



Advances of nanotechnologies for hydraulic fracturing of coal seam gas reservoirs: potential applications and some limitations in Australia

Hannah Marsden¹ · Sudeshna Basu^{1,3} · Alberto Striolo^{1,4} · Melanie MacGregor^{2,5}

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Abstract

Some of the most promising potential applications of nanotechnology to hydraulic fracturing of coal seam gas (CSG) are reviewed with a focus on Australian CSG wells. Three propitious applications were identified: (1) Nanoparticle enhanced viscoelastic surfactants (VES) fracturing fluids to prevent fluid loss by up to 30%, made possible by the formation of pseudo-filter cakes and reducing the viscosity of the VES fluids. Besides, there is no requirement of clay control additives or biocides. (2) Nano-proppants to extend fracture networks and reduce proppant embedment by introducing them prior to the emplacement of larger proppants. Fly Ash nanoparticles can be particularly effective because of their high sphericity and mechanical strength. (3) Nanoparticle-coated proppants, to mitigate the migration of particle fines by restricting them close to their source by adsorption, with MgO being the most effective. The use of nanotechnology in hydraulic fracturing applications is currently hindered due to a discordant regulatory environment compounded by the cost of the nanoparticles themselves, as well as, a lack of field data to validate the technology under real downhole conditions. Although the necessary field tests are unlikely to be conducted for as long as abundant natural gas is available, exploratory studies could pave the way for future applications.

✉ Sudeshna Basu
Sudeshna.basu@ucl.ac.uk

¹ Department of Chemical Engineering, University College London, London WC1E 7JE, UK

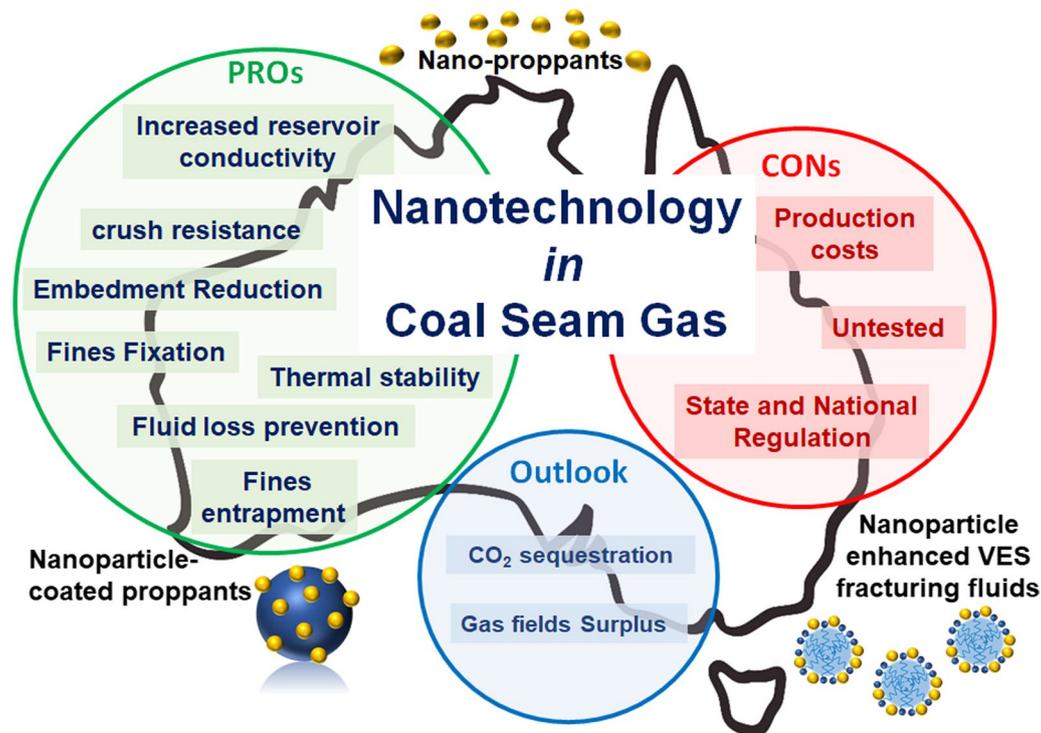
² Future Industries Institute, University of South Australia, Mawson Lakes Bvd, Mawson Lakes, SA 5195, Australia

³ Department of Earth Sciences, University College London, London WC1E 6BS, UK

⁴ School of Chemical, Biological and Materials Engineering, University of Oklahoma, Norman, OK 73019, USA

⁵ Flinders Institute for Nanoscale Science & Technology, College of Science and Engineering, Flinders University, Bedford Park, SA 5042, Australia

Graphical abstract



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1 Introduction

Coal seam gas (CSG) or coalbed methane (CBM) is an unconventional natural gas adsorbed on the internal surfaces of coal seams, in equilibrium with the free gas within the natural fractures or coal cleats. A minor proportion of CSG is also dissolved in the formation fluid present in the pore spaces (Cleveland and Morris 2015). The gas is held in place because the coals are saturated with water (Roarty 2011). Coal seams can hold larger quantities of gas per unit volume of rock than conventional reservoirs (Seidle 2011). Depending on the burial history, rank of the coal, precursor maceral composition and subsequent migration of the gas, the methane content varies from less than 1 to more than 25 m³ gas per ton of coal (Laxminarayana and Crosdale 1999; Jenkins and Boyer 2008; Zou et al. 2017). Under these scenarios, global CSG resources are estimated in excess of 256 trillion cubic metres (Seidle 2011; Islam 2015). In Eastern Australia, CSG has been linked to the development of the liquified natural gas (LNG) export industry, with its promised direct economic benefits (Simshauser and Nelson 2015; Marcos-Martinez et al. 2019).

CSG is extracted from coal seams occurring at depth of more than 200 m from the ground surface. Extending

the extraction depth from 200 to 500 m, can increase the measured gas content by 2.5 times (Esterle et al. 2006). This makes extraction from greater depth, up to 1500 m, economically attractive (Unconventional Gas Mining Submission 121 2014). The vast quantity of water extraction required to depressurize the coal seam that can lead to aquifer depletion, is one of the greatest environmental concern surrounding CSG extraction in Australia, where many regions suffers from chronic water shortages (Dingsdag 2016). As of 2016, the water produced from CSG development in Queensland was 60.5 Giga litres/year, equivalent to 1700 Giga litres of water to be produced over the life of one development (Salmachi and Yarmohammadtooski 2015; Underschultz et al. 2018). Another potential risk associated with CGS extraction is groundwater contamination with chemicals, methane and dissolved salts (Dingsdag 2016). Contamination can occur as the stray gas migrate to an aquifer above the gas deposit along poorly sealed gas production wells and along new fractures and existing faults enhanced and re-activated by fraccing (Jackson et al. 2013; Vengosh et al. 2014). The management and environmental impact of the water co-produced with CSG extraction has been the object of other reviews (Hamawand et al. 2013; Davies et al. 2015; Mallants et al. 2018), which also stressed the need for efficient

innovative technologies to achieve a sustainable CSG extraction. Such breakthrough may be brought about by existing knowledge base currently applied for other conventional and unconventional hydrocarbons extraction. We consider here the viable applications of nanotechnology to enhance CSG extraction by fracking, with focus on the Australian CSG landscape.

1.1 Coal seam gas in Australia

One of the first countries to effectively take advantage of CSG as a clean-burning fuel is Australia. As shown in Fig. 1a, production of CSG began in 1995 and has grown almost exponentially since (Evershed 2018; Roarty 2011). The majority of CSG is found in the state of Queensland, in the Bowen and Surat Basins (Fig. 1b). In 2017, CSG reserves in Australia (both proven and probable) were estimated to total nearly 40,000 PJ in Queensland alone (Gas Resources in Australia: resources assessment and operation overview 2017). CSG makes up almost one third of natural gas production in Australia (Evershed 2018), compared to only 3% in the USA (Natural Gas Explained 2019). It is an important resource to the Australian economy with strong implications for its national energy security. Indeed, the gas produced is used domestically, and the vast surplus is exported. To access the global marketplace, LNG plants (i.e. Gladstone) as well as thousands of kilometres of pipelines are being built along the Queensland coast (Roarty 2011). It is expected that as conventional gas fields such as the Cooper Basin are depleted and the mining of solid coal

drops, CSG production will play a major role in Australian energy exports (Australian Energy Update 2018 2018).

The percentage of CSG wells requiring stimulation is increasing. Mallants et al. (2018) estimated that although only 6% of the CSG wells drilled in Queensland have so far undergone hydraulic fracturing, 40% will require stimulation over the next few decades. Maximising the efficiency of the fracking process is essential to minimise the overall production costs. One major technical challenge lies in the fact that the permeability of coal seams is reduced by the closure of the cleats as the fluid pressure declines during water production and fracking (Underschultz et al. 2018). This challenge can be complicated by the complexity of both mineralogy and the natural fracture system of coal seams. Quartz, calcite, pyrite, kaolinite and other clays are typically present in different proportions (e.g. Zhang et al. 2019), resulting in high anisotropy in strength and high sensitivity to stress of the formations, compounded by differences in methane adsorption capacities of different clay minerals (Wang et al. 2020).

1.2 Hydraulic fracturing in coal seam gas

The productivity of an unconventional formation correlates with the permeability of the correspondent rocks. Coal seams permeability is determined by their cleats network, the matrix porosity being comparatively insignificant (Moore 2012; Seidle 2011), in addition to the interconnectivity between the pores and the fractures, the stress state and other physical properties. Sometimes, coal seams have sufficient permeability (> 1 mD) to allow the desorbed gas

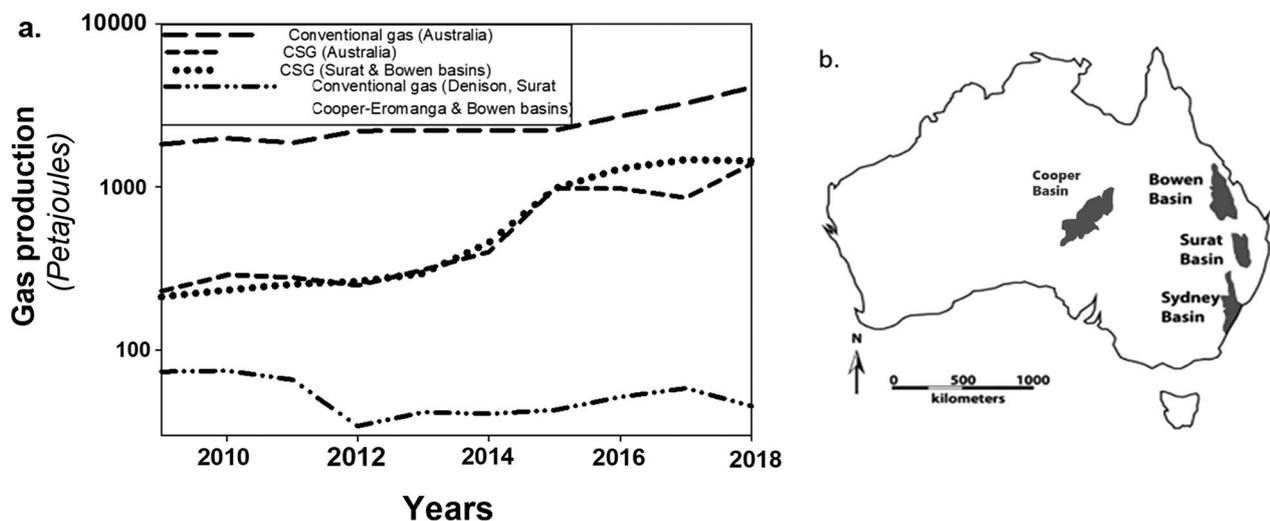


Fig. 1 **a** A significant increase in yearly production of CSG in Australia and from the Bowen and Surat basins from 2009 to 2018, as compared to conventional gas production. Data from Australian Government Geoscience Australia (2021) and Queensland Government, Open Data Portal. **b** Map of Australia, showing the most promising

ing areas for future CSG production—The Bowen and Surat Basins in Queensland and the Sydney Basin in New South Wales. Cooper Basin has also been the focus of CSG appraisal. Image modified after Moore (2012)

to flow to the wellbore. However, in most cases, hydraulic fracturing, referred to as fracing herein, or other forms of stimulation are necessary to achieve a higher fluid conductivity (APLNG; Huang et al. 2012; St John 2017; Alexeev et al. 2010; Sampath et al. 2019).

Fracing involves pumping a fluid into the formation at high pressure in order to create flow pathways in the rock (Cuss et al. 2015; Li et al. 2015a). The fracing fluid, which is typically water-based or an inert gas such as nitrogen, contains a variety of chemical additives with various functions (Cuss et al. 2015; Gottardo et al. 2016), one of which is a proppant. This crush-resistant material, often sand or ceramic, acts to keep the fractures open once the hydraulic pressure is relieved, so that fluid can flow to the wellbore (Cuss et al. 2015; Yekeen et al. 2019). After fracing, the fracture network consists of new fractures connected with existing natural ones (APLNG). In gas shales, where fracing has been successfully applied, up to 90% of the fracing fluid can be retained in the formation and the flow-back water can have very high salt content (e.g. Yethiraj and Striolo 2013). In contrast, natural gas extraction from coal seams occurs by desorption as the water is pumped out of the fractures, thereby reducing the confining pressure. While CGS recovery can be relatively slow (Roarty 2011), fracing in coal seams is less aggressive than in gas shales as it requires lower pressures (SEPA, version 121119).

Fracing can be used to recover CGS from unexploited coal beds as well as from either active or abandoned coalmines. Indeed, existing underground tunnels in coalmines offer pathways for fracing operations. Fracing can be used to increase the drainage area of an individual well (APLNG), thus increasing the yield per wellbore and reducing the number of wells required to produce a given formation. In addition to assisting in meeting the growing demand of energy, CSG extraction can also help in preventing environmental accidents and disasters due to methane gas leaks and outbursts from coal seams (Zhang et al. 2018).

1.3 Nano-fracing technology

The efficiency of hydraulic fracturing can be improved via the use of nanoparticles and of nanotechnology in general, a technique sometimes referred to as ‘nano-fracing’. Nanoparticles have unique properties due to their small size (on the order of tens of 10^{-9} m). They have high surface area to volume ratio, as well as high strength (Yekeen et al. 2019). Although, by definition, a nanoparticle has dimensions below 100 nm, in the field of hydraulic fracing the term ‘nanoparticle’ has been used to refer to both nano- and micro-scale particles (e.g. 1–1000 nm) (Gottardo et al. 2016). The physical and chemical properties of nanoparticles differ from their bulk counterparts, with significant effects below 10–20 nm (Montaño et al.

2014). For instance, stable emulsions/foams for enhanced oil recovery were achieved by using small particles with nominal diameters of 5 nm (Kim et al. 2016). Academic interest in nanotechnology applications to the petroleum industry has grown sharply in recent years (Nur Agista et al. 2018). Nanoparticles have found wide application in the hydrocarbon industry as nanosensors, drilling fluid and cement additives as well as for improving oil recovery and hydraulic fracing (Nur Agista et al. 2018). Although, so far, little effort has been targeted specifically to applying nanotechnologies to the fracing of coal seams, studies conducted in adjacent research fields may provide useful insights. For example, since the adsorption of nanoparticles on different rock types is not only controlled by the flow rate but is also strongly influenced by the presence of clay minerals (Omurlu et al. 2016; Zhou et al. 2017), titanium nanoparticles can be used as coatings on carbon nanotubes to influence residual gas adsorption/desorption (Zhang et al. 2019).

It should be recognised that the use of nanoparticles in the characterisation and eventually exploitation of unconventional hydrocarbons has been enabled by a wealth of fundamental studies. For example, Fakhri et al. (2010) quantified the transport of carbon nanotubes in porous materials, and showed that the nanotubes mobility can be controlled by tailoring their stiffness. Saha et al. (2011) showed that single walled carbon nanotubes can adsorb on porous silicate materials maintaining many of the characteristics of the pristine carbon nanotubes. Worthen et al. (2016) demonstrated how it is possible to stabilise nanoparticles dispersions in highly concentrated brines and Urena-Benavides et al. (2016) demonstrated that uniform polyelectrolyte coatings reduce the adsorption of magnetite nanoparticles in sandstone. The feasibility of advanced applications also depends on our detailed understanding of interfacial properties, facilitated by multi-scale modelling (e.g., Zheng et al. 2020; Phan and Striolo 2019; Striolo 2019).

Gottardo et al. (2016) identified 25 types of nanoparticle applications for fracing unconventional reservoirs, including 31 commercial products. Here, we first discuss the available literature on nanotechnology applied to the fracing of unconventional reservoirs, and we subsequently identify the most potentially useful applications in CSG. In the following discussion, common problems encountered during the fracing of coal seams are first identified, before reviewing nanotechnology-based materials and methods that have the potential to mitigate these issues. At last, recommendations are presented for the next steps in applying nanotechnology to fracing of CSG wells in Australia, towards improving both economic and environmental performance.

2 Nanotechnology opportunities for coal seam gas

2.1 Environmental implications of fracking leak-off during CSG operations

Sometimes, shallow coal seams, located just several hundred metres below the surface, have such low permeability (< 1 mD) to warrant stimulation (Palmer 2010; Peduzzi and Harding 2013; Seidle 2011). These coal seams may only have thicknesses on a decimetre to metre scale (Rodvelt 2014) and are often close to aquifers. Cracks induced by fracking could propagate out of the coal and into the surrounding formation, providing pathways for both fracturing fluid and produced gas to potentially contaminate groundwater, used for drinking or agriculture, as in Queensland (Espig and de Rijke 2016; Mallants et al. 2018). Also, poor environmental performance leads to a negative image for the industry compromising social acceptability (Lock the Gate 2020).

Another form of leak-off is observed when the gas migrates through the overlying formation, with methane ultimately venting into the atmosphere (Peduzzi and Harding 2013), thereby increasing the greenhouse footprint of CSG to 20–100 times that of solid coal (Roarty 2011). Furthermore, fluid leaking reduces the pressure in the formation, lowering the efficiency of the fracking as well as producing operations themselves (Cuss et al. 2015). As a result, the volume of coal drained by each well is reduced, the surface footprint of the operation increases requiring additional associated infrastructure (Peduzzi and Harding 2013) related to environmental impacts such as increased land clearing and loss of habitat, as well as an increase in cost.

Despite the risks just listed, it should be recognised that a recent investigation on the impacts of hydraulic fracturing for CSG in the Surat Basin Queensland Australia, suggests that any effect on soil, water and air quality is minimal (GISERA 2020). The study reported that elevated concentrations of salts, ammonia, metals, organic carbon and other compounds reduced to pre-fractured conditions within 40 days; in addition, current water treatment operations effectively removed most fracking and geogenic chemicals from the produced water. Yet, an overall reduction in the microbial activity was noted, which had a pronounced impact on nitrifying microorganisms (GISERA 2020). Findings being somewhat contradicting (IESC 2014; GISERA 2020), long-term evaluations may be needed to gain a better understanding of the overall environmental impact of fracking related to CSG operations.

2.2 Leakage-mitigation potential of nano-fracking fluids

There are three main types of fracking fluids; their comparative advantages and disadvantages are outlined in Table 1: (1) Slickwater, made almost exclusively of water and proppants, is very commonly used in CSG due to its low cost, and the ability of coal to tolerate large volumes of water (Barati and Liang 2014). However, because little can be done to prevent slickwater fluid leak-off, this fluid is not considered further here. (2) Polymer-based fracking fluids can display excellent fluid-leak off prevention through the formation of a filtercake, a very low permeability layer that forms on the fracture surfaces (Das et al. 2018; Vipulanandan et al. 2014). Nanoparticles can be applied to polymer-based fluids to improve several characteristics, including fluid leak-off prevention (Liang et al. 2015; Wang et al. 2017; Zhang et al. 2017). However, polymers can be adsorbed to coal surfaces; because the fractured surfaces present surface areas of up to 3 m² per cm³ (Ren et al. 2014), polymer adsorption can lead to residues that block fractures and prevent the gas migration to the wellbore (Zhang et al. 2017). As such, polymer-based fluids do not show promise in CSG applications and will not be discussed further. (3) Viscoelastic surfactant (VES) fluids are a relatively recent introduction to hydraulic fracturing technologies. It has been reported that, by using VES fluids, it is possible to limit fracture growth height (Fontana et al. 2007); VES generally do not leave a residue due to the low molecular weight of surfactants (Wu et al. 2018). Although they do not lend themselves to the formation of a filtercake (Yekeen et al. 2019), nanotechnology can provide useful properties to VES fluids used in CSG stimulation.

2.3 Nanoparticle-enhanced VES fluids

VES fluids are composed of a three-dimensional entangled network of worm-like micelles (Huang et al. 2010) (Fig. 2). The viscosity of VES fluids depends on the type of the salt solutions and the concentration of surfactants (Fig. 3), as these quantities affect the structure of the micelles. Adding nanoparticles can result in the pseudo-crosslinking of the micelles due to the high surface forces (Van der Waals) associated with the nanoparticles (Huang et al. 2010). As the micelles become physically cross linked (Fakoyah and Shah 2013), this increases the viscosity of the VES fluids (Fig. 3) even at high temperatures of up to approx. 135 °C, thus improving their proppant carrying capacity (Gurluk et al. 2013; Crews and Huang 2008). The phenomenon also leads to the formation of pseudo-filtercakes, which reduce the rate of fluid leak-off (Huang et al. 2010). Furthermore, while a polymer-generated filtercake requires chemical breakers to breakdown post-fracture, appropriate internal breakers such as polyols, for a VES filtercake can be applied

Table 1 Details of the three main fracturing fluid types and their relative advantages and disadvantages in terms of fracturing fluid leak-off

Method	Description	Advantages	Disadvantages	Sources
Slickwater	Composed primarily of water with proppant Small concentrations (<0.1%) of other chemical additives which may include polymer and friction reducer Low viscosity	Cheap and environmentally friendly—lack of chemical additives reduces the risk of groundwater contamination high stimulated reservoir volume, better fracture containment	Little to no filtercake formation (minute filtercake formation possible)—poor proppant carrier, leading to narrow fracture width and excessive water use Requires a higher injection rate which can damage the formation and create fines (see Sect. 2.6)	Rodvelt (2014), Yekeen et al. (2019), Barati and Liang (2014), Gottardo et al. (2016) and Gandossi (2013)
Polymer-based (aka gelled water)	Composed of water, proppant and a gelling agent, often guar or similar, plus other chemical additives for various purposes Viscosity increases with polymer content or with the addition of crosslinkers	Formation of a filtercake, a low permeability layer that lines the fracture surfaces and blocks pores; prevents fluids from escaping into the surrounding formation	Blocking of conductive pathways by a residue formed from the polymer sticking to the surfaces of the coal due to a high surface affinity excessive fluid loss in high permeability formation where filtercake formation is not possible	Rodvelt (2014), Yekeen et al. (2019), Barati and Liang (2014), Das et al. (2018), Gottardo et al. (2016) and Gandossi (2013)
Viscoelastic surfactant (VES)	Formed from an entanglement of worm-like micelles in a 3D network (Fig. 2) Composed of predominantly water, proppant and surfactant	Can limit fracture growth height so that induced fractures do not extend beyond the coal seam	No formation of a filtercake facilitating significant fluid leak-off	Yekeen et al. (2019), Barati and Liang (2014), Huang et al. (2010), Yan et al. (2016), Fontana et al. (2007), Gottardo et al. (2016) and Gandossi (2013)

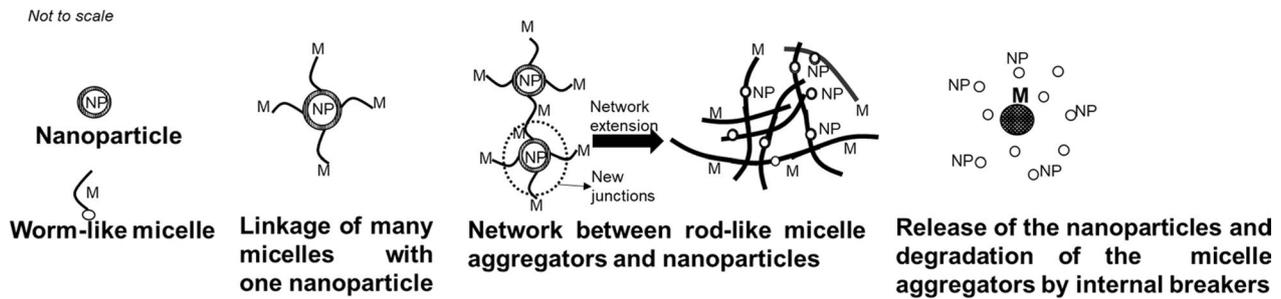


Fig. 2 Schematic representation of the mechanism of nanoparticle-enhanced VES fluid. Modified after Li et al. (2019) and Shibaev et al. (2021)

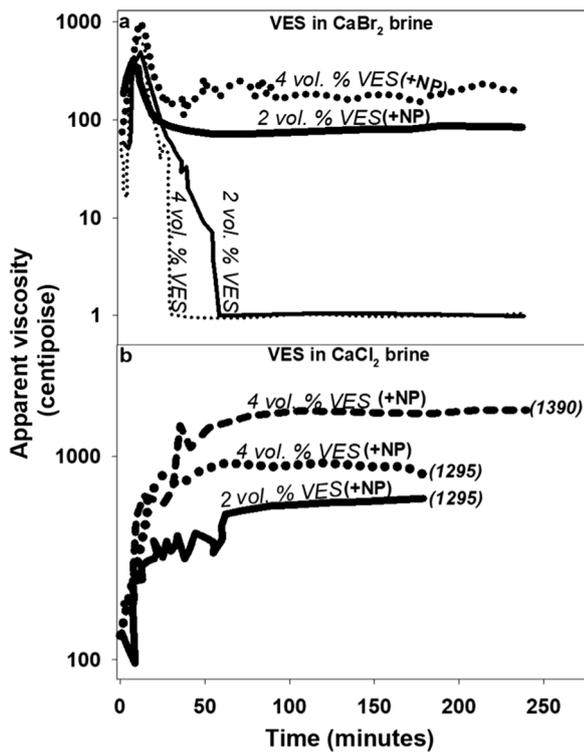


Fig. 3 The apparent viscosity of 2 vol% and 4 vol% VES in salt solutions of **a** CaBr_2 , **b** CaCl_2 . The addition of MgO nanoparticles (+NP) results in an improvement of viscosity which varies with the surfactant content, and the salt concentration indicated in parentheses in units of milligrams/cm³ against their respective curves in **b**. Modified after Gurluk et al. (2013)

within the micelles (Crews and Huang 2008; Yekeen et al. 2019). The internal breakers work by rearranging the more entangled elongated micelles to non-viscous spherical structures (Fig. 2), allowing for easy clean up and restoration of the conductive pathways (Crews and Huang 2008). As an additional benefit, VES fracturing fluids do not require clay control additives as the cationic surfactant in the system can function as a temporary clay control agent (Gomaa et al. 2011), nor biocides since they do not contain any biopolymers (Gandossi 2013; Gandossi and Von Estorff 2015).

Xiao et al. (2017) investigated a nanoparticle-enhanced VES fracturing fluid specific to coals. By introducing a low concentration (0.5%) of silica nanoparticle-modified polyester fibre to an anionic VES fracturing fluid, they were able to effectively reduce the velocity of leak-off by over 30%. The silica nanoparticles adhered to the micelles and acted as crosslinkers because of the high density of charged groups on the nanoparticle surfaces. This formed a filtercake-like structure that obstructed fluid flow. The addition of the nanocomposite fibre also reduced the amount of surfactant required to achieve the viscosity, by reducing the frictional resistance up to 20% at a shearing rate of 5000 s⁻¹; this lowers the potential of the surfactant to adsorb onto the coal surface (and thus block conductive pores and fractures for gas flow). It also reduces cost, which is often a fundamental barrier to the implementation of VES fracturing fluids, as the cost depend on the amount of nanoparticles used. Maxey et al. (2008) demonstrated that nanoparticle concentrations below 1.12 wt% can be sufficient to improve fluid loss properties. Together, these works indicate that the nanoparticle concentrations required to reduce fluid leak off are relatively low. The additional cost associated with nanoparticles used as additives may not be a deterrent if it is accompanied by a reduction in the surfactant concentration from 2.5% to 1.0% to achieve an equivalent proppant-carrying capacity (Xiao et al. 2017).

The nanoparticles can also block nano- as well as micro-sized pores (Fig. 4) that conventional fluid loss control additives could not. Abrams (1977) reported that particle diameters must be no more than one third of the size of the pore throat if they are to effectively block it, as the particles need to penetrate into the pores to cause bridging. As the permeability of coal drops with depth (Fig. 5), so does its porosity and pore throat diameter (Barati 2015). The throat radius distribution of different coal samples varies considerably, with a range of 0.72–8.66 μm (Pan et al. 2019). The simple addition of SiO_2 and polyelectrolyte complex nanoparticles to a fracturing fluid was found to reduce the volume of fluid loss by more than half, for those samples with a permeability of 0.1 mD and below (Fig. 6).

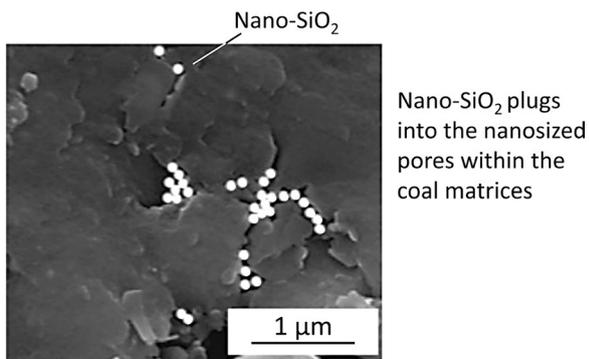


Fig. 4 SEM picture of coal sample. Nano SiO₂ (white spheres) plugs into the nanosized pores in the coal matrix. Image reproduced with permission from Cai et al. (2016)

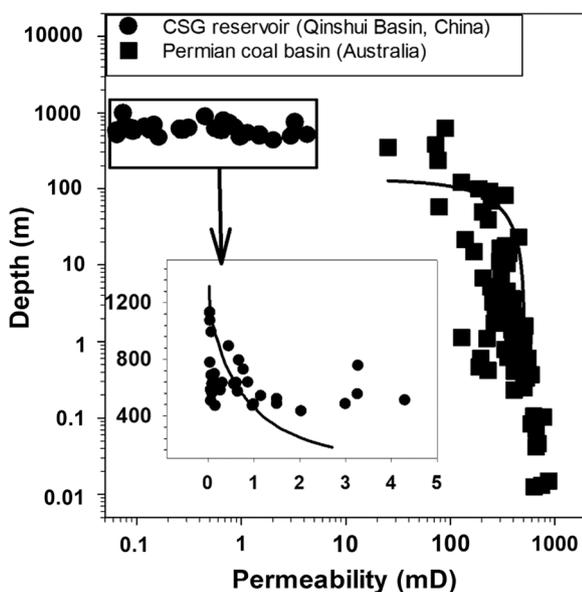


Fig. 5 Permeability against depth for an unnamed Australian Permian coal basin (square) and a CSG reservoir in China (circle). The permeability varies with depth, based on pressure and temperature conditions, as well as strata stress. The figure is reproduced from Moore (2012) and Guo and Cheng (2013)

2.4 Fluid loss volume and permeability: effect of nanoparticles

Coal seams over 2.5 km deep have a reduced permeability (Fig. 5), thereby reducing the ease of extraction (Moore 2012). A larger lithostatic load presents further complications related to fracture collapse/conductivity during hydraulic stimulation. For instance, halving a fracture width can decrease its local permeability by eight times (Seidle 2011). Using proppants is critical to maintain fracture aperture (Seidle 2011; Keshavarz et al. 2015). The proppants must be strong enough to withstand the closure stresses

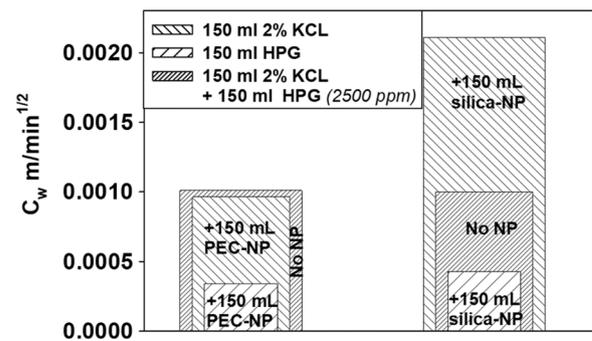


Fig. 6 Fluid loss coefficient (C_w) calculated for silica and polyelectrolyte complex (PEC) nanoparticles (NP), mixed with 2% KCl and hydroxypropyl guar (HPG) gum and, compared with the results for HPG solutions made in 2% KCl with no added NP. Modified after Barati (2015)

experienced at depth, which could be greater than 20,000 psi (approx. 140,000 kPa) in the deepest wells (Ottestad 2014; Wanniarachchi et al. 2017). For coal seams at depths less than 1200 m, quartz sand is sufficient (Rodvelt 2014). However, deeper seams may require advanced proppants with higher mechanical strength. It has been observed that SiO₂ and Fe₂O₃ nanoparticles can improve the compressive strength of cement mortar (Li et al. 2004), and that spinel (MgAl₂O₄), a nano-porous oxide, is capable of withstanding pressures of up to 13,000 psi (approx. 90,000 kPa) (Ottestad 2014). For these strategies to succeed, placing the proppants within every cleat and induced fracture is required for sustaining complex fracture networks able to effectively drain the seam. This is challenging because particle agglomeration or their attachment to the coal matrix can inhibit deep penetration of the injected proppants (Keshavarz et al. 2015).

2.5 Nano-proppants and conductivity

Coal is a relatively ductile rock, which can undergo creep (Ren et al. 2014). At high closure stresses, a proppant particle may be embedded into the fracture surface rather than holding it open, reducing the fracture aperture by 10% to 60% and the conductivity by 100 times (Zhi and Elsworth 2020). A recent study has shown that using a proppant size smaller than 16/30 Tyler Mesh Size (particle size range of 600–1180 micron) showed only 0.1 mm embedment, compared to 0.56 mm for proppants 16/30 in Mesh Size (Bandara et al. 2021). Despite having the capacity to greatly reduce methane production, proppant embedment in coals has received little academic attention.

Gottardo et al. (2016) identified three main applications of nanoparticles as proppants, as shown in Table 2. Out of these, ultra-lightweight proppants (ULWPs) appear to be the most promising in CSG, particularly in tackling the problem of embedment. A product, used in commercial

Table 2 Details of three main nanotechnology applications related to proppants

Site	Description	Applicability in coals	Sources
Ultra-lightweight proppants (ULWP)	<p>Have a much lower density than conventional proppants; often exploit highly porous or hollow materials such as spinel</p> <p>Can be transported much further into the formation without settling</p> <p>Are able to prop open a more complex fracture network and results in a more even distribution of proppant on fracture surfaces</p>	<p>Capable of being carried deeper into the fracture network, even by less viscous slickwater fluids (often used in fracturing of coal seams)</p> <p>May lead to a more even distribution of particles resulting in a lower likelihood of embedment</p>	<p>Bicerano (2012), 'FracBlack HT: Ultra-lightweight Proppant' (2018), Ottestad (2014), Smith et al. (2011) and Gottardo et al. (2016)</p>
High-strength proppants	<p>Addition of nanoparticles to coating on proppant particles</p> <p>Increases proppant strength and improves chemical and thermal stability at downhole conditions</p> <p>Low density of fibre coatings improves suspension in fracturing fluid; better proppant placement</p>	<p>Improves crush resistance of conventional proppant particles, useful for deeper coals</p> <p>Proppants likely to perform more effectively at reservoir conditions</p> <p>Potentially creates a more even distribution of proppant particles resulting in lower proppant embedment</p>	<p>Guo et al. (2012), Bustos et al. (2007) and Gottardo et al. (2016)</p>
Proppants for tight and ultra-tight formations	<p>Nano-sized proppants (on the scale of 1 nm per proppant particle)</p> <p>Able to be transported through the smallest fractures and thus prop open a highly extensive fracture network</p>	<p>Capable of propping open nano- and micro-scale fractures and cleats to improve overall conductivity</p> <p>May form a monolayer on fracture surfaces to aid in the prevention of proppant embedment</p>	<p>Bose et al. (2015), Gottardo et al. (2016) and Yekeen et al. (2019)</p>

CSG completions in China, already utilises this technology: FracBlack HT™ ('FracBlack HT: Ultra-lightweight Proppant' 2018). ULWPs are nanocomposites made of a thermoset polymer matrix and a nano-filler, which yields a density close to that of water (see Table 3). This allows for effective transport of the proppants through the fracture network ('FracBlack HT: Ultra-lightweight Proppant' 2018), which promotes even distribution, thereby lowering the likelihood of embedment (Zhi and Elsworth 2020). FracBlack HT™ is reportedly capable of withstanding closure stresses of up to 8000 psi (approx. 55,000 kPa), in part due to its particle sphericity.

Bicerano (2012) performed conductive tests on proppant particles subjected to heat treatment and, reported that using thermoset nanocomposite ULWPs can improve conductivity by up to 50%, with the most suitable nano-filler materials being fumed silica and fumed alumina or carbon black. ULWPs can also be produced as hollow materials, which have an inherently low density (Gottardo et al. 2016). For example, Oxane Materials Inc. produces a range of hollow proppants designed with a ceramic shell of sintered nanoparticle-reinforced polymers that could improve productivity by 25% (Johnson 2010; Smith et al. 2011). The resultant proppant—OxThor™—is 100 micron in size and crush resistant up to 20,000 psi (Approx. 140,000 kPa) (Garneau 2014); the—OxFrac™ proppant is 100 s of microns in size and specifically designed with controlled manufacturing process to create perfectly spherical, hollow, mono-dispersed size particles, for conditions relevant to CSG (Gottardo et al. 2016). Shilova and Rybalkin (2018) found that the effectiveness of similar hollow aluminosilicate ULWPs increases with depth. The performance can be further improved by combining ULWPs with nano-sized proppants (Fig. 7). The smaller proppants would be injected first, followed by the larger ones. In this way, the nano-sized proppants are transported into the smallest fractures without compromising the conductivity of the main proppant pack in the larger structures (Bose et al. 2015). This allows for the propping of maximum fracture length, enhancing productivity. Besides, silica and polyelectrolyte (PEC) nanoparticles can be used as fluid loss control additives during fracing of low permeability reservoirs, to improve the propagation of the hydraulic fractures

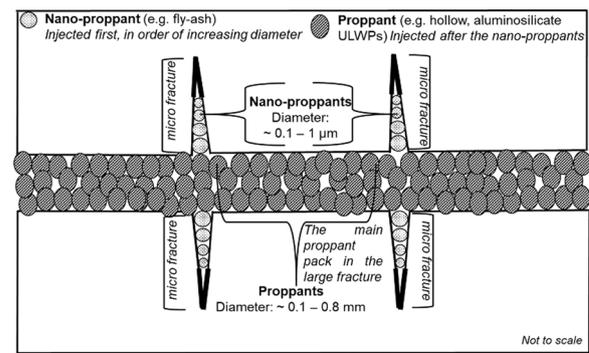


Fig. 7 Schematic representation of proppants and nano-proppants distributed in fractures and micro-fractures respectively, injected in stages during hydraulic fracing for CSG. Modified after Bose et al. (2015)

(Fig. 6). The fluid loss was reduced to zero when PEC was mixed with 2% KCl or, when silica nanoparticles were used with HPG (hydroxypropyl guar) solution (Barati 2015).

Fly-ash a by-product of coal-fired power stations, because of its properties has been suggested as an attractive nano-sized proppant (Table 3). The strength of fly-ash particles is adequate to resist typical closure stresses, and it is enhanced by a high degree of sphericity (Fig. 8), which is also favourable for securing fracture conductivity (Bose et al. 2015). Fly-ash may also have fluid loss control properties (Barati and Bose 2017), in addition to being cheap.

Other, more complex proppant technologies also utilise nanomaterials. For example, Liu et al. (2018), investigated a nanoparticle-stabilised emulsion that breaks down at depth as it interacts with the fracture surfaces, releasing the nanoparticles. The nanoparticles then adsorb onto the rock surfaces and act as proppants, with exceptional proppant placement throughout the fracture network. Likewise, carbon nanotubes offer low density coupled with strength higher than diamond (Yekeen et al. 2019), making them potentially suitable as proppants. This is promising as carbon nanotubes, now only obtained from synthetic precursors, may be obtained commercially from renewable sources such as essential oils or plant shoots (Janas 2020).

Table 3 Key properties of the FracBlack HT™ ULWP (FracBlack HT: Ultra-lightweight Proppant 2018) and fly-ash (Zhang et al. 2011; Jawed et al. 2017; Barati and Bose 2017)

Property	FracBlack HT™	Fly ash
Specific gravity (g/cm ³)	1.054	1.0–1.8
Sphericity	> 0.9	> 0.9
Median diameter	0.313 mm (30/80 mesh)	3.2 μm
Maximum closure stress	8000 psi (approx. 55,000 kPa)	1.7 × 10 ⁵ psi (approx. 1.2 × 10 ⁶ kPa)
Maximum temperature (°C)	135	100

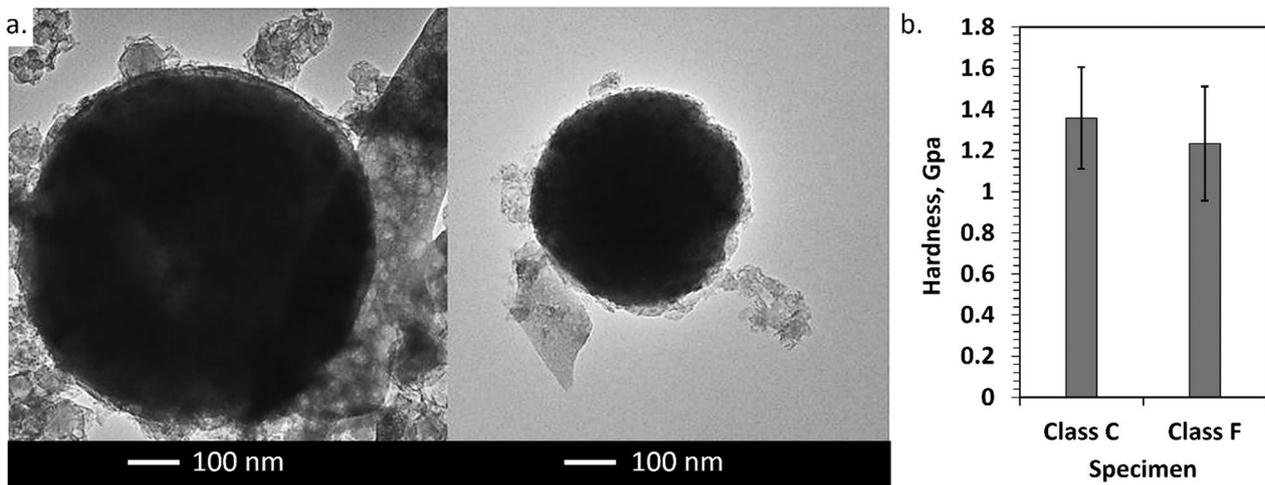


Fig. 8 **a** TEM images of fly-ash nanoparticles, illustrating their sphericity; and **b** Measured hardness of Class C and Class F fly-ash nanoparticles. Image adapted from Bose et al. (2015)

2.6 Nanotechnology for fines mitigation

Mechanical failure of coal seams is a fairly regular occurrence (Lu and Connell 2020), typically resulting in the formation of fines. Fines are small particles of the rock material that break away and can be carried by the fluids (Zheng et al. 2018). They could block channels and pore throats, reducing conductivity and productivity (Barati and Bose 2017; Moghadasi et al. 2019; Rodvelt 2014). Coal fines can migrate to the wellbore, where they both block and damage the equipment. Fines can also be created during downhole proppant crushing, a phenomenon aggravated for smaller proppants, and yet can be reduced to some extent, by using high proppant concentration (Bandara et al. 2021). To mitigate the effects of fines, nanoparticle-enhanced VES fluids may be employed.

Even with prevention measures in place, fines formation is always to be expected and requires mitigation. Nanoparticles can be applied as surface modification agents, which can effectively mitigate fines migration (Rodvelt 2014). The nanoparticle readily stick to the proppant surface and also interact with fine particles; any fine particle in the vicinity will be attracted to the nanoparticle and captured (Barati and Bose 2017). Thereby, nanoparticle-modified proppants can flocculate fines such as colloidal silica, charged and non-charged rock particles as well as clays, and restrict them closer to their source (Huang et al. 2008). This prevents blockage of critical fracture channels and stops the fines from reaching the wellbore (Belcher et al. 2010), potentially extending the life of a given well. In laboratory experiments, MgO nanoparticles have been found to be the most effective proppant-coating for fixing fines (Ahmadi et al. 2013) and TiO₂ is reported to have the highest adhesion force, i.e. better at holding the fines in place (Zheng et al. 2018). Metal

oxide nanoparticles have also been investigated as additives for fines fixation (Gottardo et al. 2016). Habibi et al. (2012) compared the performance of Al₂O₃, MgO and SiO₂ nanoparticles and found that, like with proppant coatings, MgO was the most effective at fixing fines by adsorption; fines migration was reduced by 15% when just 0.1 wt% of MgO nanoparticles were used. Nanoparticles have been successfully used for a similar purpose in drilling fluids in CSG completions (Cai et al. 2016).

3 Limitations to the implementation of nanotechnology in CSG in Australia

3.1 Regulations

Fracking is a highly regulated procedure (APLNG), which is important in building public trust. Generally, any new chemical additive, including nanoparticles, must be assessed on factors such as environmental compatibility and possible hazards to human health before they can be used in the field. A new chemical must be submitted to the Australian Industrial Chemicals Introduction Scheme and then listed on the Australian Inventory of Industrial Chemicals (AICIS Fact Sheet 2020). Toxicology is a particular concern if nanoparticles were to end up in aquifers used for drinking water and agriculture (Abbott Chalew et al. 2013; Tosco and Sethi 2018). Materials that are not harmful in their macroscopic dimensions may become so in the form of nanoparticles (Warheit et al. 2008; Yang et al. 2010). Besides size, since the toxicity of nanoparticles depend on a large number of parameters such as surface charge, shape, stability and ability to aggregate and dissolve, each nanomaterial needs individual consideration (Gicheva and Yordanov

2013). Nanoparticles stability, both short and long-term in the porous media need to be addressed with testing under real environmental conditions (Batley and McLaughlin 2010). The nanoparticles' manufacturing should also be considered when quantifying the life cycle assessment of the technology; for example, Sengul et al. (2008) identified high energy and water demands and high waste production that are related to the manufacture of nanoparticles. However, some nanoparticles such as fly-ash and carbon black are by-products of existing industrial processes and do not require additional manufacturing.

In Australia, regulation is further compounded because State and Territory governments have primary responsibility for regulating the environmental impact associated with the resource sector (Stilwell and Troy 2000), while the federal Australian government becomes involved when a CSG development is likely to have a significant environmental impact. Typically, matters of national environment significance include potential impacts on water resources. These are protected through the 2013 "water trigger" provisions of the Environment Protection and Biodiversity Conservation Act 1999 (EPBC Act amendment 2013). This means that each state can adopt its own regulations and assessment procedures, thereby possibly creating additional administrative hurdles in the nationwide implementation of new technologies. For example, in Queensland, operations must adhere to the Petroleum and Gas (Production and Safety) Act 2004, while in New South Wales, hydraulic fracturing is regulated by the Petroleum (Onshore) Act 1991 (Hydraulic Fracture Stimulation 2015). Although the National Partnership Agreement on Coal Seam Gas attempts to unify state regulations, it still devolves much of the responsibility to State and Territory governments (National Partnership Agreement on Coal Seam Gas and Large Coal Mining Development 2012). However, it is fundamentally important to take into account the expectations and concerns of local communities, as well as maintain transparency in the operations of CSG. These considerations can be integrated in the state level regulatory framework with costs and benefits, to offer flexibility in the strategic approaches to CSG development (Cronshaw and Grafton 2016).

3.2 Implementation of nanotechnology in CSG operations

Any additional monitoring and treatment of produced waste water due to the use of nanoparticles, is expected increase the gas production costs. Most of the fracking fluids will be recovered with the produced water along with the desired gas (APLNG) and the nanoparticles will likely need to be removed prior to disposal. Nonetheless, it may be the case that the nanoparticles concentration in the produced water is extremely low. It is estimated, for example, that an average

CSG well in Queensland will generate around 10,000 L of produced water per day (St John 2017). The maximum concentration of specific nanoparticles allowed in produced water before treatment processes are mandated still remains to be identified. The lower this concentration threshold is, the more expensive treatment processes are expected to be. Further, the fracking fluid that remains in the formation could leak into aquifers, potentially contaminating them with nanoparticles. Potential environmental risks due to nanoparticles both in aquifers and in produced water will have to be quantified, as well as mitigation procedures. Considering the time and resources that must be invested by each applicable state government, companies might be deterred from further developing nanoparticle-based technologies for CSG. The likelihood of this risk is expected to increase over the next few decades as regulation tightens (Seidle 2011) in line with increasingly sceptical public opinion.

3.3 Costs

Principally, the aim of introducing any new technology, including nanoparticles, is to increase profits and reduce environmental risks. Nanoparticles could achieve these goals by yielding higher production per well. However, nanoparticles add extra costs, both in the procurement and potentially in the monitoring and treatment of produced fluids. While some nanoparticles, such as fly-ash, can be acquired cheaply (Yekeen et al. 2019), others, such as carbon nanotubes, are valued at hundreds of dollars per gram (Mohamed 2012). Montgomery (2013) estimates that a typical fracturing fluid should cost no more than \$4/US gallon. Since manufacture of nanoparticles is highly energy intensive, addition of nanoparticles, even at concentrations < 1% (APLNG), would add substantial costs. As an estimate, using $(8.6\text{--}24.3) \times 10^4$ kg of nanomagnetite as a downhole contrast agent against fracturing sand and ceramic proppant would be US\$ 1–3 billion per well (Morrow et al. 2014). For nanotechnology to be feasible in CSG operations, low cost, large-scale production of custom built nanoparticles of suitable size, shape (e.g. rod-shaped vs. spherical) and surface charge, is required. Silica nanoparticles used to improve the rheological performance of VES could provide one such example (Hanafy et al. 2018).

3.4 Lack of field tests

Another barrier to the implementation of nanotechnology in CSG is the lack of field-scale tests. Yekeen et al. (2019) report that there have been no documented field trials of nano-fracking, with many possible applications still undergoing research and development. Moreover, almost all the available literature focuses on fracking in shales, and so more research is required on technologies targeted for coal

seams. Alsaba et al. (2020) reported four field trials of applications of nanoparticles in the oil and gas industry. They were related to enhanced oil recovery and shale formation stabilization, as well as evaluating the stability of carbon-based fluorescent as nano sensors under reservoir conditions. However, none of the trials were related to CSG. While laboratory studies have their uses, it is vitally important for field tests to be undertaken early in the development stage to assess how any new technology perform at reservoir conditions where pressure and temperature are high and complex stress states are present, compounded by the variability in coals properties.

One intrinsic risk related to nanoparticles is their possible agglomeration (Wang et al. 2018), with loss of intrinsic characteristics and properties, including the high surface area to volume ratio which bring about the high surface forces expected, and their ability to fit into tiny fractures and pores. One important focus of future research in this field should be on the stabilisation of common nanoparticles like silica in injected fluids and prevention of their aggregation in coal samples of different ranks (Wang et al. 2022). The laboratory findings should be applied to a larger scale in the field for testing, which would need better collaboration between researchers and CSG developers.

4 Other potential solutions

There are other options available for improving recovery of CSG that are currently cheaper and potentially more socially appealing than nanoparticles. One such alternative is chemical stimulation (Jing et al. 2020). In acid stimulation, hydrochloric acid is used to demineralise coal cleats and widen conductive pathways. Oxidant stimulation uses sodium hypochlorite and widens the cleats by dissolving the coal matrix. Both have been tested on samples of Bowen Basin coals and found to successfully increase permeability (Jing et al. 2020).

Another option is enhanced coal bed methane recovery (ECBM) which has a potential role in carbon sequestration (Godec et al. 2014). Due to its molecular shape, CO₂ has a higher adsorption affinity to coal than methane does (Moore 2012); by pumping CO₂ into the coal seam, methane is displaced and released, while CO₂ is adsorbed onto the coal surface and thus stored at depth (Gandossi 2013). In fact, nanoparticles can be applied to improve this process, as discussed by Li et al. (2015b). Other gases such as nitrogen and flue gases can also be used in ECBM (Seidle 2011), in which case, production is enhanced through the reduction of partial pressure created by the injection of gas, which stimulates diffusion of CSG towards the wellbore (Packham et al. 2012). Modelling studies are underway to better understand the molecular mechanisms responsible for these

observations (e.g. Le et al. 2015; Cole and Striolo 2019; Badmos et al. 2020).

5 Summary and conclusions

This study presented some of the most promising potential applications for nanoparticles in the hydraulic stimulation of coal seams gas as well as some barriers in implementing them:

- (1) Addition of nanoparticles such as MgO to VES fluids can improve their viscosity with the formation of a pseudo-filtercake on the surface of fractures. This prevents fluid loss into the surrounding formation, which is of particular importance in shallow and thin coal seams where fluid leakage could lead to the contamination of aquifers. For deeper coal seams, with permeability values of < 0.1 mD, the application of silica and polyelectrolyte complex (PEC) nanoparticles can be effective in controlling fluid loss.
- (2) Sequenced use of nano-proppants can help create a more extensive fracture network and reduce proppant embedment. Nano-sized proppants such as fly-ash are injected first and deposit in the smallest fractures. ULWPs are injected next and evenly prop open the main conductive channels.
- (3) Coating conventional proppant particles with nanoparticles encourages the fixation of fines, as a result of strong surface attraction. This prevents fines from blocking critical fractures and damaging equipment at the wellbore.
- (4) Meeting regulatory requirements that may become more and more stringent in the coming years, as well as the cost of the nanoparticles themselves may outweigh the benefits of implementing nanotechnology.
- (5) There is very little work focusing specifically on nanotechnology in CSG production. Because of the paucity of field studies where these technologies are tested under realistic reservoir conditions, this report advocates cautious at this time, although potential opportunities have been outlined.

In order to ensure that the nanotechnology options become viable in Australia, this study recommends the following:

- (1) Experimental laboratory study with nanoparticles on a wide range of coal samples, to account for the materials heterogeneity.
- (2) Modelling and experiments conducted at reservoir conditions, towards understanding of how nanoparticles behaves in realistic downhole scenarios.

- (3) Taking forward promising laboratory applications to small scale field experiments.
- (4) Investigations on the potential risks (or lack thereof) of nanoparticles to health and the environment, to identify produced water treatment processes in anticipation of future approval requirements.

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