The Development and Application of the Acrylamide Kind Polymers for Tertiary Oil Recovery in China

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Abstract: As many oilfields shift from high permeability and medium-high permeability to low permeability and ultra-low permeability, the complexity and specificity of their formation channels increase, and the requirements on injectivity and liquidity of polymer flooding become increasingly stricter. This paper, from the perspective of the molecular configuration of poly(acrylamide) (PAM), gives an introduction to their respective characteristics, and analyzes the applicability of PAMs in tertiary oil recovery on the basis of the applications of polymer flooding in China. The study suggests that, on the one hand, the problem that urgently needs to be solved at present is achieving the viscosification of polymers with medium-low molecular weight or low molecular weight in high-temperature and high-salt conditions; on the other hand, researchers shall change their original research thoughts, shift from large-scale design of original PAM products to one-to-one "customized" research on and development for meeting specific oilfield demands from the perspective of polymer molecular structure.

Keywords: Tertiary oil recovery, polymer flooding, acrylamide, permeability, viscosification.

1. BACKGROUND

Polymer flooding was originally used in oilfield as a compensation for water flooding. It requires addition of polymers with relative molecular weight to water, increasing the viscosity of injectant, and lowering the water phase permeability, thereby improving the mobility ratio between displacing phase and displace phase, reducing the fingering phenomenon of water, increasing the swept volume of oil-displacing agent, macroscopic sweep coefficient η and oil recovery. In recent years, many research results have shown that polymer may increase microscopic oil displacement efficiency for its high viscoelasticity [1].

Globally, different countries adopt different technologies to increase the tertiary oil recovery ratio in accordance with their respective regional and social characteristics. Specifically, the United States and Canada mainly use gas flooding, Venezuela mainly uses thermal recovery, while China mainly adopts chemical flooding since most of its lands are continental deposits and feature strong heterogeneity. However, in recent years, as is seen from practical experience, the United States, Canada, Brazil and some Middle East countries have shifted their focus to chemical flooding, indicating that chemical flooding is of great potential [2-7]. European countries including Germany and Australian have planned to adopt polymer flooding [8-10]. In Middle East countries, only Oman uses polymer flooding in its Marmul oilfield, while the Sabiriyah Maudded oilfield in Kuwait is accelerating to promote chemical flooding [11-13]. In China, Daqing Oilfield and Shengli Oilfield are at their middle and later periods, and the oil produced there has high rate of water content, for this, polymer flooding technologies are introduced in most cases in order to increase the recovery ratio [14-15]. The oil production through polymer flooding has reached 73.5 million barrels, accounting for nearly 23% of the total oil production [16].

At the beginning of the development of polymer flooding, many scholars have made a conclusion that the polymers for increasing recovery ratio must be completely linear. There are only two types of such polymers, namely xanthan gum and HPAM (partially hydrolyzed polyacrylamide). Both the polymers have high viscosity and good shear thinning capability that the colloidal solution has. Of which, the maximum temperature that xanthan gum can withstand is about 70 ˚C, lower than that of PAM (95 ˚C) [17]. The biggest disadvantage of xanthan gum is its poor performance of resistance against bacterial degradation, therefore, xanthan gum needs to be used together with formaldehyde bactericide (25 ~ 100 mg/L) [18]. Under the precondition of oilfield development on the basis of environmental protection and sustainable development, we need to focus more on the research on and

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development of acrylamide polymer flooding. Over the past 40 years of the development of polymer flooding, the oil displacement capacity of the technology has been constantly enhanced, and the composition and structure of polymer have become more complicated. Now, polymer flooding can meet the demands of oilfields with high permeability, medium-high permeability, high temperature and high mineralization. However, due to the existence of large number of oilfields with low permeability and ultra-low permeability, requirements on polymer flooding injectivity and liquidity become stricter. Therefore, how to apply acrylamide polymer flooding effectively in oilfields with medium-low permeability and low permeability has become a bottleneck at present. To sum up, this paper, from the perspective of the molecular configuration of PAM, expounds on their characteristics and actual application in China's oilfields, so as to lay a firm foundation for research researches on and development of PAM for tertiary oil recovery and finding a right approach to researches on PAMs.

2. CLASSIFICATION OF PAMs

2.1. HPAM

In 1954, the United States began to produce non-ionic HPAM commercially by using homo-polymerization. However, the non-ionic HPAM's chain link is electrically neutral and its molecules tend to curl in solution, and it has poor ability of viscosification and has an extremely strong absorbability to the surface of the stratum rock, therefore it is not applicable to the EOR field. In 1980, Muller found that the pH value and viscosity of the aged HPAM had changed, that is, the negatively-charged carboxy group increases the intermolecular repulsion, enhanced the extension of the main chain and showed visicosifying ability [19]. At present, HPAM is the most widely-used technology in polymer flooding [20-22]. Conventional HPAM shows newtonian fluid characteristics in the condition of low shearing rate, while shows non-newtonian fluid characteristics in the condition of high shearing rate. With the critical shearing rate as a splitting point, non-newtonian fluid characteristics give rise to two phenomena, namely shear thinning and shear-thickening. Specifically, shear thinning means pseudo-plastic behavior, a behavior of polymer fluid when flowing through a pipeline. In the case of the critical shearing rate, the behavior is ascribed to the extension of single dispersive cluster polymer molecules and separation between two intertwined polymer molecules that causing the apparent viscosity declines as the shearing rate increases [23]. On the other hand, shear thickening is the main form of HPAM's dilatant flow behavior, often occurs when the HPAM is in pure water or salt solution or flows through porous media [24]. Specifically, when the HPAM is in pure water or salt solution, the dilatant flow behavior is caused by new association groups as a result of the interactions between polymer molecular chains [25]; when while HPAM flows through porous media, the viscoelasticity of polymer fluid hinders the change of fluid flow direction. The shearing rate here is deemed as the change of the fluid flow direction due to the change of the pore diameter or bending of pore throat. Within a certain scope, the greater change of the flow direction, the larger the shearing rate, the larger the flow resistance caused by viscoelasticity, which is manifested by the increase of apparent viscosity [17].

The critical shearing rate is closely related to the hydrolysis degree, molecular weight, polymer concentration and solution environmental temperature of HPAM [24, 25]. Theoretically, the increase of HPAM hydrolysis degree may raise the viscosity of polymer solution, but it is not necessarily conducive to onsite applications of oilfield. The carboxylic groups in HPAM react with bivalent cations (>200 ppm) in formation water to form precipitation, resulting in degradation of the polymer molecular chain [26]. The research made by Zaitoun [27] indicates that, in the case of existence of bivalent cation, the critical hydrolysis degree of HPAM is 33% (the reaction conditions are specified). In formation water, if the bivalent cation concentration is greater than 200 ppm, the formation temperature for HPAM is 70 ˚C [28]; if the bivalent cation concentration is less than 200 ppm, the formation temperature for HPAM is up to 100 ˚C [29, 30].

Currently, there are two preparation methods for HPAM, namely hydrolysis and AM-AA (acrylamide - acrylic acid) copolymerization. Hydrolysis means addition of hydrolytic agent with a certain proportion before or after homopolymerization of acrylamide, such as sodium hydroxide, sodium carbonate and ammonium hydroxide, etc., so as to hydrolyze its side chain acylamino and generate carboxylate [31, 32]. The hydrolysis rate and hydrolysis degree of the HPAM synthesized by this method are closely related to temperature and concentration of hydrolytic agent. The hydrolysis reaction generally takes place within the temperature range of 70 – 90 ˚C, the higher the hydrolysis temperature, the higher the hydrolysis speed; the higher the pH value, the greater the hydrolysis degree. The AM-AA copolymerization means dissolving AM and AA in water for free radical polymerization under the effect of initiator [33]. As a main method for industrial production, the AM-AA copolymerization is simple, environment-friendly and has high productivity. The carboxylates hydrolyzed via the preparation methods are distributed randomly. In order to give play to the electrostatic repulsion on the carboxy groups to a greater extent, Zhang *et al*. used the technology of template polymerization, by changing the reactivity ratio between AM and AA, increase the sequential probability of carboxy groups in the HPAM molecular chain, and form micro-block structures [34, 35]. Due to the existence of coulombian force in the micro-block structures, the intermediate electric charges within each micro-block structure are under little influence of metal ions in salt solution and have higher repulsive force, thereby having relatively large hydrodynamic radius. The technique provides a possibility for enhancing the heat resistance and salt tolerance capabilities of HPAM.

2.2. Acrylamide Copolymer

2.2.1. Acrylamide- co -2-Acrylamide-2-Methylpropanesulfonic Acid (AM- co -AMPS)

In order to further increase the heat resistance and salt tolerance capabilities of polymer, 2-Acrylamide-2-methylpropanesulfonic acid (AMPS) is often used as polar anion monomer to form AM- *co* -AMPS water-soluble copolymer. The sulfonic acid group contains two S-O coordination bonds, which may enhance the ability of S to attract electrons from -OH$^-$ and make -SO$_3^-$ remain stable, thereby showing insensibility to salt (especially to bivalent metallic cations [36]), and enhancing polymer's capabilities of water solubility as well as salt tolerance and resistance against high-valent metal ions. Moradi *et al*., under the conditions of 121 ˚C and a seawater minerlization degree of 33,560 ppm, observed the solubility of AM- *co* -AMPS copolymers with different proportions, revealing that the solubility of the AM:AMPS=9:91 sample is better than that of the AM:AMPS=64:36 sample [37]. However, excessive content of AMPS will not only significantly increase the costs of copolymer, but also reduce the system viscosity. Therefore, the optimal proportion of AM:AMPS is 40:60. Furthermore, with a certain scope of reasonable proportion, AMPS is connected with a right rigid tertiary butyl group, which may help enlarge the rheological volume of polymer and achieve the result of viscosification [38, 39]. The research results of Li [40] show that the copolymerization temperature is 45.5 ˚C and the optimal synthesis proportion is AM:AMPS=8:2. At the temperature of 30 ˚C, the apparent viscosity of the copolymer is 80.5 mPa˙s and its intrinsic viscosity is 4780 mL/g. Compared with the HPAM samples of Daqing Oilfield, in the case of an environmental temperature of lower than 65 ˚C and 7.34 s^{-1}, the apparent viscosity of HPAM is slightly better than that of AM- *co* -AMPS copolymer; when the temperature rises to 90˚C, the apparent viscosity of AM- *co* -AMPS copolymer is about 15 mPa˙s higher than that of HPAM sample. Generally, the maximum temperature for oilfield applications of AM- *co* -AMPS is 90˚C [41]. Audibert [42] and Adhikary [43] also discovered that, in the aqueous solution of 30 g/L KCl (united unit), the intrinsic viscosity of AM- *co* -AMPS copolymer remained stable at a temperature of 90 ˚C after a period of 20 days; while at a temperature of 130 ˚C, the intrinsic viscosity of AM- *co* -AMPS copolymer dropped to 66% of the original value after a short period of 10 days. This shows that AM- *co* -AMPS copolymer is suitable for higher formation temperature and conditions of mineralization.

2.2.2. Acrylamide- co -Vinyl Pyrrolidone (AM- co -VP) & Acrylamide- co -Sodium p-Styrene Sulfonate (AM- co -SSS)

Besides the introduction of -SO$_3^-$ and -COO$^-$ that are insensitive to salt for the purpose of improving the heat resistance and salt-tolerance capabilities of polymer in the oil displacement process, the cyclic structure that has great steric hindrance (styrene sulfonic sodium, acryloyl morpholine and vinyl pyrrolidone) may also be used to protect the C-C main chain, enhance the rigidity of polymer molecular weight, and make curl and degradation difficult under the conditions of high temperature, high mineralization and high shearing [43]. Liu Bailin *et al*. synthesized AM/AMPS/NVP (N-vinyl-pyrrolidone) terpolymer. After the sample has aged for 130 days, the absolute values of its apparent viscosity have exceeded 6.0 mPa˙s, this is because that the NVP in copolymer can effectively inhibit the hydrolysis of acylamino in bivalent salt solution [44]. The study result of Doe [45] indicates that, at a temperature of 120 ˚C, AM- *co* -VP (1:1) copolymer remained stable in seawater for several months. However, the AM- *co* -VP copolymer is difficult to be synthesized and its viscosifying performance is poor, which needs to be improved in the future [46].

Chen [48] used dispersion polymer to prepare anti-salt and heat-resistant acrylamide- *co* -sodium p-styrene sulfonate (AM- *co* -SSS) oil-displacing agent. In the case of the same salt solution, the apparent viscosity of the oil-displacing agent is significantly better than that of PAM polymer. Moreover, the apparent viscosity of both the oil-displacing agent and PAM polymer drops as the temperature rises; differently, the viscosity retention ratio of AM- *co* -SSS is twice or triple that of PAM.

2.3. Hydrophobically - Modified Polyacrylamide (HMPAM)

Hydrophobically-modified polyacrylamide (HMPAM) refers to water soluble polymer of which the hydrophilic macro-molecular chain has a few hydrophobic groups. The macro-molecular chain of hydrophobic modified HPAM contains many charged groups and hydrophobic groups, and the macromolecular main chain still remains loose and extended in the condition of certain mineralization due to intra-molecular electrical repulsion and hydration of polar groups [49]. In 1982, Landoll used the micelle method to synthesize a series of hydrophobically modified hydroxyethylcellulose (HMHEC) polymers, which was considered the “true beginning of hydrophobic association water soluble polymer” [49]. In 1987, the Schulz team successfully synthesized the acrylamide- *co* -n-alkyl acrylamide (AM/C$_n$AM, $3 < n < 10$) in aqueous solution through free radical polymerization, and applied it in the oil displacement test for the first time [50]. In 1993, Chinese oilfield chemists Dong and Huang *et al*. began to study hydrophobic association water soluble polymer, but found that its viscosity failed to meet the application standard since the critical association

Table 1: Summary of the Correlational Research of HMPAM

System	Applicable scope	Ref.
AA/AMPS/MA MA: maleicanhydride	Applicable to oil-displacing agents for oil deposits with high temperature and high mineralization.	[54, 55]
AM/C$_{16}$DMAAC/AA) C$_{16}$DMAAC: Hexadecyl dimethylammonium chloride	The difficulties in polymer purification and after-treatment in the micellar polymerization have been overcome.	[56, 57]
AM/AMPS/AMC$_{12}$S AMC$_{12}$S: 2 - acrylamidododecanesulfonic acid)	Applicable to high temperature, high mineralization and salt-resistant performance.	[58, 59]
AM/NaAMPS-DMDA DMDA: Diallyl dimethyl ammonium chloride	Free radical polymerization, hydrophobic ampholytic copolymers, use of hydrophobic structure in the same polymer structure.	[60]
P(AM-TA) TA: Tetradecyl acrylate P(AM-HA) HA: Hexadecyl acrylate P(AM-OA) OA: Octadecyl acrylate	The series of copolymers were characterized by kjeldahl method for nitrogen determination. Use no surfactant; direct use of surface-active macro-monomer, with obvious thickening effect.	[61]
P(AM - HAD) HAD: Hexadeylacrylate copolymers	The AM - HAD polymer shows dramatic increase in apparent viscocity in 1.5% NaCl solution.	[62]
AM/AMPS/NVP NVP: N-VinyPyrrolidone	Introduction of cyclic group with high-rigid molecular chain, each monomer of non-ionic NVP (insensitive to salt) contains non-polar hydrophobic group and high-polarity hydrophilic acylamino. Its features are similar to amphiphilic features, and it can effectively inhibit -CONH2 hydrolysis.	[63, 64]
AM/NVP/DMDA	The terpolymer shows strong hydrophobic effect, and high viscosity in brine solution at low polymer concentration.	[65]
P(AM/AEBA) AEBA: 4-(2-(acryloyloxy) ethoxy) benzyl tri-ethyl ammonium bromide (AEBA)	The high performance hydrophobically associating polymers with micro - block structure characteristic is synthesized by means of microhetergenous copolymerization technique.	[66]

concentration is very high [51, 52]. In 1997, Luo [57], on the basis of the actual demands of the oil & gas extraction projects, conducted pro-production and pilot experiments on HMPAM for the first time, and obtained certain results. The results showed that the SSRAP hydrophobic associated polymers developed by the research team has good viscosifying ability in the conditions of high temperature and high mineralization, the percolation process may generate great resistance and has good mobility control ability, thereby effectively reducing the water phase permeability of porous media with medium-high permeability and increasing oil displacement efficiency [57]. The examples of the R&D work done by research institutes on HAP are shown in Table **1**.

Similar to HPAM and copolymers, under the condition of low concentration, the viscoelasticity of HAP is the same as that of ordinary HPAM. Differently, HMPAM polymer has also structural viscosity besides its own bulk viscosity [67]. The research results of Jiang [1] indicate that, by comparing HAP and HPAM that have the same molecular weight, when the concentration of polymer is higher than a certain level, the former has a higher effect of viscosification, as is shown in Figure **1**. This is because that, when the polymer concentration reaches the critical association concentration, the contact distance of hydrophobic groups on the main chain shortens, and intermolecular spatial mesh structure with certain strength is formed due to association, generating micro domain and showing higher thickening ability. Compared with HPAM, HA-PAM shows better salt-resistant performance and greater anti-shear ability. An appropriate shearing rate may help extend the hydrophilic main chain, further enhance the association between hydrophobic side chains and thereby realizing viscosification [68]. Shi [53] *et al*. prepared SSRAP hydrophobic associated polymers by taking $C_{18}H_{37}$ as hydrophobic monomer and making it copolymerize with acrylamide and acrylic acid. The oil displacement capability test result shows that at the earlier stage the recovery ratio of SSRAP is similar to that of HPAM, while at the later stage the recovery ratio of the former is 4.5% higher than that of the latter, and the injection pressure of the former is 1.5 MPa higher than that of the latter.

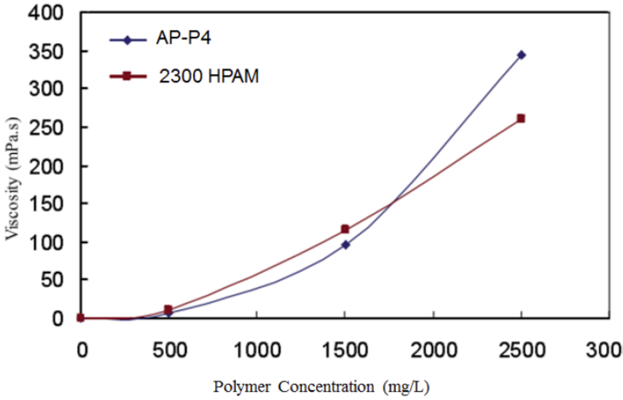


Figure 1: The viscosity of polymer solutions with different concentrations under the shear rate of polymer flooding [1].

(1) Internal Impacts on the HMPAM Association Ability

The type (hydrophobicity), quantity and distribution mode of hydrophobic group have an important impact on the association behavior of HMPAM. From the perspective of chemical structure, the quantity of hydrophobic groups determines the viscosifying ability of HMPAM. Within a certain scope, the increase of the content of hydrophobic monomers can significantly increase the viscosity of polymer. However, the excessive increase of the proportion of hydrophobic monomer may make it more difficult for polymer to dissolve and for this causes phase separation. Wang [69] conducted a research on the impact of the content of hydrophobic monomer on its aqueous solution, the research result shows that the intrinsic viscosity is $17.61 mL.g^{-1}$ when the octadecyl dimethylallyl ammonium chloride (ODAC) content is 0.4%, and the intrinsic viscosity begins to decline as the content of ODAC keeps increasing. He presume that it may be caused by the reduction of the relative molecular weight of polymer and the enhancement of intermolecular association of polymer as the proportion of hydrophobic monomer rises [69]. Yuan [70], by using the mesoscopic simulation of dissipative particle dynamics (DPD) method, studied the impact of molecular configuration changes on the behavior and nature of HMPAM in aqueous solution. By taking into account the diffusion coefficients of water molecules in HMPAM solutions with different hydrophobic alkyl chains, we find that in the system with a carbon number of 16, the diffusion coefficient of water molecule reaches its minimum value, and the system viscosity reaches its maximum value (hydrophobic modification degree~2%, hydrolysis degree~40%).

Furthermore, the position of a hydrophobic group on the molecular main chain plays a key part in associating and thickening capabilities of the hydrophobic group [71]. Researches show that the viscosifying performance of HMPAM with two-tailed hydrophobic molecules is better than that of HMPAM with single-tailed hydrophobic group [72]. HMPAM is classified into “telechelic” polymer and “multistick” polymer by the distribution location of hydrophobic group. Hydrophobic group is at the end of the polymer chain. The hydrophobic group of “telechelic” polymer is at the end of polymer, as is shown in Figure **2** [70]; for example, PAM- *co* -AVCA$_{12}$ or PAM- *co* -CAVA$_{16}$ (CAVA: 4,4’-azobis(4-cyanopentanoic acid)) [73] that is polymerized by AM and $C_{12}H_{25}OH$ or $C_{16}H_{29}OH$ via the ACVA initiated free radical reaction. For “multistick polymer” [70], hydrophobic groups are suspended irregularly on polymer main chain in the form of side group, for example, PAM- *co* -DHAM (DHAM: *N,N*-dihexylacrylamide) or PAM- *co* -DOAM (DOAM: *N,N*-dioctylacrylamide) [74], as is shown in Figure **3**. Yuan [70] is of the opinion that it is easier for telechelic to associate within the molecular chain and form larger aggregate, which is not conducive to the extension of molecular chain; while multistick conducive to viscosification. Regalado [75] is of the opinion that placing hydrophobic group on the hydrophilic main chain will help get more extended configuration and achieve higher viscosification. Nevertheless, polymers with telechelic with multisticker, such as PAM- *co* -DHAM/ACVA$_{12}$, have the greatest viscosifying ability, as is shown in Figure **4** [71].

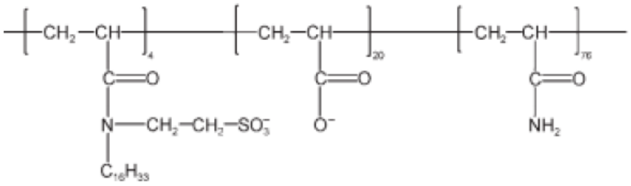
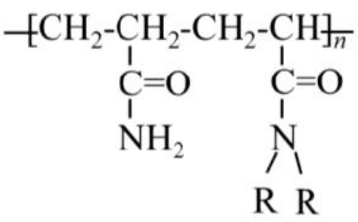


Figure 2: Schematic diagram of the molecule structures of block copolymer [70].



$$-\!\!\left[CH_2-\underset{\underset{NH_2}{|}}{\underset{C=O}{|}}{CH_2}-CH_2-\underset{\underset{\underset{R\ \ R}{/\ \backslash}}{\underset{N}{|}}}{\underset{C=O}{|}}{CH}\right]_n$$

Figure 3: Schematic of PAM- *co* -DHAM structure.

(2) External Impact on HMPAM Association Ability

Different from ordinary HPAM, since hydrophilic group and hydrophobic group exist in HMPAM, the salt that can generate free positive and negative ions in aqueous solution may also have a positive impact on the rheological performance of HMPAM, namely showing excellent salt-resistant capability in actual applications. The salt ion concentration point that may achieve HMPAM viscosification is called critical salt concentration, and its value is closed related to the type, location and quantity of hydrophobic groups in HMPAM [76, 77]. According to the intensity of the interaction between molecular chains in HMPAM, the HMPAM solution with different concentrations is divided into “dilute”, “semi-dilute” and “concentrated regime”. When the solution concentration is at the range of “dilute”, it is less likely for polymer molecules to intertwine with each other, and the viscosity has a linear relation with polymer concentration [78]. When the concentration of the solution rises to the range of “semi-dilute”, molecular chains begin to intertwine with each other, the existence of salt increases intermolecular association and causes increased viscosity [79]. When the salt concentration reaches a critical concentration, HMA polymer achieves viscosification. Zhong [80] attributes the viscosification phenomenon to the increased polarizing capability of electrolyte salt due to the existence of salt. However, the increased mineralization will enhance the intra-molecular association, lead to large-scale aggregation of hydrophobic groups, phase separation in HMPAM aqueous solution and reduction of viscosity.

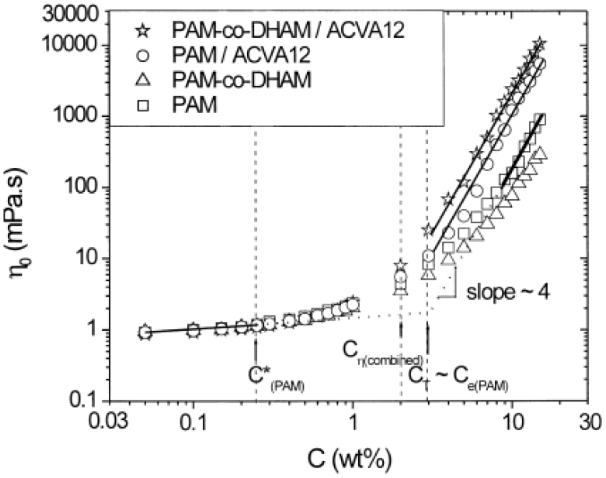


Figure 4: Comparison of viscosities of telechelic and multisticker polymers with that of the combined type on the series C12 [71].

Most HMPAM polymers have an association peak at a specific temperature [81]. Take the HMPAM copolymer poly(N-isopropylacryl-amide (PNIPAM) for example, association peak is the point where there is the largest inter-molecular force, so the temperature that is corresponding with the point (T_{max}) is correlated with the cloud point temperature of PNIPAM, while the cloud point temperature can be regulated by changing the use level of surfactant SDBS [82]. The temperature-related viscosification is attributed to the fact that the increased association is an entropy-driven endothermic process. However, when the environmental temperature is higher than the critical temperature of the solution, the reduced association will lower the viscosity [83, 84].

Zhu conducted a research on the viscoelasticity of HAP3 (HMPAM product No. 3) in porous media [85]. The results show that the associated HMPAM will increase the pressure drop significantly when passing through porous media. It is caused by blocked pore throats due to bulk effect. He inferred from this phenomena of quick pressure rise and excessive injection pressure in the onsite construction of HAP system. The results of the core displacement test conducted by Wang and Jiang [1] show that, no matter whether the association polymer has reached the critical association concentration, its final oil displacement efficiency will always be lower than that of HPAM polymer. This is because that association polymer has much higher viscosifying ability, but its corresponding elasticity is lower than that of HPAM. For example, take the association polymer with a weight concentration of 2500 mg/L for example, its viscosity is higher than that of high molecular weight HPAM solution with the same concentration, but its final oil displacement efficiency is even lower than that of 500 mg/L HPAM solution flooding. It indicates that HMPAM can not give a full play to the function of elastic recovery in strata and tends to integrate under the effect of shear stress; therefore, the association polymer does not help increase the final oil displacement efficiency [1].

Cao [86] *et al*., for the purpose of addressing the adaptation between HAP molecular aggregate and pore throat at the reservoir stratum, wrapped hydrophobic monomer with cyclodextrin, and studied the reservoir stratum and fluid at the third Southern part reservoir of Daqing Oilfield. The results show that β-cyclodextrin can effectively reduce the size of HAP molecular aggregate, enlarge the impact scope of the polymer molecular thread reservoir stratum and improve adaptability of HAP to oil reservoir.

2.4. Comb-Shaped Salt-Resistant Polymer

The research results of Zhu *et al*. show that, only when the concentration of HMPAM exceeds the first association concentration, can association occur and the oil recovery effectively enhanced; however, the polymer concentration in the core is low and not enough for association and viscosification of HMPAM polymer [87]. In order to avoid the problem of critical association concentration, Luo [90] *et al*. developed comb-shaped salt-resistant polymer for industrial use through copolymerization between acrylamide and anti-salt monomer (AHPE), with a code of KYPAM, Figure **5**. The new-type polymer has both oleophylic groups and hydrophilic groups, which repulse each other and reduce curl and intra-molecular/inter-molecular intertwinements. The macro molecular chain is comb-shaped in aqueous solution, the rigidity of the molecular chain and the regularity of the molecular structure of the polymer increase, the hydraulic radius of the molecular thread rotation increases, no association phenomenon occurred in the thickening process, and its viscosifying mechanism is similar with that of ordinary long-chain high-molecular polymers. Under the same conditions, the viscosifying ability of the comb-shaped salt-resistant polymer is 58~81% higher than that of Daqing 2B838, and 22~70% higher than that of MO-4000 produced by Mitsubishi [88]. The polymer has now been extensively applied in tertiary oil recovery and deep profile control for displacement in Daqing Oilfield, Shengli Oilfield, Karamay Oilfield, Huabei Oilfield and Bohai Oilfield, showing high oil displacement efficiency [89, 90]. In 2013, CNOOC (China national offshore oil corporation) Research Institute conducted a comparative study on polymer flooding for high temperature (83 ˚C, medium-low permeability) oilfields, and the study results showed that the comb-shaped polymer with low relative molecular weight has high capabilities of heat resistance and salt tolerance, anti-shear ability and heat stability, and is more suitable for oil reservoir with low permeability and high temperature compared with sulfonated polymers and common polymers [91]. In 2017, CNOOC Research Institute used the “comb-shaped polymer and non-ion -anion (NPAC) surfactant” to build a type of binary combination system for oil reservoir that is applicable to low permeability, 83 ˚C, 24,000 ~ 26,000 ppm high mineralization and has an average porosity of 51.1 mD. Take the Shaqi fault block of Jiangsu Oilfield for example, the recovery ratio at the natural core with a water permeability of 18.6 ~ 62.5mD (0.4PV) has been increased by about 11% [91].

In 2006, Luo [90] *et al*. drew on the idea on the design of the molecular structure of the comb-shaped salt-resistant polymer, changed the single comb-shaped side chain monomer into dual comb-shaped side-chain monomer, as a result, a reverse occurred due to the mutual repulsion between the two side chains, playing a stronger role in supporting the

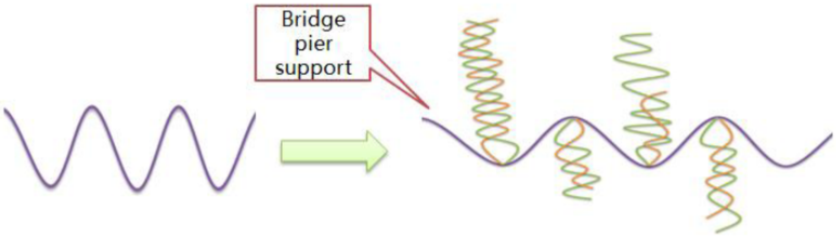


Figure 5: Schematic diagram of the comb copolymer.

molecular chain, and making the macro molecular chain take on a braided and combed shape in the aqueous solution. Compared with KYPAM, the polymer has higher rigidity and structural regularity, longer hydraulic radius of molecular thread, and stronger salt-resistant capability [90, 92, 93]. Jiang [1] said that, compared with HPAM polymer with the same molecular weight, in the case of the same weight concentration, comb-shaped polymer has higher viscosity in solution, but its elasticity is relatively low, so is the oil displacement efficiency in the case of the same quantity of capillary tube.

3. CONCLUSIONS

In the process of the researches on polymer for tertiary oil recovery, researchers used to emphasize on and development of PAMs with high molecular weight and even super - high molecular weight, so as to achieve the ability of thickening, increase the swept volume by increasing the length of the molecular chain. As crude oil has been constantly extracted, many oilfields have shifted from high permeability and medium-high permeability to low permeability and ultra-low permeability. Theoretically, the emergence of HMPAM may help solve to a certain extent such problems as excessive injection pressure and shear resistance, etc. However, it is found that the critical association concentration of HMPAM is an inevitable “threshold” for achieving effective viscosity in the process of actual onsite oilfield application. It is difficult to guarantee that the mixed concentration of the injected HMPAM and formation water is higher than the concentration for critical association, which limiting the actual applications of HMPAM polymer to a great extent in oilfields. Additionally, injectivity has also been a problem that has not been completely solved for oilfields with low permeability and ultra-low permeability. Therefore, it should be focused on the issue about the viscosification of polymers with medium-low and low molecular weights under high temperature and high salt conditions. To sum up, it is urgent for us to switch our research idea for bettering the tertiary recovery ratio, shift from large-scale design of original PAM products to one-to-one “customized” research and development for specific oilfield demands from the perspective of polymer molecular structure.

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