

Current minireview

Wettability controlling effects on the fluid occurrence and flow in shale gas reservoirs: Present problems and new sights

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Abstract:

The wettability of shale is critical for the development of shale oil and gas reservoirs. Due to its complex composition, which includes organic materials and a number of different inorganic minerals, shale's wettability may show high heterogeneity. This could significantly affect fluid occurrence and flow processes in various kinds of pores. Organic and inorganic pores may have varying capillary pressures. The methodologies to describe the capillary forces in these two varieties of pores are still lacking, though. Additionally, due to the strong capillary pressure that may prevent liquid water from entering organic pores, the mechanisms by which water and methane accumulate in inorganic pores and organic pores may differ. Therefore, the two-phase occurrence mechanisms in the various types of pores in shale continue to be difficult problems. Furthermore, because organic and inorganic pores differ in their capillary pressure and fluid occurrence, wettability can have a significant effect on relative permeability. Thus, wettability is a significant factor that impacts the exploration and development of shale gas reservoirs. The development of shale gas reservoirs could benefit significantly from a thorough understanding of wettability heterogeneity, capillary pressure, water-methane occurrence, and relative permeability.

1. Introduction

Shale gas has become an increasingly important energy resource over the past decades. Shale rocks are generally composed of complex components, such as quartz, dolomite, clay minerals, and organic matter (Zhang et al., 2021; Li et al., 2023). The composition of shale rocks significantly affects the wettability and occurrence of water and methane in the pore structure. The wettability could also affect the two-phase flow and recovery process of methane in shale gas reservoirs. However, the wettability of shale and its effects on fluid storage and flow are still challenging issues due to the complex composition and pore structure of shale.

Wettability is generally described as the ability of a fluid to spread on or contact a solid surface in the presence of other immiscible liquids. It is generally a relative preference of one fluid over another to coat the rock surface, which is commonly characterized by the contact angle at the liquid-solid interface

(Cai, 2021; Pan et al., 2022a). It is more convenient to express the trend of liquid diffusion on a solid surface by measuring the angle of contact at the liquid-solid surface. This angle, which is always measured from the liquid to the solid, is called the contact angle (θ). θ has gained significance as a measure of wettability. As the contact angle decreases, the wet characteristics of the liquid increase. Complete wettability would be evidenced by a contact angle of zero, and complete nonwetting would be evidenced by a contact angle of 180°. There have been various definitions of intermediate wettability. However, in much of the published literature, contact angles of 60° to 90° will tend to repel the liquid. Studies have shown that the wettability of natural rocks is dependent on fluid pressure (Zhang et al., 2018), roughness (AlRatrouf et al., 2018), pore geometry (Rabbani et al., 2018), and minerals (Gosiewska et al., 2002). The features of shale wettability have been extensively researched under both low-pressure and high-pressure conditions. The inorganic minerals and organic

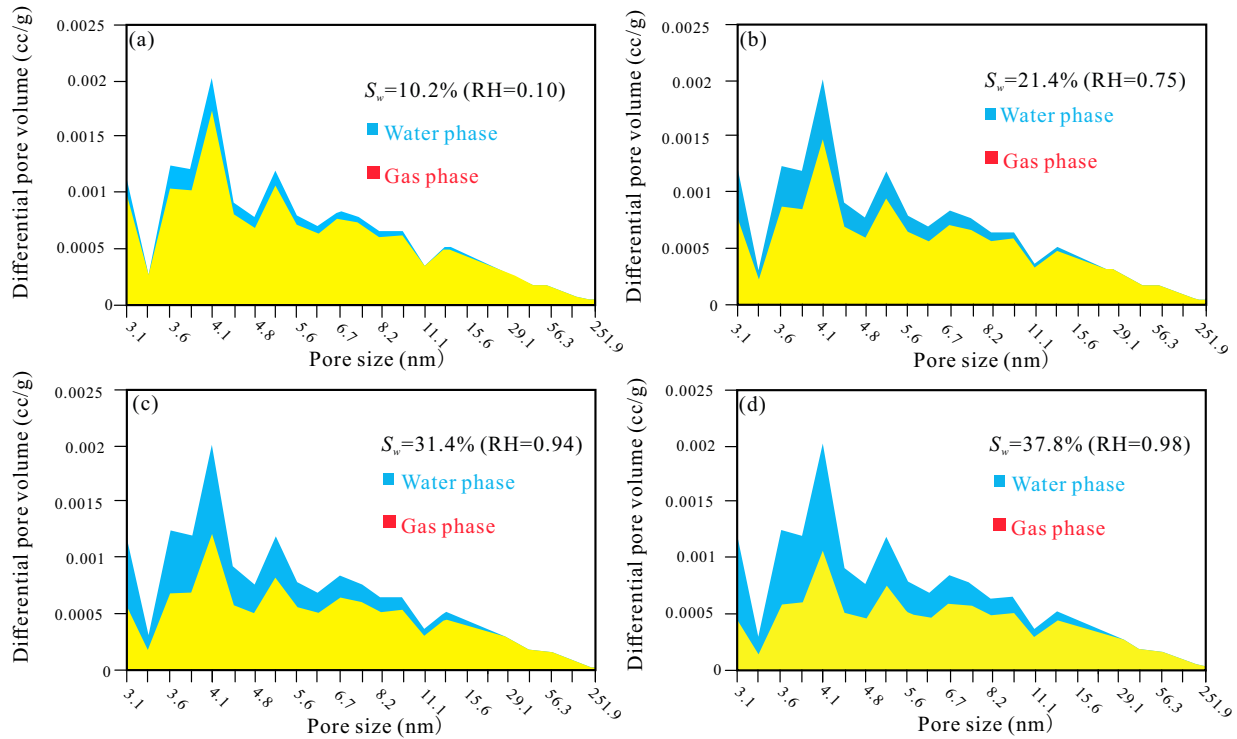


Fig. 1. Gas-water distribution characteristics in shale sample with different total S_w : (a) $S_w = 10.2\%$; (b) $S_w = 21.4\%$; (c) $S_w = 31.4\%$; (d) $S_w = 37.8\%$ (Li et al., 2016).

matter's wettability exhibit high wettability differences (Arif et al., 2021). The clay content, total organic carbon (TOC) content, pressure, and fluid composition all have big impacts on shale's wettability (Pan et al., 2022b). Shale nanopores are capable of retaining water in both the adsorbed and free states, and the ratio of the two is greatly influenced by the pore's diameter (Li et al., 2023). Thus, the gas adsorption in shale reservoirs is a "gas-water-solid" pattern instead of a single "gas-solid" pattern. Wettability could significantly affect water storage in different pores and then affect the water and gas saturations, which also affect the relative permeability.

In this paper, a few critical problems were reviewed, and new sights were proposed. According to former studies, shale has distinct micro- and macro-wettability properties; organic matter tends to be weakly water-wet, while minerals tend to be weak to strong water-wet. Shale typically has different types of pores, such as inorganic, organic, and organic-mineral pores. However, there are still some issues regarding the water-methane two-phase storage and flow mechanisms in different types of pores. This paper illustrates several problems and proposes some new sights.

2. Capillary forces in inorganic pores and organic pores

Capillary forces in shale rocks can have great impacts on the transport of oil and gas. For example, when there are two phases of immiscible liquids in the pore space, the capillary forces will result in a pressure difference between the two sides of the meniscus. The magnitude of the capillary force is

related to the surface tension of the liquid, the critical radius of curvature of the meniscus, and so on (Donnelly et al., 2016). Besides, the wettability and pore types could also affect the capillary force because of their controlling effects on water and gas distribution (Cai et al., 2014).

Unconventional oil and gas reservoirs have complicated microscopic and macroscopic wettability, and despite a variety of scientific findings, there is still a dearth of research on additional wetting phenomena, capillary force effects, and related multiphase flow mechanisms (Li et al., 2020). Considering that the meniscus could be concave or convex, respectively, the capillary pressure may act as a driving or resisting force in a multiphase flow. As the capillary force is a resistance to fluid flow, the pressure in the wetting phase is higher than that in the nonwetting phase. On the contrary, the fluid pressure in the nonwetting phase is higher than that in the wetting phase.

These pores' complicated water occurrence mechanisms are challenging to experimentally characterize in both organic and inorganic pores. It has been found that water capillary condensation occurs in hydrophilic clay mineral pores (Li et al., 2016); however, even under high humidity conditions, the pores could not be filled with water molecules due to the hydrophobic properties of the organic pores, and the water would not completely fill the shale nanopores under low pressure conditions (Sang et al., 2018). It is mainly due to the inaccessibility of water molecules to the micropores of hydrophobic organic matter. Inorganic micropores that are completely blocked by water may not be able to transport or store gases (Li et al., 2016). This study proposed a model to characterize the water storage mechanisms in different-size

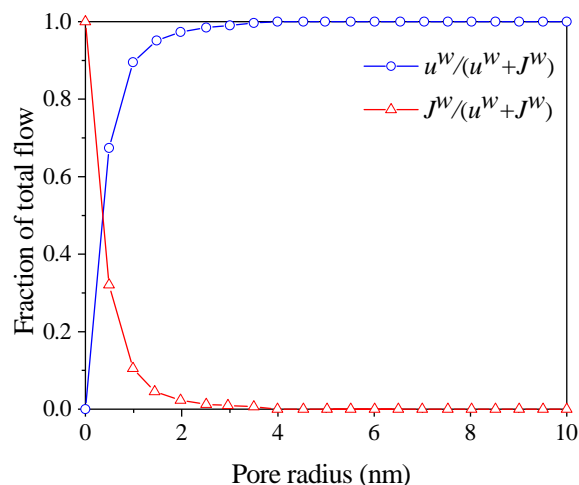


Fig. 2. Fraction of total flow due to capillarity and diffusion (Siddiqui et al., 2021).

inorganic pores in shale under various moist conditions, which can also be used to determine the gas-water distribution properties of shale samples. Even under high relative humidity (RH) settings, such as $RH = 0.98$, shale's micropores cannot be entirely filled by water (Li et al., 2016), as shown in Fig. 1. The capillary forces in organic and inorganic pores may play a significant role in water occurrence, considering the wettability heterogeneity. In addition, real shale rocks are saturated with water and methane, and the hydrophobic organic pore possibly offers an efficient place for gas storage because the capillary could inhibit the water from entering the organic pores.

Using spontaneous uptake tests, the wettability of pore systems in shales may be assessed. The capillary forces are the primary force in the uptake process for shales with nanometer-sized pore spaces. Capillary pressures commonly have a significant impact on the initial phases of water absorption (Sun et al., 2017). Furthermore, it has been demonstrated that capillary flow predominates the fluid pattern and that capillary pressure increases with decreasing pore size. Capillary pressure predominates in the imbibition of water into oil-saturated rocks, while countercurrent imbibition is responsible for the expulsion of oil from the rocks (Alvarez and Schechter, 2016; Cui et al., 2022). However, the pores in shale rocks often have different levels of wetness, especially those that are water-, oil-, and mixed-wettable (Gao et al., 2019). The wettability differences between minerals and organic matter may cause the capillary force in those pores to be completely different.

If oil-wet pores of inorganic minerals are nano-scale, the role of capillaries is relatively limited, especially due to oil molecular size, which causes low oil uptake, whereas water can still diffuse into these nano-scale pore throats. Therefore, the fewer micropores exist in the inorganic minerals of shale, the higher the rate of oil diffusion. These observations are likely to be applicable to fluid uptake in all pores (organic and inorganic) of shale rocks (Siddiqui et al., 2021). Fig. 2 shows that below 2 nm, diffusive flow starts to become significant and becomes dominant at tiny pore sizes (0.5 nm), while above 2 nm, laminar Poiseuille's flow (or Darcy's flow) prevails.

However, the capillary forces in organic and inorganic

pores are fundamentally different, and they might be revealed if the capillary forces in both are examined individually. The influences of variables like the kind of pore wettability and liquid water buildup in various pores have been frequently ignored in earlier research on capillary forces. Although some research has been done on the dynamic role of capillary forces, it is not clear from these studies how capillary forces are affected by pore wettability in shale and how they affect fluid transport. It is poorly understood how capillary forces affect fluid flow in organic and inorganic pores. In fact, it is advantageous to research capillary pressure in various pore types and comprehend the implications of capillary forces in shale reservoirs.

3. Effect of wettability heterogeneity on water storage and methane adsorption

Pore wettability may have an impact on the fluid distribution in both organic and inorganic pores. In experiments involving spontaneous experiments, inorganic pores mostly retained the liquid water. The capillary force prevents water from entering organic pores at low pressure (Zhang et al., 2020). As there is significant pressure applied, the water might enter organic pores. Water can be stored in both organic pores and the kerogen matrix. Organic pores and inorganic pores have varying water saturation levels.

Gas can be stored in pores in shale gas reservoirs as a free phase or as an adsorbed phase on the kerogen surface and mineral surface (Li et al., 2022a). The gas adsorption on the surfaces of minerals and organic pores may be impacted by the water saturation in inorganic and organic pores. The characteristics of the surface adsorption potential energy distribution and the size of a particular surface area are the most crucial adsorption properties. Adsorbent molecules always favor the "active point" with the deepest surface for adsorption. The kind and distribution of surface functional groups are two elements that affect an adsorbent's surface potential energy. The presence of numerous different oxygen-containing functional groups in shale organic matter, particularly at low maturity, is often crucial to shale wettability (Hu et al., 2013; Zou et al., 2020). Methane molecules are nonpolar, whereas water molecules are polar. Organic matter and mineral surfaces may exhibit complicated H_2O/CH_4 adsorption characteristics. The maturity also affects the wettability of kerogen, and the higher the maturity, the stronger the oil wettability index (Begum et al., 2019).

Water is an important component of shale reservoir reservoirs, and it has a negative impact on the ratio of adsorbed/free gas and methane seepage and diffusion (Wang et al., 2018b). Shale reservoirs naturally have a certain water content, which could interact with the pore surface and is difficult to remove at low temperatures. Research has shown a positive correlation between wettability and moisture content, meaning that the stronger the water wettability, the higher the moisture content (Liu et al., 2012). The interaction between carbonaceous substances (such as coal and organic shale) and water molecules is much more complex than that with non-polar molecules (such as N_2 , CH_4 , and CO_2). This complexity stems from the weak

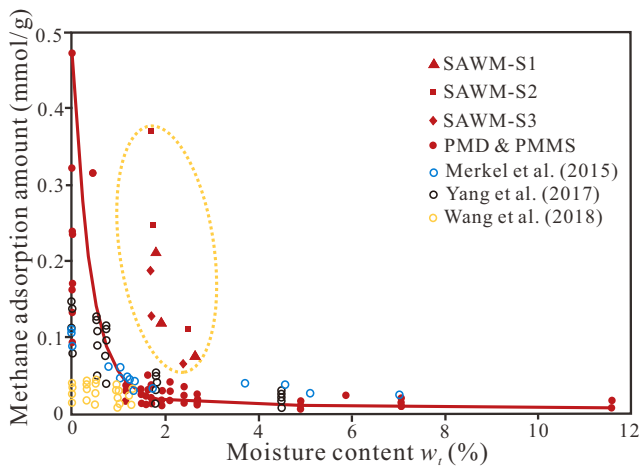


Fig. 3. Relationships between the moisture content and methane adsorption amount determined by different authors (Zhang and Yu, 2022).

dispersion between water and carbonaceous substances, while water has a tendency to form hydrogen bonds or undergo chemical interactions with other adsorbed water molecules and surface functional groups (Busch and Gensterblum, 2011). After studying the wettability and adsorption of marine shale in the Longmaxi Formation, it was found that water dramatically reduces methane adsorption capacity (Liang et al., 2016). A number of studies have investigated the adsorption process of water vapor and methane on the surface of coal and concluded that the maximum adsorption capacity of water-wet coal is significantly lower than that of dry coal, and the degree of reduction in adsorption depends on the coal rank (Nishino, 2001; Day et al., 2008). The methane adsorption capacity decreases with increasing moisture content until reaching the limit of moisture content (Day et al., 2008). Some results also indicate that clay minerals may adsorb water into interlayer pores, making it impossible for water to hinder methane molecules from reaching effective adsorption sites in clay-rich shale (Wang et al., 2018a). In addition, it was found that methane diffusion depends on the adsorption state of water, and adsorbed water forms thin layers or water clusters in pores, thereby affecting the surface pore size and methane migration (Hu et al., 2018).

These previous studies have shown that the total moisture content of shale has a negative effect on methane adsorption. They mainly investigated factors, such as material composition, porosity, temperature, pressure, or moisture on adsorption (Ma et al., 2020; Babatunde et al., 2022). Experimental results indicated that the moist content can dramatically decrease the adsorption amount of methane (Merkel et al., 2015; Yang et al., 2017; Wang et al., 2018a; Zhang and Yu, 2022), as shown in Fig. 3. However, very little research illustrates the impact of differences in liquid water saturation in different pores on shale gas adsorption.

In shale's nanopores, the capillary pressure is high, and it is a resistant force that prevents liquid water from entering inorganic pores at atmospheric pressure. However, the capillary pressure inhibits water from imbibing into organic pores, as shown in Fig. 4 (Sang et al., 2018). Thus, the

liquid water saturation in organic pores and inorganic pores could be significantly different and potentially cause different effects on gas adsorption on minerals and organic pores' surfaces. These mechanisms are still unknown due to the challenges of unknown liquid water storage mechanisms in inorganic and organic pores. A comprehensive understanding of the wettability-controlled water occurrence in organic and inorganic pores could promote the recognition of water-gas two-phase storage in shale gas reservoirs.

4. Effect of wettability on two-phase flow relative permeability

Relative permeability is the ratio of effective permeability to the absolute permeability of a rock. The relative permeability is an important index to predict the production index and dynamic change of water flooding development in an oil field, which reflects the change in the seepage capacity of the two-phase fluid in pores. The oil-wet and water-wet pores could serve as flow channels for different fluids; thus, the wettability could also influence the relative permeability.

For coal seams, absolute permeability reflects the ability of the coal itself to allow single-phase flow, which is significantly affected by the pore structure. The relative permeability of coal usually describes the competitive flow of gas-liquid and is further affected by wettability and interfacial tension (Li et al., 2022b; Qin et al., 2023). Although some progress has been made in pore structure characterization and fluid flow modeling in recent years (Wang et al., 2021, 2022). However, in the process of gas-liquid flow in coal reservoirs, the understanding of wettability and pore networks is still relatively rare.

It is widely recognized that the viscous force and the capillary force control the mobilization of the residual phase in reservoirs. The gas-water relative permeability in shale is influenced by the TOC volume and water contact angle in inorganic pores. With the increase in TOC volume, the pores tend to be oil-wet, and the water saturation corresponding to the crossing point of relative permeability decreases, as shown in Fig. 5 (Song et al., 2019). It also shows that as the TOC goes higher, the water relative permeability goes lower; however, the gas relative permeability goes higher, indicating that a higher TOC could favor the production of shale gas and inhibit water production.

The water saturation corresponding to the curves of the water-gas relative permeability is one of the indicators for evaluating water flooding. Under the hydrophilic condition, the water relative permeability increases slowly and the gas relative permeability decreases linearly with increasing water saturation; although the crossover saturation is higher, complex pore networks trap part of the gas, resulting in higher residual gas saturation. Under the hydrophobic condition, the water-gas relative permeability varies significantly, and the crossover saturation is lower, but the residual gas is less due to the hydrophobic surface (Qin et al., 2023). Research results have shown that the rock wettability heterogeneity could significantly affect the relative permeability curves (Zhao et al., 2018); however, the relative permeability in organic and in-

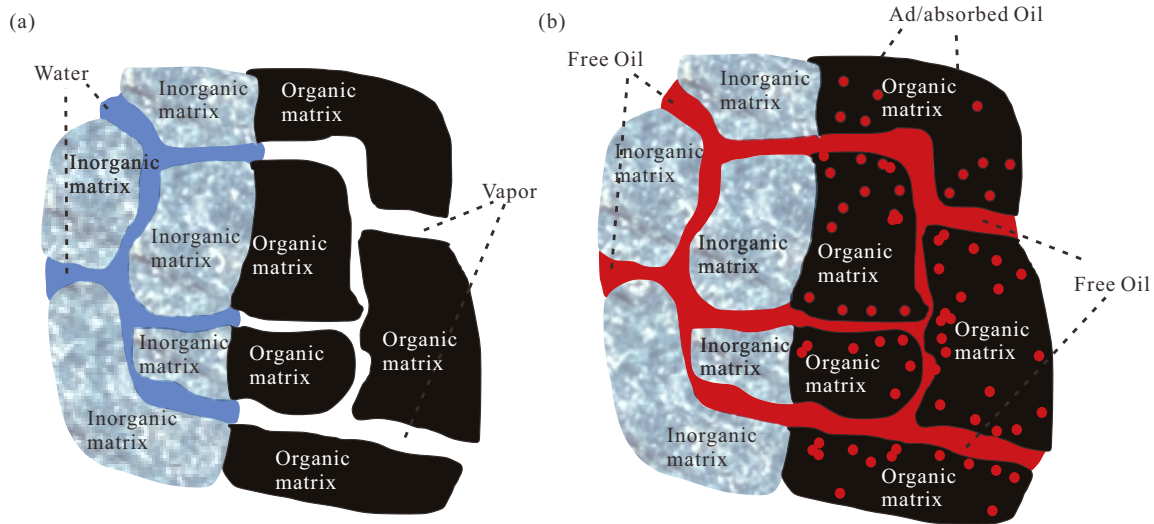


Fig. 4. Schematic of water and oil imbibition in organic and inorganic pores: (a) water only imbibes into inorganic pores; (b) oil imbibes into both organic and inorganic pores (Sang et al., 2018).

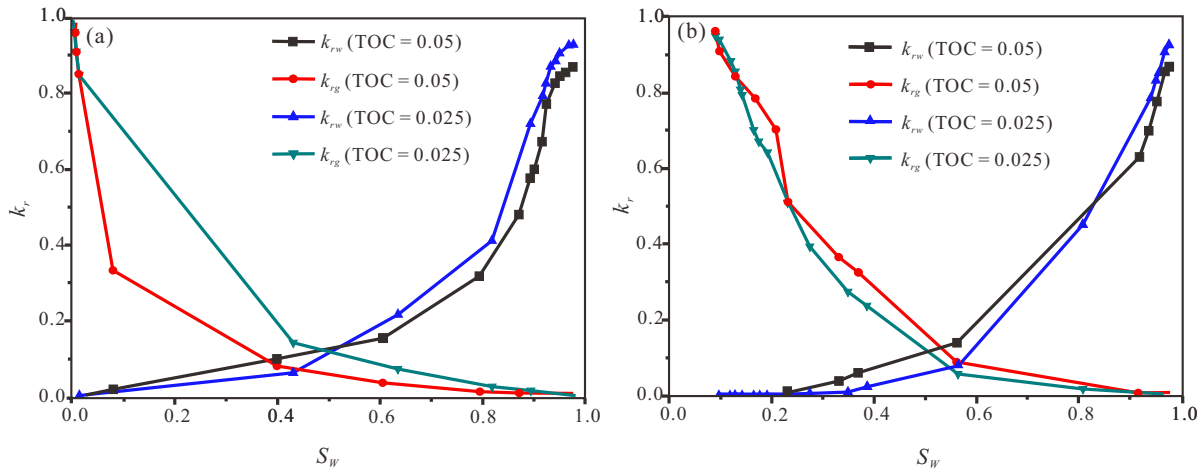


Fig. 5. Gas and water relative permeability during (a) injected water flow in and (b) injected water flow back process at different TOC in volumes (Song et al., 2019).

organic pores is different, and very little research illustrates the relative permeability differences in these two types of pores. The two-phase flow's relative permeability could dominate the gas and water flow rates in those pores. A comprehensive understanding of the relative permeability of two types of pores could benefit the evaluation and development of shale gas reservoirs.

5. Conclusion

The implications of the wettability heterogeneity of shale rocks on the occurrence and movement of water-methane were discussed in this research. Shale has considerable wettability heterogeneity due to its diverse composition, which includes different contents of inorganic minerals and organic materials. Due to the strong hydrophobic properties of the surface of organic matter, there are significant disparities between the wettability of inorganic and organic pores. The capillary pressure, which can act as a driving and resisting force for the two-

phase flow in shale pores, may be strongly impacted by the micro-wettability of the shale. It is still unclear how capillary forces work in biological and inorganic pores. As a result, wettability and capillary pressure would have a major impact on the saturation of gas and water in pores with different wettability levels. The amount of water in both organic and inorganic pores could have a big impact on how much methane can be stored, both as free gas and as adsorbed gas. The regulating effects of wettability on relative permeability remain a difficult problem. It is important to take into account the relative permeability variations between organic and inorganic pores.

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Conflict of interest

The authors declare no competing interest.

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