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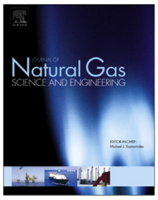

Acid-induced mineral alteration and its influence on the permeability and compressibility of coal

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ABSTRACT

The natural fracture system in coal serves as the primary conduit for water and gas flow in coal seam gas fields. For low permeability coal with highly mineralised fracture systems, the dissolution and/or modification of mineral occlusions could potentially enhance permeability and improve stress resilience. This study investigated the effect of mineral alteration by hydrochloric and hydrofluoric acid (HCl–HF) on fracture compressibility and coal permeability. Coal core immersion in 15% HF- 4% KCl solution has enhanced coal permeability to brine from 0.10 to 0.45 mD and reduced fracture compressibility from 0.020 to 0.006 bar^{-1}. Enhanced permeability and improved stress resilience were attributed to kaolinite ($Al_2Si_2O_5(OH)_4$) dissolution and hieratite (K_2SiF_6) precipitation, respectively. Geochemical speciation, simulating HF interactions with coal fracture minerals, predicted the occurrence and prevalence of both dissolution and precipitation reactions. Scanning electron microscopy-energy dispersive spectroscopy confirmed the mineral alteration phenomena. Identification of resultant structural changes and the differentiation of chemical from physical effects were elucidated using X-ray computed tomography. The overall findings show that mineral alteration by HF yielded relatively large, crystalline minerals that appeared to provide structural support to fractures, resulting in enhanced fluid flow and improved resistance to compression.

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1. Introduction

The natural fracture system in coal serves as the primary conduit for water and gas flow in coal seam gas fields. (Flores, 2013; Gray, 1987; Laubach et al., 1998; Rodrigues et al., 2014; Seidle, 2011) These fractures can be mineralised to a certain degree. (Laubach et al., 1998) In areas where fracture mineralisation is high, permeability routes are often restricted. (Gamson et al., 1992, 1993; Titheridge, 2004) During the various stages of coal seam gas (CSG) extraction, detrimental processes exacerbate the pre-existing flow restrictions. Undesirable consequences of drilling and well completion, such as fines migration and formation damage, may occur and persist though the production stage. (Civan, 2007) Mobilised particulates will likely jam these flow restriction sites, consequently decreasing coal permeability. Moreover, as effective stress increases with fluid pressure drawdown, (Alexis et al., 2015; Palmer and Mansoori, 1998) restricted flow channels risk further constriction. Thus, in order to ensure the longevity and economic viability of a CSG well, coal permeability routes must be improved, or at the least, sustained.

For low permeability coal seams with highly mineralised fractures, dissolution of mineral occlusions will likely increase fluid flow capacities. The alteration of fracture mineralogy may also influence fracture compressibility. Typically, mineral occlusions in fractures are composed of authigenic minerals, (Laubach et al., 1998; Dawson and Esterle, 2010; Ward, 2002) such as carbonates and aluminosilicates. Carbonaceous minerals dissolve in mild hydrochloric acid (HCl) whilst aluminosilicates degrade in hydrofluoric HF (HF) acid. These acids are commonly used in well workovers and formation stimulation in the conventional oil and gas industry. (Kalfayan, 2008).

In acid stimulation of sandstone formations, a sequential acid flood is typically implemented. The stimulation strategy consists of a pre-flush with HCl, a main flush of HCl–HF mixture appropriate for the targeted sector, and an overflush with HCl. (Kalfayan, 2008; Guo et al., 2007; Williams et al., 1979) The pre-flush serves to dissolve carbonates and transport mobile divalent ions away from the target sector. This is to limit fluoride precipitation during the main flush. Also, this stage serves to displace water away from the

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near-wellbore regions, minimising formation damage associated with fluorosilicate formation. (Williams et al., 1979) The main flush then enables the HF component to attack silicates whilst HCl provides sufficient acidity to maintain the reaction products in solution. Lastly, the over-flush removes spent HF from the targeted sector to prevent fluorosilicate precipitation.

It is conceivable that a similar acidising approach can be adopted for coal seam gas application. Although acidising has worked for sandstone reservoirs, the relatively more complex nature and remarkable heterogeneity of coal (Flores, 2013; Seidle, 2011; Chen et al., 1995) require that this current technology be examined in more detail. For instance, with the various reactions (Balucan et al., 2015; Steel et al., 2001a, 2001b; Steel and Patrick, 2001; Turner, 2015) that are likely to occur during coal acidising, complete coal demineralisation is practically unachievable. But even with partial mineral alteration, the dissolution of the initial mineralisation and subsequent mineral precipitation reactions (Steel et al., 2001a; Steel and Patrick, 2001; Shuchart and Ali, 1993) could potentially alter the fracture porosity (φ_f), permeability (k) and compressibility (c_f) of coal. Fig. 1 illustrates the possible effects of acid-induced mineral alteration on fracture porosity and compressibility.

In HCl–HF acid stimulation of coal fractures, mineral occlusions (Fig. 1a) will degrade once contacted with the advancing acidising fluid. (Kalfayan, 2008; Williams et al., 1979; McLeod, 1991; Portier et al., 2007) Previous studies on coal demineralisation with HF and HCl (for the purpose of producing ultra clean coal), (Steel et al., 2001a; Steel and Patrick, 2001) have shown that HF is effective in reducing coal mineralisation. Flow restrictions will then diminish as cleat minerals and jammed particulates dissolve in HF (Fig. 1b). The combination of acid-induced cavitation and interconnection of fluid pathways would likely result in enhancement of coal permeability.

The removal of cleat mineralisation may however result in collapse into newly created cavities and closure of existing fractures with the decline in fluid pressure (Fig. 1c). Further dewatering and subsequent production would eventually compress these flow channels. Although matrix shrinkage due to gas desorption (Harpalani and Schraufnagel, 1990; Pan and Connell, 2012) counteracts progressive fracture compression, volumetric reduction of fluid pathways is highly probable. As demineralised fractures will be subject to ever-increasing compressive forces during the course of resource extraction, proppant injection (Keshavarz et al., 2014a, 2014b; Khanna et al., 2013) may therefore be required to maintain these flow channels open.

With the need to sustain acid-stimulated fractures, mineral precipitation (Fig. 1d) may not be entirely counterproductive. Newly formed fluoridated precipitates (hereon referred to as neofluorides) could impart structural support to prop open newly created flow channels (Fig. 1e). Not only could HF serve as an effective fluid for mineral dissolution but could also facilitate the formation of potential proppants. Earlier studies (Steel et al., 2001a; Steel and Patrick, 2001) have shown that HF concentration influences mineral dissolution and precipitation reactions. The latter of these two reactions becomes more pronounced at higher concentrations. Table 1 lists the relevant mineral dissolution (R1-R6) and precipitation reactions (R7-R14) that could occur during HF acidising.

Neofluorides that enhance and maintain fluid flow with increasing stress may be considered as in-situ generated mineral proppants. Minerals that are larger in size and with higher mechanical strength than the initial mineralisation will permeate fluids and resist compaction better. Relatively larger structures pack less densely and will normally have larger interstitial spaces. This allows for better fluid flow as compared to that of densely packed minerals. Structurally robust minerals (influenced by crystalline habit, crystal defects, etc.), such as those having blocky (i.e., hieratite) or cubic crystal habits, are more likely to prop open permeability routes than fibrous or platy varieties (i.e., kaolinite).

With the likelihood of altering the fracture mineralisation with acidising, enhanced permeability with improved resilience to stress

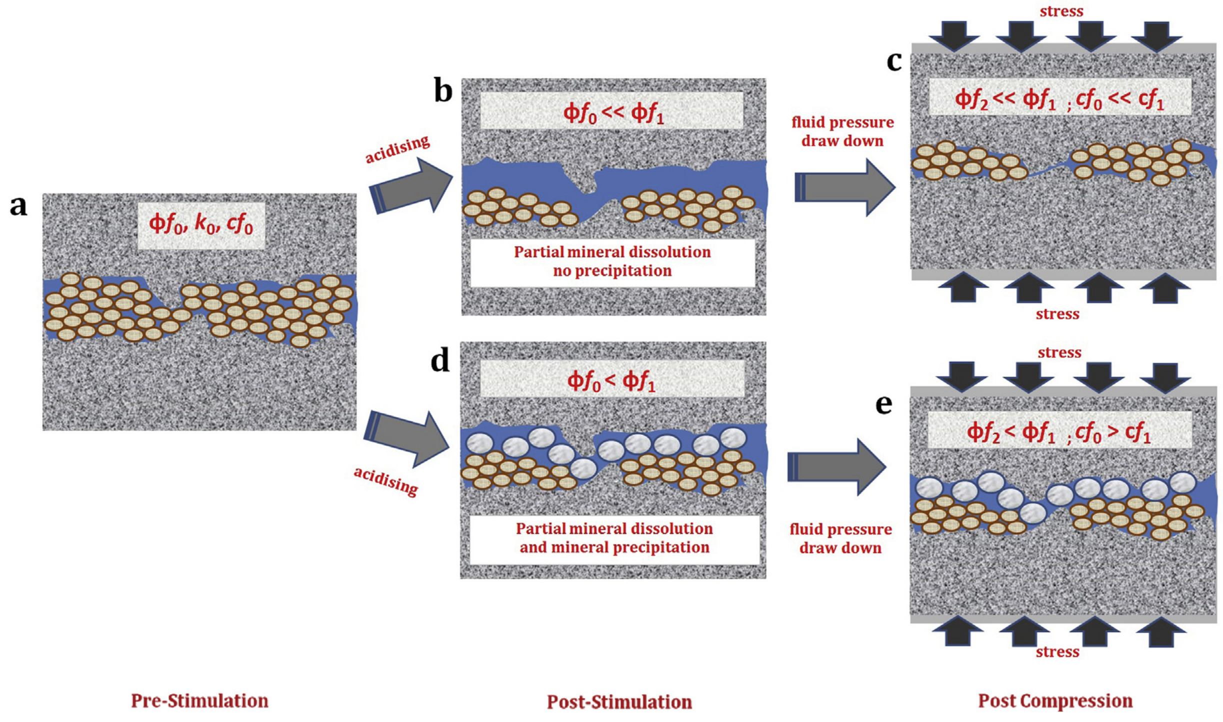

Fig. 1. Simplified diagram of a (a) mineralised coal fracture and the possible effects of (b, d) acidising and (c, e) compression on fracture porosity (φ_f) and compressibility (c_f). The possible outcomes for acidising without precipitation (b–c) and acidising with precipitation (d–e) during fluid pressure draw down are indicated.

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