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Synthesis and Prospect of Amphoteric Surfactants

Jianhua Zhao[a], Lijing Pan[b], and Yang Yang

College of Engineering, Suthwest Petroleum University, Nanchong, Sichuan, 637000, China

[a]191086593@qq.com, [b]1806551655@qq.com

Abstract

With the deepening of oil exploration and development, the difficulty of exploitation is gradually increasing. For secondary oil recovery failed to produce residual oil, adding surfactant to chemical flooding is an important technical means to improve oil recovery. However, most of the existing clean fracturing fluids use single-chain surfactant as thickener, which has poor temperature resistance and is limited in use. To solve this problem, according to the different molecular structures of amphoteric surfactants, the amphoteric surfactants synthesized in recent years can be divided into two types : single-chain type and double-chain type. Amphoteric surfactants are more closely arranged between molecules, more healthy and environmentally friendly, biodegradable, and have broad research space. The synthesis and performance application of zwitterionic Gemini surfactants were emphatically introduced. Finally, the shortcomings of the current research on amphoteric surfactants were pointed out, and the future research direction and focus were prospected.

Keywords

Amphoteric Surfactant; Synthesis; Performance; Progress.

1. Introduction

Amphoteric surfactants are surfactants consisting of long-chain hydrophobic alkyl groups and single- or double-stranded molecules that have both electrically hydrophilic groups. Due to its polar head group with two different charge properties, the interaction repulsion between the hydrophilic head groups is smaller than other anionic types, cationic types, and the active agent molecules are more closely arranged between the molecules, and this tissue structure makes it have a variety of different functional characteristics compared with traditional surfactants, and the excellent surface chemistry makes it have better practical application performance, such as reducing the water-oil interface activity, antifouling, anti-corrosion, wetting, sterilization, antistatic, etc. The Gemini surfactant molecule is produced by the combination of amphoteric surfactants and Gemini surfactants, which are composed of two single-chain amphoteric surfactants on the head base or near the head group position by connecting groups together, the use of connecting groups can be a variety of action groups or rigid or flexible [1].This unique molecular structure makes it have a unique electron density, reduce the aqueous solution surface interface tension value, higher surface interface activity, can form micelles at lower concentrations, the value of Krafft point is also lower, pH dependence behavior, calcium soap dispersion performance is excellent, etc., compared to single-chain amphoteric surfactants to the human body is low harm [2], and more healthy and environmentally friendly, easy to biodegradable, has a broad development research space, market prospects. It can be applied to people's daily production and life, industrial chemical industry, agricultural production, especially in some special industrial production areas.

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2. Single-chain Amphoteric Surfactant

People's application of surfactants as early as the 8th century BC, which has been recorded in historical records, the working people of China used grass and wood ash to wash clothes, and cleaned them with soap horns and pig pancreas. At the beginning of the 20th century, modern scholars began to conduct scientific research on the synthesis and properties of amphoteric surfactants from a chemical point of view. The Bondi S[3] study found that amino acids and fatty acids and their derivatives react to synthesize a single-stranded, two-electric surfactant. The classification of single-stranded amphoteric surfactants can be classified according to the different types of cations after the dissociation of hydrophilic groups, which can be roughly divided into four categories:

2.1 Imidazoline Type Amphoteric Surfactants

Imidazoline type amphoteric surfactants are classified according to the different hydrophilic anionic structures, which can be roughly divided into amide type, sulfonic acid type, alkyl hydroxyethyl type and carboxylic acid type, etc., which have excellent performance, high efficiency surface activity, low irritation, low toxicity and non-toxicity, and excellent biochemical degradability, often added to detergents, corrosion inhibitors and rust removers and other formulations.

Hua Man et al.[4], the hydroxyethyl ethylenediamine, lauric acid in 30% sodium hydroxide solution 150 ℃ reaction 5h ~ 10h to obtain intermediate products, continue to react with acrylic acid or its derivatives, control the inverse time of 3h ~ 8h, temperature 65 ℃~ 85 ℃, and then add NaOH aqueous solution hydrolysis can obtain a light yellow unsalted imidazoline products, the reaction needs to be carried out in an anaerobic environment, by increasing the proportion of acrylic derivatives in ingredients to study the change of product conversion rate, When the amount ratio of the substance is 2:1 acrylic derivative and intermediate reactant, the conversion rate of the product can be as high as 96%, Zheng Shue et al. [5], using a similar way to synthesize a new type of imidazoline laurate amphoteric surfactant, adding sodium 1 chloroacetate: 2 imidazoline laureate intermediate, control the reaction temperature at 85 °C, with NaOH as the catalyst, Controlling the PH at 7.5-8.5, gradually adding sodium chloroacetate dropwise for quaternary ammonium reaction, the reaction time is 4h, and the final synthetic surfactant has superior antistatic properties, and its specific resistance value is 7.45 MΩ·cm. The reaction equation is shown in Figure1:

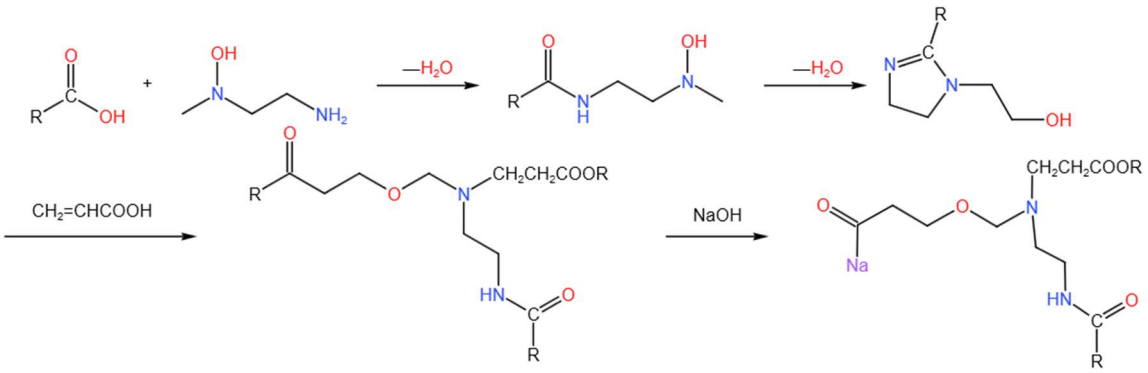

Figure 1. Synthesis of an imidazoline

Jiao Xiaofang [6], with diethylenetriamine, lauric acid and sodium chloroacetate as reactants, to study the synthesis path of a variety of different imidazoline amphoteric surfactants, in the vacuum of 0.07MPa environment, the amount ratio of the added substance lauric acid and diethylenetriamine acylating reaction, the reaction time is 2h, the reaction temperature is 140 °C -160 °C, the temperature is raised to 200 °C -210 °C, so that the product undergoes a cyclization reaction, about 4h after the end of the reaction. The reaction equation is shown in Figure2:

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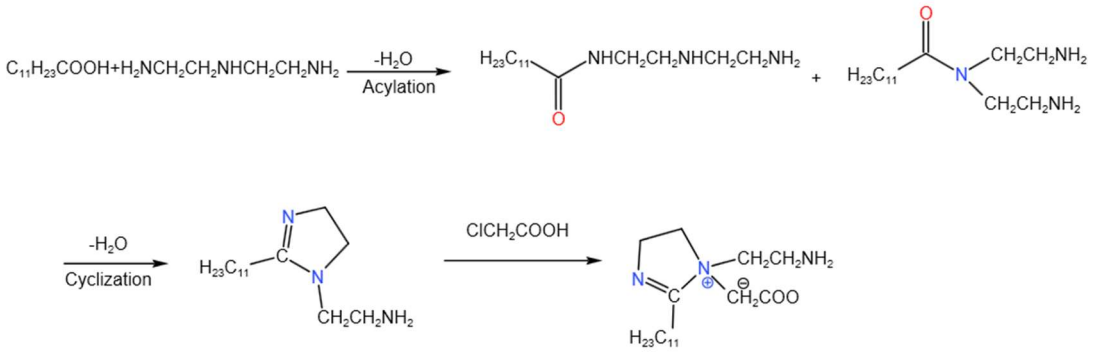

Figure 2. Synthesis of imidazoline amphoteric Surfactant

Guan Huilei [7], using fatty acids, organic amines and sodium chloroacetate as ingredients, divides the synthesis route of amphoteric imidazoline sodium acetate surfactant into two steps, first reacting hydroxyethyl ethylenediamine or diethylenetriamine with fatty acids or fatty acid derivatives to synthesize imidazoline intermediates, and then reacting imidazoline intermediates with sodium chloroacetate to synthesize imidazoline amphotericacetate. Under different reaction conditions, the reaction will generate 3 intermediates and 9 products with different structures, which makes the viscosity of different batches of products vary greatly and the quality is not easy to control. One of the reaction equations is shown in Figure3, and the product causes a high viscosity:

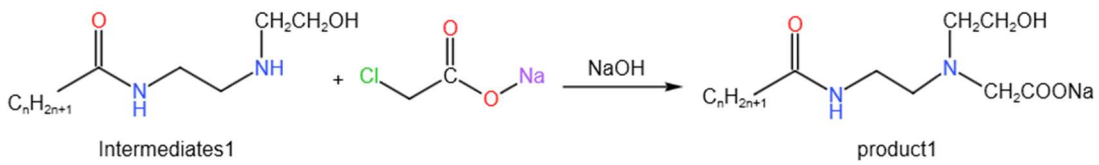

Figure 3. Synthesis of imidazoline amphoteric sodium acetate

2.2 Betaine Type Amphoteric Surfactant

Betaine type amphoteric surfactants are mainly divided into four types: betaine sulfate, sulfonyl betaine, carboxylic acid betaine and phosphate betaine surfactants. The acid and alkali resistance of this type of surfactant is more prominent, the dissolution performance is excellent, the decontamination ability is strong, the static generation can be reduced, the surface penetration is stronger, and it is often used as the main ingredient of antistatic agent in life, and added to the fungicide, shampoo and other personal care products.

Ding Wei et al. [8], with dimethylamine, epichlorohydrin, nonylphenol and sodium 3-chloro-2-hydroxypropanesulfonate as the reactants, at 85 °C, pH = 8 ~ 9, the epichlorohydrin oxide, nonylphenol through the action of catalysts for etherification reaction synthesis in the middle, and then added dimethylamine in the NaOH alkaline environment, producing tertiary amination reaction, the intermediate product and 3-chloro-2-hydroxypropanesulfonate sodium quaternary ammonium co-reaction, a total reaction of 24h, and finally the synthesis of a nonylphenol betaine amphoteric surfactant, The results show that the product has good salinity resistance and temperature resistance. Compared with other synthesis reactions at this stage, the reaction time of this synthesis route is less, the operation is simple, the time is shorter and the yield is high. Its synthesis route is shown in Figure4:

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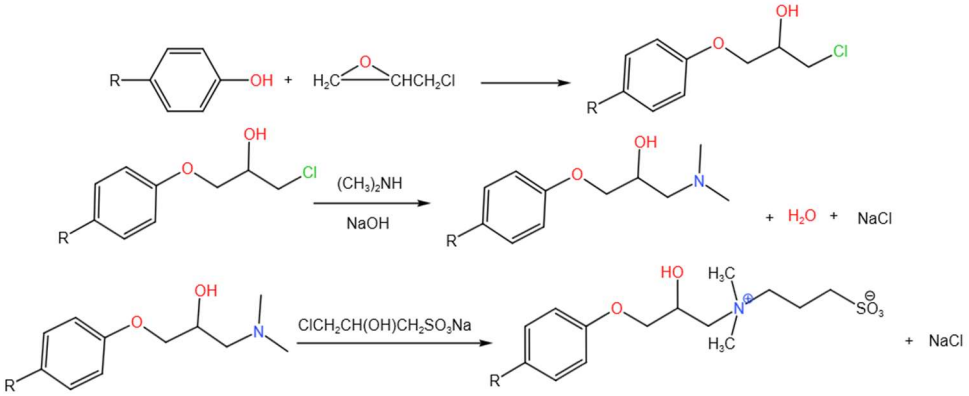

Figure 4. Synthesis of amphoteric Surfactant of Nonylphenol Betaine

Li Shujie [9], using fatty alcohols (dodecyl alcohol, tetradecyl alcohol, hexadecanol), sodium 3-chloro-2-hydroxypropanesulfonate and epichlorohydrin as ingredients, synthesized alkyl ether betaine amphoteric surfactants in three steps. The first step is to dehydrate and condense fatty alcohol and epichlorohydrin under alkaline conditions to obtain alkoxy glycidyl ether, and then add methylamine, and then synthesize alkoxy tertiary amine, and finally prepare an alkyl ether betaine series system by sodium 3-chloro-2-hydroxypropanesulfonate and tertiary amine. The synthesis route is shown in Figure5:

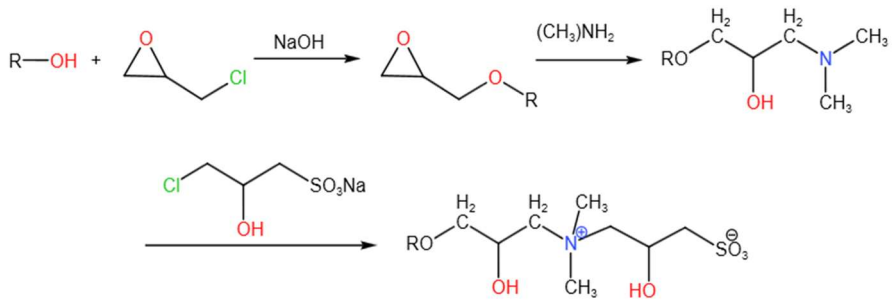

Figure 5. Synthesis of Alkyl Ether Betaine

Zhang Jiayu et al. [10], using dehydroabitaic acid extracted from a class of natural products in rosin as the main ingredient, after a series of processing to produce a new type of rosin-sulfo betaine surfactant, the product has a strong interface adsorption capacity and aggregation capacity, solubility of 2.30mmol/L, critical micelle concentration of 1.26mmol/L, maximum adsorption capacity of 2.51μmol/m2. The compounded mixing system was mixed with three kinds of crude oil (density 0.845g/cm3) in Daqing Oilfield, and it was found that the oil-water interface tension could be reduced, and the minimum could reach 0.0003mN/m. The synthesis route is shown in Figure6:

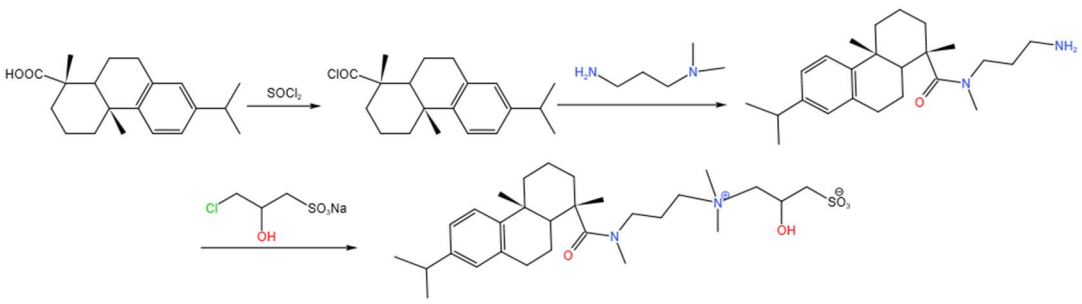

Figure 6. Synthesis of rosin-based Sulfobetaine Surfactant

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Wang Xinrui et al. [11], N,N-dimethyl-1,3-propanediamine and palmitic acid in the NaOH alkaline environment for amide dehydration condensation reaction, the synthesis of intermediate product-palmamidopropyl dimethyl tertiary amine, and then the intermediate product with sodium chloroacetate to synthesize amphoteric surfactant, the conversion of palmitic acid in the synthesis experiment was as high as 92.4%, and the critical micelle concentration CMC was found to be 1.12×10-4mol/L, and γCMC was 31.63 mN/m. The synthesis route is shown in Figure7:

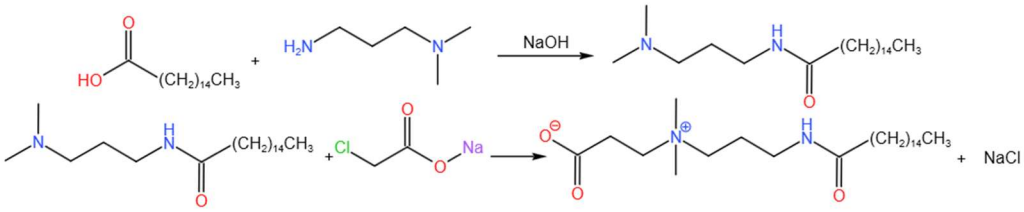

Figure 7. Synthesis of novel Amphoteric Surfactant

2.3 Amino Acid Type Amphoteric Surfactants

The amphoteric substances in the hydrophilic group with cations as amino and anionic hydroxyl groups as amino acid amphoteric surfactants, the most commonly used are α-amino acid type and β-amino propionic acid type, which have the excellent characteristics of many amino acids, such as being more gentle and safe, less irritating to the skin, etc., have a certain hard water resistance and good compatibility performance, making it widely used in medicine, textile clothing, advanced dyes, cosmetics, toiletries and other special fields [12].

Xin Yujuan [13], with straight carbon chain alcohol and twelve amine as the main ingredients added maleic anhydride for synthesis reaction, a series of new ester-containing amino acid surfactants 2-dodecanamine succinate monoalkyl esters were prepared, the synthesis route is simple and easy to operate and the utilization rate of atoms is high, and the synthesis route is shown in Figure8:

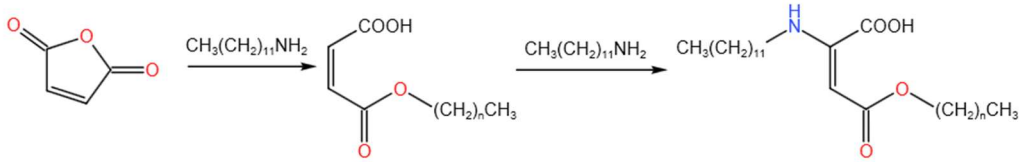

Figure 8. Synthesis of Surfactant containing ester amino acids

Gu Xuexian[14] Using long-chain aliphatic alkyl primary amines and ethanol amines as ingredients, a new type of amino acid amphoteric surfactant - N- long-chain alkyl-β-amino-acrylate ethanolamine-α-sodium aminoacetate was prepared through multi-step reaction. The product has excellent surface activity, good wetting properties, a wide equivalence area and produces less foam, less skin irritation, and can be better applied to cosmetics and washing products. The reaction is shown in Figure9:

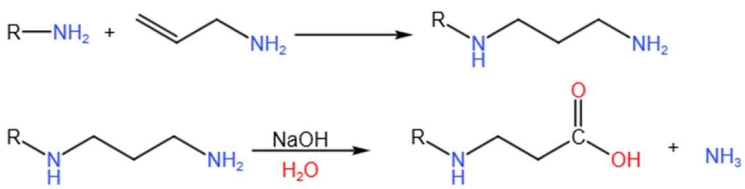

Figure 9. Synthesis of new amino acid amphoteric Surfactant

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Zhao Xiaodong et al. [15], using acrylonitrile and primary amine as materials, synthesized a new amphoteric surfactant by hydrolysis reaction in an alkaline environment manufactured by NaOH. In the first step, the acrylonitrile was added to produce an intermediate product 1, an ethanolamine was reacted to produce an intermediate product 2, and further reacted with sodium chloroacetate to give the product N- alkyl-β-amino propionic acid amphoteric surfactant. The synthesis products were found to be poor in pH=2~6.5, good at pH ≥8, excellent in temperature resistance, salt resistance and calcium resistance, which can be used as the main component of foam fluid for workover circulation in the foam drilling process. The main steps are shown in Figure10:

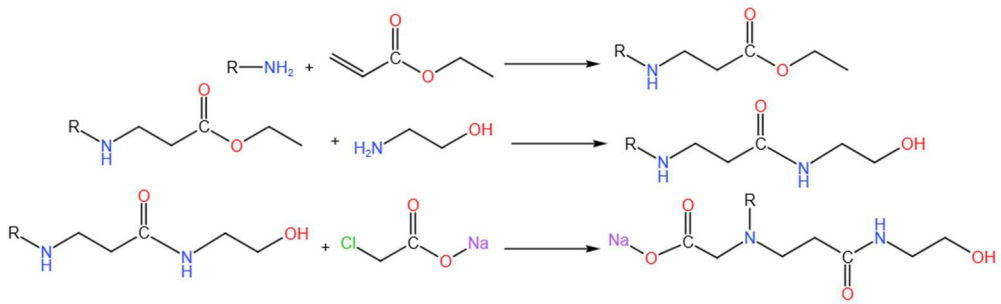

Figure 10. Synthesis of a new type of Surfactant

2.4 Other Types

Including modified type, oil-soluble polymer, fluorine-containing type, silicon-containing type, etc. [16].

Pan Hong [17], taking the cellulose extracted from the natural substance as ingredient, first modified it in the aqueous solution of NaOH, the intermediate product cellulose sulfate was obtained, and then the hydrophobic modification was made, and it was reacted with propyl oxide dimethyl octyl ammonium chloride, propyl oxide dimethyl dodecyl ammonium chloride, and epoxypropyl dimethyl tetradecyl ammonium chloride to obtain amphoteric cellulose polymer surfactant, and the synthesis route was shown in Figure11:

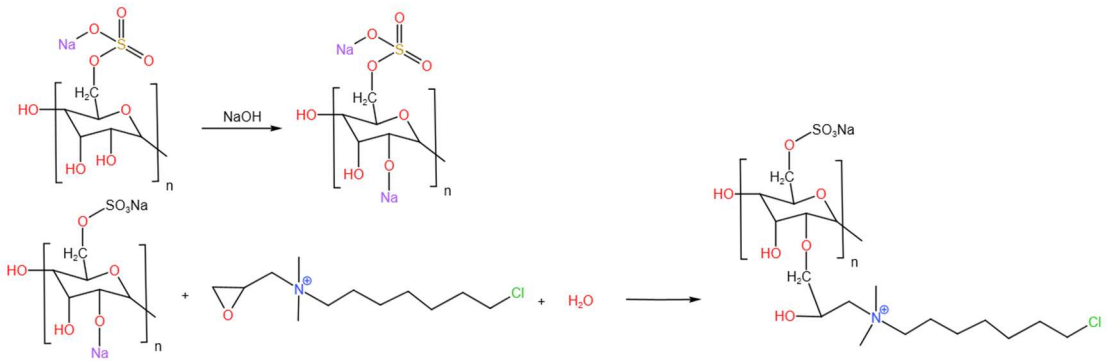

Figure 11. Synthesis of amphoteric cellulose polymer Surfactant

3. Gemini Chain Amphoteric Surfactants

A new type of surfactant that forms a molecule by chemically bonding the hydrophilic head group of two single-stranded amphoteric surfactant molecules or their adjacent head groups by means of a coupling group.

3.1 Anionic Gemini Surfactant

Hydrophilic group surfactants that hydrolyze to anions and cations are called Anionic Gemini surfactant. Due to the different electrical properties of the hydrophilic group, the Electrostatic repulsion mutual between the two hydrophilic head groups is low, so that the surfactant molecules

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are closely arranged between the two interfaces of water and oil, and its structural characteristics make the efficiency of reducing the surface tension higher. Since most of the cations are quaternary ammonium salts, they can be divided into sulfonate type, carboxylate type, phosphate ester, and sulfate amphoteric Gemini surfactants according to the hydrolyzed anions [18].

In 1998, the first amphoteric Gemini surfactant was first synthesized by Renouf et al. [19], the hydrophilic group of the active agent is sulfonate and ammonium root, so that its head base has positive and negative two kinds of electricity, compared to the molecule with only one electric head group, because the electrostatic repulsion between the two head groups in the heterosexual phase absorption molecule is much lower than the molecule with only one electrical head group, which greatly improves its surface activity.

Li Xiaojun et al. [20] with N,N'-Dimethyl ethylenediamine as the linking base skeleton, using fatty alcohols with different chain lengths (octanol, dodecyl alcohol and Sixteen alcohol) reacted with epichlorohydrin to generate alkyl glycidyl ether, and then aminated with N, N'- Dimethyl ethylenediamine into the hydrophobic chain, and finally with sodium 3-chloro-2-hydroxypropanesulfonate by quaternary ammonium reaction to obtain a series of structurally symmetrical sulfo betaine Gemini surfactants, the synthesis route was shown in Figure12:

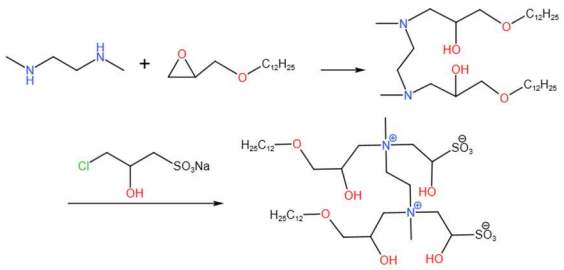

Figure 12. Synthesis of Sulfonic Betaine Surfactant

Yoshimura T et al. [21], with long carbon chain alkylamine and sodium 3-chloro-2-hydroxypropanesulfonate with different carbon content under alkaline conditions, ethanol as a solvent substitution reaction, the reaction product and dibromo alkanes react in a NaOH alkaline environment to form intermediate products, and then add bromoethane for quaternary ammonium reaction to obtain a plurality of similar sulfonate Gemini surfactants, the synthesis route was shown in Figure13:

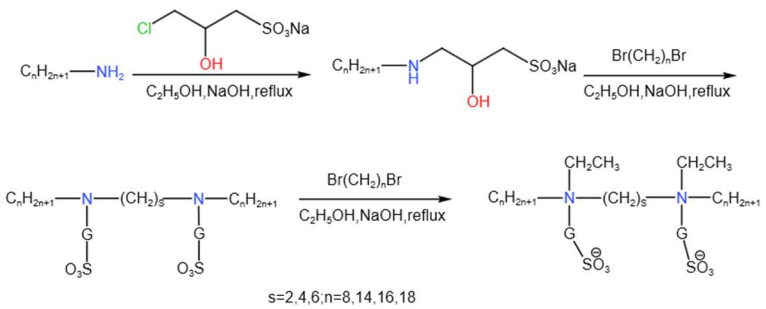

Figure 13. Synthesis of Sulfonic Gemini Surfactant

Lv Bin et al. [22] synthesized phosphate ester amphoteric Gemini surfactant, is to phosphorus oxychloride as the ingredient to provide phosphate ester base, can be replaced by two substitution reaction synthesis of the target product, the process is simple, the ingredient is easy to obtain, more suitable for large-scale promotion and production, the synthesis route was shown in Figure14:

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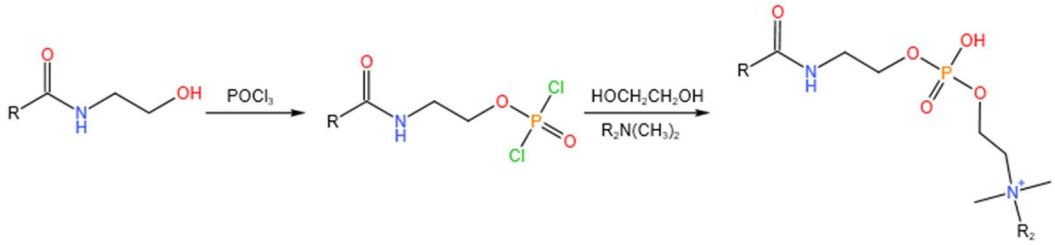

Figure 14. Synthesis of Phosphoester amphoteric Surfactant

CZhou T H et al. [23], with tetrabutylammonium bromide as the catalyst, the long chain fatty alcohol with different chain lengths and propylene oxide in the alkaline environment with NaOH as the ingredient for substitution reaction, followed by a ring-opening reaction with tertiary amines to make the epoxy group open the ring, the intermediate products at low temperature hydroxyl esterification, and finally the formation of asymmetrical sulfate amphoteric surface surfactants, detection found that it has stable thermodynamic properties, excellent positive temperature liquid crystal phase temperature range is extremely wide, Another sulfate-based amphoteric Gemini surfactant available after amidation has better solubility properties, but the synthesis steps are more complex and the cost is increased, the synthesis route was shown in Figure15:

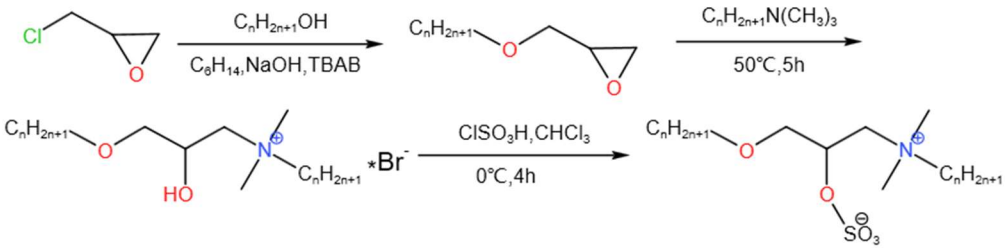

Figure15. Synthesis of Amphoteric Surfactant sulfate

Wang Yan [24], the ethylene glycol, epichlorohydrin reacted together to form an intermediate product, different from other synthesis is the second step in the HCl acidic environment, the addition of dimethyl n-dodecyl tertiary amine for ring opening reaction, and finally by Chlorosulfonic acid sulfonation reaction to synthesize the target product. Through the three-step synthesis, the synthesis steps are few, the reaction conditions are easy to achieve, and the final yield is 96%. The main steps of synthesis are as Figure16:

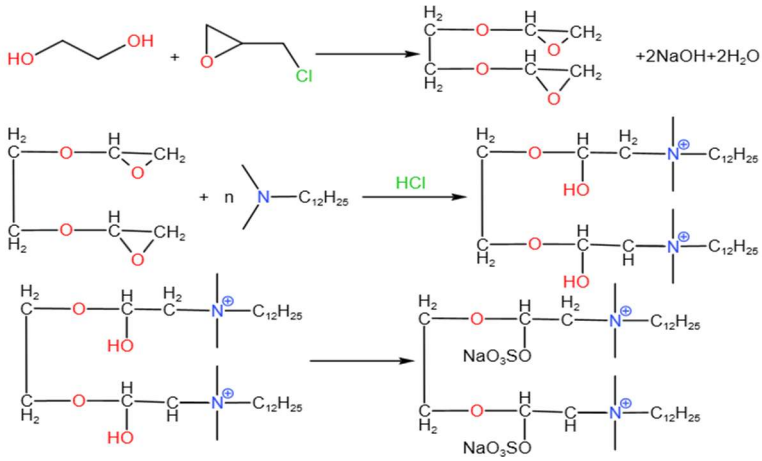

Figure 16. Synthesis of sulfonated quaternary ammonium salt Surfactant

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3.2 Anionic Type Gemini Surfactant

The nonionic groups in the anionic nonionic surfactant hydrophilic group that will not be dissociated in water can be polyoxyethylene ether chains, polyurethane chains and fluorocarbon chains, etc., but most of the surfactants of this type of surfactant currently studied are mainly polyoxyethylene ether chains. These surfactants not only have excellent emulsification properties and significant resistance to hard water of nonionic surfactants, but also combine the high interfacial activity of Gemini surfactants, neutralizing the advantages of two types of surfactants.

Alam E et al. [25], using Oil alkenyl nitrile, polyethylene glycol and polyoxyethylene ether chains as reactants, synthesized a new anionic surfactant. The Oil alkenyl nitrile was oxidized, followed by the addition of polyethylene glycol (molecular weight 350, 55,750, respectively) to open the epoxy group, and the hydroxyl and polyoxyethylene ether chains were introduced at the same time, and the final product was obtained after esterification, with fewer reaction steps, the synthesis route was shown in Figure17:

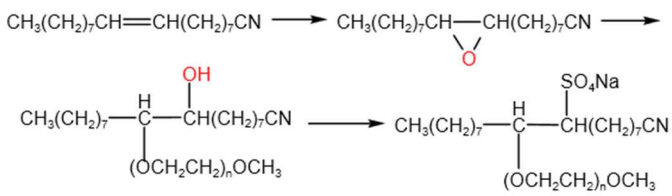

Figure 17. Synthesis of new anion-nonionic Surfactant

In recent years, by the domestic scholar Gu Yishi [26] and others, the synthesized carboxylate type anionic Gemini surfactant has excellent performance, the reaction process needs N2 to be filled, the whole process is isolated from oxygen, and ultrasonic oscillation is required, the synthesis steps are more cumbersome, and the possibility of large-scale production is small. The main steps are as Figure18:

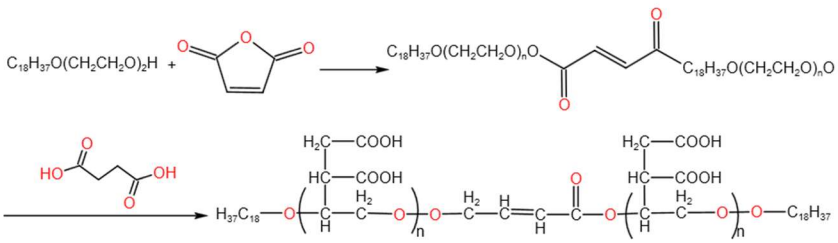

Figure 18. Synthesis of carboxylate anionic-nonionic Gemini Surfactant

3.3 Cationic-Non-Ionic Amphoteric Gemini Surfactants

Biswa et al. [27] synthesized a cationic-nonionic Gemini surfactant with polyethylene glycol chains as spacers; in 2013, Zhao Tianhong et al. [28] prepared a cation-non-ionic amphoteric Gemini surfactant DAEQ9-C1 after halogenation and quaternary ammonium, and the reaction was as Figure19:

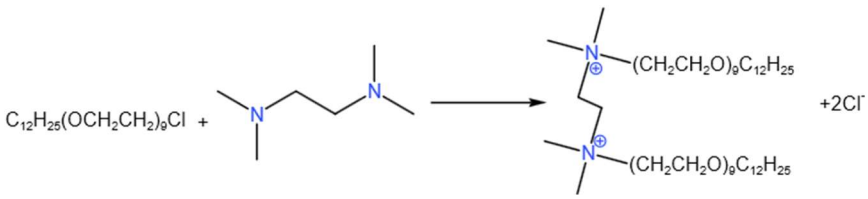

Figure 19. Synthesis of cationic-nonionic amphoteric Surfactant

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There are few reports on the synthesis of cationic-non-ionic Gemini surfactants at home and abroad, which may be due to the high cost of ingredients and the complex reaction steps and difficult to control.

4. Conclusion

Amphoteric surfactants have a unique structure and carry two electrical properties at the same time, making them have a wide range of application prospects in various fields. For single-chain amphoteric surfactants, the current technology is relatively mature and has long been put into large-scale production and use. However, the research on Gemini chain amphoteric surfactants has only been studied by scholars at home and abroad since 2000, and currently stops at the laboratory synthesis stage, research focuses on the synthesis of different new varieties, basic properties and other research, the main application areas focus on drug delivery, biosensing, protein carrier biology This medical field.

Due to the complexity of the synthesis of the amphoteric surfactant of the Gemini chain type, the harsh conditions, the technical difficulty of purification and the expensive price of ingredients, the development of the double chain type is slower than that of the single chain amphoteric surfactant, with the continuous and in-depth development of computer technology in structural chemistry, the use of molecular simulation technology for molecular design, the introduction of some functional groups for the application needs, the analysis and prediction of its molecular properties, the guidance of the synthesis of new products, can promote the development of the Gemini chain type of amphoteric surfactants. In particular, in-depth exploration in many fields such as the preparation of new materials, the exploitation of oil and gas fields, and environmental protection.

Therefore, in the face of the gap in the actual production and life of the Gemini chain amphoteric surfactant, the following aspects can be studied: (1) the use of computer technology to simulate the synthesis of the amphoteric Gemini surfactant compound, calculate its molecular structure, electron density, and then predict the performance of the synthetic compound in all aspects to select excellent varieties, in order to reduce the current research focus on a large number of synthesis phenomenon; (2) more natural substances as ingredients, synthetic reactions, in order to reduce costs, To achieve the purpose of large-scale commercial production; (3) to green chemistry as the concept, the synthesis of non-toxic or low-toxic products, according to good natural degradation capacity, friendly to the environment and pollution-free; (4) to the structure and performance of the correspondence as the theme of research, the establishment of correspondence, so as to guide the production and use; (5) improve the existing purification methods, optimize and simplify the purification route, reduce unnecessary losses; (6) expand the field of application, increase the research on the production and application of synthetic Amphoteric Gemini surfactants, In particular, as an oil repellent, it is used to reduce the water-oil interface activity and improve the efficiency of oil recovery in the tertiary oil recovery.

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