

Research Progress of eco-friendly Sugar-based Surfactants

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Abstract: Surfactant is a common industrial product widely used in chemical industry, food production, biomedicine and other fields. A variety of glucose-based bisubstituted surfactants have been developed, which not only contain the advantages of other classes of sugar-based surfactants, but also have the properties of mild conditions of use, good bactericidal ability, and non-irritation of the skin, etc. In addition, sugars are widely sourced and renewable, and the production of sugar-based surfactants with degradability has become a hot spot of the current research. This paper summarises the types of common sugar-based surfactants, the test methods of related properties and the application prospects.

Keywords: Surfactants; sugar-based surfactants; eco-friendly; performance testing.

1. Introduction

Surfactants are a class of chemical substances with both hydrophilic and hydrophobic groups in their molecular structure. Lowering the surface tension of a liquid is their primary property and they form an adsorbent layer at the liquid-gas or liquid-solid interface. The hydrophilic groups of surfactant molecules interact with water molecules, while the hydrophobic groups interact with non-polar substances.

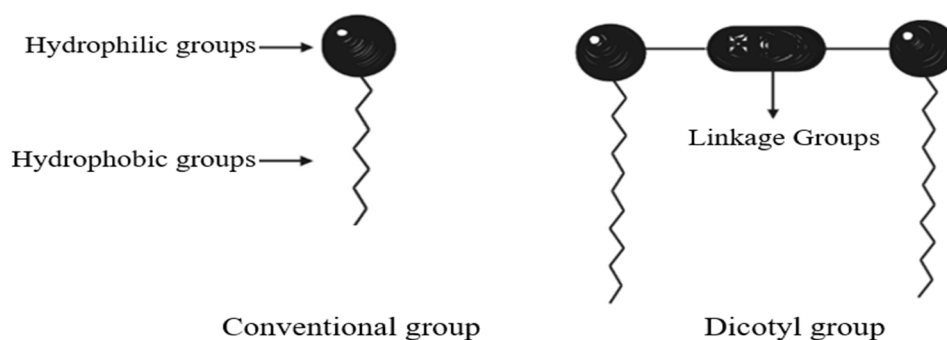


Figure 1. Comparison between Traditional Surfactants and Gemini Surfactants

In surfactants[2] Sugar-based surfactants have many advantages. Introducing sugar groups as hydrophilic groups into anionic surfactants not only improves their toxicity and irritation, but also makes them biodegradable and more ecological and environmentally friendly. Good solubility and emulsification are also conducive to the formation of a stable solution system. At the same time, the sugar base is composed of natural polysaccharides or synthetic sugar alcohols, which are renewable resources and therefore have high biodegradability. Sugar-based surfactants can be degraded in the natural environment through the action of microorganisms; microorganisms can use sugar-based surfactants as a carbon source for metabolic enzyme degradation to break them down into harmless small molecules, such as carbon dioxide, water and non-toxic compounds, which can effectively reduce their accumulation in the environment through natural degradation pathways. Sugar-based surfactants are usually highly biocompatible and biodegradable, and are therefore safer for humans, animals and plants, and are considered to be an

environmentally friendly alternative.

According to the ions coming out of the shop of hydrophilic groups in aqueous solution can be classified as anionic, cationic, amphoteric and nonionic surfactants. Amphoteric surfactants[1] It is a new type of surfactant, which is mainly achieved by the formation of chemical bonds between the linking group and the hydrophilic group. Baryonic surfactants have stronger surface activity than conventional surfactants, and its structure comparison with conventional type surfactants is shown in Fig. 1.

environmentally friendly alternative.

2. Classification of Sugar-based Surfactants

Based on the chemical structure and properties of sugar-based surfactants, we can classify them into nonionic, cationic, anionic and amphoteric types according to the type of charge after dissociation in aqueous solution. At present, a variety of new sugar-based surfactants with different functions have been developed at home and abroad. Among them, the first three are widely used.

2.1. Sugar-based nonionic surfactants

Alkyl glycoside-type surfactants, advanced fatty acid ester-type surfactants, and glycosyl-linked chain surfactants are common sugar-based nonionic surfactants in our daily life.

Alkylglycoside (APG) surfactants, as key precursors for the synthesis of sugar-based surfactants, are conventional single-head-based surfactants prepared by utilising natural

renewable resources such as sugars and vegetable oils as the basic raw materials. The study of alkyl glycosides as a base for surfactants has received extensive attention worldwide. Currently, many alkyl glycoside derivatives have been developed for pharmaceutical applications. As an example, C8-C12 alkyl glycosides have shown strong antimicrobial activity against many bacteria and fungi, which makes it a potential for a wide range of applications in medical disinfection and cleaning processes.

In recent years, fatty acid glycol esters have been studied a

lot and achieved certain results by researchers because of their better degradability and so on. Wang Kui[3] and his team successfully prepared a charcoal-based solid base catalyst K2O/C by vacuum impregnation with the assistance of ultrasonic waves, and then they let it catalyse the reaction between fibre oligosaccharides and methyl oleate to synthesize the fibre oligosaccharide fatty acid ester surfactant. The transesterification reaction mechanism of fibre oligosaccharides with methyl oleate is shown in Figure 2 below:

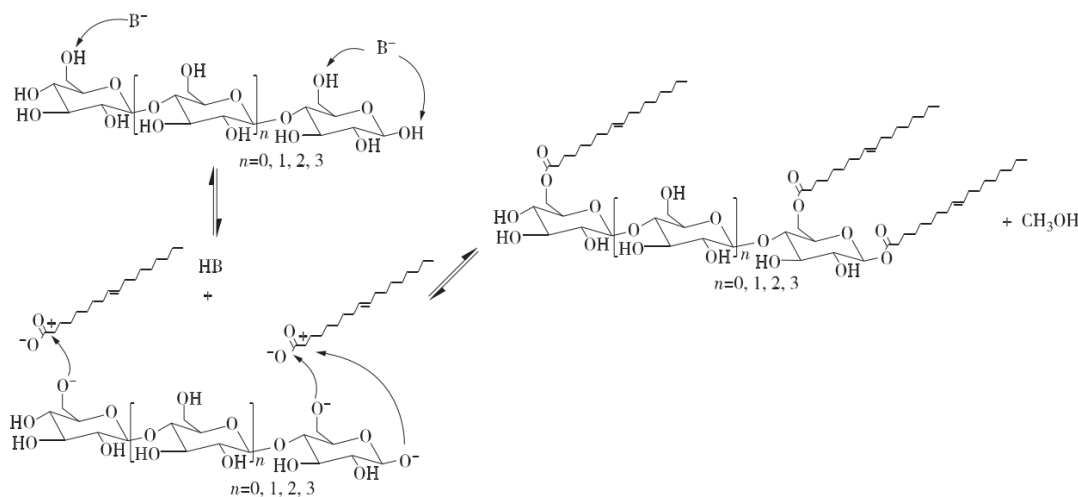


Figure 2. Mechanism of ester exchange reaction between fibre oligosaccharides and methyl oleate

Yanmei Ren^[4] et al. synthesised glucose laurate ethylene glycol maleate (GME), they used glucose, ethylene glycol, acid anhydride and lauric acid as the raw materials to combine the reaction to obtain the product: glucose laurate ethylene glycol maleate (GME) in a vacuum environment at 120 °C with phosphoric acid as the catalyst. The crude product obtained was separated and purified by chromatographic column separation, and the final product could be obtained with a yield of up to 76.67%. Measurement of the properties showed that the CMC of the GME solution was $5.77 \times 10^{-3} \text{ mol} \cdot \text{L}^{-1}$ at 20 °C, corresponding to a surface tension of 23.6 $\text{mN} \cdot \text{m}^{-1}$. Characterisation of the synthesised target compounds showed that the GME bis(maleic acid) surfactant has a significant ability to reduce surface tension and meets the "green chemistry" criteria. The results of the characterisation of the synthesised target compounds showed

that the GME bidentate surfactants have a significant ability to reduce surface tension and meet the "Green Chemistry" criteria, mainly due to the presence of degradable sugar structures in the products.

2.2. Sugar-based cationic surfactants

Sugar-based cationic surfactants can be classified into monosaccharide cationic surfactants, disaccharide cationic surfactants, and polysaccharide cationic surfactants.

Among cationic surfactants, quaternary ammonium bisglycosyl surfactants are widely used in various fields due to their high activity, high charge of cation and good biodegradability. Nai-Ni Guo^[5] et al. synthesised a novel cationic surfactant by using trimethylamine, epichlorohydrin, lauryl alcohol and stearic acid as raw materials under ultrasonic conditions and the synthetic route is shown below in Figure 3:

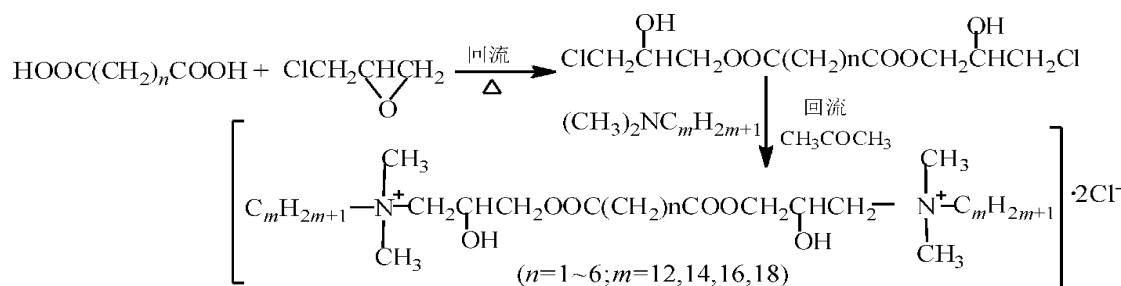


Figure 3. Synthesis method of ester bond quaternary ammonium salt Gemini surfactant

The main groups in disaccharide cationic surfactants are lactose and alginose. Menger et al. synthesised two different properties of Gemini surfactants using alginose as the raw material, one is a non-ionic surfactant with poor water

solubility; the other is a cationic surfactant with better solubility, which is more prone to form micelles and has a larger critical micellar concentration than the conventional surfactants. Although both surfactants use alginate as a

linking group, they introduce different groups, the latter one mainly introduces cation as a group to make it better soluble in water; therefore, there is a difference in the colloidal

behaviour observed in the two series, which results in the difference in water solubility. The general structural formula of alginate is shown below in Figure 4:

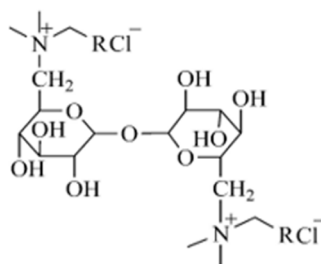


Figure 4. The structural formula of trehalose cationic surfactant

Polysaccharide cationic surfactants are mainly chitosan-based surfactants. Pei Lijun^[6] et al. synthesised a new type of chitosan derivatives, dehydrofiryamine-chitosan cationic surfactants, using chitosan and dehydrofiryamine with different relative molecular masses as raw materials. The

results of their properties were as follows: at 25 °C, the CMC of dehydrofiryamine-chitosan cationic surfactant was 39.8 mmol/L, the surface tension corresponding to CMC was 38.6 mN/m, and the emulsification time was 44 s. The synthetic route is shown in Fig. 5 below:

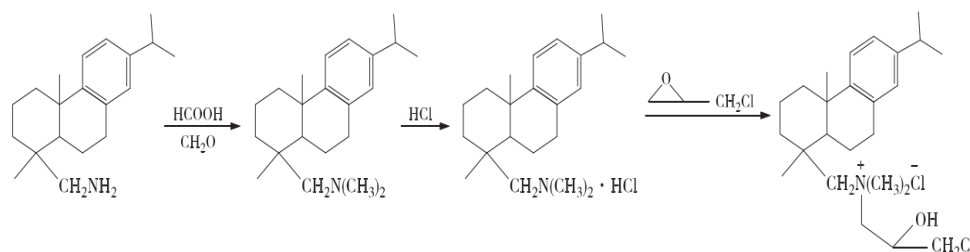


Figure 5. Synthesis route of dehydroabietylamine chitosan cationic surfactant

Zhan Yanhui et al.^[7] Magnetic chitosan/zeolite was prepared and mixed with cationic surfactant solution using magnetic stirring in a constant temperature water bath to obtain a new adsorbent. Magnetic chitosan/cationic surfactant modified zeolite adsorbent has the advantages of high utilisation rate, good separation effect, and economic durability.

2.3. Sugar-based anionic surfactants

Sugar-based anionic surfactants are a class of sugar-based anionic surfactants with good biocompatibility and environmental friendliness. The research on sugar-based anionic surfactants has received extensive attention in recent years and a series of important progress has been achieved.

Pang Shujing^[8] et al. synthesised a rosin-based anionic

surfactant, sodium 6-dehydrofiramido hexanoate (R-6), obtained using a series of reactions with dehydrofiry acid (DA) and 6-amino hexanoic acid as the raw materials. The results of the performance of R-6 showed that: at room temperature, the solubility of R-6 in water was excellent, and the solubility reached more than 800 mmol/L; the critical micelle concentration (CMC) of R-6 was 5.89 mmol/L, and the corresponding surface tension (γ) was 40.46 mN/m. The concentration of R-6 had a great influence on the foaming property, and the foam half-life was as high as 1966 min at a concentration of 10 mmol/L. The concentration of R-6 had a great influence on the foaming property. of R-6 is shown in Figure 6 below:

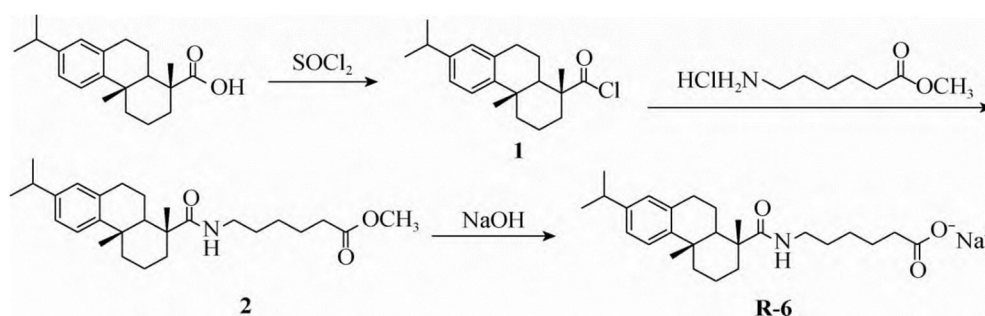


Figure 6. Synthesis route of R-6

Sugar-based anionic surfactants have a broader market potential^[9]. Its main advantages include: sugar-based anionic

surfactants have good solubility in water, can quickly form a stable solution system, easy to use and handle; can effectively

emulsify and disperse oily substances, improve the stability of emulsions and dispersions, so that it is uniformly distributed in the water; can enhance the solubility of other substances in the water, improve their activity and effect, in cosmetics, detergents and other fields by the It is widely concerned; it is mild and less irritating to the skin and eyes, suitable for the cleaning and maintenance of sensitive skin and the area around the eyes; at the same time, sugar-based anionic surfactants can be degraded by microorganisms, which has less impact on the environment and meets the requirements of green development of the country, and therefore it is widely used in the field of green chemistry.

3. Properties and Test Methods of Sugar-based Surfactants

Hydrophilicity and lipophilicity are the main properties of surfactants, in addition to this, sugar-based surfactants have many other properties which have a greater impact on the use of surfactants.

3.1. Critical micelle concentration (CMC)

At low surfactant concentrations, surfactant molecules

exist as monomers in solution and intermolecular interactions are relatively weak. As the concentration increases, the surfactant molecules begin to interact and form aggregates, or micelles, through hydrophobic and hydrophilic interactions. The critical micelle concentration is the process in which the surfactant concentration in solution reaches the lowest value that can form micelles, referred to as the CMC value. At this concentration, a series of physicochemical properties of the solution will change dramatically with the concentration near the critical micelle concentration, as shown in Figure 7.^[10] Using this property, we can measure its conductivity or surface tension to measure the CMC of the surfactant. critical micelle concentration is often used as a measure of the surface activity of the surfactant, because a small CMC value indicates that the lower the concentration of the surfactant required to form micelles, the lower the concentration required to reach the surface (interfacial) saturated adsorption and thus change the surface properties to play a role in wetting, emulsification, solubilisation and foaming, the lower the concentration required. The lower the concentration required to change the surface properties to wetting, emulsifying, solubilising and foaming, etc.^[11] The lower the concentration required to form micelles.

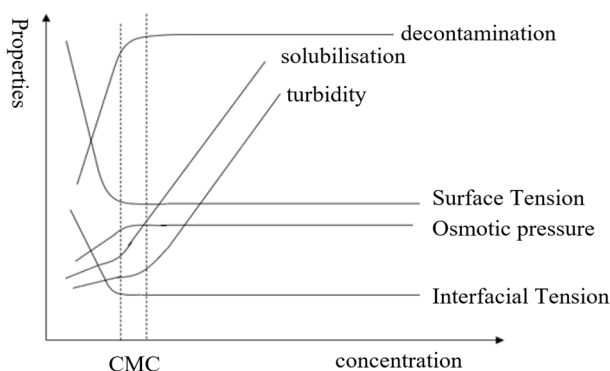


Figure 7. The variation of various properties of surfactants with concentration

Conductivity method: The magnitude of the conductivity of a surfactant solution depends on the concentration of all ions of the surfactant dissolved as single molecules in water. Ions and counter sign ions in the solution produce conductivity due to electrophoresis at the poles. Below the CMC value, the concentration of surfactant increases with the increase of surfactant and hence the conductivity undergoes a linear increase with the increase of surfactant concentration. After reaching the CMC value, the increase in conductivity with increasing surfactant concentration decreases due to the

decrease in the charged amount and the decrease in electrophoretic speed. If a conductivity meter is used to determine the conductivity at different surfactant concentrations, Fig. 8 can be obtained, and the concentration corresponding to the intersection of the two straight lines with different slopes of the conductivity curve is the CMC value of the surfactant.^[10] The concentration corresponding to the different intersection of the two slopes of the conductivity curve is the CMC value of surfactant.

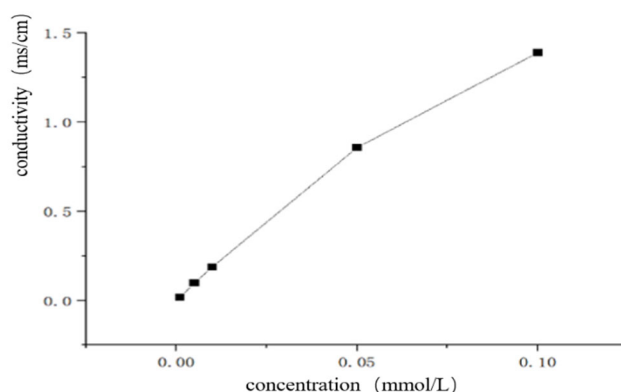


Figure 8. The conductivity of sugar based surfactants at different concentrations

Surface tension method: Increasing the concentration of surfactant in aqueous solution, the surface tension will decrease sharply around the CMC value. The surface tension at different surfactant concentrations is determined by interfacial tensiometer and plotted, and the curve shown in

Fig. 9 will be obtained, and the concentration corresponding to the sudden decrease in the curve is the CMC value of the surfactant.^[10] The concentration at which the curve suddenly decreases is the CMC value of the surfactant.

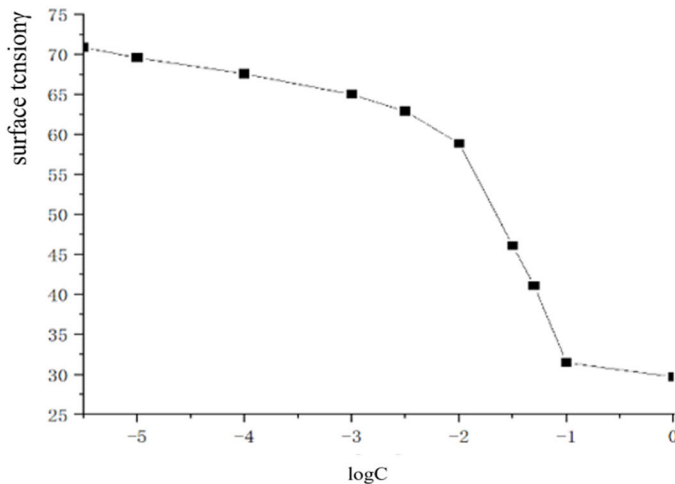


Figure 9. Surface tension of sugar based surfactants at different concentrations

3.2. HLB values

HLB (Hydrophilic-Lipophilic Balance) value is an indicator used to assess the balance between hydrophilic and hydrophobic properties of surfactants. It represents the relative proportion of hydrophilic and hydrophobic parts in

the surfactant molecule. different surfactants have different HLB values, and the HLB value of surfactants generally ranges from 1 to 40. the smaller the HLB value, the stronger and larger the adsorption of oil; the larger the HLB value, the larger the affinity with water. the reference role of HLB value^[10] See Table 1:

Table 1. Reference function of HLB value of surfactants

HLB value	reference role
1.5 to 3	defoaming action
3.5-6	Emulsification (W/O)
7 to 9	wettability
8 to 18	Emulsification (O/W)
13-15	washing effect
15-18	solubilisation

Critical Micelle Concentration (CMC) method is a common method to determine the HLB value: the HLB value is determined based on the chemical structure of the surfactant molecule and the number, size and relative position of the hydrophilic and hydrophobic groups. Therefore, the

HLB value of the surfactant can be calculated by determining the CMC value, and the HLB value of the surfactant can be calculated by the formula. The formula of the correlation between HLB and CMC is shown in Table 2:

Table 2. Related formulas for calculating HLB of surfactants

serial number	formula	Scope of application
1	$HLB = 7 + 4.02 \log (1/[CMC])$	Nonionic surfactants
2	$HLB = A \times \log CMC + B$	Anionic surfactants
3	$HLB = 1.362 \log [CMC] + 22.189$	Fluorocarbon anionic surfactants

3.3. Foaming

Surfactant foamability is the ability of a surfactant to generate stable foam when in contact with air. Surfactant solutions, when in solution, can form an adsorbent layer of surfactant that adsorbs to the surface of the liquid. This adsorbent layer reduces the surface tension of the liquid

surface and forms an elastic film that surrounds the gases, resulting in a stable foam structure.^[12] The foam structure is stabilised by forming an elastic film around the gas. Generally, the oscillation method is used^[10] Foaming and foam stabilisation test of the product: Prepare a surfactant solution with a mass fraction of 0.1% and place it in a constant

temperature water bath at 25 °C for 2 h. Take 20 mL of the solution in a 100 mL stoppered cylinder, cover the stopper, shake it vigorously up and down for 20 times, place it on a flat surface for static, and then observe and record the volume of the foam in different moments, V. The larger the volume, V, the stronger the ability of surfactant to reduce the surface tension and the greater the ability of surface tension to be reduced. The larger the volume V, the greater the ability of the surfactant to reduce surface tension, the more favourable the formation of foam and the better the performance of the surfactant.

3.4. Emulsification properties

Fats and oils layering on contact with water is due to the fact that fats and oils have a high surface tension on water, so when stirred vigorously with a glass, they are crushed into fine beads and then mixed to form an emulsion, but then re-layering occurs when the stirring stops. Emulsification is the action of adding surfactant when two solutions are layered due to surface tension and do not layered for a very long time^[10] Emulsification. The emulsifying property of surfactant is determined by its molecular structure and nature. By interacting with water and oil, it forms a film and micelle structure that reduces the interfacial tension, thus achieving the dispersion and stabilisation of grease particles. Determination of emulsification performance is generally used paraffin oil method: under the condition of 25 °C, the first preparation of the mass fraction of 0.1% surfactant, remove 20 mL in 100 mL of the cylinder with a stopper, then add an equal volume of liquid paraffin oil to the cylinder, cover the stopper tightly, and shake the cylinder up and down violently for 20 times, and then stop the cylinder will be static, and then immediately start the timekeeping, recording the time to be separated from the cylinder to 10 mL of the aqueous phase. Record the time used when the cylinder is divided into 10 mL of aqueous phase. The longer the emulsification time, the higher the emulsification performance of the surfactant.

3.5. Krafft Point

The temperature at which an ionic surfactant reaches a critical micelle concentration is called the Krafft Point.^[9] It is mainly used to characterise ionic surfactants. Generally speaking the higher the Krafft Point, the smaller the critical micelle concentration. Krafft Point directly reflects the solubility of surfactants. Generally the higher the Krafft point, the worse the solubility of the surfactant^[13] The higher the Krafft point, the poorer the solubility of the surfactant.

The specific test procedure of Krafft point is as follows: firstly, prepare 100 mL of 1.0% surfactant, secondly, measure 25 mL of aqueous solution in a 100 mL beaker, finally, insert a thermometer into the beaker and place it in ice water for slow cooling, and when crystals precipitate out of the aqueous solution, read the temperature of the moment immediately, which is the Krafft point. Repeatedly measured 3 times, take the average temperature is the Krafft point of the ionic surfactant.

4. Application of Sugar-based Surfactants

Sugars are one of the typical representatives of biological raw materials, which are prepared from starch materials with simple preparation process, wide range of raw materials and low price.^[14] Sugar-based surfactants have hydrophilic effect,

which also gives the surfactant more excellent performance: (1) It gives the surfactant good biodegradability and toxicological properties, and non-toxic and harmless, gentle to human skin without any stimulation; whether or not oxygen, it is very easy to biodegrade. (2) Sugar group contains a number of hydroxyl groups, so it has a stronger hydrophobic and hydrophilic properties. Therefore, in the oil-water mixing system, sugar-based surfactants have more excellent interfacial chemical properties. (3) When the sugar-based surfactant is in contact with water, its sugar group will form complex ions with good solubility with metal ions in water, so that the sugar-based surfactant has better resistance to hard water. (4) Sugar-based surfactants can also be used in agricultural applications such as pest control, weed control and inhibition of bacterial activity. The synthesis of glycosyl surfactants may greatly improve their irritation, toxicological properties, biodegradability, hydrophilicity and other properties, and the diversity of groups leads to a wide variety of glycosyl surfactants. Due to the diversity of groups, there is a wide range of sugar-based surfactants. Also, due to the different sugar groups, most of the surfactants are green, easy to degrade, harmless to human body and other excellent properties, so the sugar-based surfactants have a wide range of prospects for application in many fields.^[15] Therefore, sugar-based surfactants have a wide range of application prospects in many fields.

4.1. Sugar-based surfactants in detergents

Sugar-based surfactants are compounds consisting of sugars and hydrophobic groups, and are often used as one of the important ingredients in cleaning products because they have good oil-water separation properties and degreasing and cleaning abilities. In the field of care products and detergents, sugar-based surfactants can effectively remove grease and dirt and make them dispersed in water, have excellent surface activation and emulsification properties, and have very strong cleaning ability to effectively remove grease or stains.

For example, alkyl glucoside, which has the advantages of common nonionic and anionic surfactants, has glucose residues as hydrophilic groups in the amphiphilic structure of surfactants, so it has excellent surface activation, rich foam, strong detergency and other properties, and is widely used in household cleaning and cosmetic production.^[16] Alkyl glucosides are widely used in the fields of household cleaning and cosmetic production, as well as detergents, oral hygiene cleaners, bactericides, enzyme detergents and many other fields, because of their excellent surface activation, rich foam, and strong decontamination ability. Alkyl glucosamine surfactants have mild and low irritation properties, and in different applications can show delicate and long-lasting foam, improve the stability of formulations, high penetration, and do not leave traces, etc., which has a broad application prospect in baby wash formulations.^[17] Oleyl glucosamine was used in 2016 by foreign chemical companies in the field of handmade dishwashing, home wash and care^[18].

4.2. Application of sugar-based surfactants in food production

Sugar-based surfactants also have many applications in the food industry. They are often used as emulsifiers, stabilisers, thickeners, etc., and are widely used to improve the texture, mouth feel and stability of foods. Sugar-based surfactants usually consist of a hydrophilic sugar group and a hydrophobic alkyl carbon chain, and their properties can be

adjusted by modifying the sugar headgroup or the non-polar tail chain, and this structural diversity makes them have a great potential for application in the food industry.[19] This structural diversity gives it a great potential for application in the food industry.

Sugar-based surfactants have a wide range of applications in the food industry due to their good biocompatibility and environmental friendliness. They can replace traditional synthetic surfactants, reduce the adverse effects on the environment and human body, and contribute to the sustainable development of the food industry.

4.3. Sugar-based surfactants in biomedical applications

Because of their biocompatibility and environmental friendliness, sugar-based surfactants are often used in drug delivery systems, preparation of medical materials and research in the biomedical field. Sugar esters have excellent biosafety and biodegradability, and they can be converted to sugar and fatty acid residues by pancreatic lipase in vivo, so they have been widely used in the field of medicine^[20]. Alkyl glycosides are used in the pharmaceutical industry due to their advantages of surfactants, low toxicity, higher biodegradability and low production costs^[21]. Alkyl Glycosides

In drug delivery systems, sugar-based surfactants can be used to prepare nanocarriers to encapsulate and deliver drugs to the body where they are needed, which greatly improves the bioavailability of drugs and reduces toxicity while dramatically improving stability. In addition, sugar-based surfactants can also be used in the fields of biomarker detection and gene delivery, to improve the performance of biomedical materials, and to prepare degradable medical materials.

Glycoside-based surfactants are also often used in the preparation of medical devices and medical materials to improve the surface properties of the materials and to enhance the biocompatibility and degradability of the materials, thereby reducing adverse effects on the human body. Alkyl Glycoside Surfactants Function as drug carriers in pharmaceutical applications.

5. Prospect

Currently, sugar-based surfactants are widely used in various fields due to their high biocompatibility and biodegradability^[22]. The design and synthesis of some sugar-based surfactants with excellent performance and novel structure to expand their structure type and application range is the main development direction of the current research and application of sugar-based surfactants.

As the preparation process of surfactants is too complicated and long, and the production cost is too high, so in recent years, scientists are committed to the use of simple preparation methods and efficient and convenient catalytic system to synthesise surfactants, and research on more concise and economical preparation process to promote the industrial production of sugar-based surfactants. At the same time, some scientists have prepared various mixed products with different effects according to the characteristics of surfactants and different additives, which greatly increase the synergistic effect between different surfactants, effectively improve the economic efficiency, reduce the cost of industrial production, and improve the strong favourable conditions for

the market application of sugar-based surfactants. However, in the actual production and preparation process, sugar-based surfactants are also limited by the reaction conditions, and the manufacturing cost is expensive, so it is necessary to further explore a more economical and efficient synthesis route. Although sugar-based surfactants have good physiological activity, bio-greenness and biodegradability, and are widely used in industry, agriculture and people's daily life, they need to be further expanded in new fields.

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