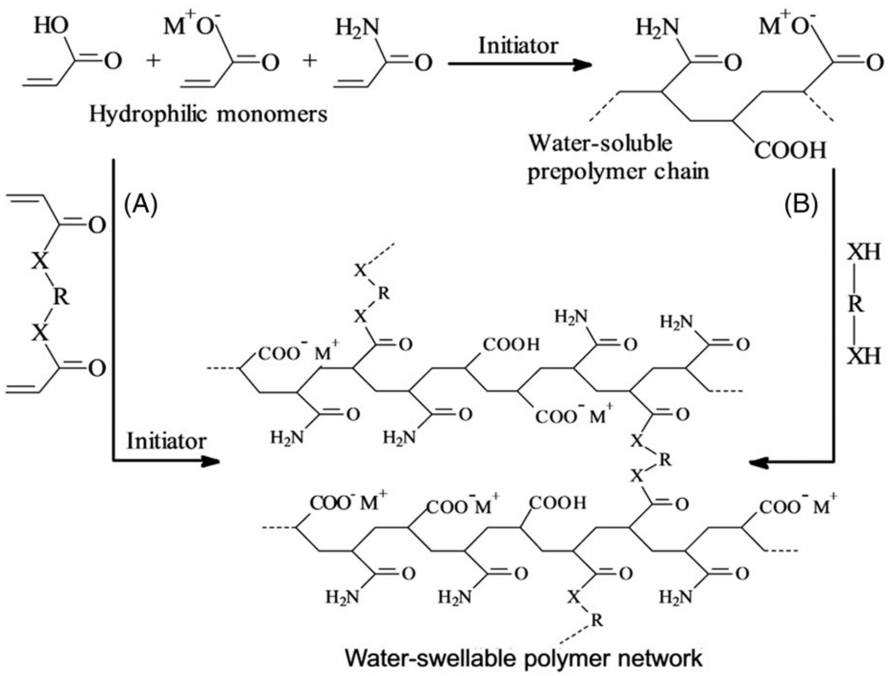

FIGURE 5 Chemical structures of the reactants and pathways to prepare an acrylic superabsorbent polymer network[12]

partially or totally replacing the synthetics by "greener" alternatives. To offset this scenario, companies are investing in the development of bio-based market.[16]

According to previous information from manufacturers of sanitary products, bio-based SAP has a lower absorption capacity than SAP of fossil origin. There is a great deal of research and development work underway in this area, and this is expected to result in a wider range of commercial products offering better quality and environmental performance in the near future.[17]

The bio-based SAP disposal products offer environmental benefits such as increasing the use of renewable resources due to the introduction of highly performing bio-based SAPs, which will reduce the use of oil-based absorbent materials,[18] and also, these types of absorbent materials have the benefits of reducing negative effects of synthetic SAP on the human body.

Making recyclable disposable diapers, napkins, hospital bed sheets, sanitary towels, and other similar products is one of the main targets for modern industries. Bio-based SAP is manufactured using renewable raw materials such as starch, cellulose, natural gums, and chitin.[16]

A novel solution to solve this problem involves the use of cellulose-based hydrogels, which are totally biodegradable. New types of bio-based hydrogels containing sodium carboxymethylcellulose and hydroxyethyl cellulose cross-linked with divinyl sulfone can swell like

SAPs. These improvements were achieved by introducing microporous and nanoporous structures into the hydrogel, which increases water retention and swelling kinetics due to capillarity effects.[19,20]

5 | ENVIRONMENTAL IMPACTS OF SAP HYGIENIC PRODUCTS

Most of the personal care products are made from the superabsorbent material, which is mainly composed of sodium polyacrylate, because of those diapers have a high water absorbing quality and can absorb 200 to 300 times its weight. Unlike natural polymers that are broken down by microorganisms to get smaller molecules, the C—C single bond of polymers found in diapers cannot be easily degraded by most microorganisms.

Most of the diapers are not easily biodegradable and have an estimated period of up to 500 years to decompose. Disposable diapers thus have bad effects on the environmental, as they are a form of solid waste. Disposable diapers are the third largest contributor to municipal solid waste in the United States of America accounting for 1.5% to 4% of the total waste.[21]

Apart from the solid waste problem, chemicals released by decomposing solid wastes can leak from dumping sites and landfills to groundwater. They can contaminate water wells, soil, and nearby water streams.[22] Also, they release some harmful gases, drainage clogging, and water contamination.

Only disposable baby diaper requires a lot of natural resources and energy. It has been estimated that over 136 kg of wood, 23 kg of petroleum, and 9 kg of chlorine are used to produce disposable diapers for one baby each year.

Assuming that one child will use at least 6500 diapers in a lifetime, this means that it takes about 1625 quarts of oil to diaper one baby for 30 months. It has been stated that it takes more oil to keep one baby dry for two and a half years than it does to lubricate all the cars an individual will ever own in a lifetime.[23]

6 | HEALTH IMPACTS OF SAP HYGIENIC PRODUCTS

Diapers can be classified as hazardous waste. Such waste has immediate or long-term negative health effects. It emerged from the laboratory results that diapers contain harmful chemicals such as dioxins. According to World Health Organization report on 2014, dioxins are persistent environmental pollutants that can cause an array of health problems including developmental delays, damaged immunity, hormone interference, skin diseases, and certain types of cancer.[22]

Wambiu et al indicated that the inner absorbent layer of a diaper is treated with chemicals, which can trigger allergic reactions among babies. When released into the environment, the dioxins can accumulate in humans.[24]

Dioxins are a class of persistent polyhalogenated aromatic hydrocarbons that induce a wide spectrum of toxic responses in experimental animals including reproductive, endocrine, developmental, and immunologic toxicities as well as carcinogenicity. The presence of an active human aryl hydrocarbon (Ah) receptor suggests that humans may respond to dioxins in a manner similar to experimental animals. In fact, there is mounting evidence of the health effects of dioxins in humans.[25] Evidence suggests that increased exposures to dioxins are associated with increased incidence of endometriosis in humans.[26]

On the other hand, some viscose rayon tampons contain toxic amounts of the chemical dioxin, because viscose rayon fibers are made from wood pulp. In the regeneration process, the wood pulp is bleached for a long time. The bleaching agents are a potential source of trace amounts of dioxin in tampons. Therefore, although the methods used for manufacturing tampons are considered to be dioxin-free processes, traces of dioxin may still be present in the wood pulp raw materials used to make viscose tampons.[27]

Another dangerous compound of conventional disposable diapers is sodium polyacrylate. This is the chemical added to the inner pad of a disposable that makes it superabsorbent. This material has some bad effects on human body. For example, it causes severe skin irritation, oozing blood from perineum and scrotal tissues, fever, vomiting, and staph infections in babies. Also, it can stick to baby's genitals and cause allergic reactions. In addition, some disposable diapers contain tributyltin and other heavy metals. Tributyltin spreads through the skin and has a hormone-like effect in the tiniest concentrations. It can harm the immune system and impair the hormonal system.[28]

Women, who are menstruating, are at risk of toxic shock syndrome (TSS) due to the release of poisonous substances from an overgrowth of bacteria called *Staphylococcus aureus*. The toxin can be absorbed through vaginal walls into bloodstream leading to TSS. The body responds with a sharp drop in blood pressure that deprives organs of oxygen and can lead to death.[29]

The use of high absorbency tampons has been closely associated with TTS. A saturated tampon, especially tampons with SAPs, with blood is a suitable place for overgrowing of bacteria.[30]

The link between TSS and tampons was not intuitive. Tampons had become trusted products in upwards of 70% of women's hygiene routines. However, the problem emerged when manufacturer shifted from cotton to synthetic materials. Companies often used cheaper ingredients, rayon, and absorptive material in some tampons.[31]

The incidence of TSS increased in the late 1970s and early 1980s, probably as a result of using superabsorbent tampons because of the relationship between the incidence of TSS, absorbency, and chemical composition of certain tampons. Rely as a manufacturer of high-absorbency tampons voluntarily removed the product from the market in September 1980. After 1985, tampons containing polyacrylate rayon, which was used to increase absorbency, were no longer manufactured.[32]

7 | NATURAL SUPERABSORBENTS: BENEFITS AND LIMITATIONS

Nowadays, using biodegradable polymers have been increased for the preparation of hydrogels because of their safety, biocompatibility, biodegradability, hydrophilicity, nontoxicity, low cost, and limited environmental problems.[33] In fact, the recognized hazards associated with the synthetic superabsorbents have spurred many researchers to develop greener alternative materials for personal care usage. Therefore, the best alternatives include natural superabsorbents such as polysaccharides-based and protein-based superabsorbents.

An important area of textiles is the health care and hygiene sector among other medical applications, because a wide range of disposable and nondisposable products such as surgical gown, mask, surgical drapes, towels, gloves, baby diapers, sanitary napkins, and others are used in hospitals. Since diapers come in contact with the skin, it is necessary to characterize the probable side effects such as skin inflammation and dermatitis. Also, parameters such as absorption capacity, absorption ability under load, and strike-through time are important as regards the wet comfort of diapers.[34]

In recent years, the increasing consumer focus on sustainable products is contributing to the growing popularity of bio-based superabsorbent polymers, which are generally based on modified cellulose, chitin, alginates, and silk fibroin. These products are an eco-friendly and sustainable alternative to various nonrenewable petroleum-based materials.

However, natural polysaccharide and protein-based superabsorbent have positive properties rather than synthetic polymers with their serious limitations, and they also display some disadvantages for use as biomaterials in personal care products. For example, their purification and low stability might be problematic. Also, when samples from natural sources are used, it is difficult to obtain identical compositions.[35]

Generally, an ideal superabsorbent for personal care products must have some functional properties, which are listed in Table 2.[12]

According to the requirement features for a superabsorbent, it is clear that the bio-based materials such as cellulose-based superabsorbent can simultaneously fulfill all the abovementioned required features.

However, it is necessary to make a balance between the properties. For instance, a hygienic superabsorbent used in personal care products must have the highest absorption rate and the lowest rewetting.[12]

This section will cover the details on the members of polysaccharides and proteins that are being widely investigated as a natural superabsorbent.

7.1 | Polysaccharide-based superabsorbent

Polysaccharide is the most attractive type for investment in the market due to increase in focus of manufacturers on bio-based SAPs and growth in awareness among consumers. Polysaccharides are the least expensive and most abundant available renewable organic material. Therefore, the use of polysaccharides in cosmetic products is growing, together with the increasing attention of manufacturing companies toward green materials. Polysaccharides are the most abundant organic materials that are available, renewable, and cheap. Some of the most widely used polysaccharide as superabsorbents are chitosan, cellulose, starch, alginate, and carrageenan (CG). In personal care products, these polymers are used as thickener and stabilizer or as moisturizing ingredients that can also improve the skin feel of the product.[36]

Generally, the synthesis of polysaccharide-based superabsorbent is performed by 2 main methods:

a) graft copolymerization of suitable vinyl monomer on polysaccharide in the presence of a cross-linker, and
b) direct cross-linking of polysaccharide.

In the first method, a polysaccharide enters reaction with an initiator. Therefore, functional groups of polysaccharide and initiator interact to form redox pair-based complexes. Then, by homogeneous cleavage of the saccharide C—C bonds, carbon radicals on the polysaccharide substrate are produced.

These free radicals initiate the graft polymerization of the vinyl monomers and cross-linking agent on the substrate.

In the second method, direct cross-linking of polysaccharides, polyvinylic compounds like divinyl sulphone, or polyfunctional compounds like glycerol, epichlorohydrin, and glyoxal is often performed.[12]

7.1.1 | Chitosan-based superabsorbent

Chitosan with its unique properties such as nontoxicity, biocompatibility, biodegradability, biological activity, and low cost is considered as a good superabsorbent.[37] Chitosan is a modified biopolymer derived from chitin, which has been found in a wide range of natural sources such as crustaceans, crabs, shrimps, and cell walls of fungi.[38] It is composed of β-(1-4)-linked D-glucosamine and *N*-acetyl-D-glucosamine randomly distributed within the polymer. The term of chitosan usually refers to a family of polymers obtained after chitin deacetylation to varying degrees.[37-39]

Sadeghi et al synthesized a superabsorbent hydrogel on the basis of chitosan-g-poly (acrylic acid-co-acrylonitrile) as a new natural-based polymer with pH-responsiveness properties. The swelling of the obtained hydrogels exhibited a high sensitivity to pH in 3 and 8. This novel superabsorbent can be used for hygienic and drug delivery system.[40] Barleany et al produced chitosan-graft-poly(acrylic acid) superabsorbent hydrogel with a notable antimicrobial activity. The obtained product is a good candidate for baby diapers and sanitary pads. Because for hygiene products applications, superabsorbent materials with antimicrobial activities are needed to prevent skin irritation.[41]

TABLE 2 Properties of an ideal superabsorbent for personal care products

Properties of an Ideal Superabsorbent	
The highest absorption capacity	Photo-stability
The highest absorbency under load	The highest durability in the swelling medium
Desired rate of absorption	The lowest price
The lowest soluble content	Odorlessness
The highest biodegradability	Colorlessness
Nontoxicity	pH-neutrality after swelling

7.1.2 | Alginate-based superabsorbent

Alginate is a naturally derived water-soluble polysaccharide that is composed of (1 → 4)-linked β-D-mannuronic acid and α-L-guluronic acid with a nonregular pattern along a polymer chain.[42] It is one of the most available biomaterials, derived from brown and bacteria and located in the intercellular matrix as a gel containing salts like sodium, calcium, magnesium, strontium, and barium. Alginate is widely used in biomedicine and personal care because of its interesting abilities like gelling property, viscous nature, stabilizing properties, and high water absorption.[43] There are some researches about using alginate-based superabsorbent for various hygienic and medical applications.

Wang et al prepared a series of sodium alginate-g-poly (sodium acrylate-co-sodium p-styrenesulfonate)/attapulgite superabsorbent composites by graft copolymerization using a cross-linker and initiator. According to the results, the swelling behaviors of the superabsorbent composites were remarkably influenced by various parameters such as pH, salt medium, and solvents.[44] Ma et al introduced an eco-friendly superabsorbent composite on the basis of sodium alginate and organo-loess with high swelling properties. According to the performance of the eco-friendly superabsorbent composite, it can be used as a promising candidate for various fields of application.[45]

In addition, the combination of alginate and chitosan fibers can be used in the hygienic products. In fact, they can be used as the hybrid-contact layer. Therefore, their antimicrobial properties reduce leakage and prevent contamination and the transmission of infectious diseases. In addition, it can provide a fresh and dry interface between the body and the absorbent.[46]

Khong et al reported a new biocompatible hydrogel on the basis of the chitosan and alginate containing mannuronic acid (poly-M), which is used in medical and hygienic applications. Figure 6 shows a chitosan-alginate gelling system.[47]

7.1.3 | Starch-based superabsorbent

Starch is 1 of the 3 most abundant polysaccharides in plants and exists in the chloroplast of leaves and the amyloplast of seeds, pulses, and tubers and other parts of plants. Starches consist of a number of monosaccharides or glucose molecules joined together with a -D-(1 → 4) and/or a -D-(1 → 6) bonds. Amylose and amylopectin are 2 main structural components of starch, and their proportion is a function of the starch source like corn, potato, tapioca, and wheat. Starch contains about 70% amorphous and about 30% crystalline regions. Starch has various applications in different fields due to its unique advantages. Biodegradability, availability, renewability, low cost, high swelling capacity in water, and easy processing are the most important features of starch. Regardless of the positive aspects of using starches, they have some disadvantages such as low surface area and surface modification to enhance sorption capacities.[48]

One of the most widely known starch-based superabsorbent on the market is Mater-Bi. Mater-Bi is a cornstarch-based material, which is marketed in 3 grades (A, Z, and V), and its application includes disposable nappies, feminine hygienic products, medical disposable, and others.[49]

The earliest commercial superabsorbent was produced from starch and AN monomer by the graft copolymerization method without using a cross-linker. The starch-g-PAN copolymer was then treated in an alkaline medium to produce a hybrid superabsorbent.[12]

Recently, a team of North Carolina researchers has developed a bio-based SAP on the basis of cornstarch instead of petroleum-based ingredients. They produced an environmentally friendly and cost-effective superabsorbent.[50] In addition, Erizal et al synthesized a superabsorbent of poly(acrylamide-co-acrylic acid)-starch by radiation copolymerization reaction. Experimental results showed that the hydrogels could absorb water very fast with a high swelling ratio at 3 minutes. The synthesized samples can be used in the personal care and hygienic products such as surgical pads, hot and cold herapy packs, medical waste solidification, disposable diapers, and sanitary napkins.[33]

7.1.4 | Bamboo-based superabsorbent

Bamboo is an Asian plant, which is found in tropical or subtropical regions. Bamboo is one of the most notable natural resources because it is a fast-growing plant with interesting properties. Bamboo is a highly water absorbent and has a natural effect of sterilization, moisture vapor transmission property, and easy drying. In addition, it has a

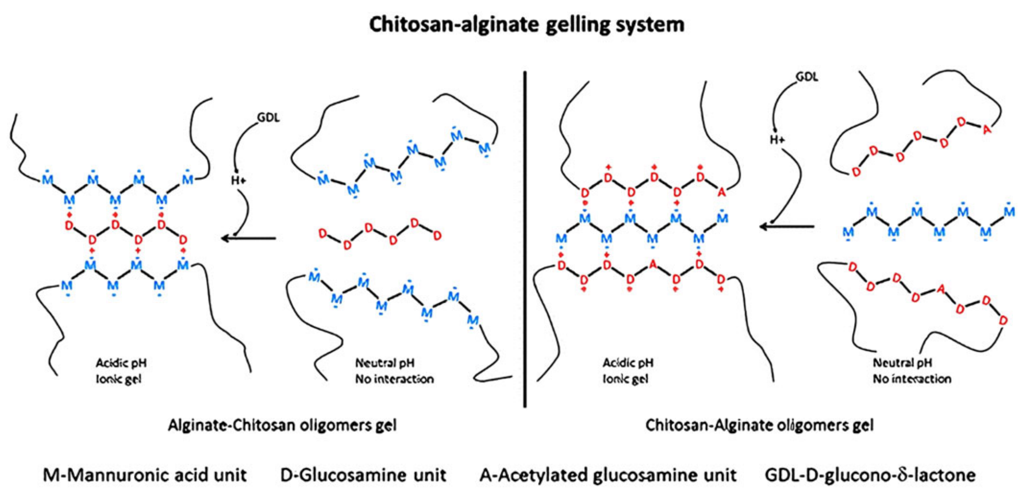

FIGURE 6 A chitosan-alginate gelling system[47]

unique antibacterial and bacteriostatic bio-agent named "Bamboo Kun." So bamboo fibers can be used in different hygienic applications such as baby diapers, feminine sanitary protection, adult incontinence products, sanitary towels, and absorbent pads.

In a research, Shanmugasundaram et al characterized the baby diapers made from 4 different fibrous compositions, namely, pure bamboo, pure cotton, bamboo/cotton (70/30), and bamboo/cotton (50/50). Antibacterial evaluation is performed on baby diapers against *S aureus* and *Escherichia coli*. The prepared diapers were characterized in terms of absorption capacity, liquid strike-through, acquisition time under load, and diaper rewet under load. On the basis of the results, the performance of diapers produced from a bamboo/cotton (70/30) fibers blend is superior in comparison with the other ones.[34]

7.1.5 | CG-based superabsorbent

Carrageenan is a high molecular weight linear polysaccharide consisting of repeating galactose units and 3,6-anhydrogalactose joined by alternating α-(1,3) and β-(1,4) glycosidic links. Carrageenan is soluble in water above 60°C and is considered as a thermosensitive physical hydrogel. The presence of sulfate groups in CG and its chemical affinity to mammalian glycosaminoglycan are the most important factors determining properties such as antiviral, anticoagulant, antioxidant, and anticancer activities. Carrageenan hydrogels are widely used in drug delivery, immobilization of enzymes, and various types of pharmaceuticals.[43] For example, Hosseinzadeh proposed a new full-polysaccharide superabsorbent hydrogel via chemical cross-linking of CG and sodium alginate (Alg) using epichlorohydrin as a cross-linker. The swelling of obtained hydrogels in solutions with various pH exhibited a high sensitivity to pH.[42]

In addition, using CG superabsorbent for producing the hygienic product has been increased in recent years. Salimi et al introduced a new smart CG-based superabsorbent hydrogel hybrid. The obtained product exhibited high water absorption and absorbency under load, which are substantial for baby diaper and feminine personal care absorbent products.[51]

7.1.6 | Cellulose-based superabsorbent

Cellulose-based products are used for the absorption of water and other aqueous fluids. Among cellulose-based products, paper towels and tissue papers are some of the most widely used absorbents. Fluff pulps are the most common products for disposable diapers. In fact, baby diaper uses cellulose fluff combined with a SAP to create the absorbent core, which acts as the storage structure in the product.[52]

According to the increased environmental awareness and serious problems of synthetic superabsorbents, it seems that cellulosic fibers for producing disposable diapers such as incontinence products and feminine hygiene products can be considered as a good candidate. Cellulose-based superabsorbents are explained in Section 8 in details.

7.2 | Protein-based superabsorbent

The protein-based superabsorbent is composed of isolated proteins from a natural extracellular matrix. Controllable bioactivity, biocompatibility, and biodegradability make these hydrogels promising candidates as smart biomaterials for drug delivery, tissue engineering, personal care products, regenerative medicine, and other applications.[35] This part will focus on proteins derived from natural sources, such as silk fibroin and gelatin and their application in personal care products.

7.2.1 | Collagen- and gelatin-based superabsorbent

Most of the collagens are fibrous proteins, responsible for important mechanical functions throughout the body. Collagens are particularly present in articular and bone tissues, where they provide most of the biochemical properties essential for proper functioning.[35] Gelatin is a heterogeneous mixture of polypeptides derived from collagen. It is soluble at the temperature above 35°C and gels below. Easy isolation and solubilization, low cost, biocompatibility, and biodegradability are the main properties of gelatin, which expand its applications.[53]

Marandi et al produced a series of hydrogel nanocomposites by grafting AA and AM on gelatin in the presence of Na-montmorillonite nanoparticles. The hydrogel nanocomposites exhibited good thermal strength, swelling in salt solutions, and also de-swelling behavior in different saline solutions that can make them useful in various fields such as hygienic product, medical, and agriculture.[54] Pourjavadi et al reported an efficient synthesis of hydrolyzed collagen-g-poly (sodium acrylate-co-2-hydroxyethyl acrylate) hydrogel through chemical cross-linking by graft copolymerization of these 2 monomers onto the protein backbone in the presence of a cross-linking agent. The hydrogel exhibited high water absorbency under load, which is considered as a good property for baby diapers and female hygiene absorbents. In addition, because of the use of protein as a natural backbone, it is more compatible with the human body.[55]

7.2.2 | Silk fibroin–based superabsorbent

Silks are natural structural proteins produced by insects such as orb-weaving spiders and silk worms. Silk fibroin is a fibrous protein produced by the silkworm *Bombyx mori*, which contains 2 protein components, fibroin and sericin. Fibroin consists of polypeptides that are joined by a disulfide bridge, and the sericin is hydrophilic protein surrounding the fibroin.[35] Recently, silk fibroin has become a popular biomaterial because of its excellent biocompatibility, tunable degradation, mechanical properties, ease of processing, and sufficient supply.[56] So the silk fibroin–based superabsorbent can be used in various personal care applications.

Bellas et al invented a silk fibroin–based composite comprising nonhydrolyzed silk fibroin and a humectant agent. The obtained silk fibroin–based compositions can be used for personal care products, cosmetic products, skin care products, body care products, and/or hair products.[57] Also, Pushpa et al prepared the nano silk sericin–based hydrogels from silk industry waste. The hydrogels having sericin protein are bio-acceptable and antimicrobial. Therefore, they can be used in a wide range of applications, for example, in agriculture for soil conditioners, seed germination, water retention, plant growth, and in biomedical fields as wound dressing membrane and as antimicrobial water absorption layer in personal care products like baby diapers and sanitary napkins.[58]

8 | CELLULOSE-BASED SUPERABSORBENTS FOR HYGIENIC PRODUCTS

With the appearance of superabsorbent hydrogels in the 1950s, considerable interest has been attracted within the scientific community as well as the industrial world. That is why rapid progress has been made in the past few decades because of the demand for superabsorbent materials in the sanitary industry. As mentioned before, superabsorbent hydrogels are networks with a high capacity of water uptake, which can absorb, swell, and retain aqueous solutions up to hundreds of times their own weight.[59,60] Even though most of the superabsorbent hydrogels are produced from synthetic polymers (essentially acrylics) for their acceptable properties, it has been suggested to replace synthetic polymers with the green alternatives because of their degradability and biocompatibility.[61]

One of the green materials that can be used for the production of superabsorbent hydrogels is cellulose. Cellulose, as one of the carbohydrate polymers, is the most abundant resource in nature. Cellulose products are biocompatible, biodegradable, nontoxic, inexpensive, and renewable. Cellulose, which has abundant hydroxyl groups, can be used to prepare superabsorbent hydrogels easily with fascinating structures and properties.[62]

Cellulose superabsorbent hydrogels can absorb and retain extremely large amounts of a liquid relative to their own mass. These cellulose hydrogels absorb aqueous solutions through hydrogen bonding with water molecules.[63] For example, Rouhani et al used *Luffa cylindrica* fibers as a cellulose-enriched material for preparation of cellulose nanowhiskers-based hydrogel as a surface modifying system with enhanced water absorption property. They applied the obtained hydrogel on the polyester fabric to achieve the high water absorption. It was found that because of the participation of hydroxyl groups of luffa nanohydrogel, the water uptake ability of treated fabrics in comparison with control polyester significantly increased.[64]

Many factors can affect the capacity of cellulose superabsorbents. The total absorbency and swelling capacity are controlled by the type and degree of cross-linkers used to make the superabsorbent product. Low-density superabsorbent hydrogels generally have a higher absorbent capacity and swell to a larger degree. On the other hand, high cross-link density polymers exhibit lower absorbent capacity and swell, but the gel strength is firmer and can maintain particle shape even under modest pressure.[65]

One of the largest uses of cellulose superabsorbents is found in personal disposable hygiene products, such as baby diapers, adult protective underwear, and sanitary napkins.[61]

Different methods of preparation of cellulose superabsorbent hydrogels and their usage in personal health products are explained below.

One of the advantages of cellulose-based superabsorbent hydrogels is their ability to overcome the routine obstacles of synthetic polymers. Most recently, cellulose-based superabsorbent hydrogels have become accessible and essential materials in many applications. Introducing cellulose series materials into the superabsorbent hydrogels can eliminate the disadvantages of synthetic-based superabsorbent hydrogels to obtain an acceptable product.[6] In this way, a product, which is a multifunctional and efficient superabsorbent product, can be achieved. Compared with the synthetic superabsorbent hydrogels, cellulose-based superabsorbent hydrogels have high absorbency, high strength, good salt resistance, excellent biodegradable ability and biocompatibility, and other special functions that promise a wide range of applications in many fields (Figure 7).[19,66]

8.1 | Preparation methods

There are many preparation methods for production of cellulose-based superabsorbent hydrogels. These methods are divided to 2 main groups, which are chemical and physical methods. Some of the methods are described in the following.

8.1.1 | Physical methods

Unlike chemical synthesis methods, physical synthesis methods always refer to the molecular assembly cross-link by the hydrogen bond or the ionic bond between the polymers, or by the interaction between the polymers.

8.1.2 | Chemical methods

The stable structure and effective swelling of cellulose-based hydrogels often require a chemically cross-linked network. Some difunctional molecules are used as the cross-linker for cellulose or its derivatives to covalently bind different polymer molecules in a 3-dimensional hydrophilic network.[66] Chemical synthesis methods are widely used to fabricate the cellulose-based superabsorbent hydrogels for the formation of covalent linkage, in essence. Typical chemical synthesis methods are stated as below.

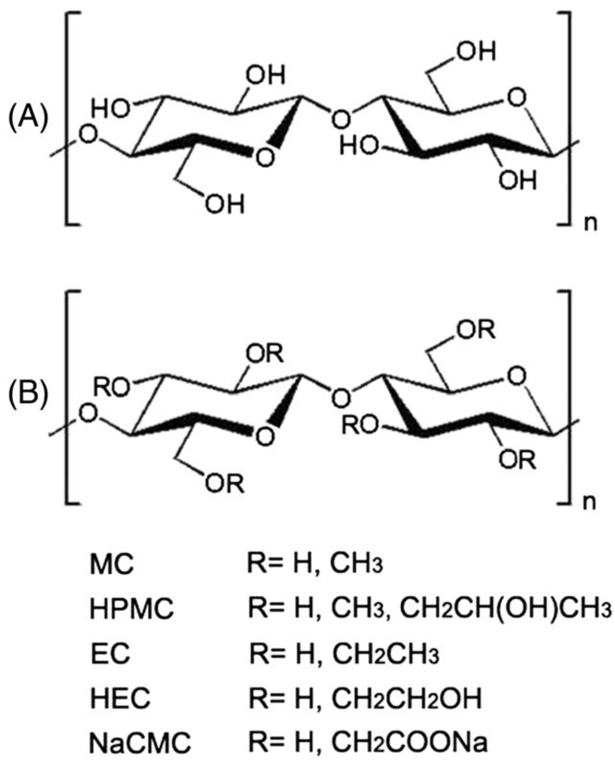

FIGURE 7 Cellulose-based hydrogels and cross-linking strategies.[19] HEC, hydroxyethyl cellulose; NaCMC, sodium carboxymethylcellulose; EC, Ethylcellulose; HPMC, hydroxypropyl methylcellulose; MC, Methylcellulose

- Polymerization

One of the best ways for fabrication of superabsorbent hydrogels is the solution polymerization due to its better control of the heat of polymerization, lower cost, and more convenient. Most of the cellulose-based superabsorbent hydrogels are produced in this way. Generally, the cellulose series macromolecular, monomer(s), initiator, and cross-linker(s) are freely soluble in water, or have good solubility in water. Once the initiator is induced by the temperature or the radiation, the polymerization process starts. After a certain time, the product of this reaction can be dried out and pulverized for various applications (Figure 8).[67]

Several mechanisms have been suggested to explain the synthesis of cellulose-based superabsorbent hydrogels. One of the mechanisms for solution polymerization synthesis of cellulose-based superabsorbent hydrogels is based on the free radical–induced polymerization. The free radical polymerization is a process in which monomers are polymerized through the initiation of initiators. This kind of polymerization has been used so extensively because it has high polymerization rate and happens in an aqueous medium, which is safe and harmless. The cellulose macromolecules produce the free radical initiated by the initiator, then interact with the monomers forming the graft copolymer. The main induce approach is chemical induce, containing mono-induce system,[68] bi-induce system,[69] and even ternary-induce system.[70] There are also other induce systems that are proven in different papers.

- Inverse-phase suspension polymerization

Inverse-phase suspension polymerization is conducted in the dispersed and continuous phases. The dispersed phase is aqueous, and the continuous phase is organic. The monomer is usually dissolved in the dispersed phase, and a surfactant is used to help the monomer and other aqueous reagents to be effectively dispersed throughout the continuous phase. Although particles with desirable sizes can be obtained by this technique, removal of the organic solvents, such as *n*-hexane and toluene, is a very challenging problem. This is one of the appropriate techniques for the polymerization of highly hydrophilic monomers, such as salts of AA and methacrylic acid and AM.[71] These significant results have been achieved by inducing a microporous structure in the hydrogel, by means of a phase inversion desiccation technique in acetone (ie, a nonsolvent for cellulose), which increased the water absorption, as well as the swelling kinetics, due to capillarity effects.[19]

- Microwave irradiation polymerization

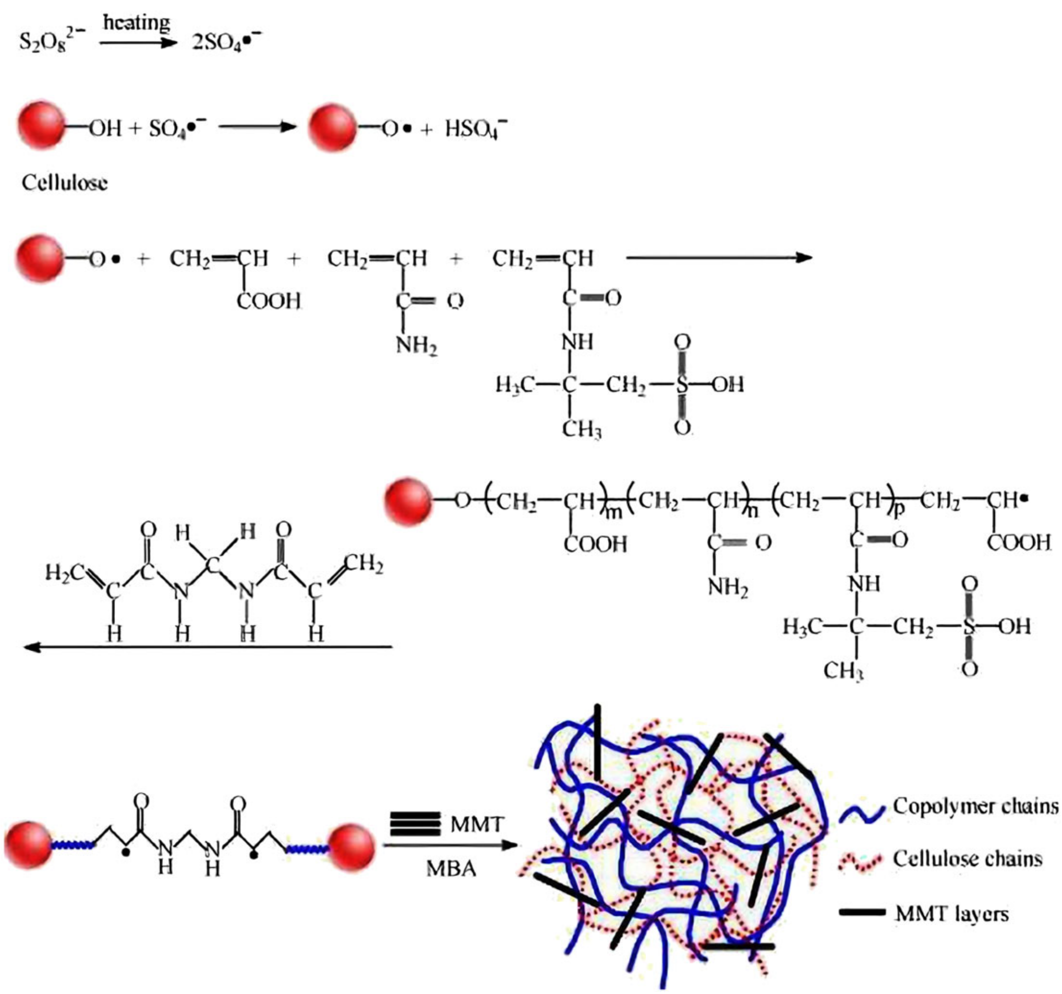

FIGURE 8 Mechanism for synthesis of cellulose superabsorbent hydrogel.[67] MMT, montmorillonite; MBA, methylenebisacrylamide

Microwave irradiation technology as an emerging polymerization skill, compared with the tradition approaches, displays stronger penetrating ability, faster heating, cleaner, and higher efficiency.

One of the promising methods for the cleaner production of cellulose-based superabsorbent hydrogels is using microwave irradiation polymerization. Giachi reported that the microwave-synthesized product possessed faster swelling and shrinking kinetics in comparison with the superabsorbent hydrogels prepared by conventional methods.[72]

8.2 | Applications of cellulose-based hydrogels

Applications of superabsorbent hydrogels can be divided to several groups including agriculture and horticulture, water treatment, biomedical, drug delivery systems, and personal health care. Since the focus of the review is on personal health care products, it will be discussed extensively.

A number of superabsorbent hydrogel products with cellulose based have been either available commercially or in the process of development. In addition, many patents about cellulose-based superabsorbent hydrogels have been granted for various possible applications.[69] In cases where biodegradability of a hydrogel is required or recommended, cellulosics are appealing hydrogel precursor materials, due to their low cost, the large availability and biocompatibility of cellulose, and the responsiveness of some cellulosics to variations of external stimuli.[62] Most of these used for agricultural and horticulture, personal health care field, water treatments, biomedical fields, and the stimuli-response smart behavior applications (Figure 9).[67] Additionally, many promising applications as protective barriers for volatile organic compounds spilled in the environment and as absorbents for waste oil had also been explored.

Superabsorbent hydrogels are widely used in the hygienic field like disposable diapers and female napkins for their ability to absorb and retain a large amount of secreted fluids, such as urine and blood. It was reported that the first generation commercial superabsorbent hydrogels were produced in Japan in 1978 as a component of female napkins. Then it was rapidly extended its market all over the world for the ability to retain the secreted liquids under pressure. In a word, superabsorbent hydrogels have caused a huge revolution in the personal health care industry. At present, superabsorbent hydrogels that contained in sanitary napkin are also primarily polymerized by AA or AM, which are costly, poorly degradable, and environmentally unfriendly. Liu et al provided novel tactics by incorporation of flax yarn waste into superabsorbent hydrogels for sanitary napkin applications.[73] Their results showed that the product exhibit excellent biodegradability, superabsorbent, and retention ability of artificial blood solution compared with those of the currently marketed sanitary napkin products.

Although more convenient, suitable, and comfortable disposable health care products have extensively been applied in modern time, biodegradable health care products have not either been industrialized or been commercially available. In view of the foregoing, the key technique of converting the cellulose-based superabsorbent hydrogels into the core layer of health care products needs to be broken through.[74,75]

Most of the cellulose-based hydrogels, which are used in health care products, have the same structure: an envelope of nonwoven tissue, a plastic cover material, and an absorbent fluff of wood pulp cellulose, mixed in most cases with one of the superabsorbent products. The basic idea of diaper recycling is to recover separately the cellulose, which is biodegradable and recyclable. The plastic cover material can also be recycled for some other applications.[19]

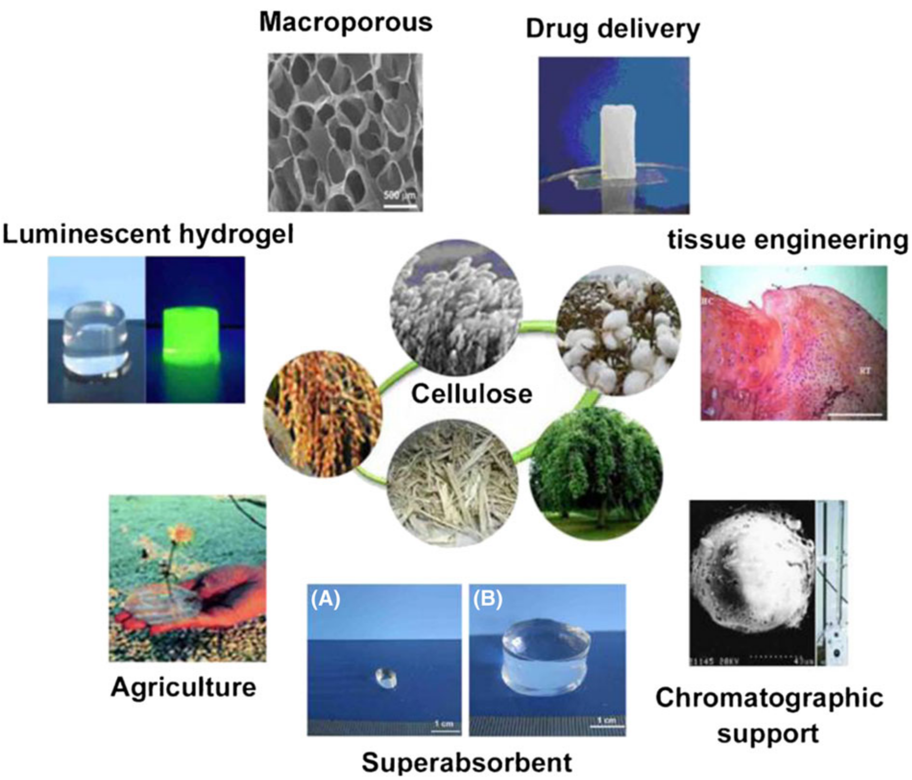

FIGURE 9 Applications and developments of cellulose-based hydrogels[66]

On the basis of the high water absorption properties of cellulose-based products, they have been used for the absorption of water and other aqueous fluids throughout recent history. Among such products, the paper towels, tissue papers, and diapers are some of the most widely used absorbents. For example, fluff pulps, which are generally based on kraft pulps, are commonly used for disposable diapers. Obtaining recyclable disposable diapers, napkins, hospital bed sheets, sanitary towels, and other products is one of the vital targets for the modern industry. Recently has been proposed an innovative solution to this problem, which involves the use of cellulose-based superabsorbent that are totally biodegradable.[76]

9 | FUTURE TRENDS AND CHALLENGES

A guide published by the Environmental Health Association of Nova Scotia cites a study reported in the "Archives of Environmental Health" that found that diapers release volatile organic chemicals. Toxic chemicals such as toluene, ethylbenzene, xylene, and dipentene have been linked to adverse health effects in humans with long-term exposure. The inner absorbent layer of a common disposable diaper (acrylate-based hydrogels) is also treated with chemicals, which can trigger allergic reactions.

Bio-based polymers still hold a tiny fraction of the total global hygienic products market. The worldwide interest in bio-based polymers has accelerated in recent years due to the desire and need to find non–fossil fuel-based polymers.

On the other hand, Because of the increasing attention for environmental protection issues, biodegradable hydrogels such as cellulose-based ones arise lively interest for potential commercial application in health care.

Cellulose-based superabsorbents offer important contributions in reducing the dependence on fossil fuels and through the related positive environmental impacts such as reduced carbon dioxide emissions.

Challenges that need to be addressed in the coming years include management of raw materials, performance of cellulose-based hydrogels, and their cost for production. Performance of cellulose-based superabsorbent products is well known, and it is relatively easy to replace the existing product with similar performance.

However, despite of these advancements, there are still some drawbacks, which prevent the wider commercialization of cellulose-based superabsorbent in personal care applications. This is mainly due to performance and price when compared with their conventional counterparts, which remains a significant challenge for cellulose-based personal care products.

10 | CONCLUSION

Increasing global demand for improved absorbent materials for personal care products creates an incentive for new basic research and development of efficient absorbent materials and systems with additional benefits such as high absorption capacity, biodegradability and biocompatibility. Therefore, new generation of personal care products based on the natural SAPs such as various types of polysaccharide and protein-based hydrogels was introduced. These materials have some benefits compared with the conventional polyacrylate superabsorbent, and they do not have any negative health and environmental effects. Because in the personal care articles such as baby diapers, feminine hygiene, and adults incontinence products, skin care has a special importance; using materials with no skin allergy symptoms such as rashes and inflammation is preferred.

Cellulose-based hydrogels are biocompatible and biodegradable materials, which show promising future for a number of industrial uses such as hygienic products, especially in cases where environmental issues are concerned. Several water-soluble cellulose derivatives can be used, singularly or in combination, to form hydrogel networks possessing specific properties in terms of swelling capability and sensitivity to external stimuli, which can be used in various health care products.

In this review, basic information about superabsorbent hygiene products was reviewed, and new superabsorbent materials based on the natural polymers, their properties, and applications in the personal care articles were introduced.

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