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Reservoir Science
http://dx.doi.org/10.62762/RS.2025.494074

RESEARCH ARTICLE

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Effects of Crosslinking Agents and Reservoir Conditions on the Propagation of Fractures in Coal Reservoirs During Hydraulic Fracturing

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Abstract

Low fracturing efficiency and high permeability filtration present substantial challenges during the fracturing development of coalbed methane (CBM), significantly hindering its efficient exploitation. In this study, a cross-linker featuring specific polar functional groups on its side chains was synthesized, and a multi-functional coupling evaluation apparatus was developed to systematically investigate the performance characteristics of water-based fracturing fluids. Furthermore, molecular dynamics simulations were employed to elucidate the microscopic mechanisms by which the modified cross-linkers and various external factors influence CBM extraction efficiency. The results indicated that the modified water-based fracturing fluid enhances fracture propagation and reduces fluid filtration into the reservoir. Increasing the concentration of the cross-linker (0wt% to 0.3wt%) improves both gas production efficiency and fracture expansion capacity (20m of crack length to 36m). Conversely, elevated reservoir temperatures (383K to 433K) markedly decrease gas recovery efficiency while significantly increasing fluid seepage (5.8ml to 7.5ml) and expansion capacity (26m of crack length to 42m). In contrast, higher reservoir pressure demonstrates an opposite trend by enhancing extraction efficiency and mitigating fluid filtration. The alteration of chemical bonding interactions between fluid molecules is identified as a key microscopic mechanism influencing CBM development, thereby offering a theoretical foundation for optimizing water-based fracturing strategies in low-permeability coal reservoirs.

Keywords: enhanced oil recovery, reservoir transformation, coalbed methane mining, water-based fracturing, low permeability reservoir.

1 Introduction

Global energy shortages have emerged as a critical challenge, restricting regional economic growth and international trade [1]. Addressing this issue requires either developing alternative energy sources or improving extraction efficiency, primarily through renewable energy exploration and the optimization of conventional extraction

Submitted: 22 August 2025
Accepted: 18 October 2025
Published: 28 October 2025

Vol. 1, **No.** 1, 2025.
10.62762/RS.2025.494074

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Citation
Cao, L., Lv, M., Li, C., Sun, Q., Wu, M., Xu, C., & Dou, J. (2025). Effects of Crosslinking Agents and Reservoir Conditions on the Propagation of Fractures in Coal Reservoirs During Hydraulic Fracturing. *Reservoir Science*, 1(1), 36–51.

technologies [2]. (1) Development of Renewable Energy Sources: The harnessing and conversion of renewable energy forms such as wind, solar, and hydrogen energy have emerged as promising alternatives to fossil fuels. These energy sources not only offer practical and effective pathways for alleviating energy shortages [3], but also contribute to mitigating environmental concerns such as the greenhouse effect and global pollution. Nevertheless, despite their environmental advantages and potential to ease the global energy crisis, several critical challenges continue to hinder the widespread implementation of renewable energy technologies [4]. Firstly, the lack of standardized specifications and technical norms for energy conversion equipment introduces uncertainty and risk into the industrial deployment of renewable systems [5]. Secondly, limitations in energy storage capacity remain a major bottleneck, restricting the ability to maintain stable energy supply and impeding grid integration. Finally, the inherent intermittency and variability of certain renewable sources—particularly solar and wind energy—pose significant challenges to their reliable large-scale utilization in industrial and commercial sectors [6]. (2) Technological innovation in reservoir transformation [7]. The mature operations of reservoir transformation for traditional energy sources have established a complete energy extraction measure for conventional reservoirs, but it has posed a challenge to the extraction efficiency of new energy sources such as coalbed methane or shale gas. The innovation of reservoir transformation presents a very significant extraction difficulty due to the differences in reservoir conditions of coalbed methane, which obviously requires geological engineers to improve reservoir transformation methods based on the unique reservoir conditions of coalbed methane [8]. However, the existing reservoir transformation measures that can be used for conventional energy sources are difficult to meet the advantages of coalbed methane or shale gas extraction, which brings severe application challenges to geological engineers or chemical engineers.

Water-based fracturing is the most widely used reservoir transformation technology, with excellent application effects on the extension of reservoir fractures and the evolution of rock damage [9]. The present methods of coalbed methane extraction and conventional reservoir fracture expansion are achieved by water-based fracturing technology [10]. The capacity for effective fracture extension, efficient sand carrying and suspension ability has had a significant impact on reservoir transformation and EOR. However, it should be noted that many defects of water-based fracturing fluids are also accompanied by many processes of reservoir transformation [11]. The alteration of fluid properties represents the primary impediment to the stable transformation of reservoir fractures by water-based fracturing fluids [12]. The elevated temperature environment associated with coalbed reservoirs will inevitably lead to a rapid change in rheological index and fluid viscosity of water-based fracturing fluids in reservoir fractures [13]. This, in turn, will result in a continuous reduction of coalbed fracture expansion behaviour of water-based fracturing fluids on the macroscopic level. Concurrently, the reduction in viscosity engendered by the elevated temperature of the reservoir environment will concomitantly augment the sedimentation rate of the proppant, a phenomenon that is deleterious to fracture support and smoothness. Furthermore, the cross-linker present within the water-based fracturing fluid is adsorbed on the fracture surface, thereby reducing reservoir permeability and hindering the convergence of coalbed methane and reservoir protection. The most significant factor that must be addressed in order to rectify the aforementioned defects of water-based fracturing fluid is the structural modification of the cross-linker, in order to adapt to the specific reservoir conditions of the coal seam.

Thus, this study developed a novel cross-linker, based on the deficiencies of the current water-based fracturing fluid. These include the variable rheological parameters and the ease with which the cross-linker molecular structure is destroyed. Concurrently, the study also developed a series of coupled evaluation devices capable of assessing the various performance characteristics of water-based fracturing fluids. This development is intended to enhance the precision of evaluation data and ensure the reliability of fluid performance. Furthermore, the performance influence mechanism of the novel cross-linker was elucidated under diverse coal seam conditions through molecular dynamics theory, facilitating the efficient exploitation of coalbed methane and providing fundamental scientific and technological support for maintaining energy security.

2 Materials and methods

2.1 Materials and Equipment

Materials: Methylene glycol, boric acid, ethanol, ammonium persulfate and triethylenetetramine were all of analytical grade and were provided by Aladdin

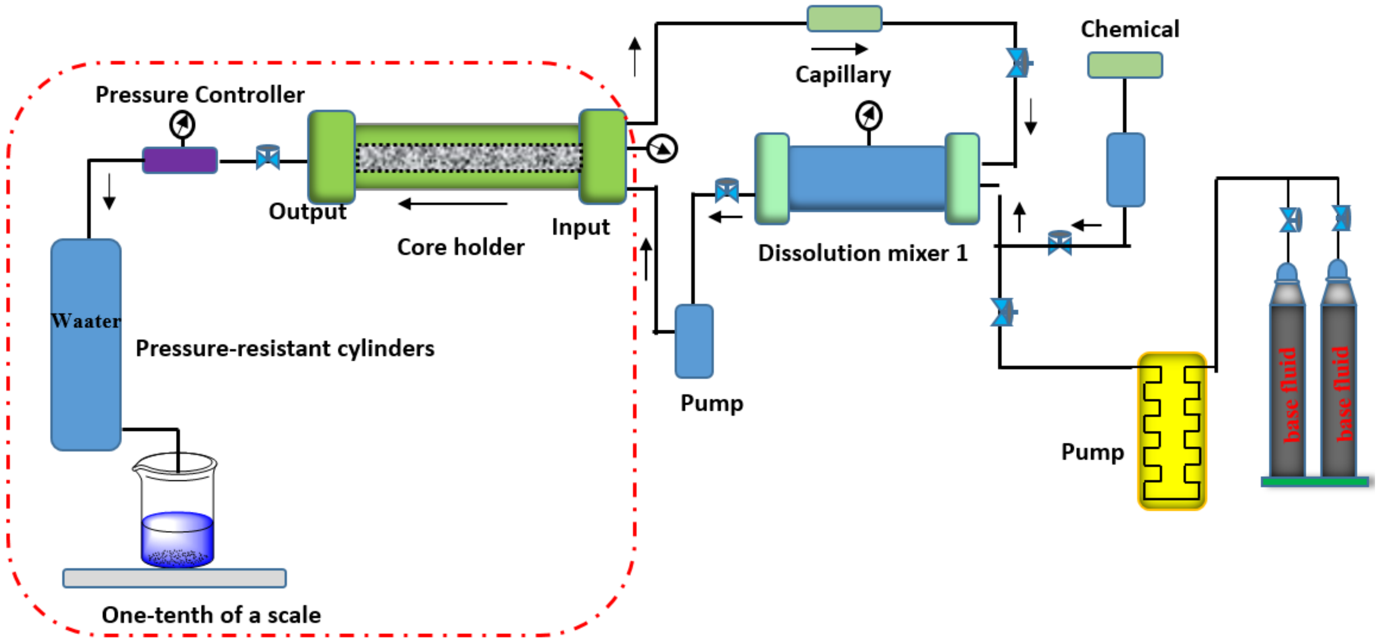

Figure 1. Multi-performance coupling device for water-based fracturing fluid.

Reagent (Shanghai) Co., Ltd. Sodium hydroxide was chromatographically pure (99.8%) and was provided by Xilong Chemical Co., Ltd. Acrylamide grafted guar gum was provided by Nanjing Chemical Reagent Co., Ltd. The commercial organic boron crosslinking agent is purchased from Dongying Fukos Petroleum Technology Co., Ltd. It presents a yellow uniform liquid with a crosslinking time of 30-240 seconds.

Equipment: The multifunctional coupling evaluation device (see Figure 1) is self-assembled in the laboratory, and its airtightness and precision have been tested and qualified. The device incorporates a pressurisation and pressure stabilisation module, a temperature control module, a fracturing and expansion module, a coalbed methane recovery module, and a data processing module. Firstly, the fracturing fluid, composed of water and proppant, is injected into a pressure-stabilised and pressure-resistant steel cylinder for temporary storage. This cylinder can also be used to pressurise to a reservoir pressure similar to that of the coal seam. Concurrently, resistance wires are positioned on the exterior of the pressure-resistant steel cylinder and the pipeline to facilitate real-time temperature detection, simulating the reservoir temperature. This approach enables the simulation to approach the actual coal seam environment. The fluid viscosity of the water-based fracturing fluid is measured in a capillary (r=0.4mm, L=10m), while the fracture expansion and coalbed methane recovery are in the core holder.

2.2 Calculation of fluid viscosity

The viscosity of water-based fracturing fluid, characterized as a non-Newtonian fluid exhibiting shear-thinning behavior, can be quantitatively assessed using a capillary viscometer. The corresponding viscosity calculation for non-Newtonian fluids is presented in Equation 1, which facilitates the analysis of viscosity variations under different coal seam conditions.

$$\eta = \frac{\tau_w}{\gamma_w} = \frac{D\Delta p/4L}{8\nu/D} \tag{1}$$

where η presented the fluid viscosity, $Pa \cdot s$; τ_w showed a wall shear stress, Pa; $\dot{\gamma}_w$ displayed the apparent shear rate, s^{-1}; D was the capillary diameter, m; the pressure difference of capillary was indicated by Δp, MPa; and L was capillary length, m; v was the flow velocity of Water-based fracturing fluid, $m \cdot s^{-1}$.

In addition, Equation 2 can also be employed to analyze and characterize the rheological index and consistency coefficient of water-based fracturing fluids, parameters that are directly linked to the efficient and stable production of coalbed methane and associated operational performance [14].

$$\lg \tau_w = \lg K(\frac{3n+1}{4n})^n + n \lg(\frac{8\nu}{D}) \tag{2}$$

The relationship between rheological index and consistency coefficient can also be solved and calculated by Equation 2.

2.3 Fracture propagation in coal seams

The core holder depicted in Figure 1 was employed as a foundation for the exploration of the coal seam fracture expansion behaviour under various factors. The objective of this exploration is to enhance the efficiency of coal seam gas extraction. Initially, the fracture in the coal seam core is preset, thus facilitating the rock damage and fracture initiation of water-based fracturing fluid under external pressure. Concurrently, the coal seam gas recovery rate of water-based fracturing fluid under varying conditions can be exhibited in real time through CT scanning.

Furthermore, a numerical model of fracture propagation of water-based fracturing fluid in geological reservoirs was constructed, with the purpose of evaluating and simulating alterations in fracture parameters caused by the material composition of water-based fracturing fluid and the influence of reservoir environment.

2.3.1 Control equations of water-based fracturing fluid in reservoir fractures

The governing equation for the flow of water-based fracturing fluid within reservoir fractures must be explicitly defined, as it critically determines the extent of changes in fluid properties and the subsequent fracture propagation behavior. Equations 3 and 4 present the flow dynamics of water-based fracturing fluid in reservoir fractures, incorporating the effect of fluid filtration. These formulations can be utilized to elucidate the impact of fluid viscosity on fracture geometry evolution [15].

$$q_x = -\frac{w^{2n+1}}{(2n+1)K}(\frac{|\partial p/\partial x|}{2})^{\frac{1}{n-1}}\partial p/\partial x \tag{3}$$

$$\mu_{eff} = K\gamma^{n-1} \tag{4}$$

where q_x is the volume flow rate of reservoir fractures. w and ∂p are the fracture width and pressure gradient. x is the crack length direction in the crack plane. n and K are the consistency coefficient and rheological index of water-based fracturing fluid, and they are mainly affected by the fluid viscosity η. $\dot{\gamma}$ is the shear rates of the water-based fracturing fluid.

The solid-liquid coupled temperature field equation of water-based fracturing fluid and reservoir fractures is shown in Equation 5, which can explain the changes in reservoir temperature under the influence of fluid viscosity and filtration behaviour [16].

$$\rho c_p(\frac{\partial(wT)}{\partial t} + \nabla(wvT)) = \nabla(k_f w\nabla T) + \rho c_p\frac{2C_l}{\sqrt{t-\tau(x)}}(T-T_r) \tag{5}$$

where ρ and c_p are the density and specific heat capacity of water-based fracturing fluid. T and T_r are the fluid temperature and reservoir temperature. v and k_f are the fluid flow rate and thermal conductivity. C_l is the filtration coefficient of reservoir fluid.

Moreover, the pressure control equation of the coupled model of water-based fracturing fluid (Non-Newtonian fluid) in reservoir fractures is demonstrated in equation 6[17].

$$\frac{4(1-\nu^2)}{E}\frac{\partial p_e}{\partial t} - \frac{\partial\left[\frac{1}{(2n+1)K(T)}(\frac{4(1-\nu^2)}{E}p_e)^{2n+1}(\frac{|\frac{\partial p_e}{\partial x}|}{2})^{1/n-1}\frac{\partial p_e}{\partial x}\right]}{\partial x} + \frac{2C_l}{\sqrt{t-\tau(x)}} = 0 \tag{6}$$

where E and p_e are the reservoir Young's modulus and effective pressure.

2.3.2 Control equations of water-based fracturing fluid in reservoir fractures

The temperature and pressure at the reservoir fracture's entrance correspond to the injection temperature and pumping pressure of the water-based fracturing fluid. Meanwhile, the temperature and pressure at the fracture tip are approximately equivalent to the reservoir temperature and the minimum horizontal principal stress (closing stress). Concurrently, the reservoir boundary pressure and temperature are equivalent to the reservoir fracture pressure and reservoir temperature, respectively.

2.3.3 Construction and reliability verification of numerical simulation model

The expansion model of reservoir fractures by water-based fracturing fluid constructed in this study follows the hyperbolic shape criterion (Figure 2(a)), which is mainly applicable to the crack expansion of quasi-brittle materials such as coal seams or low-permeability rock formations. The hyperbolic shape criterion has been demonstrated to facilitate the realisation of crack initiation, in addition to facilitating the analysis of normal and shear strength during the damage process. Furthermore, it has been shown to enable the explanation of the evolution of elastic stiffness.

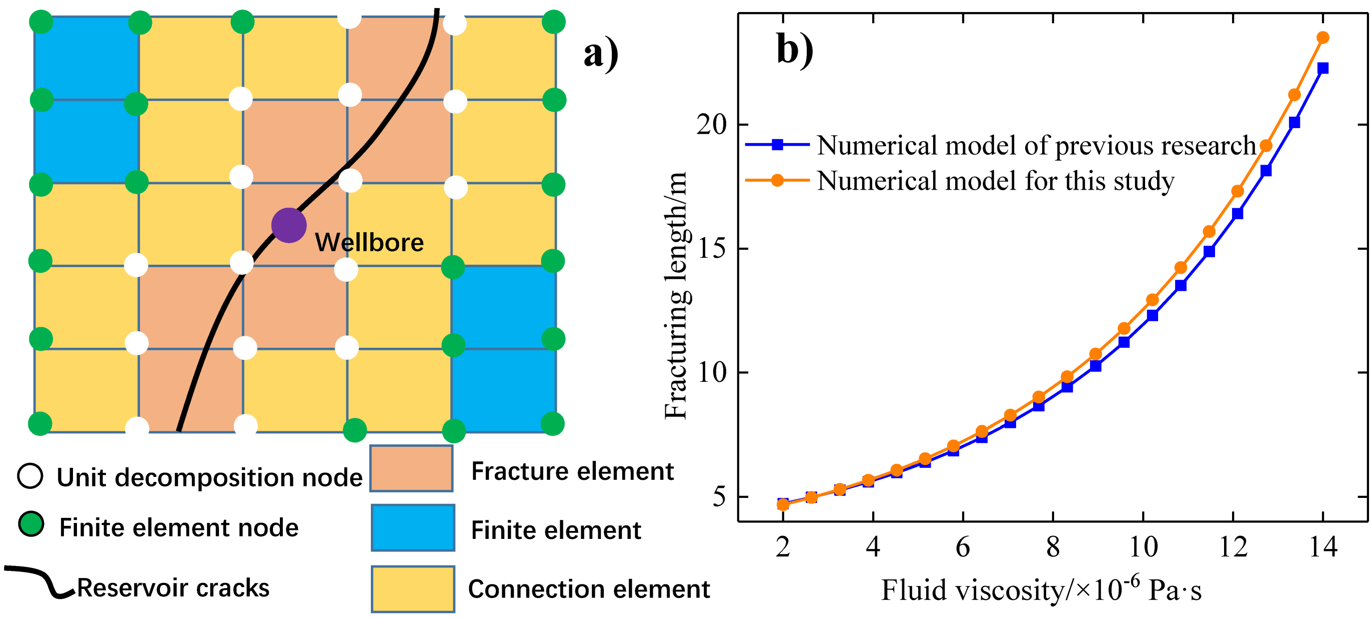

Figure 2. Numerical model and reliability analysis of reservoir fracturing fluid.

The reliability analysis investigated the comparison of data between the fracturing numerical model constructed in this study and the models in previous studies (Figure 2(b)), and this analysis enhanced the application stability and data accuracy of the numerical model in the study of the propagation behaviour of coal seam fractures. As demonstrated in Figure 2(b), the numerical model of this study exhibits a highly comparable data fit to the previously established numerical model in terms of the trend of fracture expansion [18]. However, the enhancement of the numerical model's applicability is achieved through the utilisation of a smoother curve trend.

2.4 Preparation and characterization of crosslinker

In a three-necked round-bottom flask, 18 g of methylene glycol, 15.4 g of boric acid, 18.2g of silicon dioxide, 19 mL of ethanol, and 43 ppm of catalyst were introduced. The reaction mixture was stirred in an oil bath at 80 $°C$ and 325 rpm for 4 hours. Subsequently, methylene glycol and ethanol were removed under reduced pressure. The intermediate product was then transferred to a separate flask, to which 41.4 g of triethylenetetramine was added, followed by reflux at 136 $°C$ for 3.2 hours. A clear, light-yellow liquid was obtained after separation and purification (Figure 3).

2.5 Construction of molecular dynamics models

The molecular dynamics (MD) model of the water-based fracturing fluid in a geological reservoir was established using the *Sketch* module to construct the molecular structures of water, guar gum (M = 520), and the crosslinker. Subsequently, the geometry optimization module was employed to minimize the energy of each molecule and obtain their optimized configurations. The *Amorphous Cell* module was then utilized to assemble the three optimized components and construct the fracturing system, with the composition of each component adjusted according to practical requirements, thereby generating a representative disordered molecular system.

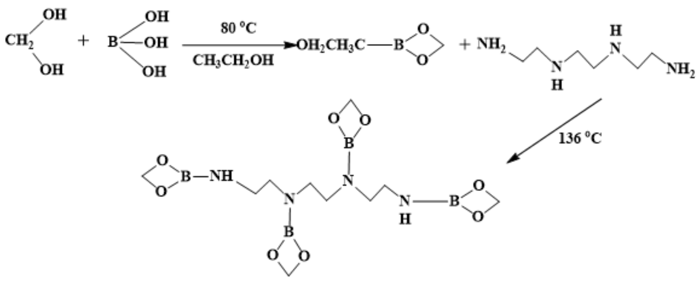

Figure 3. Preparation process of crosslinker.

Intermolecular interactions, particularly hydrogen bonding, were modeled using the *Forcite* module with the COMPASS II force field, followed by energy minimization to achieve a stable configuration. Finally, the *Forcite Dynamics* module was applied to perform molecular dynamics simulations by setting the temperature (T), pressure (P), and simulation time (100 ps) to obtain the corresponding computational results.

3 Results and Discussion

3.1 Chemical characterization of crosslinkers

The organic boron crosslinker was characterised by ^{1}H-NMR (400 MHz, $CDCl_3$) δ: 1.50 (t, 2H), 2.67 (q, 4H), 2.77 (q, 4H), 2.81 (q, 4H), 6.10 (t, 8H). The hydrogen peak of each group in ^{1}H-NMR was detected, and the boron crosslinker was successfully synthesised (Figure 4).

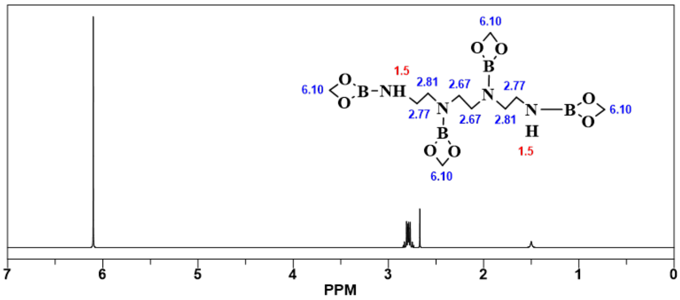

Figure 4. ^{1}H-NMR Characterization of Boron Crosslinker.

3.2 Effect of fluid viscosity on coalbed methane recovery and fracture propagation

Cross-linkers are currently employed as efficient chemicals to enhance numerous properties of water-based fracturing fluids in geological reservoirs, a process which is crucial to achieving excellent reservoir transformation capabilities. As illustrated in Figure 5, the data demonstrate a shift in trends concerning the fluid viscosity and fracture propagation parameters of water-based fracturing fluids in response to variations in cross-linker content. This analysis facilitates the exploration of the effects of cross-linker content on both reservoir fluids and reservoir fractures. As demonstrated in Figure 5, water-based fracturing fluids devoid of cross-linkers exhibit relatively low fluid viscosity, and the incorporation of 0.15% cross-linker fails to elicit a substantial viscosity enhancement in water-based fracturing fluids. Water-based fracturing fluids containing 0.15% cross-linker only demonstrate a viscosity increase of 10 mPa·s, which is evidently challenging to meet the fracturing requirements of reservoir fluids. However, it has been demonstrated that an increase in cross-linker content to 0.3% will result in a rapid and exponential increase in fluid viscosity. Such a rapid increase trend will inevitably help improve many properties of water-based fracturing fluids in geological fractures. Concurrently, Figure 4 also demonstrates the fluid viscosity relationship curve between the commercial cross-linker and the reservoir fluid. The commercial cross-linker content and fluid viscosity also demonstrate a comparable growth trend to that observed in the synthetic cross-linker. However, the commercial cross-linker exhibited a weaker viscosity growth than the synthetic cross-linker, a phenomenon that may be attributable to the molecular structure and functional groups [19].

Firstly, the synthetic crosslinker molecules displayed in Figure 3 have been shown to construct a greater number of hydrophilic groups in the side chains, thus promoting mutual attraction between the crosslinker and the guar gum and water molecules. Concurrently, the side chain crosslinkers formed in the synthetic crosslinker can promote more functional groups (hydrophilic groups such as ether groups) to collide with water molecules and interact with them to a greater extent, which effectively increases the probability of the functional groups of the crosslinker touching water molecules. Furthermore, the strong electronic magnetic field formed by the double ether groups contained in the synthetic crosslinker molecules enhances the attraction of the hydroxyl groups of the guar gum or water molecules that also have strong hydrophilicity, which can maximize the intermolecular hydrogen bonds between the crosslinker molecules and the organic macromolecules or water in the water-based fracturing fluid [20]. However, the linear structure of the commercially available crosslinker carries fewer hydrophilic groups, which weakens the intermolecular bonds between the crosslinker molecules and the hydroxyl groups in the water and guar gum molecules [21]. The low-density microscopic grid formed by fewer chemical bonds will inevitably lead to low fluid viscosity on a macro scale, and other properties of the water-based fracturing fluid will also be greatly reduced.

Furthermore, water-based fracturing fluids comprising cross-linking agents have been observed to induce a conspicuous augmentation in fracture length. However, the rate of this increase varies considerably, contingent on the specific cross-linking agent employed. When the content of the cross-linking agent is less than 0.15%, the length of the reservoir fracture will only show a gradual increase, and a fracture length increase of 5 cm will be observed when the content of the cross-linking agent increases from 0% to 0.15%. However, when the cross-linking agent exceeds 0.15%, the growth rate of the reservoir fracture accelerates, resulting in an exponentially increasing fracture length due to the increased amount of cross-linking agent. The cross-linking agent content demonstrates a change trend that is wholly consistent

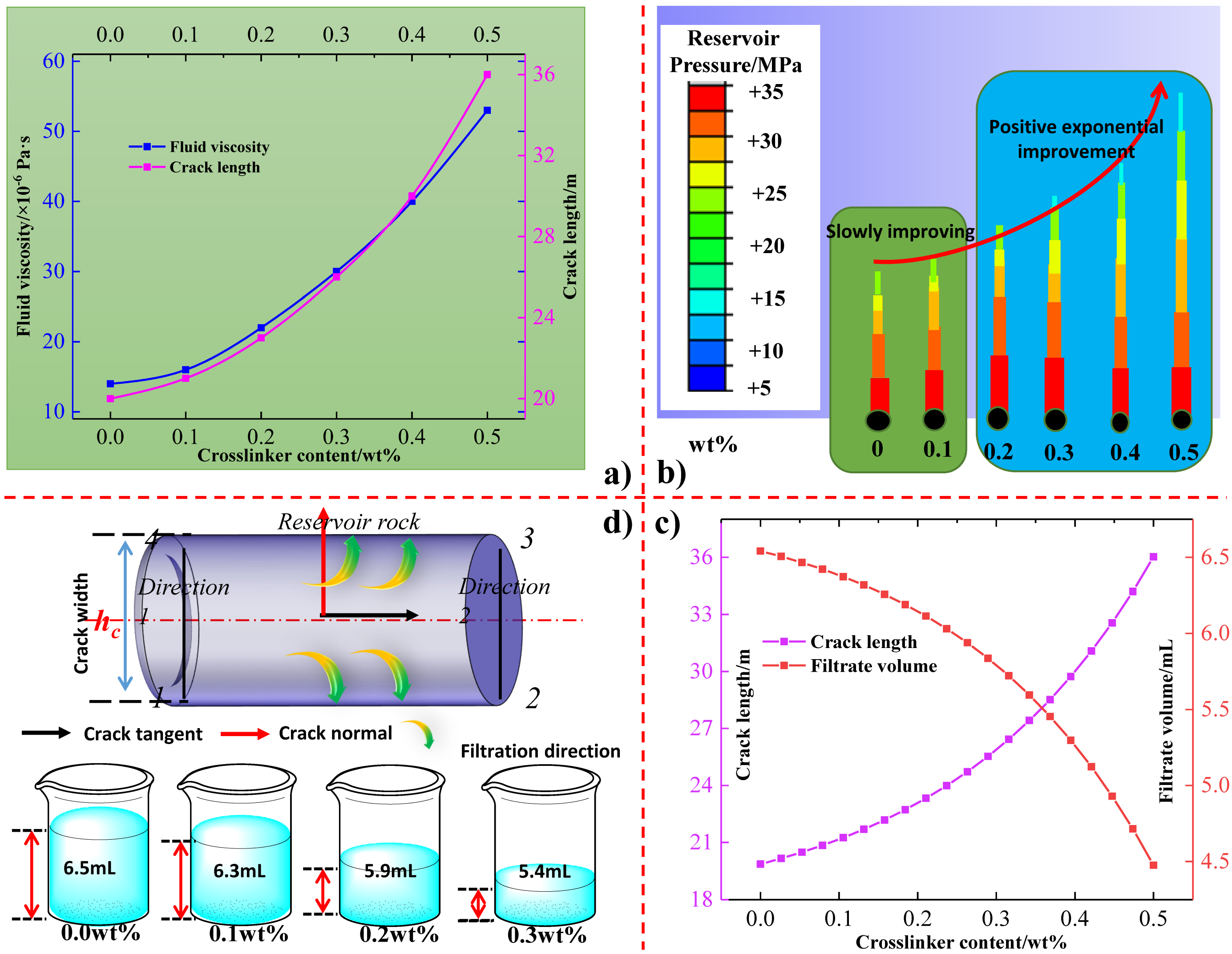

Figure 5. Relationship curve between crosslinker content, fluid viscosity, crack length and filtration capacity. a) Among crosslinker content, fluid viscosity and crack length. b) The variation trend of crack length with cross-linking agent content. c) Among crosslinker content, filtration capacity and crack length. d) Fluid filtration model within reservoir fractures.

with the fluid viscosity of the water-based fracturing fluid, a phenomenon that is principally attributed to the hydrogen bonding effect between microscopic molecules and the three-dimensional grid density in the water-based fracturing fluid [22]. In the absence of a cross-linking agent in water-based fracturing fluid, the presence of water and guar gum hydroxyl groups results in the formation of a polar effect that is found to be very weak. This, in turn, hinders the formation of strong and dense intermolecular hydrogen bonds [23]. However, water-based fracturing fluids with elevated levels of cross-linking agents have been observed to generate a substantial number of molecules that form dense and substantial intermolecular hydrogen bonds. This phenomenon has been shown to directly facilitate the rapid escalation of pressure within reservoir fractures and the consequent expansion of these fractures. The weaker filtration formed by the addition of cross-linking agents will inevitably have a positive effect on the expansion behaviour of reservoir fractures, which is also related to the construction of intermolecular hydrogen bonds. The free molecules present within the water-based fracturing fluid, characterised by a higher cross-linking agent, have the capacity to form intermolecular hydrogen bonds through the mutual attraction of polar functional groups. This process initiates the assembly of a three-dimensional grid structure by the free molecules. The reduction of free molecules weakens the fracture seepage of the water-based fracturing fluid, and the faster-growing fracture pressure can quickly break through the minimum fracture initiation pressure of the reservoir. However, water-based fracturing fluids devoid of cross-linking agents will contain a significant number of free molecules that will penetrate the micropores on the fracture surface. This will inevitably result in a slow increase in fracture pressure and fracture expansion.

3.3 Effect of Reservoir temperature on coalbed methane recovery and fracture propagation

Furthermore, it is essential to recognize the critical influence of reservoir conditions on the rheological behavior and fracture propagation capacity of water-based fracturing fluids, which directly affect the recovery efficiency of coalbed methane. Among these conditions, reservoir temperature stands out as a key geological parameter due to its pronounced impact on the performance of water-based fracturing fluids during coalbed methane extraction. As demonstrated in Figure 6, the reservoir temperature exhibits an inverse proportional relationship with the fluid viscosity in the water-based fracturing fluid. Furthermore, an increase in reservoir temperature is observed to result in an augmentation of fracture extension length. The variation trend between reservoir temperature and fluid viscosity, and fracture extension capacity, differs significantly from the relevant data presented in Figure 5(a). This indicates that the relationship between fluid viscosity and fracture extension length is not always direct.

The aforementioned disparities can be elucidated by the Arrhenius equation and molecular dynamics theory [24], which facilitate the discernment of the influence of reservoir temperature and fracture extension through molecular activity and three-dimensional grid structure. The Arrhenius equation posits the notion that the molecular activity in coal seam fractures undergoes a gradual increase in conjunction with an escalation in coal seam temperature. Concurrently, a perpetual fluctuation in motion state is exhibited. The molecules of water-based fracturing fluid exhibit reduced mobility at low temperatures, attributable to diminished molecular activity. This diminished activity is insufficient to stretch or break the pre-existing intermolecular hydrogen bonds. However, elevated reservoir temperatures will result in the transfer of heat to the fracturing fluid molecules, thereby leading to increased molecular activity and intensified irregular motion. Furthermore, an increase in the intensity of Brownian motion will serve to accelerate the process of molecular separation, thereby resulting in the dissolution of the three-dimensional grid structure that has been established, and the concomitant reduction in grid density [25]. The reduction in three-dimensional grid density is manifest in the macroscopic reduction of the apparent viscosity of water-based fracturing fluid in coal seam fractures, which is evidently not conducive to coal seam reconstruction.

Moreover, the disruption of the three-dimensional grid structure and intermolecular hydrogen bonds at elevated temperatures has been shown to result in a continuous increase of free molecules, thereby evidently enhancing the fluid filtration behaviour of water-based fracturing fluid [26, 27]. Fluid filtration and reduced fluid viscosity are not conducive to the efficient expansion of reservoir fractures, while the increasing fracture expansion trend in Figure 6 is completely opposite to the above description. It is hypothesised that the increasing trend of fracture expansion is primarily associated with the macroscopic volume expansion of water-based fracturing fluid. Conversely, fluid filtration and viscosity reduction have been demonstrated to exert a negligible hindering effect on fracture expansion. The molecular activity of water-based fracturing fluid at low temperatures is insufficient to generate the same polar repulsion force between molecules, thereby ensuring that there is no significant increase in fluid volume [28, 29]. This is supported by the bond length and energy variations under different temperatures shown in Table 1. However, elevated temperatures can markedly augment the repulsion between molecules bearing the same polar groups [30, 31]. Concurrently, the increase in molecular spacing engenders an augmentation in the volume of water-based fracturing fluid. It can be demonstrated that, in the final analysis, the greater the fluid volume at elevated temperatures, the more rapidly sufficient pressure will accumulate within the fracture. This, in turn, will promote the high-pressure fluid to rapidly breach the minimum fracture initiation pressure of the coal seam, thereby achieving rock damage and fracture expansion [32, 33].

The commercial cross-linker is unable to achieve the same viscosity retention and crack extension capabilities as the synthetic cross-linker. However, the viscosity and crack extension trends of the two cross-linkers for water-based fracturing fluids are essentially analogous (see Figure 6(b)). The disparities observed between the commercial cross-linker and the synthetic cross-linker are primarily ascribed to the variations in their molecular structure and functional groups. These structural differences inevitably result in alterations to the

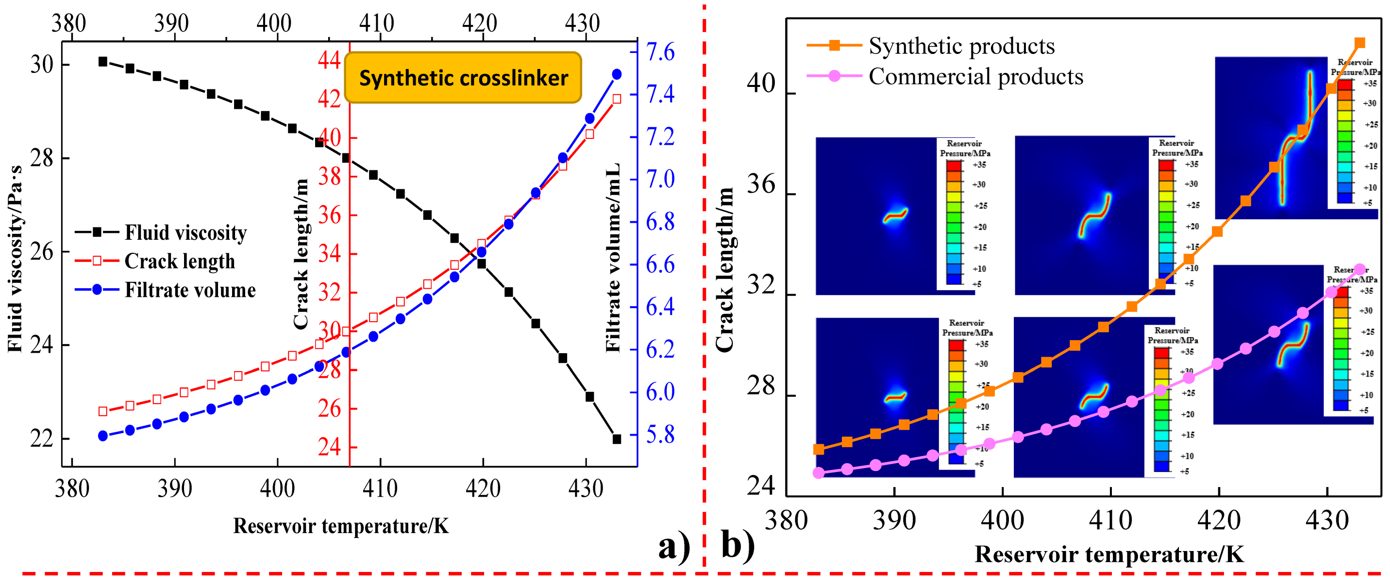

Figure 6. Effect of reservoir temperature on fluid viscosity and fracture propagation of water-based fracturing fluid. a) Synthetic crosslinker. b) Comparative data of synthetic crosslinkers and commercial products.

Table 1. The bond length and bond energy data between molecules under different Reservoir temperature.

Reservoir temperature, K	383	393	403	413	423	433
Single Bond length, nm	105	106	108	112	118	129
Single Bond Energy, 10^{-6}J/mol	267	263	258	252	243	230
Number of Bond	3547	3515	3459	3372	3286	3029

structure of intermolecular hydrogen bonds and three-dimensional grids at the microscopic level [34, 35]. The molecular structure of the commercial cross-linker is linear, which invariably restricts the loading of functional groups (polar groups) in large quantities at the molecular structure's periphery. However, the synthetic cross-linker is a branched organic boron cross-linker, which inevitably results in a large number of functional groups (ether groups) being loaded at the branched positions [36, 37]. The incorporation of a synthetic cross-linker has been demonstrated to facilitate the rapid formation of robust and substantial intermolecular hydrogen bonds with the hydroxyl groups present within guar gum and water molecules. These bonds exhibit a high degree of resistance to stretching and shearing under elevated temperatures within the reservoir. However, the presence of a limited number of functional groups on the commercial cross-linker molecules results in the formation of only a few weak intermolecular hydrogen bonds with guar gum or water molecules [38, 39]. These bonds are susceptible to stretching or shearing under conditions of irregular motion at elevated temperatures. The aforementioned disparities in molecular structure consequently result in discrepancies in the number of free molecules and filtration at a constant reservoir temperature [40, 41]. Moreover, the fracture expansion capacity is unable to increase rapidly due to the constraints imposed by the volume expansion capacity of commercially available cross-linkers.

3.4 Effect of reservoir pressure on Coalbed methane recovery and fracture propagation

As a pivotal factor with the capacity to modify the rheological parameters of water-based fracturing fluid, reservoir pressure can also influence the expansion behaviour and crack aperture of coal seam fractures. As illustrated in Figure 7, there is a demonstrable shift in trends of fluid viscosity and fracture length under varying reservoir pressures. This provides a valuable foundation for the effective exploitation of coalbed methane. It has been demonstrated that the apparent viscosity of water-based fracturing fluid increases gradually with an increase in reservoir pressure. Furthermore, the fluid viscosity increases rapidly under high pressure. However, it has been demonstrated that low reservoir pressure can rapidly enhance fluid viscosity [42], a phenomenon that may be associated with the development of microscopic intermolecular hydrogen bonds. As previously

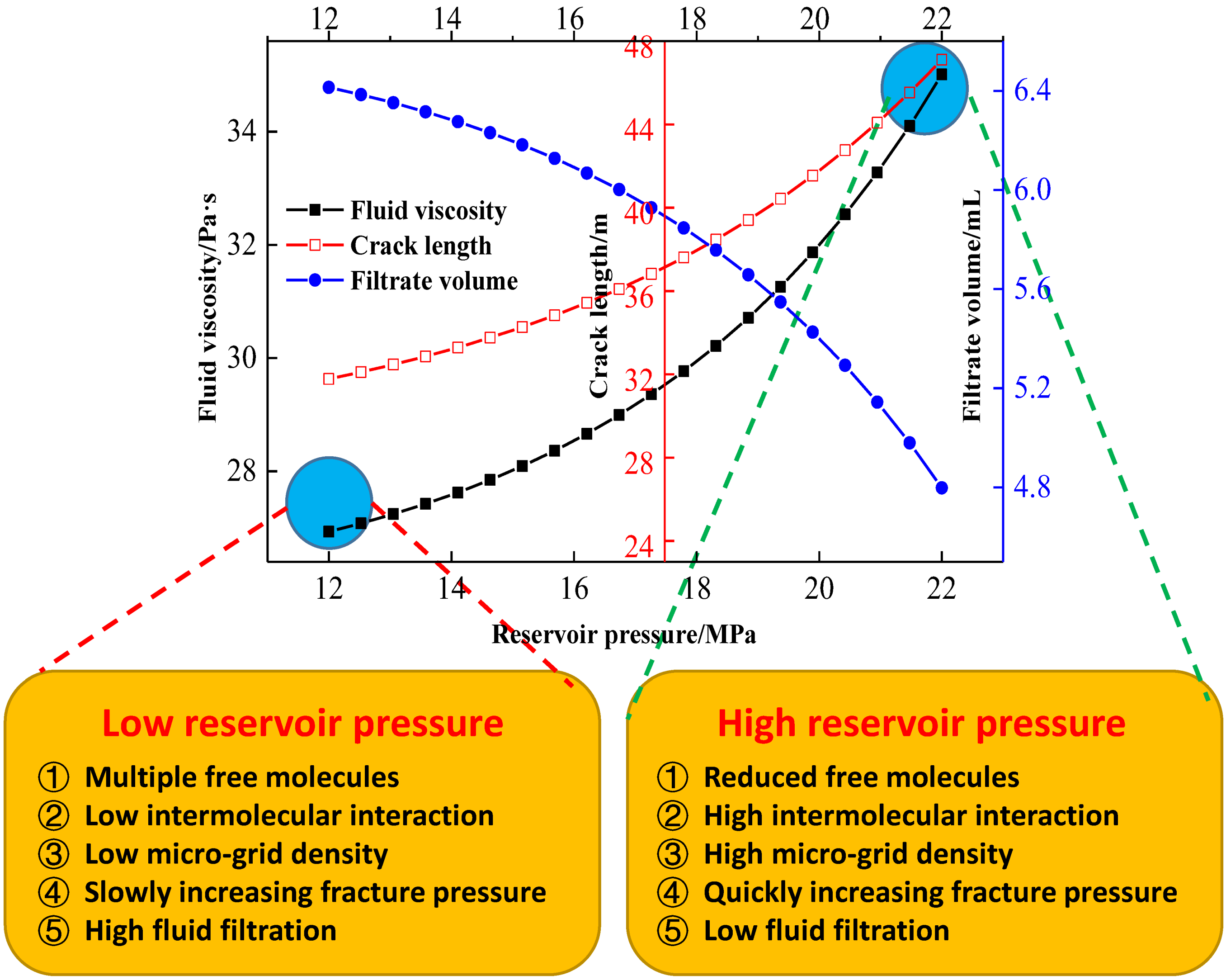

Figure 7. Effect of reservoir pressure on fluid viscosity and fracture propagation of water-based fracturing fluid.

mentioned, cross-linking agents can act as media, interacting with hydroxyl groups of guar or water molecules through polar functional groups (ester or ether groups) to form intermolecular hydrogen bonds. This is a significant microscopic theory that can express fluid viscosity [43, 44]. In conditions of low pressure, the molecules of water-based fracturing fluid are subject to a weak compression, which is insufficient to facilitate the approach of molecules in the free state and thereby enable the formation of new chemical hydrogen bonds. Furthermore, the presence of a reduced number of intermolecular hydrogen bonds in water-based fracturing fluid under low pressure renders it evident that the chemical bond density and bond energy will not be significantly increased by the squeezing process. Consequently, the number of chemical bonds and three-dimensional grid density of water-based fracturing fluid under low pressure will not cause a significant increase in fluid viscosity during the increase of reservoir pressure [45, 46]. However, an increase in reservoir pressure will have a significant effect on the fluid viscosity of water-based fracturing fluid, attributable to the rapid formation of intermolecular hydrogen bonds and the efficient base pressure of chemical bonds. Firstly, elevated reservoir pressure will result in the compression and proximity of various molecules within water-based fracturing fluid, thereby facilitating the formation of new intermolecular hydrogen bonds between molecules that were initially in a free state [47, 48]. Furthermore, the hydrogen bonds that have been constructed under low pressure are constantly squeezed due to external pressure, resulting in the formation of a short hydrogen bond with gradually increasing bond energy [49]. The two aforementioned phenomena contribute to the formation of a denser microscopic grid, which is also the key microscopic mechanism for constructing high-viscosity fluids.

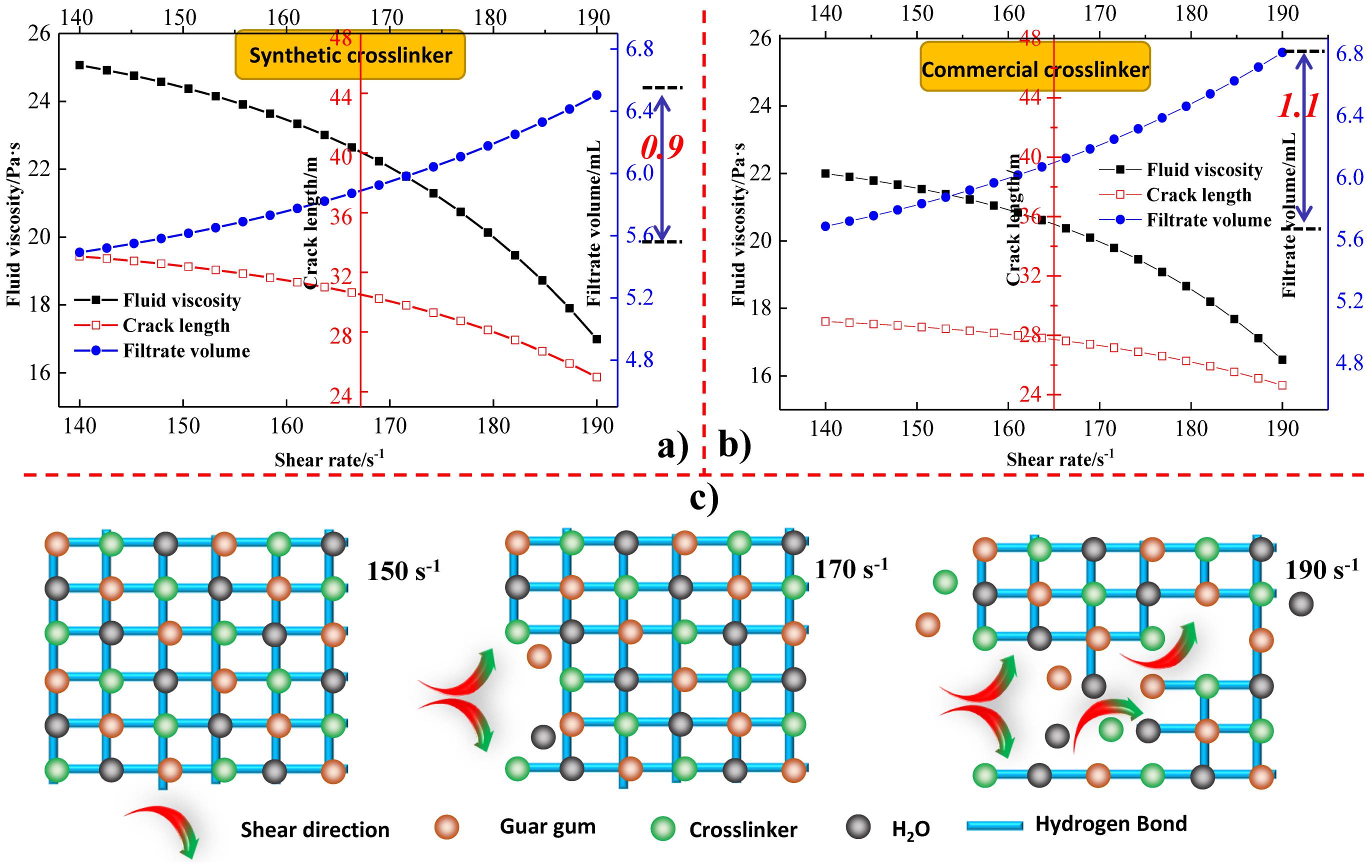

Figure 8. Effect of shear rate on fluid viscosity and crack growth behavior.

Furthermore, the pressure within the reservoir has been shown to encourage the propagation of fractures within the reservoir rock. This phenomenon is primarily attributed to the impact of reservoir pressure on fluid viscosity and the filtration process. Free molecules in water-based fracturing fluid that cannot form intermolecular hydrogen bonds through cross-linking agents are easily pressed into reservoir fractures and flow into reservoir rocks. This is not conducive to the rapid increase of fluid pressure in the reservoir to the minimum fracturing pressure. In conditions of low pressure, the molecular distance is considerable, thus hindering the prompt compression of the original free molecules into new intermolecular hydrogen bonds. This, in turn, results in an increase in free molecules seeping into the reservoir rock, which is not conducive to the expansion of fractures. However, under conditions of elevated pressure, free molecules can be rapidly compressed to approach one another, thereby forming new chemical bonds. This is an effective strategy to prevent infiltration into reservoir pores, which would otherwise result in the accumulation of a substantial volume of water-based fracturing fluid within reservoir fractures. This phenomenon is conducive to the rapid breakthrough of reservoir pressure, which can exceed the minimum fracturing pressure required for fracture expansion. Consequently, water-based fracturing fluid under high pressure is more likely to cause rock damage and fracture expansion, which is primarily attributed to the low filtration capacity of water-based fracturing fluid under high pressure.

3.5 Effect of reservoir shearing on coalbed methane recovery and fracture propagation

The intermolecular hydrogen bonds formed between molecules in the water-based fracturing fluid modify the rheological parameters and fracturing efficacy due to the shear effect during the flow process, which exerts a substantial influence on coal seam mining and reservoir transformation. As illustrated in Figure 8, the impact of fluid viscosity and fracture extension length of water-based fracturing fluid under varying shear rates can provide fundamental data for enhancing the shear resistance of water-based fracturing fluid during coal seam transformation. As demonstrated in Figure 8, the fluid viscosity of the water-based fracturing fluid exhibits a discernible inverse correlation with the shear rate within the reservoir, thereby substantiating the shear-thinning characteristics of the water-based fracturing fluid. Furthermore, the non-Newtonian fluid characteristics of the water-based fracturing fluid verify the inverse relationship between viscosity and shear in Figure 8, thereby demonstrating the accuracy of the experimental evaluation measures of fluid viscosity. Despite the demonstrated inverse proportional relationship between fluid viscosity and shear rate, a considerable disparity remains in the trend of fluid viscosity across varying shear rates.

The fluid viscosity demonstrates a reduced decline at low shear rates, while a heightened decrease in viscosity is evident at elevated shear rates. The aforementioned disparities in fluid viscosity at varying shear rates are predominantly attributable to the disparate destructive impact of shear stress on the intermolecular hydrogen bonds present within the water-based fracturing fluid (Table 2). Concurrently, the density of the three-dimensional grid structure manifests entirely disparate microscopic morphologies, a consequence of the stretching of intermolecular hydrogen bonds. The low shear stress formed by a low shear rate has been shown to exert a less destructive effect on intermolecular hydrogen bonds, and hydrogen bond stretching or exercise occurs between a few molecules. Consequently, the microscopic three-dimensional grid density remains largely unaltered due to the negligible effect of low shear stress, thereby engendering a comparatively stable fluid viscosity at low shear rates. However, elevated levels of shear stress and shear rate result in the stretching and lengthening of chemical bonds, accompanied by the rupture of intermolecular hydrogen bonds, characterised by low bond energy. It has been demonstrated by preceding studies that the three-dimensional grid density is a determining factor in the fluid viscosity of water-based fracturing fluid. Furthermore, it has been established that intermolecular hydrogen bonds directly influence the three-dimensional grid density. Consequently, a reduced shear rate can preserve the elevated viscosity of water-based fracturing fluid; however, at elevated shear rates, water-based fracturing fluid demonstrates a substantial decline in viscosity due to augmented hydrogen bond stretching and destruction.

Concurrently, the commercial cross-linker and the synthetic cross-linker exhibited comparable shear-thinning characteristics in water-based fracturing fluids, indicating that the two cross-linkers exerted analogous influences on the fluid parameters of water-based fracturing fluids. However, the

Table 2. The bond length and bond energy data between molecules under different cross-linker contents.

cross-linker contents	0	0.1%	0.2%	0.3%	0.4%	0.5%
Single Bond length, nm	105	98	98	97	97	97
Single Bond Energy, 10^{-6}J/mol	267	263	265	262	261	261
Number of Bond	3547	3596	3714	3968	4419	5346

commercial cross-linker exhibited a more rapid decrease in viscosity compared to the synthetic cross-linker. This observation is indicative of the weaker shear resistance exhibited by the water-based fracturing fluid containing the commercial cross-linker. The disparity in shear resistance between the commercial cross-linker and the synthetic cross-linker is predominantly ascribed to the discrepancy in molecular structure and the position of functional groups of the two cross-linkers. The molecular structure of the commercial cross-linker exhibits a straight chain morphology, which invariably results in the functional groups being connected exclusively to the straight chain. The presence of a reduced number of functional groups in the commercial cross-linker molecules, in conjunction with the absence of hydroxyl groups capable of interacting with a greater number of guar and water molecules, results in a diminished number of intermolecular hydrogen bonds and, consequently, a thin three-dimensional grid structure. However, the branched structure presented by the synthetic cross-linker provides a structural basis for the efficient loading of functional groups, which can enable the cross-linker to build more stable intermolecular hydrogen bonds with hydroxyl groups from multiple targets. Consequently, the synthesised cross-linker has the capacity to construct a denser micro-grid structure, which can undoubtedly reduce the hydrogen bond destruction under the same shearing action. However, under the same shearing action, the commercial cross-linker will cause less hydrogen bond destruction, which is clearly not conducive to the coal seam transformation and fracture expansion of water-based fracturing fluid.

Furthermore, the expansion behaviour of water-based fracturing fluid on coal seam fractures at varying shear rates was subjected to rigorous analysis. As demonstrated in Figure 8(b), the decline in fracture extension length is indicative of an escalating

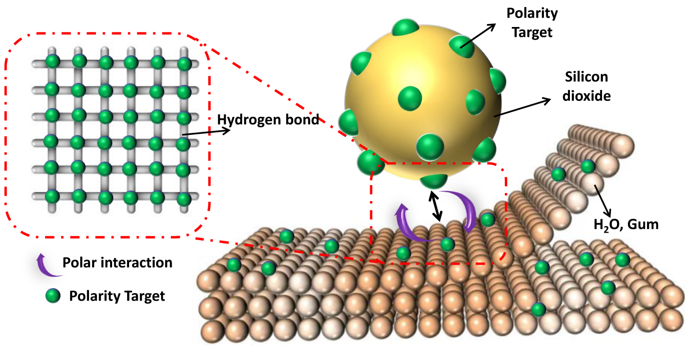

Figure 9. Chemical interactions and grid models between various substances in water-based fracturing fluids in geological reservoirs.

shear rate. The inverse relationship between the aforementioned fracture extension trend and shear rate is primarily attributed to the filtration effect of water-based fracturing fluid, which can result in alterations in pressure within the fracture and the capacity for rock damage. In circumstances where the rate of shear is minimal, there is a reduced probability of intermolecular hydrogen bonds being stretched or sheared. Consequently, a smaller number of free molecules re-enter the loose water-based fracturing fluid base fluid as a consequence of hydrogen bond breakage. However, elevated shear rates have been observed to induce a greater incidence of hydrogen bond breakage and the formation of free molecules. A significant proportion of free molecules are devoid of hydrogen bond drag and are more likely to penetrate into the reservoir rock through the fracture pores. Consequently, the free molecules formed by shear stress and the filtration effect are the primary factors contributing to the inverse relationship between shear rate and fracture extension, which will inevitably reduce the efficiency of coalbed methane extraction.

The aforementioned studies demonstrate that the incorporation and structural modification of the crosslinker promote the formation of a greater number of high-energy intermolecular hydrogen bonds among the compounds, thereby enhancing the fracture propagation capability of the modified crosslinker in water-based fracturing fluids compared with that of commercial crosslinkers. In addition, the three-dimensional spherical configuration (Figure 9) of the modified crosslinker facilitates the directional formation of dense microscopic networks between polar groups (e.g., hydroxyl groups), water molecules, and guar gum chains, which is essential for its superior fluid-loss control and fracture propagation performance. Moreover, reservoir temperature and pressure also contribute to enhanced fracture propagation, although they exert opposite effects on fluid-loss behavior. The similarity in fracture propagation under varying temperature and pressure conditions arises from the fact that both elevated temperature and pressure increase the internal fracture pressure, thereby promoting fracture initiation. However, their differing impacts on fluid loss originate from their distinct influences on intermolecular hydrogen bonding. Finally, shear action can disrupt chemical bonds and increase fluid loss, ultimately leading to a reduction in fracture pressure.

4 Conclusion

In this study, a modified boron cross-linking agent containing polar functional groups on the side chains was prepared, and the nuclear magnetic resonance hydrogen spectrum verified the accuracy of the molecular synthesis. Concurrently, a multi-coupling device was constructed for the purpose of evaluating the various properties of water-based fracturing fluids and the expansion behaviour of coal seam fractures. This device was designed to analyse the influence of different factors on water-based fracturing fluids and fracture lengths. Furthermore, the expansion model of water-based fracturing fluids in coal seam fractures was verified through reliability analysis, and the numerical model has a realistic basis for coal seam fracture expansion. The intermolecular hydrogen bond theory and the three-dimensional grid theory provide a theoretical basis for predicting the effects of other factors on fluid viscosity and fracture expansion trends by explaining the changes in the microscopic interactions between molecules in the fracturing fluid under different factors. However, the present study did not further investigate the hydrogen bonding interactions among the molecular components of the water-based fracturing fluid, nor the interfacial interactions between the fluid compounds and the reservoir rock, both of which are critical issues that warrant deeper analysis and clarification. In addition, the influence of water-based fracturing fluids on the adsorption behavior and permeability characteristics of geological reservoirs has emerged as a major research challenge in reservoir engineering.

Data Availability Statement

Data will be made available on request.

Funding

This work was supported in part by the Heilongjiang Postdoctoral Financial Assistance under Grant LBH-Z24245; in part by the Academic Success, the Introduction of Talent Research Start-up Program of Heilongjiang Bayi Agricultural University under Grant XYB202003; in part by the Young Innovative Talents Project of Heilongjiang Bayi Agricultural University under Grant ZRCQC202313.

Conflicts of Interest

The authors declare no conflicts of interest.

Ethical Approval and Consent to Participate

Not applicable.

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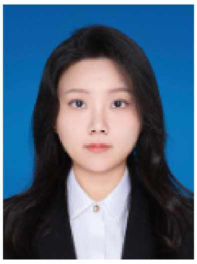

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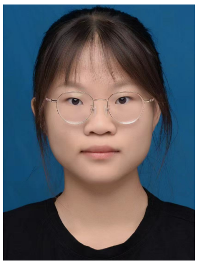

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