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Effects of plasma-treated rock asphalt on the mechanical properties and microstructure of oil-well cement

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HIGHLIGHTS

- The adhesion between the plasma-treated rock asphalt and matrix cement improved.
- The treated rock asphalt can improve the toughness of the oil-well cement paste.
- The treated rock asphalt can deflect cracks and restrain crack propagation.
- The rock asphalt has a positive effect on the pore structure within the cement paste.

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ABSTRACT

In this study, natural rock asphalt with a high softening point was selected as an oil-well cement admixture. The effects of the asphalt, which was subjected to a low-temperature plasma treatment, on the mechanical properties and microstructure of the cement paste were investigated. The results show that the hydrophilicity of the surface of the rock asphalt, and the interfacial bonding between the asphalt and cement matrix were improved with the use of the low-temperature plasma technology. The addition of the treated rock asphalt reduced the compressive strength of the cement paste but improved its tensile strength. Triaxial stress-strain curves, obtained under static and multicycle loads, showed that the rock asphalt improved the toughness of the oil-well cement paste. The addition of the 3% rock asphalt obviously enhanced the mechanical deformability of the cement paste. The results of the scanning electron microscopy analysis showed that the rock asphalt can deflect cracks and prevent crack propagation. The ability to restrain crack propagation improved as the adhesion between the treated hydrophilic asphalt and matrix cement improved. The results of the nitrogen-adsorption tests and mercury-intrusion porosimetry tests showed that the incorporation of the rock asphalt reduced the number of harmful pores present within the cement composite. The results of the X-ray diffraction (XRD) and thermogravimetric (TG) analyses performed on the asphalt-cement composites showed that the asphalt slightly retards cement hydration.

1. Introduction

Cementing is the key to oil and gas exploitation. It is the process of injecting cement slurry into the annular space that exists between the casing and formation within a wellbore, during drilling operations. The hardened cement slurry within the annular space is known as the cement sheath, and it provides structural support and zonal isolation [1]. The cement sheath is a brittle material, with low tensile strength and poor fracture toughness [2,3]. The effects of the cementing conditions, as well as the downhole temperature and pressure conditions, on the cement sheath can cause destabilization, and expansion of the cracks within the cement stone. These effects can also impair the integrity of the cement sheath, and result in poor adhesion between the casing and formation [4,5]. The failure of the cement sheath induces annular channelling and can even trigger huge economic losses and endanger life. Therefore, it is important to improve the toughness and tensile strength of cement paste by tailoring its microstructure to prevent crack propagation.

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Studies have shown that asphalt can improve the toughness of cement-based materials. Liu et al. [6] studied the mechanical properties of cement-emulsified asphalt mortar (CEAM) with varying asphalt-to-cement (A/C) ratios. The results showed that the toughness and 28-day elastic modulus of the CEAM increased and decreased, respectively, as the A/C ratio increased. Tian et al. [7] reported that the elastic modulus of a hardened cement asphalt binder decreased as the A/C ratio increased. Huang et al. [8] studied the effect of reclaimed pavement material containing asphalt and aggregate, also known as reclaimed asphalt pavement (RAP), on the mechanical properties of concrete. The results indicated that the toughness and strength of the concrete increased and decreased, respectively, as the RAP content increased. Rutherford et al. [9] found that CEAM possessed the properties of high-strength Portland cement mortar combined with the energy-dissipation capability of asphalt. Many existing reports provide examples of the successful use of asphalt as a toughening material in road concrete. However, considering the field of cementing engineering, few studies have used asphalt to improve the mechanical properties of well cement stone. This may be because the wellbore environment is more severe than that of a road, which includes atmospheric temperature and pressure. However, the wellbore possesses a high-temperature and high-pressure environment. In the field of road engineering, petroleum asphalt, with a relatively low softening point, can be used to improve durability. While in the field of cement engineering, asphalt will soften, deform, and flow under the high-temperature and high-pressure conditions of the wellbore. This reduces dramatically the strength of cement paste, and therefore does not toughen the oil-well cement (OWC) paste. Therefore, the key to toughening OWC paste is to select an appropriate asphalt material that satisfies the requirements of the service conditions. We previously studied the effects of sulfonated asphalt on the properties of OWC [10]. It was determined that a suitable quantity of sulfonated asphalt can greatly improve the tensile strength and toughness of OWC; however, the sulfonate group in the sulfonated asphalt, which has a strong retardation effect on hydration, will greatly reduce the early compressive strength of the cement paste. Therefore, it is necessary to identify other asphalt materials with high softening points that result in a lower degree of retardation of the toughening of OWC. The literatures have shown that natural rock asphalt is formed by petroleum that is exposed to heat, pressure, oxidation, catalysts, and bacteria, and undergoes geological deposition for hundreds of millions of years [11–13]. Natural rock asphalt has a high softening point compared with that of the petroleum asphalt. Therefore, natural rock asphalt was selected as the oil-well cement admixture in this study. The effects of the asphalt on the mechanical properties and microstructure of cement paste were studied.

The surface of rock asphalt is oil-friendly and cannot easily be dispersed in cement slurry. Thus, the hydrophilicity of the rock-asphalt surface should be improved initially. Low-temperature plasma technology can change the chemical composition and structure of a surface of a material. This is based on a multiphase chemical reaction that occurs at the interface between the solid polymer and gas phase [14,15]. Low-temperature plasma technology can be used to improve the properties of a polymer surface (such as its hydrophilicity, bonding, dye, and biocompatibility) via surface etching, the formation of a crosslinked structure, and the introduction of specific functional groups [16–18]. This technology can be used to improve the surface properties of a material while maintaining its original bulk characteristics [19–22]. Therefore, in this study, low-temperature plasma technology was used to improve the hydrophilicity of the surface of the rock asphalt. Subsequently, the feasibility of using natural rock asphalt to improve the toughness of oil-well cement paste was investigated.

2. Materials and method

The properties of raw materials are described in this section, along with details on the preparation and characterization of the specimens.

2.1. Raw materials

Class-G high sulfate-resistant cement was provided by Jiajiang Guiju Texing Cement Co., Ltd., Sichuan, China. The mineral composition of the cement is presented in Table 1. Rock asphalt was obtained from Mingzhi Industrial Co., Ltd., Shanghai, China. The particle-size distribution of the rock asphalt and cement powder was tested using a Mastersizer-2000 laser granulometer; the results are shown in Fig. 1. As shown, the particle sizes of the rock asphalt range from 3 μm to 300 μm, and the average particle size, d (0.5), is 53.58 μm. The particle sizes of the cement primarily range from 1 to 180 μm, and the average particle size, d (0.5), is 31.10 μm. Fig. 2 shows a scanning electron microscopy (SEM) photograph of the rock asphalt powder. As shown, the rock asphalt powders exhibit an uneven size distribution. The asphalt has a block-like shape with irregular edges; many bumpy lines exist on its surface. The main composition of the asphalt is shown in Table 2. During the experiment, some additives were incorporated into the oil-well cement, such as fluid-loss additives (G33S), dispersing agents (SXY-2), and defoaming agents.

2.2. Experiment

2.2.1. Surface modification of the rock asphalt

The surface modification of the rock asphalt was performed using low-temperature plasma equipment (model DT-03), manufactured by Suzhou Aopus Plasma Technology Co., Ltd, China. The samples were treated for pre-determined periods of 1, 3, and 5 min, under a flow of oxygen at a rate of 30–50 cm^3/min. A power of 120 W was used.

2.2.2. Contact angle test and infrared analysis

The hydrophilicity of the rock asphalt surface was analysed prior to and following modification, using a contact-angle measurement instrument (XG-CAMA, Shanghai Xuanyichuangxi Industrial Equipment Co., Ltd., China). The asphalt was first pressed into the form of small, round tablets, which were subsequently placed on the platform. Subsequently, a drop of water was dropped onto the surface of each tablet. Images were captured following the stabilization of the water droplet. As shown in Fig. 3, a tangent line is created along the droplet surface at the junction of the three phases, namely the solid, liquid, and gas. The contact angle is represented by the angle (θ_c) between the tangent line and the solid-phase boundary line in the liquid. Finally, the 'JY-82 series video contact-angle operating system software' of the contact-angle measurement instrument was used to measure the contact angle (θ_c) between the asphalt and water.

To characterize the changes in the functional groups of the rock asphalt prior to and following the plasma treatment, a Fourier-transform infrared spectrometer (FTIR, Nicolet-6700, USA) was used. To conduct the test, potassium bromide was mixed with the sample powder. A wave number range of 4000–500 cm^{-1} was used, along with a resolution of 4.

2.2.3. Specimen preparation

The oil-well cement slurry system consists of cement, water, asphalt, and various admixtures. The formula of the cement slurry is shown in Table 3. Cement slurries were prepared and cured according to American Petroleum Institute (API) Specification RP 10B-2 [23]. A water-to-cement ratio of 0.44 was adopted. The cement slurries were prepared by adding the powder to water, which was subsequently blended using a Waring blender at a speed of 4000 rpm for 15 s, and 12000 rpm for 35 s.

Table 1
Mineral composition of cement.

Composition	C_3S	C_3A	Fe_2O_3	MgO	$2C_3A + C_4AF$	SO_3	Loss	Insoluble residue
Content	57.33	2.34	4.76	1.42	18.21	2.52	1.03	0.55