

Original Paper

Glucose derived carbon spheres grafted with an oil-absorbing polymer as a composite plugging agent for oil-based drilling fluids

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ABSTRACT

A composite plugging agent, glucose-derived carbon spheres grafted with poly (methyl methacrylate-co-octadecyl methacrylate-co-styrene) (GCS-g-PMOS), was synthesized by grafting an oil-absorbing polymer onto glucose-derived carbon spheres through suspension polymerization for application in oil-based drilling fluids. This material was designed to overcome the limited adaptability of conventional rigid plugging agents by integrating the structural support of carbon spheres with the swelling and filling capabilities of the polymer phase. Response surface methodology was used to optimize the preparation parameters, and the optimum formulation was determined to be 0.75 wt% glucose-derived carbon spheres, 1.5 wt% *N,N*-methylenebisacrylamide, and 2.0 wt% benzoyl peroxide. Under these conditions, the oil absorption capacity reached 9.02 g/g. Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM), thermogravimetric analysis (TGA), and wettability analyses confirmed successful grafting of the polymer phase and showed that the composite possessed good thermal stability and strong oil affinity. After aging at 160 °C, the addition of 6 wt% GCS-g-PMOS reduced the American Petroleum Institute (API) filtrate loss from 4.4 to 2.4 mL and decreased the filter cake thickness from 2.0 to 0.9 mm. The material also exhibited effective plugging performance in media of different scales and reduced the permeability of the filter cake, with a maximum permeability reduction rate of 63.53%. These results demonstrate that the integration of rigid carbon spheres and an oil-absorbing polymer is beneficial for forming a dense sealing structure and enhancing fluid-loss control in oil-based drilling fluids under high temperature conditions.

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1. Introduction

With the continuous development of unconventional oil and gas resources, drilling operations are increasingly extending into deeper and more complex shale formations (Gao, 2024; Mahmoud et al., 2025; Sun et al., 2025). Under such conditions, wellbore instability has become one of the major challenges affecting drilling safety and operational efficiency (Gautam et al., 2022; Li et al., 2025a; Sudharsan and Khare, 2024). Shale formations usually contain abundant nano-scale pores, micro-scale pores, and natural

fractures. These seepage channels provide pathways for drilling-fluid invasion and pressure transmission, which are widely recognized as key factors inducing shale weakening, borehole enlargement, and collapse (Abdullah et al., 2022; Li et al., 2024; Xu et al., 2023). Therefore, effective control of filtrate invasion and pressure transmission is essential for maintaining wellbore stability in shale drilling.

Oil-based drilling fluids (OBDFs) are widely used in shale formations because of their excellent inhibition performance, lubricity, and thermal stability (Pu et al., 2022). Compared with water-based drilling fluids (WBDFs), OBDFs can significantly reduce hydration damage and are therefore considered more suitable for drilling complex shales. However, in formations with multiscale pore-fracture systems, OBDFs still cannot completely prevent filtrate invasion. Once the filtrate enters formation pores and fractures, pressure can be transmitted into

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the near-wellbore zone, resulting in deterioration of wellbore stability. For this reason, improving the sealing and plugging performance of OBDs remains an important issue in deep and complex shale drilling (Sun et al., 2026; Yan et al., 2025b; Zhao et al., 2025).

Among various wellbore stabilization technologies, adding plugging agents to drilling fluids to form a low-permeability sealing barrier on the borehole wall is regarded as one of the most effective methods. The plugging process involves particle entry, bridging, filling, deformation, and compact sealing within formation pores and fractures (Huang et al., 2022). Conventional plugging agents such as calcium carbonate and asphalt-based materials mainly rely on mechanical bridging (Xu et al., 2025; Zhao et al., 2024). Although these materials can reduce fluid loss to some extent, their relatively large particle size and limited adaptability make them less effective for sealing nano-scale pores and irregular microfractures. As shale reservoirs become more heterogeneous and their pore-fracture structures more complex, conventional rigid plugging agents are no longer sufficient to meet the sealing requirements.

In recent years, nanomaterials have been better able to match multiscale pore and fracture systems (Davoodi et al., 2024; Gokapai et al., 2024). Bai et al. (2024b) studied graphene nanosheets as sealing agents in OBDs and showed that the nanosheets improved plugging performance in shale microfractures and reduced fluid invasion. Xu et al. (2023) prepared a zwitterionic polymer and nano silica composite and found that the material could enhance shale stability through both hydration inhibition and physical plugging. Ibrahim et al. (2024) further showed that the filtrate control of silica nanoparticles was closely related to particle size, concentration, and surface properties, which means that nano silica can improve filtration only when the interfacial characteristics are properly controlled. More recently, Lv et al. (2025) synthesized carbon nanospheres using glucose as the carbon source and applied them as plugging agents in OBDs. Their results showed that the nanomaterials reduced high temperature high pressure fluid loss and lowered filter cake permeability. However, most of these materials still depend mainly on particle accumulation, rigid bridging, or surface adsorption after entering the formation (Geng et al., 2021; Li et al., 2022; Zhang et al., 2023b). When the pore structure is irregular or the fracture width changes under pressure, their deformation capacity is limited, which makes it difficult to form a dense and stable sealing layer.

Polymeric microspheres have emerged as promising plugging agents because they can deform under pressure and adapt to complex pore fracture geometries (Meng et al., 2023; Zhang et al., 2023a; Zhao et al., 2023). Zhao et al. (2023) prepared polymer nanospheres and microspheres for OBDs and found that the microspheres had good compatibility with the oil phase, a broad particle size distribution, and effective plugging performance over a range of media. Meng et al. (2025) further developed polystyrene acrylic resin microspheres for OBDs and showed that the particles could still retard pressure transmission and improve wellbore stability after ultra-high temperature aging. In addition to polymer microspheres, oil-absorbing resin materials have also been introduced into OBDs (Bai et al., 2025a, 2025b; Su et al., 2025). Bai et al. (2022a) prepared a ternary self-swelling oil-absorbing resin and showed that the material had good oil absorption capacity and could be used for lost circulation control during drilling. In a subsequent study, Bai et al. (2022b) applied an oil-absorbing polymer gel synthesized from rigid and flexible monomers to control fluid loss of OBDs and found that the material exhibited good plugging performance after oil absorption and swelling. More recently, Bai et al. (2024a) reported a ternary composite

oil-absorbing resin for OBDs and showed that the resin could be dispersed in the oil phase and reduce fluid loss through swelling and filling. These results indicate that polymer-based materials can improve sealing through particle migration, deformation, swelling, and void filling. However, conventional polymer microspheres and oil-absorbing resin materials still face limitations in deep formations (Du et al., 2025; Li et al., 2025b). Their thermal stability is not always sufficient, their mechanical strength may be inadequate under high differential pressure, and their long-term sealing stability within seepage channels may decrease after deformation and swelling.

Despite these advances, it remains challenging for a single polymer-based plugging material to simultaneously provide sufficient mechanical support, thermal stability, and adaptive sealing performance under deep-formation conditions. Oil-absorbing resins can swell after contacting the oil phase and thus improve pore filling, but excessive softening after swelling may weaken the supporting effect of the plugging structure. In contrast, rigid particles can provide structural support during bridging, but their sealing performance is often limited when used alone because they cannot effectively fill irregular pore-fracture structures. Therefore, constructing a rigid deformable composite plugging agent is a feasible strategy to combine bridging support with swelling induced filling. Zhong et al. (2025) prepared hydrothermal carbon nanospheres from glucose and introduced them into drilling fluids, and their results showed that the carbon nanospheres could improve rheological and filtration properties, indicating that glucose derived carbon spheres (GCS) are structurally stable and compatible with drilling fluid systems. Guo et al. (2012) modified the surface of carbon spheres and improved their dispersion in a polymer matrix, which demonstrated that carbon spheres can be functionalized and used as reactive substrates for further interfacial construction. Fei et al. (2022) further constructed carbon spheres wrapped with a two dimensional covalent organic polymer, confirming that carbon spheres can serve as rigid cores for subsequent polymer related surface assembly. These studies suggest that GCS are suitable rigid supports for composite plugging agents and that grafting polymer chains onto their surface is a feasible route. Compared with conventional core-shell microspheres and physically blended inorganic/polymer composites, chemical grafting provides a more stable interfacial connection between the GCS core and the oil-absorbing polymer phase. In physically blended systems, the rigid particles and polymer phase may migrate or deform independently under differential pressure, which can weaken the long-term stability of the sealing layer. By contrast, the grafted structure enables the rigid carbon spheres and deformable polymer network to act cooperatively during oil absorption, swelling, and pore filling. In the monomer formulation, methyl methacrylate and styrene contribute to the polymer skeleton and thermal resistance, octadecyl methacrylate introduces long alkyl chains for oil affinity and swelling, and MBA regulates the crosslinked network density.

In this study, glucose-derived carbon spheres grafted with an oil-absorbing polymer (GCS-g-PMOS) were designed and prepared as a composite plugging agent for OBDs. The design aimed to integrate structural support, oil affinity, and adaptive filling into a single material. Preparation conditions were optimized using response surface methodology (RSM), and the resulting material was characterized in terms of structure, morphology, thermal stability, and wettability. Its effects on rheology, emulsion stability, filtrate control, and multiscale plugging were then evaluated, and the corresponding plugging mechanism was discussed.

2. Materials and methods

2.1. Chemicals

All chemicals used in this study were of analytical grade, and deionized water was used throughout. Glucose, poly (vinyl alcohol) (PVA, type 1788), methyl methacrylate (MMA), octadecyl methacrylate (ODMA), styrene (St), *N,N'*-methylenebisacrylamide (MBA), benzoyl peroxide (BPO), and absolute ethanol were purchased from Shanghai Aladdin Bio-Chem Technology Co., Ltd. All reagents were used as received without further purification.

Commercial plugging agents, including calcium carbonate (CaCO_3), elastic graphite (FGR), silicate fiber (SF), and modified resin (HLKY), were used for comparison. All commercial plugging agents were used as received without further purification.

2.2. Synthesis

2.2.1. Preparation of GCS

GCS were prepared by a hydrothermal method. A 10 wt% glucose aqueous solution was transferred into a Teflon-lined autoclave with a filling ratio of 80%. The autoclave was heated at 180 °C for 16 h and then cooled to room temperature. The solid products were collected by centrifugation at 5000 rpm for 30 min, washed three times with ethanol and deionized water, and dried at

80 °C for 24 h to obtain brown-black GCS powders. The yield of GCS was 46%.

2.2.2. Preparation of GCS-g-PMOS

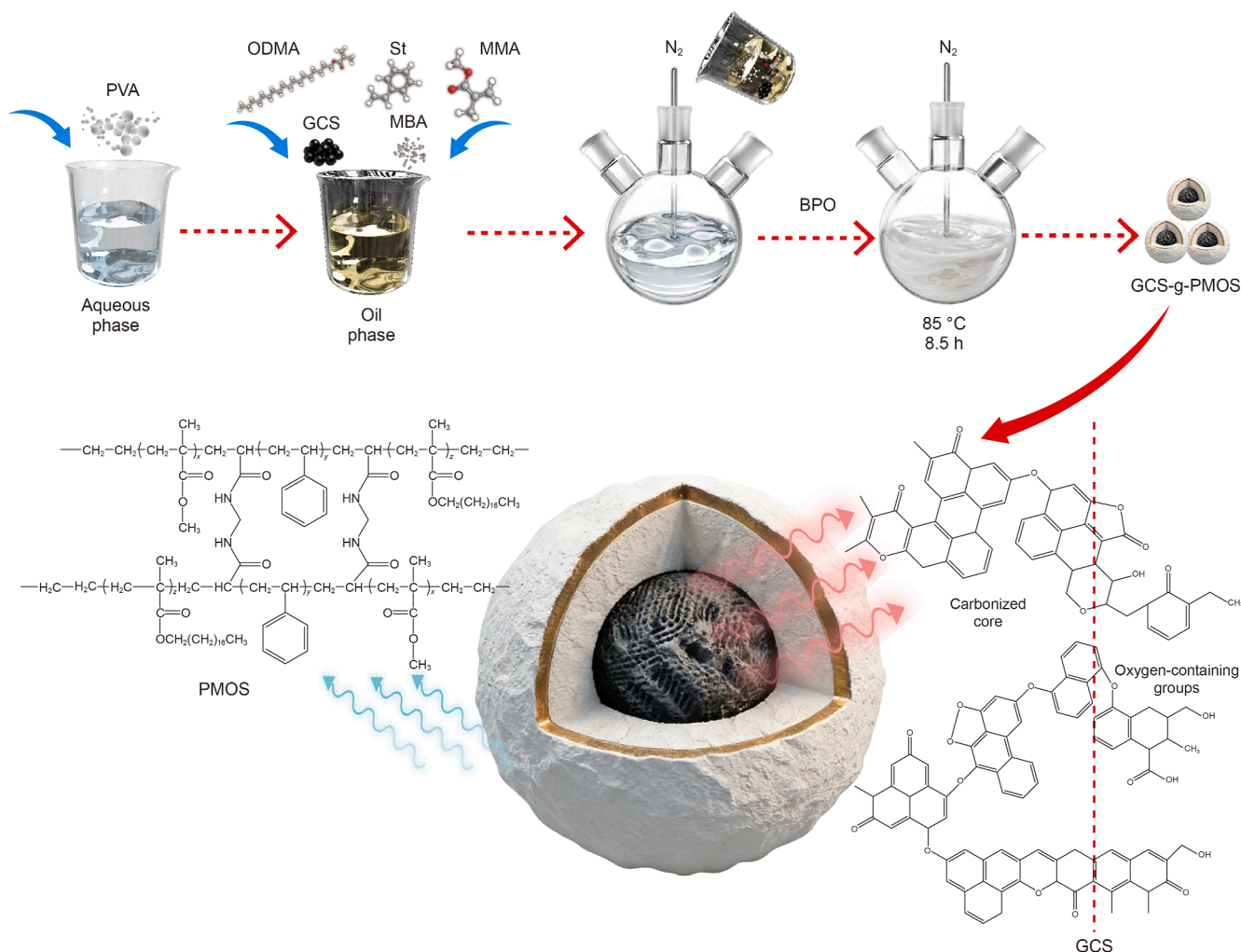
GCS-g-PMOS was prepared by suspension polymerization. First, 1.5 wt% PVA was dissolved in 200 mL of deionized water to form the aqueous phase, which was transferred into a three-necked flask under mechanical stirring.

The oil phase consisted of MMA, ODMA, and St with a fixed total monomer mass of 20.0 g. The mass fractions of MMA, ODMA, and St were 40, 30, and 30 wt%, respectively. MBA was dissolved in a small amount of ethanol and then added to the oil phase under stirring. GCS was subsequently introduced, and the mixture was ultrasonicated to ensure uniform dispersion.

After nitrogen purging for 30 min, the oil phase was added to the aqueous phase, BPO was then introduced under continuous stirring in a nitrogen atmosphere, and the polymerization was carried out at 85 °C for 8.5 h. The resulting products were washed with deionized water and ethanol and dried at 60 °C for 24 h (as shown in Scheme 1).

2.3. Oil absorption test

The oil absorption capacity of GCS-g-PMOS is essential for evaluating its applicability in OBDFs and predicting its plugging



Scheme 1. Schematic illustration of the preparation process of the GCS-g-PMOS.

performance. A gravimetric method was adopted to determine the oil absorption capacity (W) of the product. Approximately 1 g of pre-weighed sample was placed on a sieve (15 cm $\times$ 5 cm), fully submerged in a glass dish (18 cm $\times$ 8 cm) filled with white oil, and then sealed with plastic wrap. The sample was allowed to absorb oil for 24 h to ensure saturation. The oil-absorbing resin was subsequently removed and reweighed. The oil absorption capacity was calculated using the following equation:

$$W = \frac{m_2 - m_1}{m_1} \quad (1)$$

where m_1 and m_2 are the masses of GCS-g-PMOS before and after oil absorption, g, respectively; and W is the oil absorption capacity, g/g.

2.4. Characterization

The morphology of GCS and GCS-g-PMOS was characterized by scanning electron microscopy (SEM; SU8220, Hitachi, Tokyo, Japan). Functional groups were analyzed by Fourier transform infrared spectroscopy (FTIR; Nicolet iS20, Thermo Fisher Scientific, Waltham, MA, USA) in transmission mode over 400–4000 cm^{-1} using KBr pellets prepared from 2 mg of sample and 200 mg of anhydrous KBr. Thermal stability was investigated by thermogravimetric analysis (TGA; Mettler Toledo, Greifensee, Switzerland). Approximately 8–10 mg of sample was loaded into alumina crucibles and heated from 30 to 800 $^{\circ}\text{C}$ under nitrogen (50 mL/min) at 10 $^{\circ}\text{C}/\text{min}$. Surface wettability was evaluated by static contact-angle measurements using an OCA 25 instrument (DataPhysics Instruments, Filderstadt, Germany).

2.5. Preparation and evaluation of oil-based drilling fluid

The base oil-based drilling fluid (OBDF) was formulated with an oil–water ratio of 80:20 and a target density of 1.8 g/cm^3 . The composition of the fluid was as follows: 5 wt% emulsifiers in total, consisting of Span 80 and Tween 80, 2.5 wt% organoclay, 3 wt% CaO, and 25 wt% CaCl_2 brine (internal phase). To achieve the required density, barite was incorporated as a weighting agent until the density reached 1.8 g/cm^3 . The GCS-g-PMOS concentration was expressed as a mass percentage relative to the total mass of the drilling fluid.

Rheological measurements: the rheological properties of the OBDF and plugging fluid formulations were evaluated using a six-speed rotational viscometer in accordance with API RP 13B-2 issued by the American Petroleum Institute (API). Apparent viscosity (AV), plastic viscosity (PV), and yield point (YP) were calculated as follows:

$$\eta_a = \frac{\theta_{600}}{2} \quad (2)$$

$$\mu_p = \theta_{600} - \theta_{300} \quad (3)$$

$$\tau_0 = \theta_{300} - \eta_a \quad (4)$$

where θ_{600} and θ_{300} are viscometer readings at 600 and 300 rpm, respectively; η_a is the apparent viscosity, $\text{mPa}\cdot\text{s}$; μ_p is the plastic viscosity, $\text{mPa}\cdot\text{s}$; and τ_0 is the yield point, Pa.

API filtration measurement: The API fluid loss of the base OBDF and the drilling fluids containing different concentrations of GCS-g-PMOS was measured using a standard API filter press in accordance with API RP 13B-2. The prepared drilling fluid was first sheared thoroughly and then tested either before aging or after hot rolling at 160 $^{\circ}\text{C}$ for 16 h, depending on the

experimental conditions. For each test, the fluid was poured into the filter cell equipped with standard API filter paper, and the filtration test was carried out under a pressure of 0.69 MPa for 30 min at room temperature. The collected filtrate volume was recorded as the API fluid loss, denoted as FL_{API} . After filtration, the thickness of the filter cake was measured at several positions and the average value was reported. All measurements were performed at least in triplicate, and the average value was used for analysis.

Electrical stability measurement: The electrical stability of the OBDF was measured to evaluate emulsion stability. The fluid was prepared according to the specified formulation and sheared at 10,000 rpm for 20 min to ensure uniform dispersion. Electrical stability was measured both before aging and after aging at 160 $^{\circ}\text{C}$ for 16 h. The aged samples were cooled to room temperature prior to testing. Electrical stability was measured using an OFITE electrical stability tester (Model 131-50) by applying an increasing AC voltage until a sharp rise in current occurred, indicating emulsion breakdown. The corresponding voltage was recorded as the electrical stability value in volts V. All measurements were performed in triplicate, and the average value was reported.

Plugging performance measurement: The plugging performance of GCS-g-PMOS was evaluated by two complementary tests, namely multiscale high-temperature high-pressure (HTHP) filtration tests and an artificial filter cake permeability reduction test. Unless otherwise specified, the drilling fluids were aged at 160 $^{\circ}\text{C}$ for 16 h and then cooled to room temperature before testing.

For the multiscale HTHP filtration test, the base OBDF and the OBDF formulations containing different concentrations of GCS-g-PMOS were tested using a HTHP filter press at a differential pressure of 3.5 MPa for 30 min. Four filtration media were used to simulate seepage channels of different scales, including a 0.45 μm microporous membrane, a 3 μm sand ceramics, standard API filter paper with a maximum pore size smaller than 20 μm , and a 2 mm filter crevice. The cumulative filtrate volume was recorded to evaluate the plugging performance of the OBDF. For the 0.45 μm microporous membrane, the cumulative filtrate volume was recorded as a function of time. For the sand ceramics, API filter paper, and filter crevice, the final filtrate volume after 30 min was used for comparison because filtrate breakthrough occurred rapidly in the larger channels.

To evaluate the effect of temperature on plugging performance, the same HTHP filtration procedure was repeated after aging the drilling fluids at different temperatures, while keeping the differential pressure, filtration time, and filtration media unchanged.

Permeability reduction measurement: The permeability reduction ability of GCS-g-PMOS was evaluated using an artificial filter cake. For each test, a fresh artificial filter cake was first prepared from the base OBDF without GCS-g-PMOS using an HTHP filter press at 3.5 MPa for 30 min after hot rolling at 160 $^{\circ}\text{C}$ for 16 h. After cooling to room temperature, the loosely attached surface layer was carefully removed, and the filter-cake thickness was measured at several positions and averaged. White oil was then used as the permeating fluid to determine the initial permeability of the artificial filter cake. The white-oil permeability test was conducted under the same differential pressure for 30 min, and the cumulative filtrate volume was recorded every 5 min. The cumulative filtrate volume obtained in this step was denoted as $\text{FL}_{\text{before}}$ (mL). The same artificial filter cake was subsequently treated with a plugging fluid containing 0, 1, 2, 4, or 6 wt% GCS-g-PMOS under 3.5 MPa for 30 min, followed by a second white-oil permeability test under the same conditions. The cumulative filtrate volume obtained after treatment was denoted as FL_{after} (mL).

The permeability of the artificial filter cake was calculated according to Darcy's law:

$$K = \frac{Q\mu L}{A\Delta P} \quad (5)$$

where K is the permeability of the artificial filter cake, 10^{-4} mD; Q is the filtration rate obtained from the slope of the cumulative filtrate volume versus time curve, cm^3/s ; μ is the viscosity of white oil, mPa·s; L is the thickness of the filter cake, mm; A is the effective filtration area, cm^2 ; and ΔP is the differential pressure, MPa.

The permeability reduction rate was calculated as follows:

$$k_r = \frac{K_0 - K_1}{K_0} \quad (6)$$

where K_0 and K_1 are the permeability values of the artificial filter cake before and after treatment with the plugging fluid, respectively, 10^{-4} mD; and k_r is the permeability reduction rate.

To further evaluate the relative plugging performance of GCS-g-PMOS, several commercial plugging agents, including CaCO_3 , elastic graphite (FGR), silicate fiber (SF), and modified resin (HLKY), were selected for comparison. The additive concentration was fixed at 4 wt%, and all drilling fluids were hot rolled at 160 °C for 16 h before testing. API fluid loss, filter-cake thickness, and multiscale HTHP fluid loss through the filter crevice and sand ceramics were measured under the same conditions.

2.6. Experimental design and response surface methodology

To optimize the preparation conditions of GCS-g-PMOS, response surface methodology (RSM) based on a Box–Behnken design was employed. According to the preliminary experiments, GCS content, MBA content, and BPO content were selected as the independent variables, and oil absorption capacity was used as the

Table 1
Independent variables and levels used in the Box–Behnken design.

Factor	Symbol	Levels		
		−1	0	+1
GCS content, wt%	A	0.30	0.75	1.20
MBA content, wt%	B	0.50	1.50	2.50
BPO content, wt%	C	1.00	2.00	3.00

Table 2
Box–Behnken experimental matrix and response values for oil absorption.

Run	Factor			Oil absorption, g/g		Residual
	A	B	C	Experimental value	Predicted value	
1	0 (0.75)	0 (1.50)	0 (2.00)	8.82	8.89	−0.0680
2	+1 (1.20)	0 (1.50)	+1 (3.00)	8.12	8.16	−0.0387
3	+1 (1.20)	−1 (0.50)	0 (2.00)	7.20	7.02	0.1788
4	−1 (0.30)	0 (1.50)	−1 (1.00)	7.60	7.56	0.0388
5	0 (0.75)	0 (1.50)	0 (2.00)	8.95	8.89	0.0620
6	−1 (0.30)	0 (1.50)	+1 (3.00)	7.90	7.82	0.0813
7	0 (0.75)	0 (1.50)	0 (2.00)	9.02	8.89	0.1320
8	+1 (1.20)	0 (1.50)	−1 (1.00)	7.45	7.53	−0.0812
9	0 (0.75)	−1 (0.50)	−1 (1.00)	6.85	6.95	−0.0975
10	−1 (0.30)	−1 (0.50)	0 (2.00)	7.18	7.12	0.0588
11	+1 (1.20)	+1 (2.50)	0 (2.00)	7.78	7.84	−0.0587
12	0 (0.75)	0 (1.50)	0 (2.00)	8.75	8.89	−0.1380
13	0 (0.75)	+1 (2.50)	+1 (3.00)	8.05	7.95	0.0975
14	0 (0.75)	−1 (0.50)	+1 (3.00)	7.15	7.29	−0.1400
15	0 (0.75)	+1 (2.50)	−1 (1.00)	7.55	7.41	0.1400
16	−1 (0.30)	+1 (2.50)	0 (2.00)	7.25	7.43	−0.1787
17	0 (0.75)	0 (1.50)	0 (2.00)	8.90	8.89	0.0120

response. The GCS, MBA, and BPO contents were expressed as mass percentages relative to the total monomer mass. Table 1 lists the factors and their levels used in the experimental design. Table 2 presents the Box–Behnken experimental matrix together with the corresponding oil absorption values obtained in each run.

Each factor was investigated at three levels coded as −1, 0, and +1. A total of 17 experimental runs, including five replicated center points, were generated using Design-Expert 13 software. The relationship between the response and the independent variables was described by a second order polynomial model, as expressed below:

$$Y = \beta_0 + \sum \beta_i X_i + \sum \beta_{ii} X_i^2 + \sum \beta_{ij} X_i X_j + \varepsilon \quad (7)$$

where Y represents the predicted response; β_0 is the intercept term; β_i , β_{ii} , and β_{ij} are the linear, quadratic, and interaction coefficients, respectively; X_i and X_j are the coded independent variables; and ε is the random error.

The adequacy and significance of the model were evaluated by analysis of variance (ANOVA), including F -value, p -value, coefficient of determination (R^2), adjusted R^2 , predicted R^2 , and lack of fit tests. These statistical parameters were used to validate the reliability and predictive capability of the model.

3. Results and discussion

3.1. Response surface analysis of oil absorption

3.1.1. Experimental design and regression model

Table 2 shows the experimental design matrix and the corresponding oil absorption. The oil absorption ranged from 6.85 to 9.02 g/g, indicating that the selected factor levels covered an appropriate experimental range for model construction. The experimental data were fitted to a quadratic polynomial equation. The regression model in terms of the actual factors is given below:

$$\begin{aligned} \text{Oil absorption} = & 2.76244 + 3.66204A + 2.82325B + 2.13808C \\ & + 0.283333AB + 0.205556AC + 0.050000BC - 2.88395A^2 \\ & - 0.951599B^2 - 0.536500C^2 \end{aligned}$$

where A , B , and C represent the contents of GCS, MBA, and BPO, respectively.

Although the fitted equation suggested a local positive tendency for GCS content, its linear term was not significant according to ANOVA ($p = 0.2254$), whereas its quadratic term was significant ($p = 0.0002$). This indicates that GCS content did not have a simple monotonic effect on oil absorption, and an intermediate GCS content was more favorable because excessive GCS could restrict polymer chain expansion and reduce the effective swelling volume.

3.1.2. Analysis of variance and model diagnostics

Table 3 shows the analysis of variance for the quadratic model. The model F -value reached 34.04, and the corresponding p -value was lower than 0.0001, indicating that the model was highly significant. Among the model terms, MBA content, BPO content, and the three quadratic terms showed significant effects on the response. By contrast, the linear term of GCS content and the interaction terms AB , AC , and BC were not significant within the studied range.

Table 3 presents the ANOVA results for the quadratic response model. The lack of fit p -value was 0.0965, indicating that the lack of fit was not significant relative to the pure error. Therefore, the quadratic model was considered adequate to describe the relationship between the preparation variables and the oil absorption capacity within the selected design space.

Table 4 lists the fit statistics of the model. The coefficient of determination reached 0.9777, and the adjusted coefficient of determination was 0.9489, indicating good agreement between the experimental values and the fitted values. The predicted R^2 value of 0.7190 was lower than the adjusted R^2 value of 0.9489, indicating limited extrapolative predictive capability of the model. Thus, the model was mainly used for interpolation within the investigated factor ranges. Since the optimized formulation was located near the center of the BBD space and the predicted oil absorption value of 8.89 g/g agreed well with the experimental value of 9.02 g/g, the optimized point was considered reliable within the studied range.

3.1.3. Response surface analysis

Fig. 1 shows the response surface plots and contour plots describing the effects of GCS content, MBA content, and BPO content on oil absorption. As shown in Fig. 1, the oil absorption capacity generally increased first and then decreased with

increasing factor level, which is consistent with the significant quadratic terms obtained from the ANOVA.

In Fig. 1(a), the oil absorption initially increases and then decreases with increasing MBA content, whereas the effect of GCS remains relatively moderate. At low MBA content, insufficient crosslinking leads to a loosely structured network with limited oil retention. Increasing MBA promotes the formation of a stable three-dimensional crosslinked network, which optimizes the crosslink density and facilitates oil absorption. However, excessive MBA results in an overly dense structure that restricts chain mobility and reduces free volume, thereby hindering oil diffusion and lowering oil absorption. In contrast, GCS mainly acts as a rigid filler, contributing to pore formation and increased surface area, with a less pronounced effect than MBA.

Fig. 1(b) shows the interaction between GCS (A) and BPO (C). The oil absorption initially increases and then decreases with increasing BPO content. At low BPO content, the limited generation of free radicals leads to incomplete polymerization, resulting in an insufficiently developed polymer network. With increasing BPO content, the polymerization process becomes more efficient, promoting the formation of a more uniform crosslinked network, which enhances oil absorption.

However, excessive BPO accelerates the initiation process, leading to the formation of shorter polymer chains and an increase in crosslinking density, which results in a more compact network structure and restricts oil absorption. In comparison, GCS functions as a structural component within the polymer matrix, contributing to the formation of a more uniform network structure and thereby enhancing oil absorption at moderate BPO content.

Fig. 1(c) presents the interaction between MBA (B) and BPO (C). A maximum oil absorption is observed at intermediate levels of both factors, indicating a synergistic effect. MBA primarily determines the crosslinking density, whereas BPO governs the initiation efficiency of polymerization. Their combined effect regulates the formation of the polymer network structure.

At appropriate concentrations, a crosslinked network with a balanced crosslinking density is formed, which is favorable for oil absorption. However, excessive MBA combined with insufficient BPO leads to incomplete polymerization and an inadequately formed network structure. In contrast, excessive BPO accelerates the initiation process, resulting in shorter polymer chains and increased crosslinking density, which produces a more compact network structure and reduces oil absorption.

3.1.4. Optimization and validation of optimal conditions

Based on the response surface analysis, the optimal preparation conditions were determined by numerical optimization. As shown in Fig. 1(d), the oil absorption reaches a maximum at intermediate levels of the variables.

The optimal formulation obtained from numerical optimization was 0.75 wt% GCS, 1.5 wt% MBA, and 2.0 wt% BPO. According to the fitted model, the predicted oil absorption under these conditions was 8.89 g/g. In the corresponding experiment, the oil absorption reached 9.02 g/g, indicating good agreement between the model prediction and the experimental result. The contour plot shows that the region with higher oil absorption is concentrated near the center, and the contour lines are nearly elliptical, indicating that the response is mainly governed by quadratic effects with relatively weak interactions between variables, which is consistent with the ANOVA results.

At the optimal composition, the MBA content of 1.5 wt% provides a suitable crosslinked structure for oil absorption. This structure allows oil molecules to enter the grafted polymer network while avoiding excessive network compaction. Such a

Table 3
ANOVA for the quadratic response model.

Source	Sum of squares	Mean square	F-value	p-value
Model	8.33	0.9254	34.04	<0.0001
A	0.0480	0.0480	1.77	0.2254
B	0.6328	0.6328	23.28	0.0019
C	0.3916	0.3916	14.40	0.0068
AB	0.0650	0.0650	2.39	0.1659
AC	0.0342	0.0342	1.26	0.2989
BC	0.0100	0.0100	0.3678	0.5633
A ²	1.44	1.44	52.82	0.0002
B ²	3.81	3.81	140.22	<0.0001
C ²	1.21	1.21	44.58	0.0003
Lack of fit	0.1452	0.0484	4.30	0.0965

Table 4
Fit statistics for the quadratic response model.

Parameter	Value	Parameter	Value
R^2	0.9777	Adjusted R^2	0.9489
Predicted R^2	0.7190	Adeq precision	15.345
Std. Dev.	0.1649	C.V., %	2.084

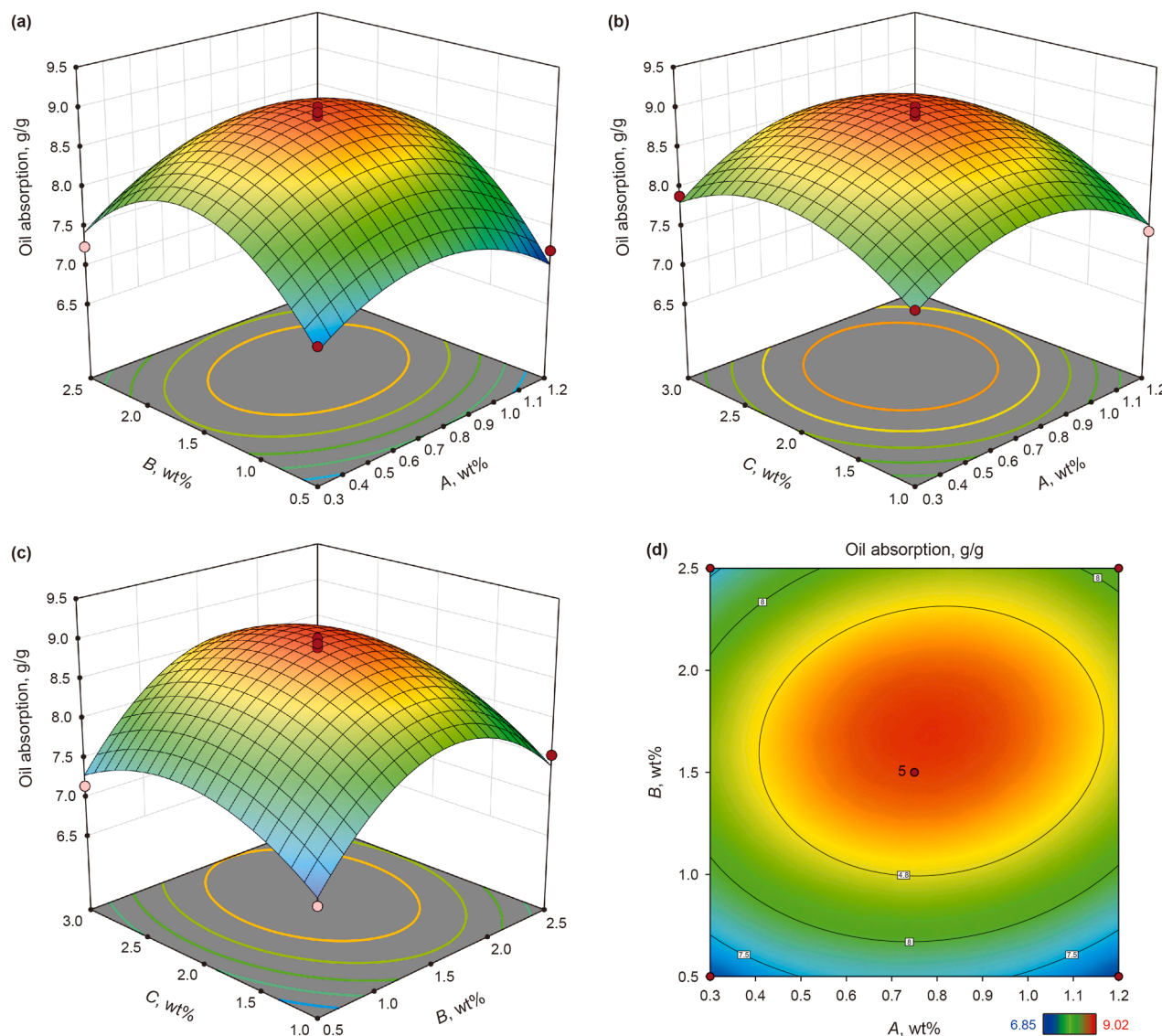


Fig. 1. Response surface and contour plots for oil absorption (Y): (a) GCS content (A) and MBA content (B); (b) GCS content (A) and BPO content (C); (c) MBA content (B) and BPO content (C); (d) contour plot showing the interaction between GCS content (A) and MBA content (B).

balanced structure is also beneficial for the subsequent swelling and filling behavior during plugging.

3.2. Microstructure and structure characterization

In the FTIR spectrum of GCS (Fig. 2(a)), the broad band at 3428 cm^{-1} is assigned to O–H stretching vibrations associated with surface hydroxyl groups and adsorbed water. The weak bands at 2978 and 2918 cm^{-1} are attributed to aliphatic C–H stretching vibrations. The band at 1706 cm^{-1} corresponds to the C=O stretching vibration of carbonyl-containing groups, whereas the bands at 1630 and 1512 cm^{-1} are mainly associated with skeletal vibrations of conjugated and aromatic carbon structures formed during the hydrothermal carbonization of glucose. The band at 1630 cm^{-1} may also contain a contribution from the bending vibration of adsorbed water (Lv et al., 2025). Compared with GCS, GCS-g-PMOS (Fig. 2(b)) exhibits pronounced absorption bands at 2921 and 2852 cm^{-1} , which are assigned to the asymmetric and symmetric stretching vibrations of methylene groups, respectively. The distinct band at

1726 cm^{-1} corresponds to the C=O stretching vibration of ester groups, while the band at 1157 cm^{-1} is assigned to the C–O–C stretching vibration of ester structures. The bands at 1446 and 1383 cm^{-1} are attributed to C–H deformation vibrations in the polymer phase. These spectral changes confirm the presence of methacrylate-derived ester groups and long-chain alkyl segments in the composite and support the formation of a PMOS-containing polymer phase in GCS-g-PMOS (Zhou et al., 2023).

The thermogravimetric behavior of GCS and GCS-g-PMOS is shown in Fig. 3. Both samples exhibit multistep weight-loss processes, indicating sequential decomposition of surface functionalities and carbonaceous or polymeric structures (Chen et al., 2021; Minakov et al., 2023).

As shown in Fig. 3(a), GCS undergoes an initial weight loss of $4.34\text{ wt\%}$ below $150\text{ }^{\circ}\text{C}$, with a derivative thermogravimetric (DTG) peak at $71.26\text{ }^{\circ}\text{C}$, which is attributed to desorption of physically adsorbed water and residual low-molecular-weight species. A continuous mass loss is then observed between 150 and $400\text{ }^{\circ}\text{C}$ owing to decomposition of oxygen-containing functional groups and partial breakdown of the amorphous carbon framework. The

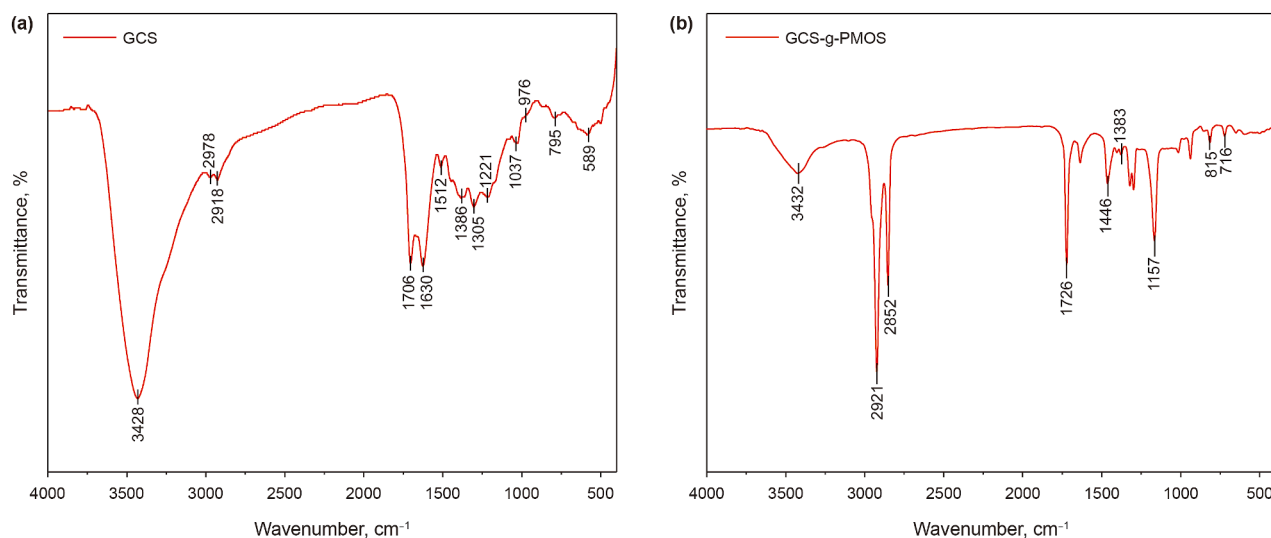


Fig. 2. FTIR spectra of GCS (a) and GCS-g-PMOS (b).

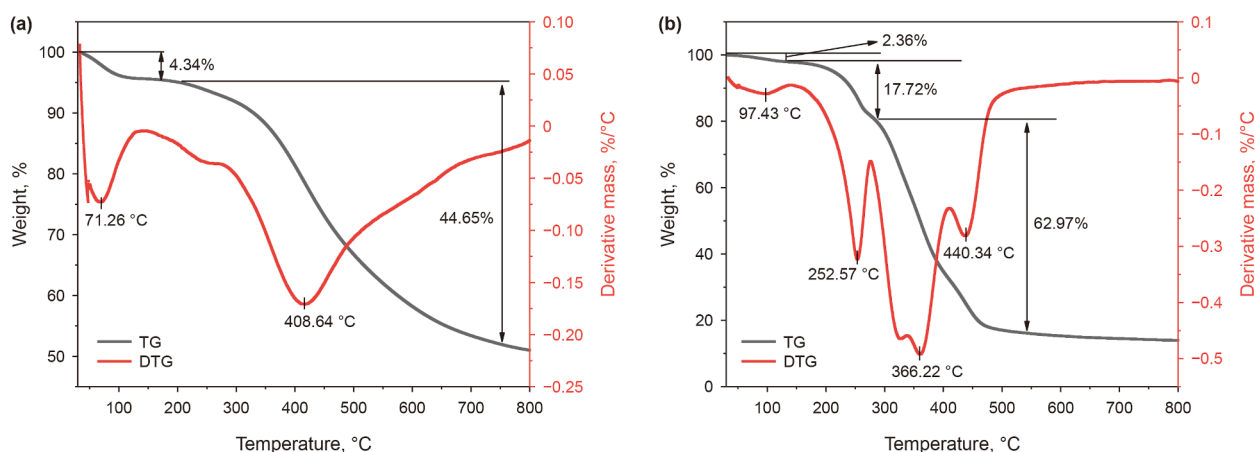


Fig. 3. Thermogravimetric (TG) and derivative thermogravimetric (DTG) curves of GCS (a) and GCS-g-PMOS (b).

major DTG peak appears at 408.64 $^{\circ}\text{C}$, and the mass loss in this stage reaches approximately 44.65 wt%. Above 400 $^{\circ}\text{C}$, the mass decreases gradually, indicating continued carbonization and structural rearrangement.

As shown in Fig. 3(b), GCS-g-PMOS exhibits an initial weight loss of approximately 2.36% below 150 $^{\circ}\text{C}$, with a DTG peak at 97.43 $^{\circ}\text{C}$. In the range of 150–300 $^{\circ}\text{C}$, a weight loss of about 17.72% is observed, accompanied by a DTG peak at 252.57 $^{\circ}\text{C}$. This stage is attributed mainly to cleavage of ester-containing side groups and partial degradation of long alkyl chains derived from ODMA (Jian et al., 2023). Between 300 and 500 $^{\circ}\text{C}$, GCS-g-PMOS shows a major weight loss of 62.97%, with DTG peaks at 366.22 and 440.34 $^{\circ}\text{C}$, corresponding to overlapping degradation of the grafted polymer network and further decomposition of the carbonaceous framework. The higher residual mass at elevated temperature indicates that the GCS core contributes to the thermal resistance and structural stability of the composite (Yan et al., 2025a).

The morphology of GCS and GCS-g-PMOS was characterized by SEM, as shown in Fig. 4. In Fig. 4(a), GCS exhibits a spherical morphology with a relatively smooth surface and a uniform size distribution. The particles are well separated, indicating that the carbon spheres maintain an intact morphology after hydrothermal treatment.

In contrast, as shown in Fig. 4(b), the surface of GCS-g-PMOS becomes rougher and the particles appear partially interconnected, suggesting the formation of a polymer layer on the surface of GCS after graft polymerization.

The wettability of GCS-g-PMOS was evaluated by contact angle measurements, as shown in Fig. 5. The contact angle for the oil droplet is 18.6 $^{\circ}$, indicating strong affinity between the material surface and oil. In contrast, the water droplet exhibits a contact angle of 31.3 $^{\circ}$, suggesting that the surface also maintains a certain degree of hydrophilicity.

The relatively low contact angles for both oil and water indicate that GCS-g-PMOS exhibits amphiphilic characteristics. This behavior can be attributed to the coexistence of hydrophilic groups on GCS and hydrophobic segments introduced by graft polymerization, which together regulate the surface wettability of the material.

3.3. Plugging performance

3.3.1. Effects of GCS-g-PMOS on rheology and filtration behavior of the OBDF

The effects of GCS-g-PMOS on the rheological and filtration properties of the base drilling fluid are summarized in Table 5.

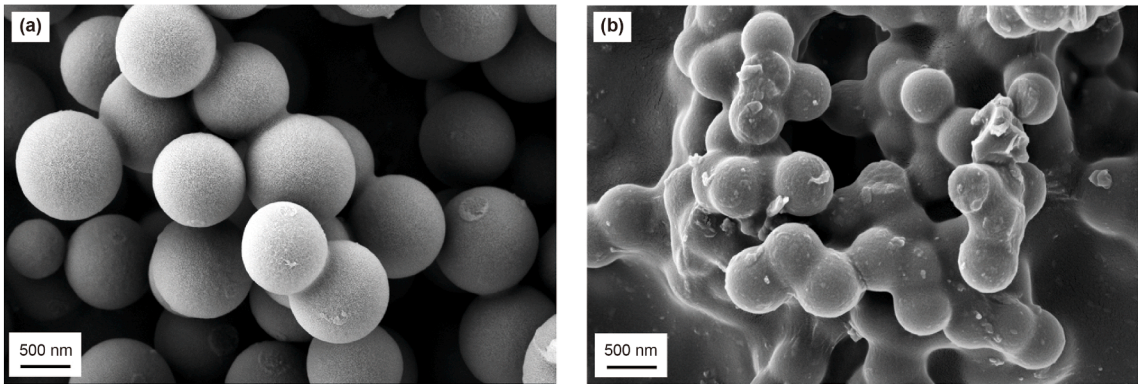


Fig. 4. SEM images of GCS (a) and GCS-g-PMOS (b).

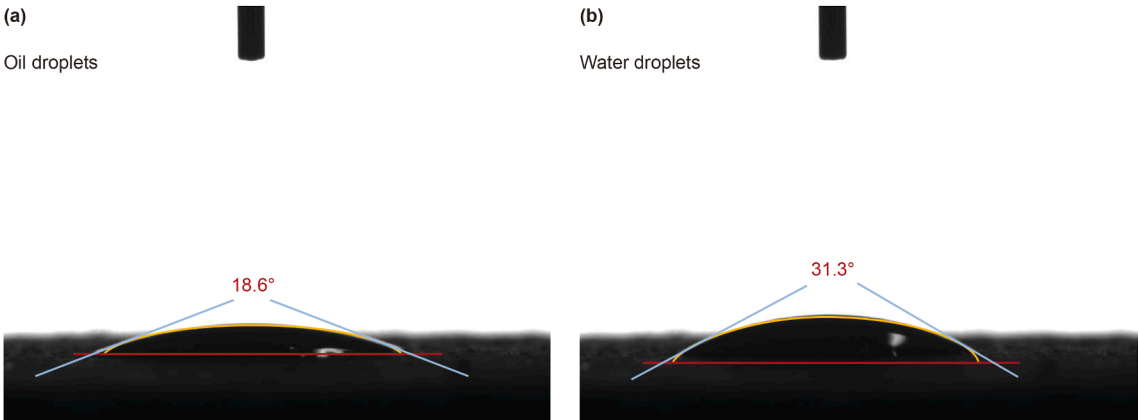


Fig. 5. Contact angle measurements of GCS-g-PMOS with oil (a) and water (b) droplets.

Table 5
Rheological properties and filtration performance of drilling fluids containing different concentrations of GCS-g-PMOS before and after aging at 160 °C.

GCS-g-PMOS concentration, wt%	Test state	AV, mPa·s	PV, mPa·s	YP, Pa	YP/PV	G _{10s} /G _{10min}	FL _{API} , mL	Filter-cake thickness, mm
0	Before aging	50.5	41.5	9.0	0.22	1.5/6.0	4.8	2.0
	After aging	53.0	44.0	9.0	0.20	2.0/6.0	4.4	2.0
1	Before aging	52.0	42.5	9.5	0.22	2.0/4.5	4.6	1.7
	After aging	52.5	42.5	10.0	0.24	2.5/6.0	4.4	1.8
2	Before aging	57.0	46.0	11.0	0.24	3.0/7.0	4.0	1.6
	After aging	59.5	47.5	12.0	0.25	3.5/7.0	4.2	1.7
4	Before aging	59.0	48.0	11.0	0.23	3.0/8.0	3.0	1.2
	After aging	61.5	50.0	11.5	0.23	3.5/9.0	3.2	1.4
6	Before aging	62.0	49.0	13.0	0.26	4.0/9.0	2.0	0.8
	After aging	65.5	53.5	12.0	0.22	3.5/8.0	2.4	0.9

With increasing GCS-g-PMOS concentration from 0 to 6 wt%, the AV, PV, and YP all increased before aging. Specifically, the AV increased from 50.5 to 62.0 mPa·s, the PV increased from 41.5 to 49.0 mPa·s, and the YP increased from 9.0 to 13.0 Pa. After hot rolling at 160 °C, a similar trend was observed, indicating that GCS-g-PMOS retained its rheological modification effect under elevated temperature conditions (Ofei et al., 2021).

The increase in viscosity and yield point can be attributed to interactions between GCS-g-PMOS particles and the continuous oil phase, which strengthen the internal structure of the drilling fluid and increase resistance to flow (Di et al., 2022; Geng et al., 2019). Importantly, the YP/PV ratio remained in the range of 0.20–0.26, indicating that the fluid maintained a reasonable balance between viscosity and carrying capacity. The gel strength also increased gradually with increasing dosage, suggesting improved suspension

capacity without excessive gelation (Fayad et al., 2021; Ghavami et al., 2018). In addition to modifying rheology, GCS-g-PMOS improved the fluid-loss control performance of the drilling fluid. The FL_{API} decreased from 4.8 to 2.0 mL before aging and from 4.4 to 2.4 mL after aging as its concentration increased from 0 to 6 wt%. Meanwhile, the filter cake thickness decreased from 2.0 to 0.8 mm before aging and from 2.0 to 0.9 mm after aging. These results indicate that GCS-g-PMOS promoted the formation of a denser and thinner sealing layer and thus enhanced fluid-loss control (Liu et al., 2025; Vasconcelos et al., 2022).

3.3.2. Effect of GCS-g-PMOS concentration on the electrical stability of OBDF

The effect of GCS-g-PMOS concentration on the electrical stability (ES) of the OBDF is shown in Fig. 6. The ES increased

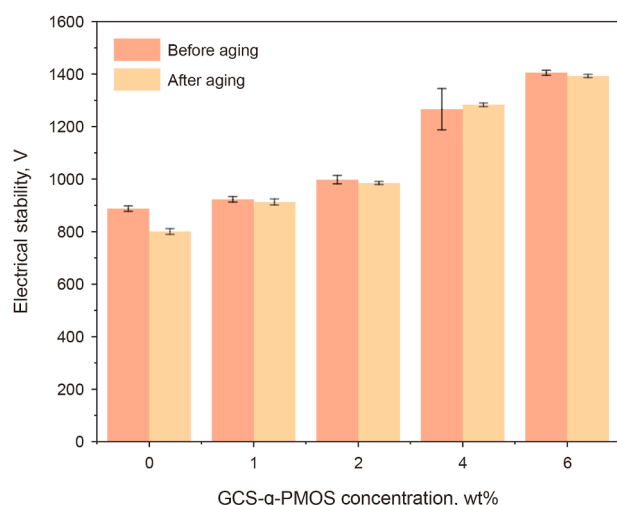


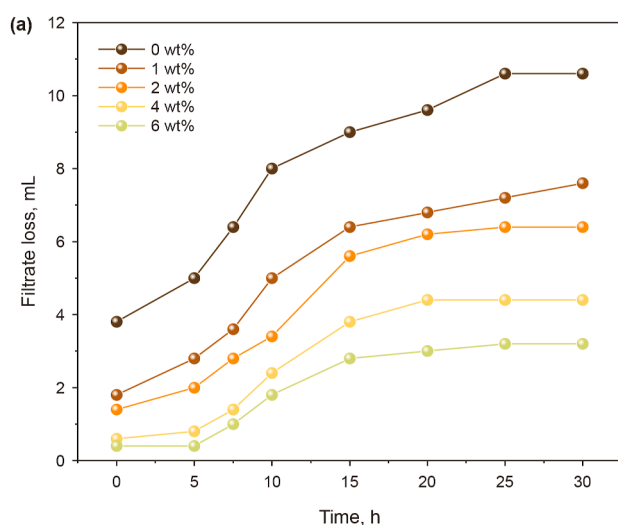
Fig. 6. Effect of GCS-g-PMOS concentration on the electrical stability (ES) of OBDF.

progressively as the concentration of GCS-g-PMOS increased, indicating that the emulsion stability of the OBDF was not impaired by the addition of the plugging agent. The increase in ES suggests that GCS-g-PMOS did not destabilize the invert emulsion.

The improvement in ES is reasonably attributed to the good oil-phase affinity of GCS-g-PMOS, as indicated by the contact angle results. This behavior is consistent with the low oil contact angle, which indicates a strong affinity of the particle surface for the continuous oil phase. Within the investigated concentration range, no deterioration of ES was observed, demonstrating that GCS-g-PMOS is compatible with the OBDF system and does not disrupt the original emulsification state. The low oil contact angle of 18.6° and water contact angle of 31.3° indicate that GCS-g-PMOS has strong oil affinity and partial amphiphilicity. Such wettability favors its dispersion in OBDF and its distribution at oil–water interfaces and pore spaces, thereby improving emulsion stability and plugging performance.

3.3.3. Filtration and plugging performance in media of different scales

Fig. 7 illustrates the plugging performance of GCS-g-PMOS in multiscale filtration media. Fig. 7(a) shows the time-dependent



filtration behavior through a $0.45\ \mu\text{m}$ microporous membrane, while Fig. 7(b) compares the final fluid loss in larger-scale media, including a $3\ \mu\text{m}$ sand ceramic, API filter paper, and a 2 mm filter crevice.

As shown in Fig. 7(a), the filtrate volume through the $0.45\ \mu\text{m}$ microporous membrane decreases markedly after the addition of GCS-g-PMOS, and the filtration curves gradually approach a plateau with time. This behavior indicates that GCS-g-PMOS can rapidly form an effective sealing layer in micro-scale pores, thereby restricting further filtrate invasion. The rapid decline in filtration rate at the initial stage further suggests that the plugging agent can quickly respond to fine seepage channels and establish an early barrier.

As shown in Fig. 7(b), the fluid loss in all three media decreases progressively with increasing GCS-g-PMOS concentration, indicating that the plugging agent is also effective in wider seepage channels. Although the absolute fluid loss remains different among the three media because of their different channel sizes and structures, the overall downward trend demonstrates that GCS-g-PMOS exhibits good multiscale plugging performance. This result suggests that the material can not only respond to micro-scale pores but also reduce fluid invasion in larger pores and fracture.

As shown in Table 6, the artificial filter cake results further confirm the plugging capability, demonstrated by the significant reduction in permeability after treatment with GCS-g-PMOS. The permeability reduction rate reaches a high level, indicating effective modification of the internal pore structure of the filter cake. This performance can be attributed to the combined effects of GCS and the oil-absorbing resin. The rigid carbon spheres provide structural support and bridging within the pore spaces, while the oil-absorbing resin swells and deforms in the oil phase, enhancing pore filling and sealing. This combined mechanism promotes the formation of a dense and stable plugging layer, which is consistent with the improved filtration behavior observed in Fig. 7.

3.3.4. Effect of temperature on the oil absorption and plugging performance of GCS-g-PMOS

The effect of temperature on the oil absorption of GCS-g-PMOS is presented in Fig. 8. As shown in Fig. 8(a), the oil absorption increases progressively with increasing temperature over 24 h. At higher temperatures such as 160 and $180\ ^\circ\text{C}$, the oil absorption is

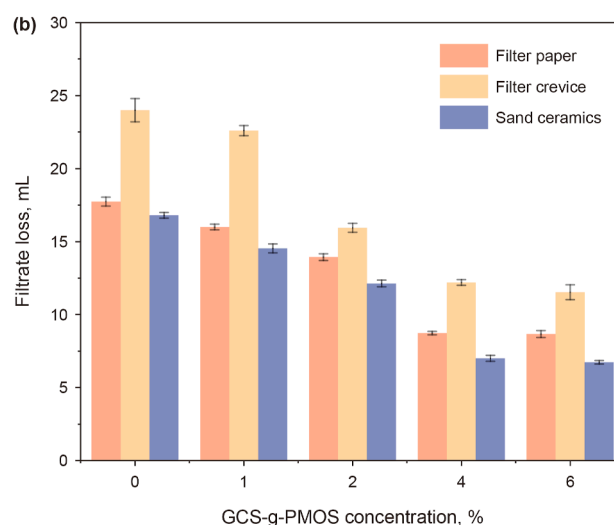
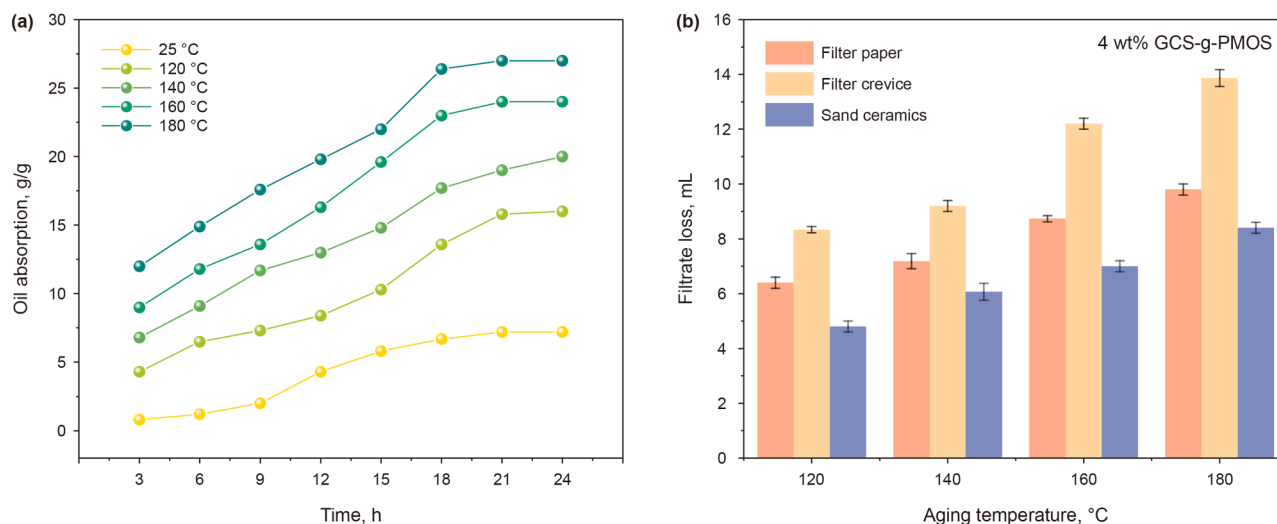


Fig. 7. Plugging performance of GCS-g-PMOS in multiscale media. (a) Filtration behavior through a $0.45\ \mu\text{m}$ microporous membrane; (b) filtrate loss through a $3\ \mu\text{m}$ sand ceramic, API filter paper ($<20\ \mu\text{m}$), and a 2 mm filter crevice.

Table 6

Filtrate volume, permeability, and permeability reduction of the artificial filter cake before and after treatment with GCS-g-PMOS.

GCS-g-PMOS concentration, wt%	FL _{before} , mL	FL _{after} , mL	K_0 , 10^{-4} mD	K_1 , 10^{-4} mD	k_r , %
0	1.58	1.60	9.34	9.00	3.64
1	1.62	1.00	9.40	7.34	21.91
2	1.56	0.82	9.53	6.18	35.15
4	1.54	0.50	9.65	4.11	57.41
6	1.60	0.50	9.68	3.53	63.53

**Fig. 8.** Effect of temperature on oil absorption of GCS-g-PMOS (a) and fluid-loss control of OBDF (b) at different aging temperatures.

significantly higher than that at 25 °C, and the growth rate is more pronounced in the early stage before gradually reaching equilibrium. This indicates that elevated temperature promotes the oil absorption process and accelerates the swelling behavior of the material (Li et al., 2025b; Wang et al., 2025).

The effect of aging temperature on the fluid loss of OBDF is shown in Fig. 8(b). It can be observed that the fluid loss increases with increasing aging temperature under all testing conditions, including filter paper, filter crevice, and sand ceramics. For example, the fluid loss under sand ceramics conditions shows a gradual increase from lower temperature to 180 °C. However, the overall variation remains moderate, and no sharp increase is observed, indicating that the drilling fluid maintains acceptable fluid-loss control even at elevated temperatures.

The temperature-dependent behavior can be attributed to the combined effects of thermal conditions on both the oil phase and the polymer structure. With increasing temperature, the viscosity of the oil phase decreases and molecular mobility is enhanced, which facilitates the diffusion of oil molecules into the polymer network. At the same time, increased chain mobility within the polymer matrix allows for more efficient swelling, resulting in higher oil absorption. In contrast, the slight increase in fluid loss at elevated temperatures may be related to the weakening of the structural integrity of the plugging layer under thermal conditions, although the overall sealing ability remains effective.

This behavior indicates that GCS-g-PMOS retains its plugging effectiveness under high-temperature conditions. The oil absorption and swelling characteristics of the resin enable it to fill pore spaces and microfractures effectively, thereby forming a sealing structure that can function under elevated temperatures. This

reflects the advantage of oil-absorbing resins in OBDFs, as their strong oleophilicity ensures good compatibility with the continuous oil phase and supports stable plugging performance.

Combined with the characterization results, the temperature-dependent plugging performance is closely related to the structure of GCS-g-PMOS. FTIR confirms the introduction of ester-containing PMOS chains, which provide oil-affinitive segments for oil absorption and swelling. TGA shows that the main degradation of the grafted polymer network occurs above 300 °C, supporting its structural integrity and fluid-loss control after aging at 160 °C. Moreover, the rough polymer-coated surface and strong oil affinity facilitate oil absorption and swelling-induced filling, thereby promoting the formation of an effective sealing structure.

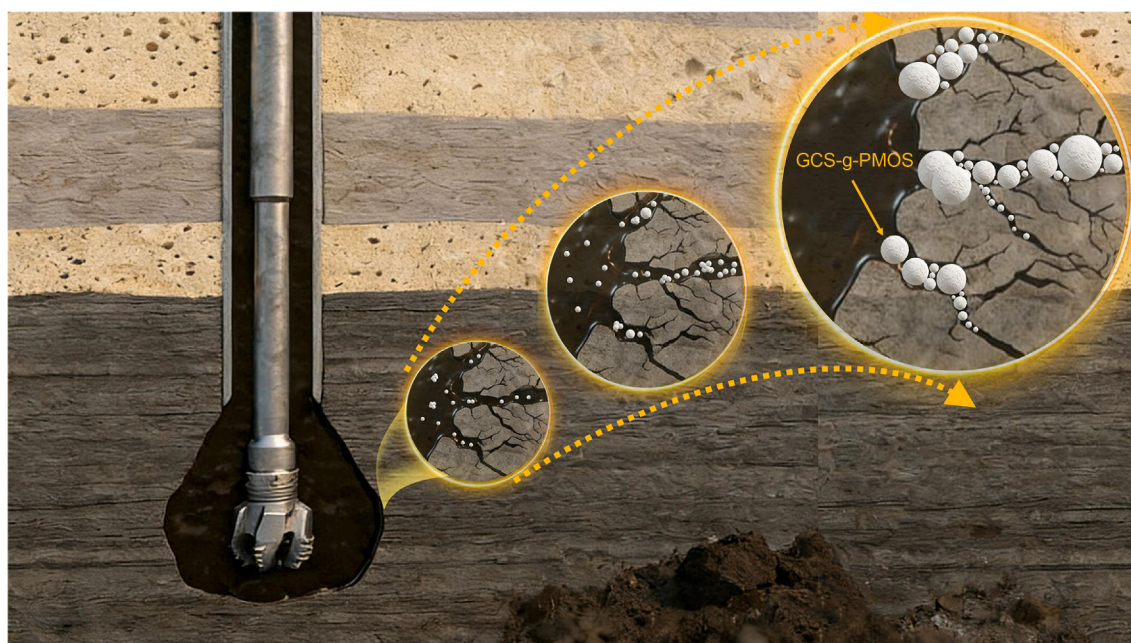
3.3.5. Comparative plugging performance with commercial agents

To further demonstrate the relative advantage of GCS-g-PMOS, its plugging performance was compared with that of several commercial plugging agents under the same conditions. As shown in Table 7, GCS-g-PMOS exhibited the lowest FL_{API}, the thinnest filter cake, and the lowest fluid loss through both the filter crevice and the sand ceramics after aging at 160 °C for 16 h, indicating its better adaptability to multiscale seepage channels. CaCO₃ mainly relies on rigid bridging, FGR and SF mainly provide mechanical filling or fibrous blocking, and HLKY primarily depends on resin deformation. In contrast, GCS-g-PMOS combines rigid GCS support with oil absorption and swelling-induced filling of the grafted PMOS phase. The interfacial coupling between GCS and PMOS enables cooperative bridging, deformation, and void filling, thereby forming a denser and more stable sealing layer. This structural advantage explains the superior filtrate control and multiscale plugging performance of GCS-g-PMOS.

Table 7

Comparative plugging performance of GCS-g-PMOS and commercial plugging agents after aging at 160 °C for 16 h.

Additive	Concentration, wt%	FL _{API} , mL	Filter-cake thickness, mm	FL _{filter crevice} , mL	FL _{sand ceramics} , mL
GCS-g-PMOS	4	3.2	1.40	12.0	6.80
CaCO ₃	4	4.6	1.90	17.8	12.0
FGR	4	4.4	1.53	13.0	9.6
SF	4	4.8	1.87	14.4	8.8
HLKY	4	3.4	1.45	14.0	7.0

**Fig. 9.** Schematic illustration of the proposed plugging mechanism of GCS-g-PMOS in OBDFs.

3.4. Plugging mechanism

Based on the results above, the plugging behavior of GCS-g-PMOS in OBDFs can be described as a combined bridging–filling–sealing process, as illustrated in Fig. 9. Under differential pressure, GCS-g-PMOS particles are transported into pores and microfractures along with the OBDF. The rigid GCS cores initially accumulate at pore throats or narrow fracture sections, forming a bridging framework through particle packing. This initial bridging effect reduces the flow channels and provides a structural framework for subsequent filling.

The grafted PMOS phase further improves the sealing effect through oil absorption and swelling. The RSM results indicate that the MBA content significantly influences the oil absorption behavior of GCS-g-PMOS. When the MBA content is too low, the crosslinked network is insufficient, which hinders oil retention and stable filling after swelling. When the MBA content is too high, the network becomes overly compact, restricting oil diffusion and swelling. Therefore, an optimized MBA content of 1.5 wt% provides an appropriate crosslinked structure that allows the polymer phase to absorb oil and swell while maintaining the stability of the plugging structure under differential pressure. During filtration, the swollen polymer phase fills the residual voids between rigid particles and further blocks seepage channels in the filter cake. Thus, the rigid GCS cores mainly provide bridging and support, while the oil-swollen PMOS phase contributes to void filling and sealing. This synergistic effect is

consistent with the multiscale plugging results and the artificial filter cake test. The permeability reduction rate increased from 3.64% in the control to 63.53% at 6 wt% GCS-g-PMOS, indicating that GCS-g-PMOS effectively reduced filter-cake permeability through the combined effects of rigid bridging and swelling-induced filling (Bai et al., 2025a).

4. Conclusions

A composite plugging agent, GCS-g-PMOS, was successfully synthesized by grafting an oil-absorbing polymer onto glucose-derived carbon spheres. This material combines the structural support of rigid carbon spheres with the swelling and filling properties of the polymer phase, providing an effective strategy for enhancing the plugging performance of oil-based drilling fluids (OBDFs) in multiscale pore and fracture systems. Response surface methodology identified the optimal preparation conditions as 0.75 wt% GCS, 1.5 wt% MBA, and 2.0 wt% BPO, under which the oil absorption capacity reached 9.02 g/g. Evaluation results demonstrated that GCS-g-PMOS had good compatibility with the OBDFs and improved rheological properties, emulsion stability, and filtrate control performance. After aging at 160 °C, the API fluid loss (FL_{API}) decreased from 4.4 to 2.4 mL at a concentration of 6 wt%, and the filter cake thickness was reduced from 2.0 to 0.9 mm. In addition, the material showed effective plugging ability in media of different scales, achieving a maximum permeability reduction rate of 63.53% in the artificial filter cake test. Compared with CaCO₃, FGR,

SF, and HLKY at the same concentration of 4 wt%, GCS-g-PMOS showed lower API fluid loss and lower filtrate loss through the 2 mm filter crevice and 3 μm sand ceramics after aging at 160 °C for 16 h. Overall, these results demonstrate that the combined effects of rigid support and swelling-induced filling contribute to the formation of a dense sealing layer, indicating that GCS-g-PMOS has strong application potential in high-temperature OBDs.

CRediT authorship contribution statement

Jia-Ning He: Writing – review & editing, Writing – original draft. **Yang Bai:** Investigation, Funding acquisition, Conceptualization. **Tuo-Ming Zhang:** Investigation, Data curation. **Deng Gu:** Visualization, Methodology. **Rui-Fang Wang:** Writing – review & editing, Project administration.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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