

## Article

# Polyethylene Composite Particles as Novel Water Plugging Agent for High-Temperature and High-Salinity Reservoirs

Bo Deng<sup>1,2,†</sup>, Ning Yang<sup>1,†</sup> , Jiangan Li<sup>2</sup>, Chenwei Zou<sup>1</sup> , Yunpu Tang<sup>2</sup>, Jianwei Gu<sup>1</sup>, Yifei Liu<sup>1,\*</sup>   
and Wei Liu<sup>1,\*</sup>

<sup>1</sup> School of Petroleum Engineering, China University of Petroleum (East China), Qingdao 266580, China; b17020080@s.upc.edu.cn (B.D.); yangning0332@163.com (N.Y.); zou\_chenwei@yeah.net (C.Z.); gjwlc@upc.edu.cn (J.G.)

<sup>2</sup> Tincy Group Energy Resources Ltd., Panjin 124013, China; li.jiangan@citictincy.com (J.L.); tang.yunpu@citictincy.com (Y.T.)

\* Correspondence: liuyifei@upc.edu.cn (Y.L.); liu.wei@upc.edu.cn (W.L.)

† These authors contributed equally to this work.

**Abstract:** Water channeling has always been one of the urgent problems during oilfield development. Especially for fractured reservoirs with high temperature, high salinity, and severe heterogeneity (e.g., deep carbonate reservoirs), it is difficult for the existing plugging agents to realize effective water plugging. In this paper, chemically stable polyethylene (PE) was selected as the main component to prepare multiscale PE composite particles that can be easily dispersed in water as a novel water plugging agent for fractured reservoir with high temperature and high salinity. The characteristics of the prepared PE composite particles, including thermal stability, salinity resistance, dispersibility, coalescence properties, and microscopic morphology, were systematically studied. Finally, the plugging performance of the particles was evaluated through visual physical simulation experiments. The prepared PE composite particles can be pulverized to a minimum of 6  $\mu\text{m}$ , and the particle size is controllable within 6  $\mu\text{m}$  to 3 mm by adjusting the pulverization parameters. The PE composite particles are easily dispersed in water by adding the dispersant, which is conducive to injectivity during the field application process. The particle size remains unchanged under the condition of salinity of 0–3.0  $\times 10^5$  mg/L, which indicates that the prepared particles have good salt-resistant stability. After high-temperature aging, the particles adhere to each other, and the size of the agglomerations reach a size dozens of times larger than the initial size of the particle, which is conducive to effective plugging in fractures. Thermal degradation behavior analysis shows that the PE composite particles could theoretically withstand a temperature of 434.4 °C. It can be seen from the SEM images that after high-temperature melting and kneading with other components, the microstructure changes from a fibrous structure to a dense flake structure. Physical simulation experiments show that the PE composite particles accumulate in fracture after injection and form effective plugging through coalescence and adhesion of the particles, thereby realizing water flow diversion.

**Keywords:** polyethylene composite particles; enhanced oil recovery; water plugging; high temperature and high salinity reservoirs



**Citation:** Deng, B.; Yang, N.; Li, J.; Zou, C.; Tang, Y.; Gu, J.; Liu, Y.; Liu, W. Polyethylene Composite Particles as Novel Water Plugging Agent for High-Temperature and High-Salinity Reservoirs. *Processes* **2023**, *11*, 3044. <https://doi.org/10.3390/pr11103044>

Academic Editors: Haiping Zhu and Qingbang Meng

Received: 31 August 2023

Revised: 10 October 2023

Accepted: 18 October 2023

Published: 23 October 2023



**Copyright:** © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

## 1. Introduction

With the increase in global energy demand and the deepening of oil exploration and development, carbonate reservoirs are increasingly important, with more than 50% of global oil and gas resources [1–3]. Carbonate reservoirs are mainly fractured reservoirs with high production, which is one of the important fields for increasing oil reserves and production in the world [4,5]. With the development of fractured carbonate reservoirs, many shortcomings such as early water breakthrough, rapid water cut rise, high natural

decline rate, and low recovery factor are gradually exposed [6]. The main reason for the above problems is the serious heterogeneity of fractured carbonate reservoirs. In the process of water flooding development, water is easy to break through along high permeability fractures, resulting in low sweep efficiency, which brings many technical problems to the development [7–9]. Therefore, the key to the efficient development of fractured carbonate reservoirs is finding a suitable water plugging agent for plugging high-permeability fractures so as to reduce water cut and improve oil recovery.

At present, the main water plugging agents used in fractured carbonate reservoirs are crosslinked polymer gels and foams [10–16]. However, many carbonate reservoirs currently developed are under harsh conditions of high temperature and high salinity [17,18]. In such reservoirs as the Tahe Oilfield in northwest China, the reservoir temperature is generally above 130 °C, and the salinity of formation brine is higher than  $2.2 \times 10^5$  mg/L [19–21]. Under such harsh conditions, crosslinked polymer gels and foams are difficult to maintain stability, resulting in a short validity period and a poor water plugging effect [22,23]. During the investigation, it was found that some thermoplastic materials exhibited excellent stability properties and strong resistance to temperature and salinity. Therefore, thermoplastic materials are intended to be applied to fracture plugging to achieve efficient water plugging under harsh conditions. Polyethylene (PE) is a typical thermoplastic material. Due to its excellent chemical stability, temperature resistance, salt resistance, and processability [24–27], the PE is selected as the main component for the preparation of a water plugging agent used in fractured carbonate reservoirs with high temperature and high salt. In addition, with the development of industry, a large amount of waste PE has been produced in past decades [28,29]. The treatment of waste PE mainly includes burial and incineration. Burying and incineration will cause serious pollution to the environment. Therefore, reusing waste PE in water plugging is of great significance for environmental protection and reducing oilfield costs.

The PE must be modified to be applied to water plugging. On the one hand, due to the large difference in fracture size (micron to millimeter scale) in the reservoir, PE needs to be pulverized into multiscale particles to meet the injection and plugging requirements for fractures of different sizes [30–32]. On the other hand, due to the hydrophobicity of PE, it is difficult to disperse into water without adding other additives, which is not conducive to injection during field application. Therefore, it is necessary to add modifiers to make the PE surface hydrophilic. At present, other components are usually introduced by melt blending to change the physical properties of thermoplastic materials [33].

The main pulverization methods include ball mill pulverization, air stream pulverization, and ultrafine pulverization [29,34]. However, it should be noted that a large amount of heat will be produced during the pulverization process, resulting in softening of the PE and difficulty in continuing pulverization. In order to solve this problem, on the one hand, the tenacity of the PE could be reduced by adding a tenacity reduction agent to make it easier to be crushed. On the other hand, it is necessary to choose a suitable pulverization process, such as the low-temperature pulverization process, to neutralize the heat generated during the pulverization process [34].

In this study, low-cost PE was used as the main raw material to prepare the modified PE composites by introducing a tenacity reduction agent, a dispersant, and a cosolvent through the melt blending method. Then the multiscale PE composite particles, which can be easily dispersed in water as a novel water plugging agent for a fractured reservoir with high temperature and high salinity, were prepared by the low-temperature pulverization process. The salt tolerance and dispersion stability, thermal stability, coalescence properties under high temperature, and microstructural characteristics of the prepared PE composite particles were evaluated. Finally, the plugging performance and water-plugging mechanism of the PE composite particles were revealed through visual physical simulation experiments. This study is of great significance for water plugging in high-temperature and high-salinity reservoirs.

## 2. Experimental

### 2.1. Materials

The polyethylene (PE) used as the raw material was provided by Beijing Zhonglai Chemical Co., Ltd., Beijing, China. The petroleum resin used as the cosolvent was provided by Shandong Tianyi Chemical Co., Ltd., Weifang, China. The calcium carbonate ( $\text{CaCO}_3$ ) used as the tenacity reduction agent was provided by Guangzhou Zhenwei Chemical Technology Co., Ltd., Guangzhou, China. The sodium dodecyl sulfate ( $\text{C}_{12}\text{H}_{25}\text{NaO}_4\text{S}$ ) used as the dispersant was provided by Linyi Lvsen Chemical Co., Ltd., Linyi, China.

### 2.2. Preparation of PE Composite Particles

#### 2.2.1. Preparation of PE Blend

The PE blend used as the intermediate products used to prepare the PE composite particles was prepared by the heating-compounding process [35–37]. The equipment used was the twin-screw extruder (Korke, Nanjing, China). The preparation process is shown in Figure 1. Firstly, the raw materials were dried in an oven at 70 °C for 12 h. Then the PE particles, cosolvent, tenacity reduction agent, and dispersant were stirred evenly in a specific proportion, and the twin-screw extruder was started at the same time. The twin-screw extruder was preheated for 30 min at 120 °C. The stirred material was added, and the temperature of each heating zone and the feeding speed of the twin-screw extruder were set as shown in Table 1. The twin-screw extruder produced the PE blend at a speed of 100 r/min, and product collection was begun after the twin-screw extruder, producing a uniform cylinder-shaped product. The collected uniform cylindrical blends (shown in Figure 2) are intermediate products used to prepare PE composite particles.

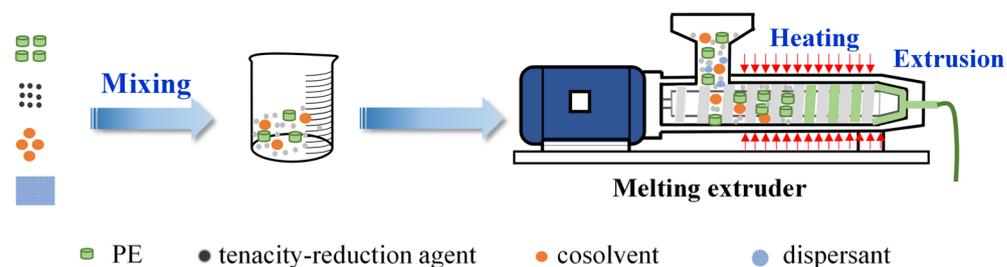


Figure 1. Schematic diagram of the preparation of PE blend.



Figure 2. Photograph of the prepared PE blends (the intermediate products used to prepare PE composite particles).

**Table 1.** The temperature setting of each heating zone of the twin-screw extruder.

Heating Zone	Zone 1	Zone 2	Zone 3	Zone 4	Zone 5	Zone 6	Zone 7	Outlet
Temperature, °C	80	130	140	150	150	140	140	140

### 2.2.2. Preparation of the Multiscale PE Composite Particles

Firstly, the prepared strip-shaped PE blends, which were used as the intermediate products for preparing the PE composite particles, were granulated into small pieces of about 3 mm in diameter by a dicing machine. Then the small pieces were poured into the pulverizer, and the shredding parameters were adjusted to prepare the PE composite particles with different sizes.

### 2.3. Characterization and Evaluation of the PE Composite Particles

#### 2.3.1. Salt Tolerance and Dispersion Stability

The PE composite particle dispersion with 1.0 wt% was prepared using brine with different salinity. The particle size of the dispersion was measured by a laser particle size analyzer (Bettersize-2000, Bettersize Instruments Ltd., Dandong, China) to study the effect of salinity on the particle size.

In addition, brine with a high salinity of  $2.5 \times 10^5$  mg/L was used to prepare the PE composite particle dispersion. The prepared dispersion was placed at room temperature for a long time, and the particle size was measured at specific time intervals to observe the change in particle size.

#### 2.3.2. Thermal Stability

Firstly, the heat resistance of the thermoplastic PE composite particles was evaluated by bottle tests. The PE composite particle dispersion was prepared with brine with a salinity of  $2.5 \times 10^5$  mg/L and sealed in thermo-resistant and pressure-resistant glass bottles, and then placed in thermostats at 120 °C and 200 °C, respectively. The change in the macro morphology of particles with aging time under high temperatures was observed.

Moreover, a simultaneous thermal analyzer (SDT-Q600, TA Instruments, New Castle, DE, USA) was applied to study the thermal stability of the prepared PE composite particles. The sample was heated from 25 °C to 800 °C at a rate of 10 °C/min, and the high-purity nitrogen was used as shielding gas as well as an atmosphere gas at the rate of 20 mL/min.

#### 2.3.3. Coalescence Characteristics

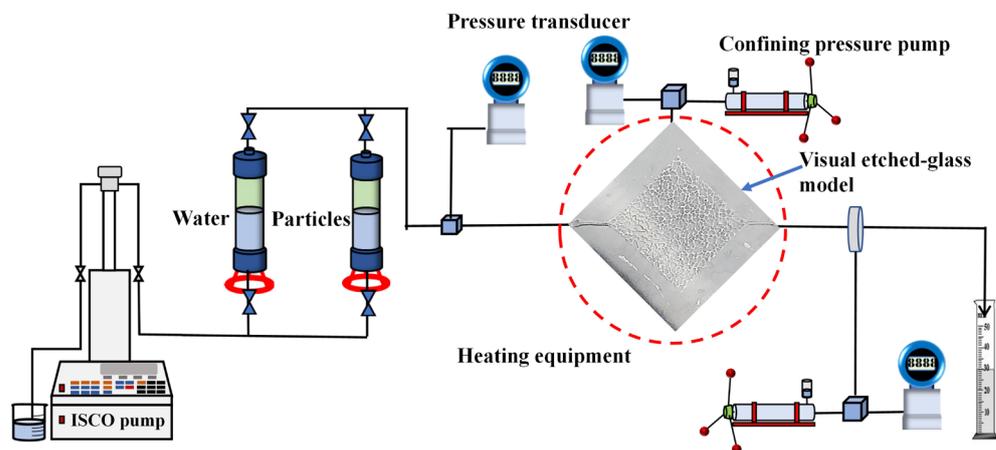
The PE composite particle dispersion with 1.0 wt% was prepared and sealed in thermo-resistant and pressure-resistant glass bottles. The bottles were placed in a thermostat at 150 °C. After aging for a specific time, the average particle size of the dispersion was measured, and the microscopic morphology of the particles was observed by a Zeiss microscope.

#### 2.3.4. Plugging Characteristics

The plugging characteristics of the prepared PE composite particles were evaluated using the visual etched-glass model. The average width of the etched channels was about 200 µm to simulate the fracture network. The experimental schematic diagram is shown in Figure 3, and the experimental process was as follows.

1. The model was cleaned and dried at 120 °C for 48 h, and the dry weight of the model was recorded.
2. The model was saturated with brine water with a salinity of  $2.5 \times 10^5$  mg/L. The weight of the saturated model was recorded, and the flow channel volume (FV) was calculated by the mass difference.
3. The heating device was set to 120 °C. Then the simulated oil was injected at a rate of 0.001 mL/min to make the model saturated with oil.

4. Brine water flooding using 0.001 mL/min until oil production was negligible.
5. The PE composite particle dispersion was injected at a rate of 0.001 mL/min with an injection volume of 1 FV. Then both ends of the model were sealed, and the model was aged at 120 °C for 24 h.
6. Subsequent brine water flooding was performed at a rate of 0.001 mL/min. Images of retention and plugging characteristics of the PE composite particles were recorded.



**Figure 3.** Experimental schematic diagram for plugging characteristics evaluation using etched-glass model.

### 3. Results and Discussion

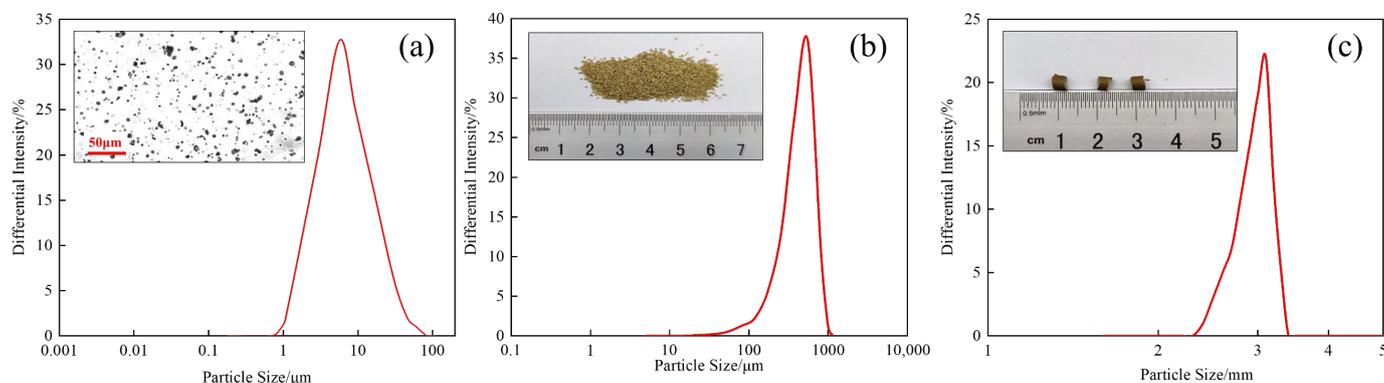
#### 3.1. Preparation of PE Composite Particles

Based on the previous experimental results we found that when the concentration of the tenacity reduction agent is above 10%, it is more conducive to the pulverization and granulation of PE composite particles. Moreover, the concentration cosolvent should be no less than 40% to ensure that the PE composite particles have good oil solubility to realize the oil–water selective plugging. After continuous optimization in the laboratory, the mass ratio of the various components in the PE composite particles was optimized as PE main agent:cosolvent:tenacity reduction agent:dispersant = 40%:40%:10%:10%.

Small fractures in carbonate reservoirs are tens of microns, and large fractures can reach several millimeters or even tens of millimeters, which requires that the particle size of the PE composite particles can be adjusted to meet the injection and plugging requirements for fractures of different scales. During the experiments, a ball mill, an ultrafine pulverizer, and a low-temperature pulverizer were used to pulverize the PE composite particles. Due to the rapid collision between the particles during the pulverization process, a large amount of heat was generated, which made it difficult to further pulverize the composite particles. Therefore, the ball mill pulverization process and the ultrafine pulverization process cannot pulverize the composite particles for a long time to meet the requirements of industrial application. The low-temperature pulverization process can quickly neutralize the heat generated by the particle collision, and the pulverization effect is significantly better than the former two. Meanwhile, the particle size can be adjusted by changing the grinding speed of the pulverizer.

Figure 4 shows photographs and particle size distributions of the PE composite particles prepared with three typical preparation parameters using the low-temperature pulverization method. The results show that the particle size of the PE composite particles presents a normal distribution, and the particle size of most particles is in a similar range. When the PE blend was pulverized with a high grinding speed of 50 Hz, the PE composite particles can be pulverized to a minimum particle size of about 6.0  $\mu\text{m}$ , as shown in Figure 4a. When the grinding speed decreased to 30 Hz, the average particle size of the prepared PE composite particles was about 380.0  $\mu\text{m}$ , as shown in Figure 4b. When the intermediate products were granulated only by a dicing machine without being pul-

verized by the pulverizer, the prepared PE composite particles were with a maximum particle size of about 3.0 mm, as shown in Figure 4c. It can be seen from the above results that the particle size range of the prepared PE composite particles is controllable from 6.0  $\mu\text{m}$  to 3.0 mm, which meets the injection and plugging requirements for fractures of different scales.



**Figure 4.** Photographs and particle size distributions of the PE composite particles prepared with different preparation parameters: (a) the PE composite particles prepared with grinding speed of 50 Hz; (b) the PE composite particles prepared with grinding speed of 30 Hz; (c) the PE composite particles prepared only using a dicing machine without being pulverized.

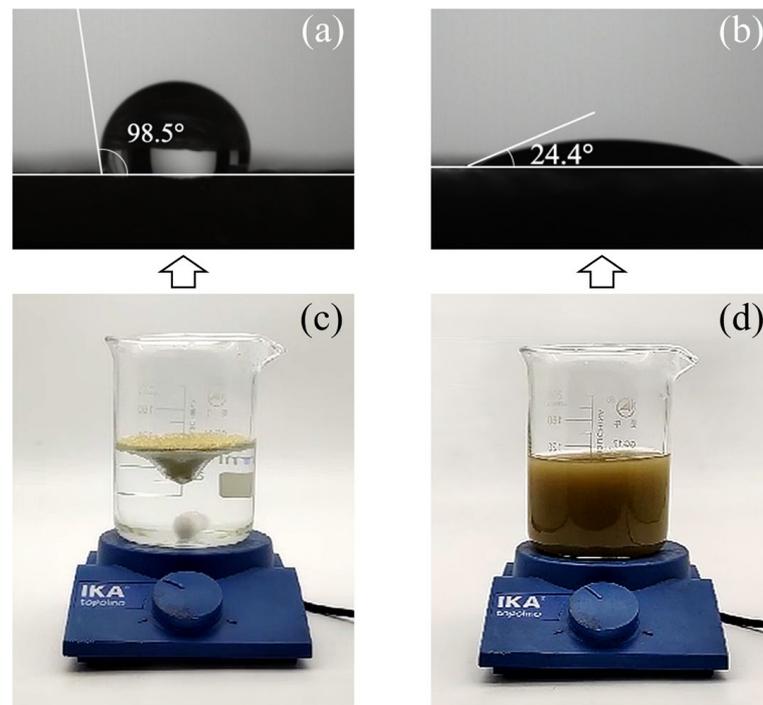
The PE particles are difficult to disperse in water, especially when the particles are pulverized to the micron scale, due to the hydrophobicity of PE main agent, which is not conducive to the field application. Therefore, the dispersant was added during the preparation process of the PE composite particles to improve the dispersibility in water. The surface hydrophilicity and the dispersibility of composite particles before and after adding the dispersant were studied. The smooth sheets of PE composites before and after the addition of the dispersant were prepared using a tableting machine, and the surface hydrophilicity was characterized by the water contact angle using the contact angle measurement instrument. The results are shown in Figure 5. It can be seen from Figure 5a the contact angle was  $98.5^\circ$  before adding the dispersant, which indicated that the surface of composite particles was hydrophobic in this case. The hydrophobicity of the surface makes it difficult for the composite particles to be dispersed in water, as shown in Figure 5c. In addition, the water contact decreased from  $98.5^\circ$  to  $24.4^\circ$  after adding the dispersant, as shown in Figure 5b, which indicated that the hydrophilicity of the composite particles increased significantly after the dispersant was added. The improvement in surface hydrophilicity was conducive to the dispersion of the composite particles in water. The PE composite particles prepared by adding the dispersant could be uniformly dispersed in water after several seconds of stirring, as shown in Figure 5d.

### 3.2. Salt and Temperature Tolerance of the Prepared PE Composite Particles

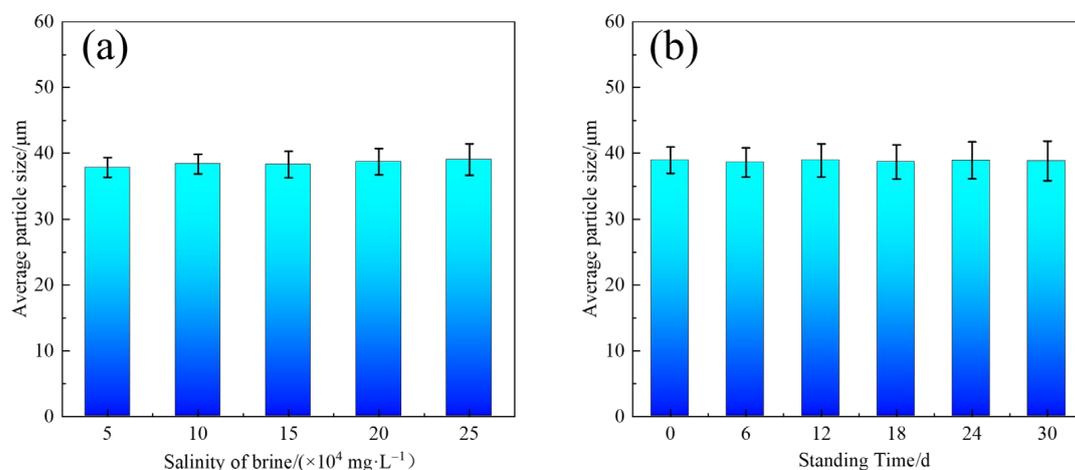
#### 3.2.1. Salt Tolerance and Dispersion Stability

During the field application process, the PE composite particles are often prepared by formation brine with high salinity. For example, in Tahe Oilfield in northwest China, the salinity of brine used for preparing the dispersions exceeds  $2.0 \times 10^5$  mg/L. The salinity of the brine for preparing the dispersion has a significant impact on the dispersion stability. The salt ions can compress the electric double layer to make the particles aggregate, which is not conducive to the field injection of PE composite particle dispersion. Therefore, it is necessary to study the salt resistance and dispersion stability of the PE composite particles. The effects of salinity and standing time on the average particle size of the dispersions are shown in Figure 6. The results in Figure 6a show that with the increase in salinity, the average particle size in the dispersions remained basically unchanged, indicating that the PE composite particles did not aggregate. Even if the salinity of the brine increased

to  $2.5 \times 10^5$  mg/L, the average particle size of the prepared dispersion was still basically consistent with that of the dispersion prepared with ultrapure water, indicating that the prepared PE composite particles had good salt tolerance. The results of long-term resting experiments are shown in Figure 6b. The PE composite particles have good dispersion stability under high-salinity ( $2.5 \times 10^5$  mg/L,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+} > 1.0 \times 10^4$  mg/L) conditions with no significant change in particle size after 30 days of standing. Therefore, PE composite particles can be formulated directly with output water for field application without affecting their particle properties.



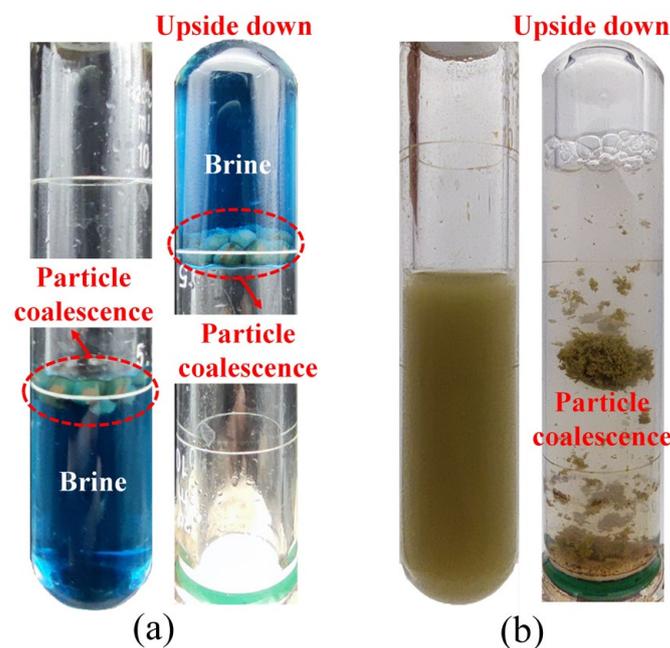
**Figure 5.** Surface hydrophilicity and dispersibility of composite particles before and after adding the dispersant: (a) contact angle before adding the dispersant (b) contact angle after adding the dispersant; (c) poor water dispersion before adding dispersant; (d) excellent water dispersion after adding dispersant.



**Figure 6.** Effects of salinity and standing time on the average particle size of the dispersions. (a) The effect of salinity of brine on average particle size; (b) the effect of standing time on the average particle size.

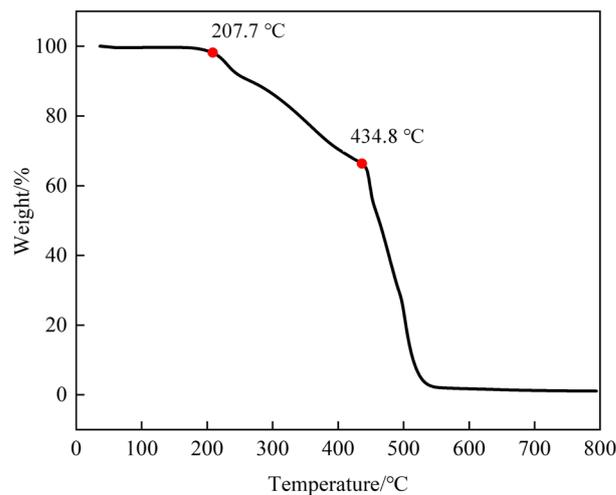
### 3.2.2. Thermal Stability

The temperature of deep carbonate reservoirs generally exceeds 120 °C, and even some ultra-high temperature reservoirs exceeding 180 °C have been found, which puts forward higher requirements for the temperature resistance of water-plugging agents. In this part, the thermal stability of the PE composite particles was evaluated. The PE composite particles were dispersed in brine with a salinity of  $2.5 \times 10^5$  mg/L and sealed in thermo-resistant and pressure-resistant glass bottles. The dispersions were placed in 200 °C and 120 °C thermostats, respectively, to observe the change in particle morphology. It can be seen from Figure 7 that after 30 days of high temperature aging, the particles did not decompose, but gathered together to form agglomerates. Especially after aging at 200 °C, the formed agglomerate could even seal the brine at the bottom of the inverted glass bottle, as shown in Figure 7a. The preliminary bottle test results showed that the prepared PE composite particles had excellent thermal stability, and the temperature tolerance could reach above 200 °C.



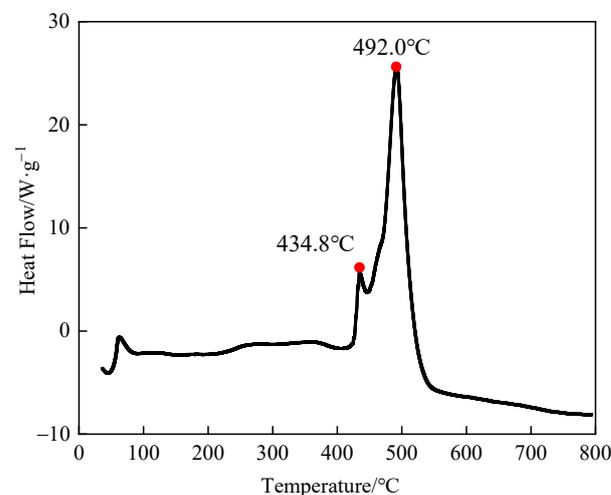
**Figure 7.** Particle morphology photos before and after high temperature aging: (a) aging at 200 °C (b) aging at 120 °C.

In order to further evaluate the thermal stability of PE composite particles, thermal degradation behavior was investigated. Thermal degradation methods, including thermogravimetric (TG) analysis and differential scanning calorimetry (DSC), are commonly used to evaluate the temperature resistance of materials [38,39]. From the TG curve in Figure 8, it can be seen that PE composite particles began to decompose slowly at 207.7 °C and decomposed rapidly at 434.8 °C. The TG curve showed two obvious inflection points because PE composite particles were processed by multiple components. Since the decomposition temperature of the dispersant (sodium dodecyl sulfate) in the PE composite particles was about 200 °C, it can be inferred that the first inflection point of the TG curve was caused by the decomposition of the dispersant. However, the PE main agent and the cosolvent (petroleum resin) with high proportions of mass began to decompose rapidly after 400 °C. The TG results indicated that the temperature resistance of the prepared PE composite particles reached above 200 °C.



**Figure 8.** The TG curve of the PE composite particles.

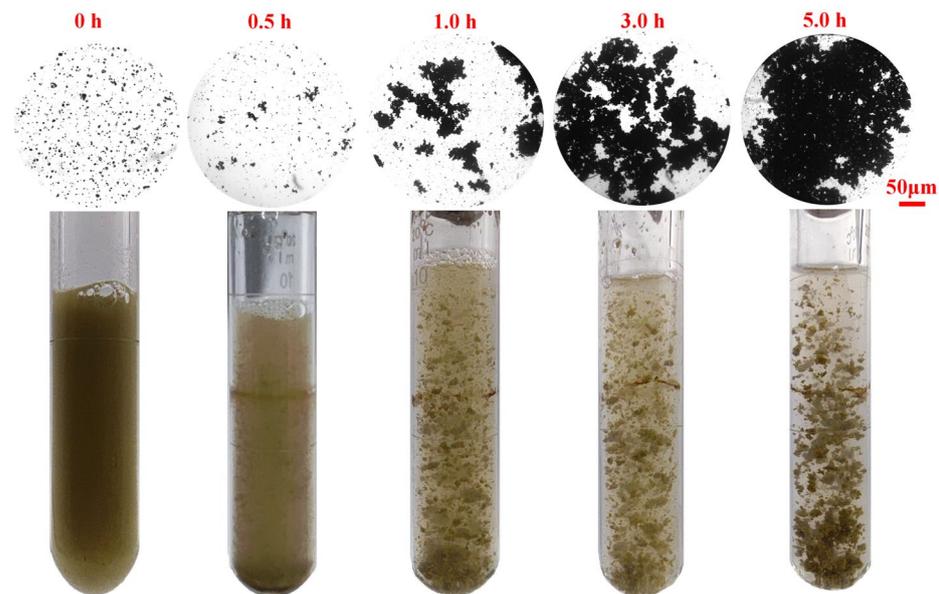
In addition, it can be seen from the DSC curve shown in Figure 9 that there were two obvious endothermic peaks during the heating process of the PE composite particles, which were at 434.8 °C and 492.0 °C, respectively. This is because the main agent and the cosolvent in the composite particles are both thermoplastic materials, and both of them absorb a lot of heat after being heated, resulting in structural changes. Combined with the TG data, although the mass had a certain loss, the main structure was not destroyed after the PE composite particles were heated to above 207.7 °C. This indicates that the prepared PE composite particles have excellent temperature resistance of above 200 °C and fully meet the application requirements of ultra-deep reservoirs with high temperature.



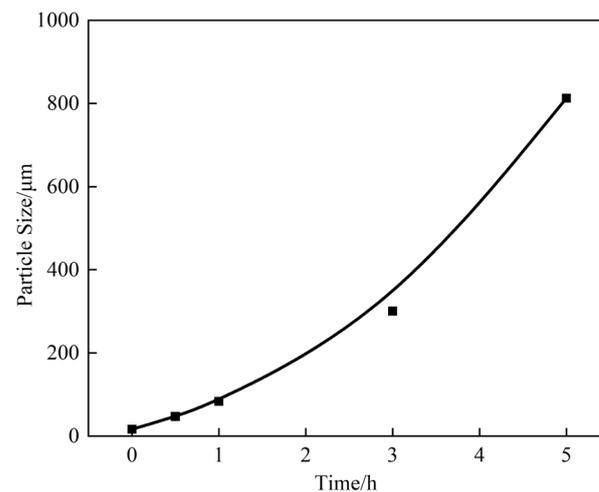
**Figure 9.** The DSC curve of the PE composite particles.

### 3.3. Coalescence Characteristics of the Prepared PE Composite Particles

Because the prepared PE composite particles are temperature sensitive, the state change process of particles after heating needs to be studied, because this has a great influence on the plugging characteristics of the particles after being injected into the high-temperature reservoir. Particles with an average size of 16.5  $\mu\text{m}$  were used to prepare the dispersions with a mass concentration of 1.0%. The brine used to prepare the dispersions had a salinity of  $2.5 \times 10^5$  mg/L. The sealed dispersions were placed at 150 °C to investigate the changes in particle size and morphology with the aging time. The results are shown in Figures 10 and 11.



**Figure 10.** Changes in macroscopic and microscopic morphology with aging time at 150 °C.

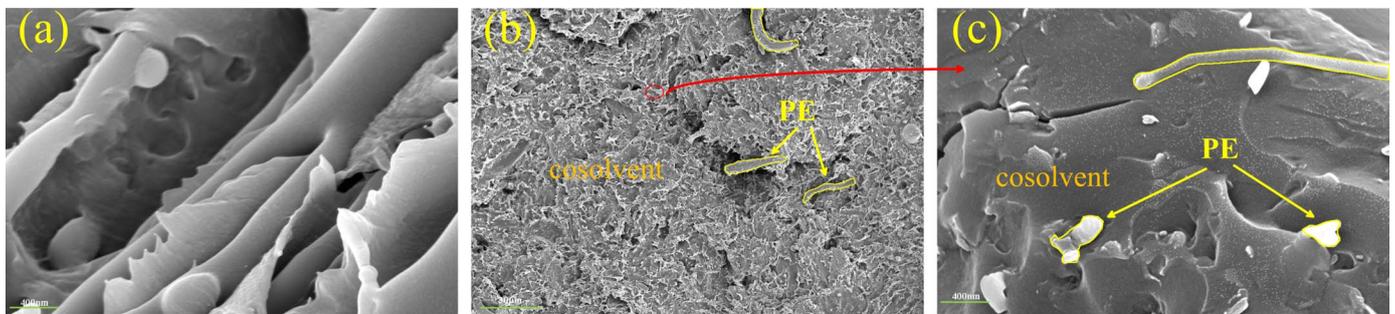


**Figure 11.** Change in average particle size with aging time at high temperature.

From the macroscopic photograph of the dispersions in the glass bottles in Figure 10, it can be seen that the particles were uniformly dispersed at the beginning and gradually agglomerated after aging for about 1 h. The dynamic process of particle aggregation can be seen more accurately from the microscopic photos. After high-temperature aging for 0.5 h, only a small number of particles agglomerated, and the diameter of the agglomerations was about 50–100 μm. After high-temperature aging for more than 1 h, the aggregation rate increased significantly, and a large number of agglomerations with diameter of about 200 μm were observed. With the further extension of high temperature aging time, the size of the agglomerations reached hundreds of microns. Furthermore, the change in average particle size in dispersion with aging time was measured, as shown in Figure 11. It can be seen that the average particle size increased from 16.5 μm to more than 812.7 μm, and the coalescence expansion ratio reached dozens of times, which was consistent with the results shown in the photos in Figure 10. The coalescence characteristics of the PE composite particles at high temperature are conducive to effective plugging of fractures after the particles have been injected into high-temperature reservoirs. It is worth noting that the PE composite particles coalesce rapidly only under static conditions. In the flow state, the particles are basically not agglomerated, so there is no need to worry about the problems of particle agglomeration and injection difficulties during the injection process.

### 3.4. Microstructure

The microstructure of the composite material determines its performance characteristics [40]. In this part, the microstructure characteristics of the PE main agent and the prepared PE composite particles were analyzed by the scanning electron microscope (SEM). The SEM images are shown in Figure 12. It can be seen that the micro-morphology of the PE main agent is with a fibrous structure, while that of the prepared PE composite particles is with a dense flake structure. This indicated that the addition of additives significantly changed the microstructure of the PE main agent. At the same time, it can be seen from Figure 12b,c that the PE and the cosolvent (petroleum resin) are mixed uniformly, and small-sized fibrous PE is uniformly dispersed in the cosolvent, indicating that the PE and the cosolvent have good compatibility. In general, material with a fibrous structure has high tenacity, which is not conducive to its pulverization [41–43]. However, after adding the cosolvent and the tenacity-reduction agent, the structure of the PE composites changed from a fibrous structure to a dense flake structure, which significantly reduced the tenacity of the material. This is the main reason why the prepared PE composites could be pulverized to the micrometer scale.



**Figure 12.** The SEM images: (a) the PE main agent with 30,000 $\times$  magnification; (b) the PE composite particle with 500 $\times$  magnification; (c) the PE composite particle with 30,000 $\times$  magnification.

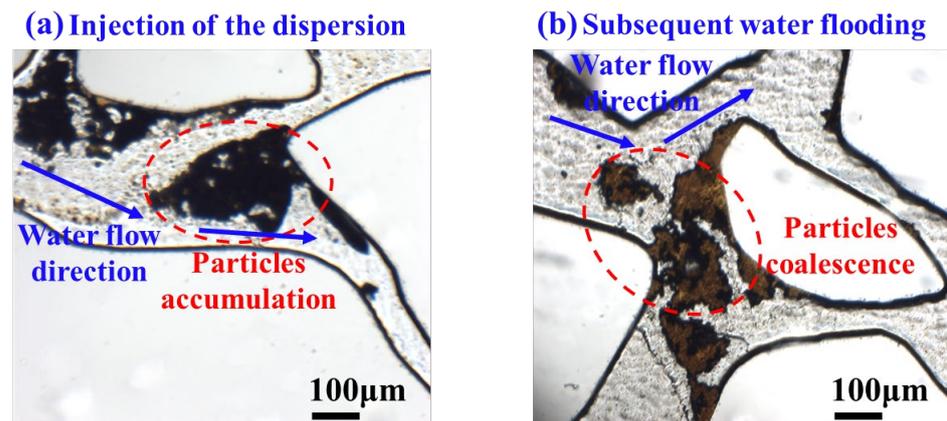
### 3.5. Water Plugging Mechanism

The previous discussions showed that the prepared PE composite particles have obvious coalescence characteristics under high temperature, and the size of the agglomerations is dozens of times larger than the initial size of the PE composite particles, which is conducive to effective plugging after the particles have been injected into reservoirs. In this part, the etched-glass model was used to evaluate the plugging characteristics of the PE composite particles.

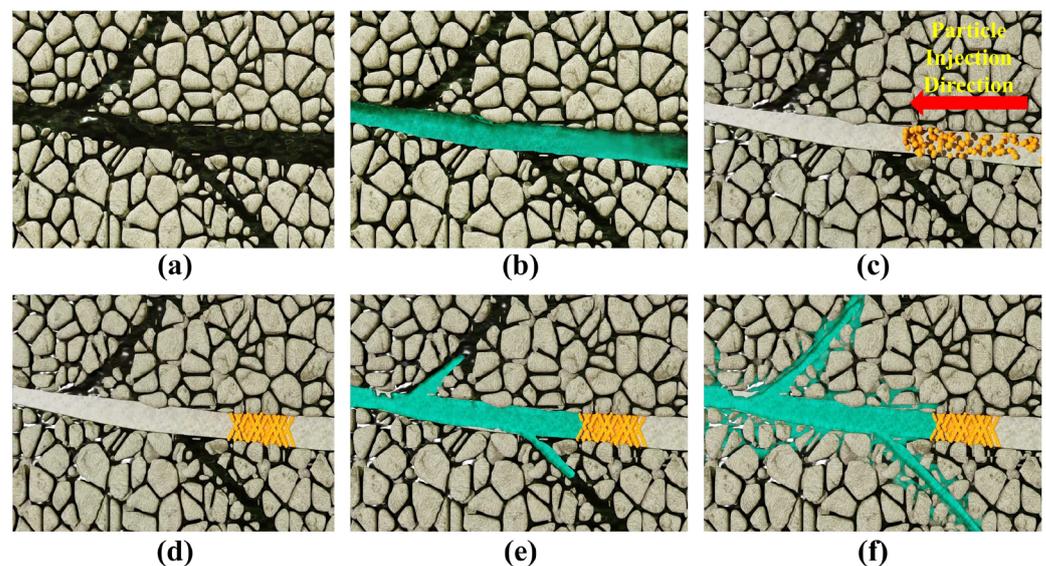
The premise of effective plugging by the PE composite particles is the retention and accumulation of the particles in flow channels. It was found during the experiments that the PE composite particles were easier to accumulate at positions where the fracture width changed, as shown in Figure 13a. After the injection of the particles, under the action of high temperature, the particles coalesced with each other or adhered to the surface of the fracture. The coalescence and adhesion of the particles formed effective plugging in the channeling fracture, which caused the subsequently injected water to be diverted to other regions and thus improved the sweep efficiency of subsequent water flooding, as shown in Figure 13b.

Based on the above results and discussions, the water plugging mechanism by the prepared PE composite particles is proposed, as shown in Figure 14. Fractures of different sizes, either naturally or formed by hydraulic fracturing, exist in the reservoir, as shown in Figure 14a. During the water flooding process, the water breakthrough forms in the large-sized fracture, which results in a low sweep efficiency of the injected water and a high remaining oil saturation in the un-swept zones. The water flooding becomes ineffective. The remaining oil cannot be displaced out any more, as shown in Figure 14b. The PE composite particles are then injected into the reservoir and accumulated in the large-sized

fractures. Under the action of high temperature in the reservoir, the particles coalesce with each other, which forms effective plugging in the large-sized fracture, as shown in Figure 14c,d. During the subsequent water flooding process, the water is diverted to the previously un-swept zones with high remaining oil saturation (i.e., small-sized fractures or pores) because of the plugging of the PE composite particles in the large-sized channeling fracture. Thus, the remaining oil in the small-sized fractures or pores can be efficiently displaced by subsequent water flooding, as shown in Figure 14e,f, which reduces the water production and increases the oil production in the subsequent production process of the oil well.



**Figure 13.** The accumulation and plugging characteristics of the PE composite particles.



**Figure 14.** Schematic diagram of the water plugging mechanism by the PE composite particles: (a) initial state reservoir; (b) water flooding; (c) injection of PE composite particles; (d) plugging of large-sized fracture; (e) subsequent water flooding; (f) remaining oil recovery.

#### 4. Conclusions

In this paper, multiscale PE composite particles as a novel water plugging agent for a fractured reservoir with high temperature and high salinity were prepared. The characteristics of the prepared PE composite particles, including thermal stability, salinity resistance, dispersibility, coalescence properties, and microscopic morphology, were systematically studied. And the plugging performance of the particles was evaluated through visual physical simulation experiments. The major conclusions are summarized as follows:

1. The prepared PE composite particles can be pulverized to a minimum of 6  $\mu\text{m}$  and the particle size is controllable within 6  $\mu\text{m}$  to 3 mm by adjusting the preparation parameters.
2. The prepared PE composite particles have good salt resistance and dispersibility. The particles can be easily dispersed in water and stably dispersed in brine with a salinity of up to  $3.0 \times 10^5$  mg/L.
3. The prepared PE composite particles have excellent thermal stability. The temperature resistance can reach above 400 °C.
4. After high-temperature aging, the particles adhere to each other, and the size of the agglomerations reach dozens of times larger than the initial size of the particle, which is conducive to effective plugging in fractures.
5. Physical simulation experiments show that the PE composite particles accumulate in a fracture after injection and form effective plugging through coalescence and adhesion of the particles, thereby realizing water flow diversion.

**Author Contributions:** Data curation, B.D., J.L. and Y.T.; Funding acquisition, Y.L.; Investigation, B.D., N.Y. and J.L.; Methodology, Y.L. and W.L.; Supervision, Y.L. and W.L.; Visualization, N.Y. and C.Z.; Writing—original draft, B.D. and N.Y.; Writing—review & editing, Y.L. and J.G. All authors have read and agreed to the published version of the manuscript.

**Funding:** The authors acknowledge the financial support of the National Natural Science Foundation of China (52104061), the Shandong Provincial Natural Science Foundation (ZR2021QE075), and the Fundamental Research Funds for the Central Universities (20CX06090A, 23CX06007A).

**Data Availability Statement:** Data are contained within the article.

**Conflicts of Interest:** The authors declare no conflict of interest.

## References

1. Zou, C.; Yang, Z.; Tao, S.; Li, W.; Wu, S.; Hou, L.; Zhu, R.; Yuan, X.; Wang, L.; Gao, X.; et al. Nano-hydrocarbon and the accumulation in coexisting source and reservoir. *Pet. Explor. Dev.* **2012**, *39*, 15–32. [[CrossRef](#)]
2. Thomas, S. Enhanced Oil Recovery—An Overview. *Oil Gas Sci. Technol. Rev. IFP* **2008**, *63*, 9–19. [[CrossRef](#)]
3. Gao, M.; Liu, P.; Xue, Q.; Zhao, M.; Guo, X.; You, Q.; Dai, C. Non-ionic polar small molecules induced transition from elastic hydrogel via viscoelastic wormlike micelles to spherical micelles in zwitterionic surfactant systems. *J. Mol. Liq.* **2022**, *359*, 119343. [[CrossRef](#)]
4. Allan, J.; Sun, S.Q. Controls on recovery factor in fractured reservoirs: Lessons learned from 100 fractured fields. In Proceedings of the SPE Annual Technical Conference and Exhibition, Denver, CO, USA, 5–8 October 2003. [[CrossRef](#)]
5. Yan, X.; Kang, Y.; Xu, C.; Shang, X.; You, Z.; Zhang, J. Fracture plugging zone for lost circulation control in fractured reservoirs: Multiscale structure and structure characterization methods. *Powder Technol.* **2020**, *370*, 159–175. [[CrossRef](#)]
6. Zhao, R.; Zhao, H.; Yue, X.A.; Wu, Y.; Ke, W. Enhanced heavy oil recovery from fractured vuggy carbonated reservoir of Tahe oilfield with  $\text{N}_2/\text{CO}_2$  gas mixture. In *International Oil and Gas Conference and Exhibition in China*; OnePetro: Richardson, TX, USA, 2010. [[CrossRef](#)]
7. Xiao, Y.; Zhang, Z.W.; Jiang, T.W.; Cai, Z.Z.; Deng, X.L.; Zhou, J.F. Dynamic and static combination method for fracture-vug unit division of fractured-vuggy reservoirs. *Arab. J. Sci. Eng.* **2018**, *43*, 2633–2640. [[CrossRef](#)]
8. Xu, C.; Zhang, H.; Kang, Y.; Zhang, J.; Bai, Y.; Zhang, J.; You, Z. Physical plugging of lost circulation fractures at microscopic level. *Fuel* **2022**, *317*, 123477. [[CrossRef](#)]
9. Lu, J.; Goudarzi, A.; Chen, P.; Kim, D.H.; Pope, G.A. Surfactant enhanced oil recovery from naturally fractured reservoirs. In Proceedings of the SPE Annual Technical Conference and Exhibition, San Antonio, TX, USA, 8–10 October 2012. [[CrossRef](#)]
10. Zou, C.; Dai, C.; Liu, Y.; You, Q.; Ding, F.; Huang, Y.; Sun, N. A novel self-degradable gel (SDG) as liquid temporary plugging agent for high-temperature reservoirs. *J. Mol. Liq.* **2023**, *386*, 122463. [[CrossRef](#)]
11. Falls, A.H.; Hirasaki, G.J.; Patzek, T.W.; Gauglitz, D.A.; Miller, D.D.; Ratulowski, T. Development of a Mechanistic Foam Simulator: The Population Balance and Generation by Snap-Off. *SPE Reserv. Eng.* **1988**, *3*, 884–892. [[CrossRef](#)]
12. Schramm, L.L. Metals and applications in the petroleum industry. *Adv. Chem.* **1992**, *231*, 3–24.
13. Farajzadeh, R.; Lotfollahi, M.; Eftekhari, A.A.; Rossen, W.R.; Hirasaki, G.J.H. Effect of Permeability on Implicit-Texture Foam Model Parameters and the Limiting Capillary Pressure. *Energy Fuels* **2015**, *29*, 3011–3018. [[CrossRef](#)]
14. Conn, C.A.; Ma, K.; Hirasaki, G.J.; Biswal, S.L. Visualizing oil displacement with foam in a microfluidic device with permeability contrast. *Lab Chip* **2014**, *14*, 3968–3977. [[CrossRef](#)] [[PubMed](#)]

15. Diwu, P.; Jiang, B.; Hou, J.; You, Z.; Wang, J.; Sun, L.; Ju, Y.; Zhang, Y.; Liu, T. Experimental Study on Expansion Characteristics of Core-Shell and Polymeric Microspheres. *J. Nanotechnol.* **2018**, *2018*, 7602982. [[CrossRef](#)]
16. Liu, Y.-F.; Zou, C.-W.; Lou, X.-G.; Gao, M.-W.; Zhao, G.; Zhao, M.-W.; Dai, C.-L. Experimental investigation on migration and retention mechanisms of elastic gel particles (EGPs) in pore-throats using multidimensional visualized models. *Pet. Sci.* **2022**, *19*, 2374–2386. [[CrossRef](#)]
17. Wang, Z.; Wang, Z.; Zeng, F.; Guo, P.; Xu, X.; Deng, D. The material balance equation for fractured vuggy gas reservoirs with bottom water-drive combining stress and gravity effects. *J. Nat. Gas Sci. Eng.* **2017**, *44*, 96–108. [[CrossRef](#)]
18. Long, Q.L.; Zhu, H.J.; Xie, H.X.; Bai, F.L.; Chen, Q.F. Laboratory study of water shutoff in the fracture-cavity carbonate reservoir. *Pet. Explor. Dev.* **2009**, *36*, 108–112. [[CrossRef](#)]
19. Li, Z.B.; Fan, X.M. Investigation of Well Logging Evaluation Methods for Carbonate Reservoirs in Tahe Oilfield, Tarim Basin. *Mar. Orig. Pet. Geol.* **2002**, *112*, 104104. [[CrossRef](#)]
20. Jin, Z.; Zhu, D.; Hu, W.; Zhang, X.F.; Zhang, J.T. Mineralogical and geochemical features of hydrothermally dissolved carbonate reservoir rocks in Tarim Basin, NW China. *Geochim. Cosmochim. Acta Suppl.* **2005**, *69*, A847.
21. Li, J.F.; Zhao, Q.; Hao, S.L.; Li, Z.H. Physical modeling of the fracture-cave systems of carbonate reservoirs in Tahe oilfield. *Geophys. Prospect. Pet.* **2005**, *44*, 428. [[CrossRef](#)]
22. Fang, J.; Wang, J.; Wen, Q.; Fang, S.; He, X.; Ma, Y.; Wu, Y.; Dai, C. Research of phenolic crosslinker gel for profile control and oil displacement in high temperature and high salinity reservoirs. *J. Appl. Polym. Sci.* **2018**, *135*, 46075. [[CrossRef](#)]
23. Kamal, M.S.; Sultan, A.S.; Al-Mubaiyedh, U.A.; Hussein, I.A.; Feng, Y. Rheological Properties of Thermoviscosifying Polymers in High-temperature and High-salinity Environments. *Can. J. Chem. Eng.* **2015**, *93*, 1194–1200. [[CrossRef](#)]
24. Oladele, I.O.; Oladejo, M.O.; Adediran, A.A.; Makinde-Isola, B.A.; Owa, A.F.; Akinlabi, E.T. Influence of designated properties on the characteristics of dombeya buettneri fiber/graphite hybrid reinforced polypropylene composites. *Sci. Rep.* **2020**, *10*, 11105. [[CrossRef](#)] [[PubMed](#)]
25. Gong, J.; Jia, R.L.; Hou, Y.F. Study on Turbulent Ultrafine Pulverization of Low Density Polyethylene at Room Temperature. *J. Lanzhou Univ. Technol.* **2006**, *32*, 3. [[CrossRef](#)]
26. Li, H.; Ma, Z.X. Crushing of waste plastics. *China Plast.* **2001**, *4*, 88–92.
27. Zhang, X.; Wang, Z.; Cong, L.; Nie, S.; Li, J. Effects of Fiber Content and Size on the Mechanical Properties of Wheat Straw/Recycled Polyethylene Composites. *J. Polym. Environ.* **2020**, *28*, 1833–1840. [[CrossRef](#)]
28. La Mantia, F.; Morreale, M. Mechanical properties of recycled polyethylene ecocomposites filled with natural organic fillers. *Polym. Eng. Sci.* **2006**, *46*, 1131–1139. [[CrossRef](#)]
29. Goodarzi, V.; Ahmadi, Z.; Saeb, M.R.; Hemmati, F.; Ghaffari, M.; Formela, K. Weldability of pipe grade polyethylenes as realized from thermal and mechanical properties assessments. *J. Polym. Eng.* **2016**, *36*, 853–860. [[CrossRef](#)]
30. Wang, X.; Zhang, H.; Li, Y.; Yang, Y.; Shan, H.; Xiao, Z. Laboratory development and field application of novel cement system for cementing high-temperature oil wells. In Proceedings of the SPE Canada Unconventional Resources Conference, Calgary, AB, Canada, 19–21 October 2010. [[CrossRef](#)]
31. Cheng, T.; Hou, J.; Yang, Y.; You, Z.; Liu, Y.; Zhao, F.; Li, J. Study on the Plugging Performance of Bilayer-Coating Microspheres for In-Depth Conformance Control: Experimental Study and Mathematical Modeling. *Ind. Eng. Chem. Res.* **2019**, *58*, 6796–6810. [[CrossRef](#)]
32. Liu, J.; Li, L.; Xu, Z.; Sun, Y.; Wu, Y.; Dai, C. Biomimetic functional hydrogel particles with enhanced adhesion characteristics for applications in fracture conformance control. *J. Ind. Eng. Chem.* **2022**, *106*, 482–491. [[CrossRef](#)]
33. He, X.; Zha, X.; Zhu, X.; Qi, X.; Liu, B. Effect of short chain branches distribution on fracture behavior of polyethylene pipe resins. *Polym. Test.* **2018**, *68*, 219–228. [[CrossRef](#)]
34. Chen, Y.; Li, Y.; Zou, H.; Liang, M. Effect of solid-state shear milling on structure and properties of HDPE/UHMWPE blends. *J. Appl. Polym. Sci.* **2013**, *131*, 1001–1007. [[CrossRef](#)]
35. Salehiyan, R.; Yoo, Y.; Choi, W.J.; Hyun, K. Characterization of Morphologies of Compatibilized Polypropylene/Polystyrene Blends with Nanoparticles via Nonlinear Rheological Properties from FT-Rheology. *Macromolecules* **2014**, *47*, 4066–4076. [[CrossRef](#)]
36. Michalska-Požoga, I.; Rydzkowski, T.; Mazur, P.; Sadowska, O.; Thakur, V.K. A study on the thermodynamic changes in the mixture of polypropylene (PP) with varying contents of technological and post-user recyclates for sustainable nanocomposites. *Vacuum* **2017**, *146*, 641–648. [[CrossRef](#)]
37. Chiang, T.-C.; Liu, H.-L.; Tsai, L.-C.; Jiang, T.; Ma, N.; Tsai, F.-C. Improvement of the mechanical property and thermal stability of polypropylene/recycled rubber composite by chemical modification and physical blending. *Sci. Rep.* **2020**, *10*, 2432. [[CrossRef](#)] [[PubMed](#)]
38. Ahmetli, G.; Kocaman, S.; Ozaytekin, I.; Bozkurt, P. Epoxy composites based on inexpensive char filler obtained from plastic waste and natural resources. *Polym. Compos.* **2013**, *34*, 500–509. [[CrossRef](#)]
39. Liu, X.; Wang, Y.; Yu, L.; Tong, Z.; Chen, L.; Liu, H.; Li, X. Thermal degradation and stability of starch under different processing conditions. *Starch-Stärke* **2013**, *65*, 48–60. [[CrossRef](#)]
40. Pärpärä, E.; Darie, R.N.; Popescu, C.-M.; Uddin, A.; Vasile, C. Structure–morphology–mechanical properties relationship of some polypropylene/lignocellulosic composites. *Mater. Des.* **2014**, *56*, 763–772. [[CrossRef](#)]
41. Zhao, Y.; Zhu, Y.; Sui, G.; Chen, F.; Fu, Q. Tailoring the crystalline morphology and mechanical property of olefin block copolymer via blending with a small amount of UHMWPE. *Polymer* **2017**, *109*, 137–145. [[CrossRef](#)]

42. Du, M.; Guo, B.; Liu, M.; Jia, D. Preparation and Characterization of Polypropylene Grafted Halloysite and Their Compatibility Effect to Polypropylene/Halloysite Composite. *Polym. J.* **2006**, *38*, 1198–1204. [[CrossRef](#)]
43. Dufresne, A.; Castaño, J. Polysaccharide nanomaterial reinforced starch nanocomposites: A review. *Starch-Stärke* **2017**, *69*, 1500307. [[CrossRef](#)]

**Disclaimer/Publisher’s Note:** The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.