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Full Length Article

An experimental study of the agglomeration of coal fines in suspensions: Inspiration for controlling fines in coal reservoirs

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ABSTRACT

Coal fines are among the key issues that restrict the highly efficient production of coalbed methane (CBM) because they cause blockages in reservoirs and accidents in wellbores. Coal fines always agglomerate to migrate and plug flow paths in formation water. Therefore, an investigation of their agglomerate behaviour in deionized, standard saline and $NaHCO_3$ suspensions was conducted on different rank coals (81.95 and 92.22% Cdaf) collected in the Ordos and Qinshui basins. The particle size distributions (PSD) of the suspended coal fines in suspensions over time were monitored to analyse the sizes and number of aggregations using a particle size analyser. The dispersive coal fines that were mostly less than 10 μm in size aggregated into several ranges of aggregations larger than 10 μm in a rapid process until reaching a stable period of sedimentation, inducing the average sizes of the suspended particles to increase rapidly and their quantities to suddenly decrease. As observed under a scanning electron microscope, the dispersive fine particles, which had quasi-ellipsoid, plate and flake shapes, clustered together. The average sizes of the suspended particles in the $NaHCO_3$ suspensions were the greatest, and their quantities were the smallest after the rapid agglomeration, exhibiting a capacity for agglomeration somewhat better than that in the standard saline suspensions and much better than that in the deionized suspensions. The PSD differences of the suspended bituminous coals in the three suspensions were more remarkable than those for anthracites because the former, which had higher ζ potentials measured using a microscopic electrophoresis instrument, were more easily impacted by different types of ions. Bituminous coal has more oxygen-containing functional groups than anthracite, which forms thicker hydration layers that prevent coal fines from agglomerating. According to extended DLVO (Derjguin-Landau-Verwey-Overbeek) theory, cations (Na^+ and H^+), especially H^+ ionized by HCO_3^-, can neutralize the charge of electrical double-layers and help coal fines to agglomerate. In addition, OH^- hydrolysed by HCO_3^- can reduce the viscosity of suspensions. Therefore, in CBM reservoirs, $NaHCO_3$ may fix some portions of coal fines at their sources by agglomeration and contribute to the other fine particles migration by reducing the viscosity. $NaHCO_3$ may be a potential additive in fracturing fluid, which needs to be further researched.

1. Introduction

Typically, a coalbed methane (CBM) production process consists of three stages: dewatering, stable production and decline, and those stages correspond to three flow phases: single-phase water flow, gas-water two-phase flow and single-phase gas flow [1]. During progressive dewatering (i.e., depressurizing), gas flows in the pore and cleat networks, and production rises to a peak for months or years [2]. Production from good gas wells can be high and stable for a long time. Although gas production is influenced by various factors, blockage by fines that plugs flow paths (i.e., pore throats, cleats and proppant packs) is a non-negligible issue that results in a significant decline in gas production [3]. The fines issue occurs throughout the entire process of gas production, although the concentration and size of fines gradually decrease [4,5]. Zhao et al. investigated the sizes of coal fines produced in CBM wells in the southern Qinshui basin and found the primary sizes ranged from 10 μm to 100 μm for a production time of less than 1 year, and few fines exceeded either 1 μm or 1000 μm [6].

Fines generation has been summarized into five aspects: 1) the tectonic effect, 2) fracturing, drilling and perforation, 3) hydraulic drag and lift due to fluid flow, 4) desorption of gas, and 5) increases in effective stress [7]. Due to the tectonic effect, coals with severe brittle deformation and ductile deformation contain significant amounts of fine grains or powders in seams [8,9]. This type of CBM reservoir is

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always avoided on the scene. During hydraulic fracturing, coal failure is more likely to occur near wellbores that have the highest pressures [10]. Coal fines generated during hydraulic fracturing are massive and larger in size than those found in production [11]. Those fines cannot be discharged completely, and some portion of them will deposit in the proppant pack and block gas seepage [12]. When gases are desorbed from the coal matrix, desorption-induced shrinkage not only opens the coal fissures, increasing permeability but also induces stress anisotropy and shear failure in coals [13]. Coal fines are produced by shear failure primarily due to exposed microstructures, cleat elbows and micro-facture tips [14,15]. These fines migrate through and deposit in pores, cleats and fractures, which reduces their conductivity and permeability [16]. Therefore, the trend of large increases in permeability during this period is sometimes followed by a sudden drop in permeability [17].

In addition to the above issues, fluid velocity sensitivity (FVS), which induces massive fines in coal reservoirs, is an important issue that impacts gas productivity. The FVS effect mainly occurs in the two-phase gas and water stage [18]. As the fluid velocity increases, coal fines will rearrange and migrate into pores, cleats and the proppant pack. They may be deposited in fissures [19], particularly in narrow flow paths [20], which results in blockages and decreasing permeabilities. Fines migrations in flow paths are controlled by mineralogy, salinity, changes in PH and flow velocity [21].

These factors governing fines migration also impact the agglomeration of coal fines [22]. Coal fines favour aggregation and detainment in flow paths when compared with fines in sandstone reservoirs (Fig. 1). Aggregations of coal fines also influence the rheology of formation water [23]. However, little research has been conducted on the behaviours of agglomerates and their impacts on CBM reservoirs.

In this work, the agglomerate behaviours of coal fines are investigated in three types of suspension systems: deionized suspensions, standard saline suspensions and sodium bicarbonate ($NaHCO_3$) suspensions. The particle size distributions (PSD) of the suspended coal fines were compared among all solutions and coal types. The mechanisms responsible for those differences are discussed by employing extended DLVO theory.

2. Coal samples and methods

2.1. Coal samples and their geological setting

The experimental coal samples were collected from the No. 3 coal seam of the Shanxi group (in the Permian), from the Ulan colliery located in the Ordos basin and Sihe colliery in the Qinshui basin (Fig. 2). According to the ASTM D388-15 international standard, the samples were bituminous coal and anthracite coal with primary banded structures and a predominant clarain lithotype (Fig. 2). Table 1 shows their basic properties, and the proximate and ultimate analyses followed the ASTM D3172-13 and ASTM D3176-15 international standards, respectively.

The Sihe colliery is located in the southern margin of the Qinshui basin, which is among the most successful areas of commercial CBM production in China (Fig. 2a). The basin's structures are relatively simple, and the coal-bearing strata are stable, with an average tip of 4° [24]. The Taiyuan formation (Upper Carboniferous) and Shanxi formation (Lower Permian) consist of approximately 5 mineable coal seams, the total thickness of which is approximately 150 m (Fig. 2b). The No. 3 coal seam is the only mineable seam in the Shanxi formation (Lower Permian) and has an average thickness of 6.11 m. This coal seam is the primary target formation for CBM development [25]. Most of the coals in the southern Qinshui basin are anthracite, and the gas contents generally range from 10 to 37 m^3/t [26].

The Ulan colliery, which contains medium volatile bituminous coals, is situated on the east wing of the Hulusitai syncline, which is on the northwest edge of the Ordos basin (Fig. 2a). The Taiyuan formation (Upper Carboniferous) and Shanxi formation (Lower Permian) are also main coal-bearing strata, with approximately 17 coal seams in this area (Fig. 2b). The total thickness of the coal seams is approximately 30.74 m, and the No. 3 coal seam has an average thickness of 9.7 m. In that coal seam, the relative amount of gushing gas is 16.15 m^3/t.

2.2. Experiments and methodology

Fig. 3 shows a sketch of the experimental process. The −80 mesh (< 185.41 μm) coal samples were smashed, and their true particle size distributions (PSD) were measured using an Ls100Q Particle Size Analyzer (PSA) (Beckman Coulter, Inc., USA). To investigate the effects of ion concentration and pH on the agglomerate behaviours of the coal fines, four coal-water suspension systems including a deionized suspension, standard saline (mass ratio of $NaCl:CaCl_2:MgCl_2 \cdot 6H_2O$ = 7.0:0.6:0.4) suspension, $NaHCO_3$ suspension and alcohol suspension were prepared using the −80 mesh coals. The pH values of the first three were 6.3, 6.25 and 8.5, respectively, and the ion concentrations of the standard saline suspension and $NaHCO_3$ suspension were both 1500 mg/L. The concentrations of the coal fines in the suspensions were 0.5 g/ml, and each suspension's volume was 200 ml.

A S400 particle size analyser (PSA) (Mettler Toledo, LLC., Redmond, USA) was used to monitor the particle size distribution (PSD) of the suspended fines in the analytical process of the suspensions after they were sufficiently stirred (Fig. 3). This equipment can monitor the particle sizes of suspended solids and count them in real time based on the reflection theory of laser light. The measurement range of this device is 1–1000 μm. The PSDs of the deionized, standard saline and $NaHCO_3$ suspensions were monitored during the first standing time of 20 min and at the 1st, 3rd and 6th hours of standing.

Ethyl alcohol can be used as a dispersant to effectively prevent coal fines from agglomerating. An alcohol suspension was utilized to investigate the true PSDs of the suspended coal fines and to compare the differences among the PSDs for the original smashed and suspended coal fines in the suspensions.

The true PSDs of the original smashed coal fines were measured using an Ls100Q PSA made by Beckman Coulter. That instrument was used to measure the dried particle sizes from 0.4 to 1000 μm and to count them using diffracted light. Small amounts of dispersant (Coulter Dispersant Type IIA anionic) were used to disperse the coal fines.

A Quanta TM 250 scanning electron microscope (SEM) (FEI, Oregon, USA) was used to observe the coal fine aggregates (Fig. 3). The SEM's maximum image resolution is 3 nm. To discuss the mechanisms of the agglomeration behaviours of the coal fines, the functional groups of the coal samples and the ζ potentials of the coal surfaces in the suspensions were analysed in this work (Fig. 3). The functional groups were analysed using a Vertex 80v Fourier transform infrared

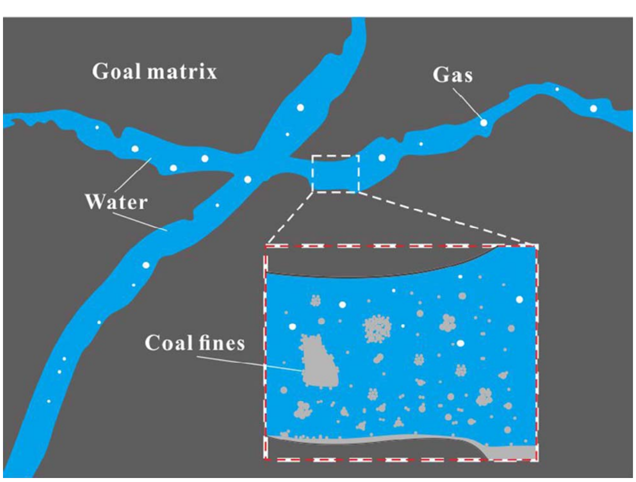

Fig. 1. Schematic of the agglomeration and migration of coal fines in reservoirs.