A review: hydrophobicity and surface charge of coals and coal-associated minerals

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Abstract. The hydrophobicity and surface charge of coals and coal-associated minerals play important roles in froth flotation and dewatering in coal preparation plants. These properties are also important for respirable dust control and the application of surfactants. This paper discusses techniques for the measurement of hydrophobicity and surface charge and then uncovers literature values for contact angles and surface charge for coals of varying rank, including some maceral analyses. It also provides a review of literature data for contact angle and surface charge for the major minerals in coal. Readers can use these data for understanding mineral removal or recovery using various preparation techniques, including fine desliming, froth flotation, selective agglomeration, and other processes that are influenced by hydrophobicity and surface charge as we continue to develop the need for ultra clean coals.

1 Introduction

The interaction of coal and coal-associated minerals can be assessed using zeta potential and contact angle measurements to provide an understanding of surface charge and hydrophobicity, respectively. These properties affect the behaviour of froth flotation and thickening/dewatering processes in coal preparation plants. In addition, the application of dust suppressants, of growing concern with the incidence of “black lung disease” or pneumoconiosis increasing in many areas, also relies on these properties [1]. Having information about these properties for coal and coal-associated minerals can assist both researchers and plant engineers and operators in their understanding and selection of dust suppressants, flotation reagents, flocculants, and other dewatering aids.

The authors reviewed the literature to obtain data regarding the zeta potential and contact angle for coals as well as for several primary coal-associated minerals. The zeta potential, in millivolts (mV), measures the “apparent” surface charge [2] and can be positive or negative or zero. The zeta potential is affected by pH as well as the presence of other ions in solution as shown by Arnold and Aplan for coals and clay minerals [3]. When the zeta potential is 0, also known as the isoelectric point or point of zero charge, the repulsions are weak, and particles tend to aggregate and settle. Similarly charged particles will repel each other while oppositely charged particles can be attracted to each other [2]. Since different

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minerals behave differently at different pH values or in the presence of other ions, a slurry with coal and minerals can be dispersed or flocculated depending on slurry conditions. This paper provides zeta potential data for coals as well as for the minerals highlighted in Table 1.

Table 1. Minerals in coal [after 4].

Group Type	Mineral
Shale	**Illite, Montmorillonite,** Bravaisite, Hydromuscovite, Muscovite
Kaolin	**Kaolinite**, Levisite, Metahalloysite
Sulphide	**Pyrite,** Marcasite
Carbonate	**Calcite,** Siderite, **Dolomite,** Ankerite
Chloride	Sylvite, Halite
Accessory Minerals	**Quartz, Gypsum,** Chlorite, Rutile, Hematite, Magnetite, Sphalerite, Feldspar, Garnet, Hornblende, Apatite, Zircon, Epidote, Biotite, Augite, Prochlorite, Diaspore, Lepidocrocite, Barite, Kyanite, Staurolite, Topaz, Tourmaline, Pyrophyllite, Penninite

Contact angle measurements are used to assess hydrophobicity, or the "water-fearing" nature of a particle. Measured in degrees (°), a mineral with a large contact angle will be more hydrophobic, which means it is repelled by water, while a small contact angle indicates a hydrophilic mineral. This becomes a key property for froth flotation during particle/bubble interactions, for the addition of dust suppressants for during particle/water droplet interactions, and in dewatering to allow the shedding of water from surfaces through applied dewatering forces [5]. When contact angles are high and zeta potentials are at the isoelectric point, dewatering is most efficient [6]. If the surface charge and hydrophobicity is understood, it is easier to predict how particles will react to a water-based spray system being used to control dust [7].

Zeta potential and contact angle are both used to understand how coal and coal-associated minerals interact. With real world applications in froth flotation, dewatering, and respirable dust suppression, this paper consolidates zeta potential and contact angle information to create easier access to these data.

2 Contact angle and zeta potential data

Kaolinite, illite, and montmorillonite are common clay minerals found in coals. Contact angle values for kaolinite are given in Table 2 with zeta potential values found in Figure 1. The literature results for illite are found in Table 3 and Figure 2, while those for montmorillonite are given in Table 4 and Figure 3. As shown, the contact angle values are generally low, except for some values for montmorillonite that would suggest that these minerals would not

report to the froth flotation concentrate unless carried over by water in the froth phase [8]. The zeta potential values are generally negative until very low pH values.

Table 2. Literature summary of contact angles found for kaolinite [9-11]

Contact Angle (°)	Source	Other Information
Around 20	[9]	
19.56	[10]	
16.9 ± 0.7	[11]	33% Relative Humidity

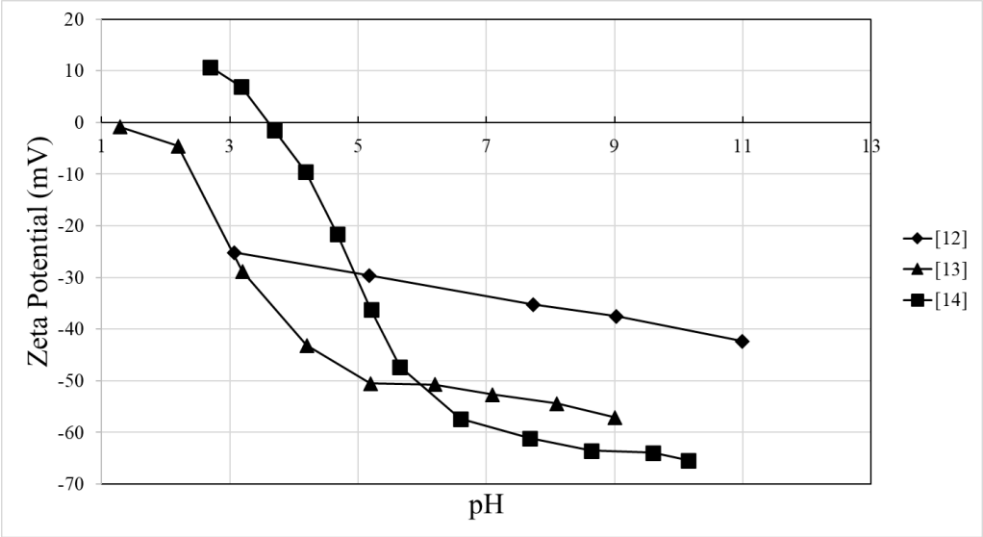

Fig. 1. Graphical representation of the variation of the zeta potential with pH for kaolinite [12-14].

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Table 3. Contact angle values for illite, as reported in the literature [11, 15, 16].

Contact Angle (°)	Source	Other Information
34.2	[11]	33% Relative Humidity
34 ± 3	[15]	
Around 30	[16]	Water/oil

Understanding the zeta potential of these clay minerals is extremely important in understanding the potential for slime coatings in coal flotation. It should be noted that water quality, the presence of various cations and anions along with pH, has a major role in determining the ability of the clays to attach to coal or other mineral surfaces. In some instances, the clays will stay dispersed and not cause coal depression, while in other cases, clay slimes will attach to the coal surface, depressing flotation recovery [3].

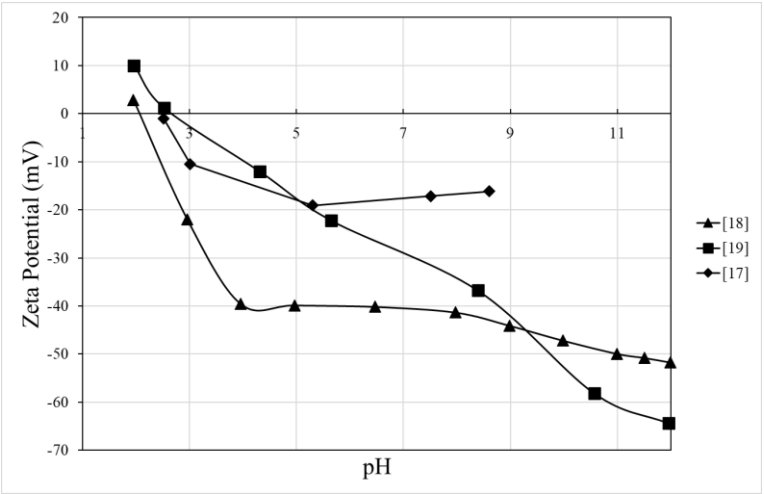

Fig. 2. Graphical representation of the variation of zeta potential with pH for illite [17-19].

Table 4. Montmorillonite contact angles in the literature [20-22].

Contact Angle (°)	Source	Other Information
52.8	[20]	Raw Mt
52± 0.5	[21]	-Surfactant: cetyltrimethylammonium ammonium bromide, -Organic Mt (1.5), 1.5 meaning the surfactant amount is 1.5 times the cation exchange capacity of the Mt, which was 106.4 mmol/100 g -Preparation: wet
35.8°	[22]	Not modified, Pure hydrophilic

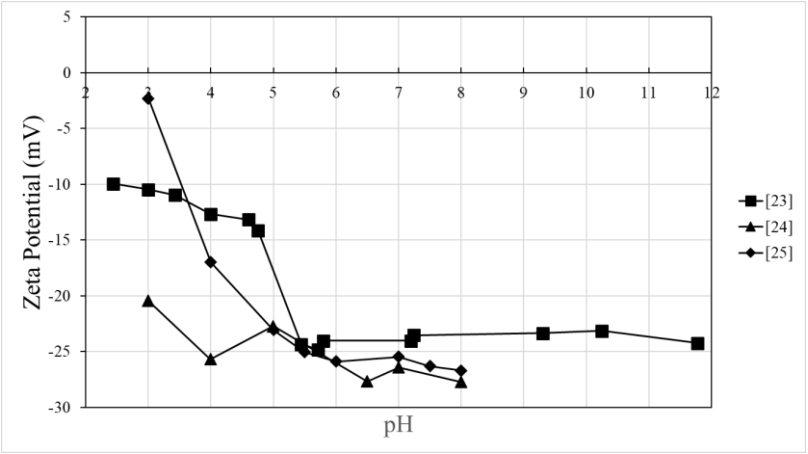

Fig. 3. Graphical representation of the variation of zeta potential with pH for montmorillonite [23-25].

Table 5 gives the literature values for the contact angle of quartz are low, indicating a lack of natural hydrophobicity. Figure 4 gives the zeta potential value of quartz found in several literature sources. As shown, the zeta potential of quartz is negative across the wide range of pH values with an isoelectric point at pH 2. Quartz is one of the main culprits in causing pneumoconiosis and silicosis, with surface properties being of great interest as researchers develop methods of reducing its toxicity [26].

Table 5. Quartz contact angles from the literature [27-31].

Contact Angle (°)	Source	Other Information
27.8–50.3	[27]	Reservoir Rock
35	[28]	
43 ∓ 2	[29]	
26.8	[30]	
26.15	[31]	

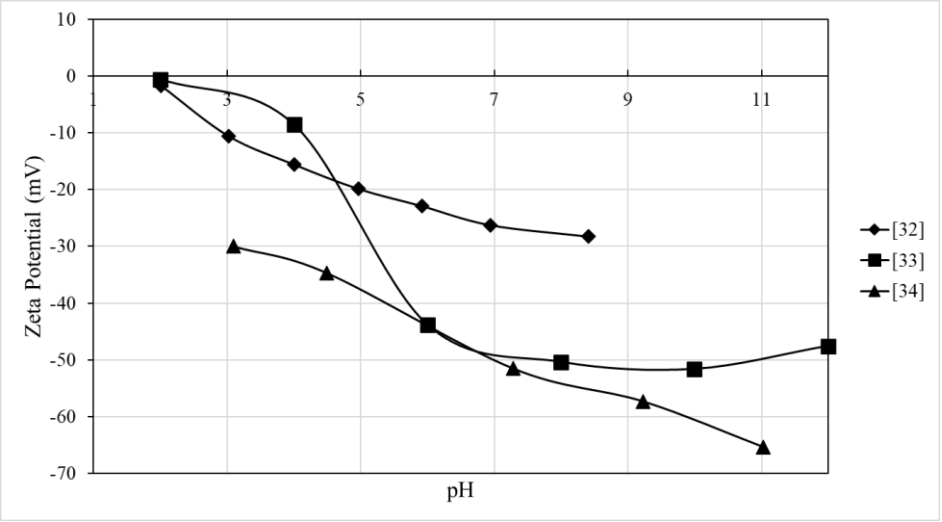

Fig. 4. Graphical representation of the variation of zeta potential with pH for quartz [32-34].

Dolomite contact angles, some of the lowest reported for the coal-associated minerals, are shown in Table 6. The literature values for the zeta potential of dolomite are not consistent (Figure 5), suggesting variations in dolomite composition or errors in the measurements.

Table 6. Dolomite contact angles from the literature [35-37].

Contact Angle (°)	Source
6.6	[35]
Around 15	[36]
23	[37]

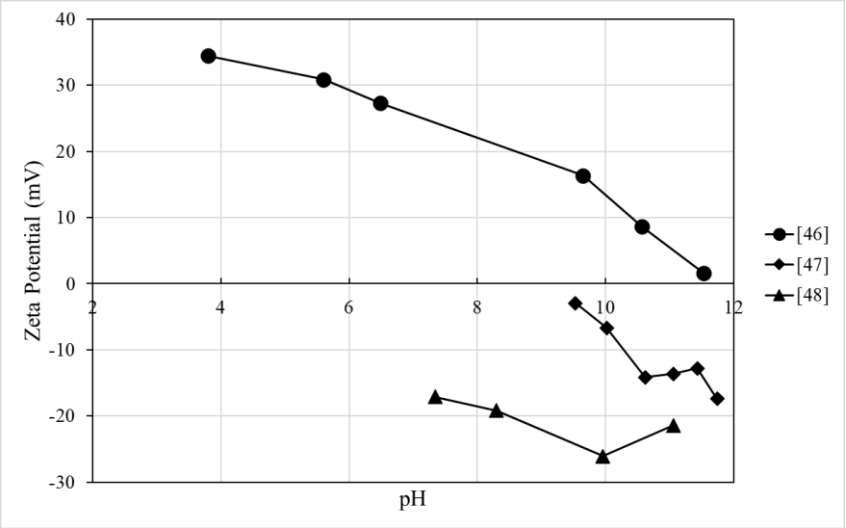

Fig. 5. Graphical representation of the variation of the zeta potential with pH for dolomite [38-40].

Only one source for the gypsum contact angle was found as shown in Table 7. This is also a very low value. The gypsum zeta potential values are relatively inconsistent in the literature are shown in Figure 6. The zeta potentials are very low, ranging from 15 to -15 mV.

Table 7. Gypsum contact angles from the literature [28].

Contact Angle (°)	Source
17	[26]

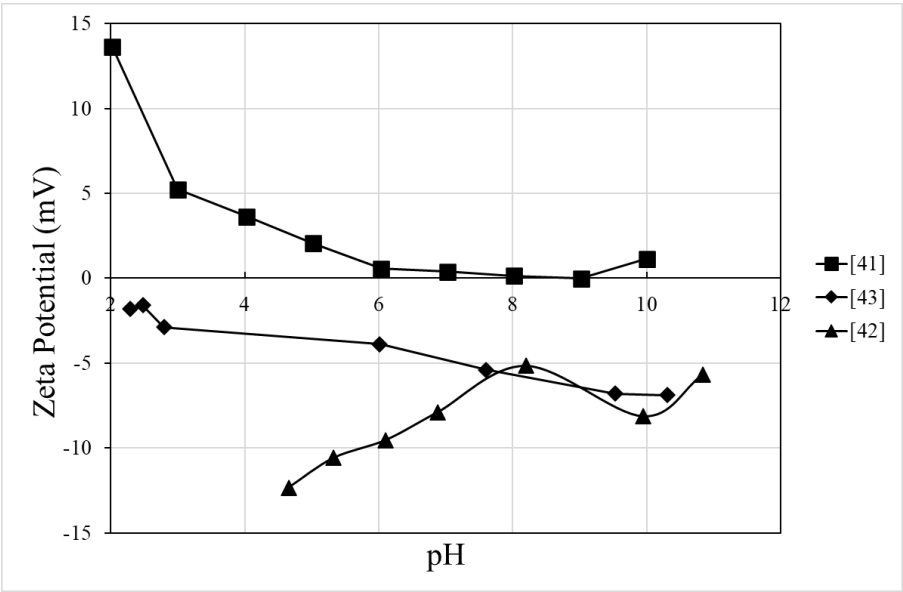

Fig. 6. Graphical representation of the variation of zeta potential with pH for gypsum [41-43].

Contact angle values for pyrite are given in Table 8, indicating that this mineral could be considered hydrophobic with these higher values, approaching those found for some coals. Indeed, the natural hydrophobicity of pyrite has been an issue in coal flotation that has been noted for many decades [44], with researchers studying a variety of pyrite depressants. The zeta potential curves in Figure 7 indicate isoelectric points between pH 3 and 4.

Table 8. Pyrite contact angles from the literature [45-47].

Contact Angle (°)	Source
62.5	[45]
79.47	[46]
76.4	[47]

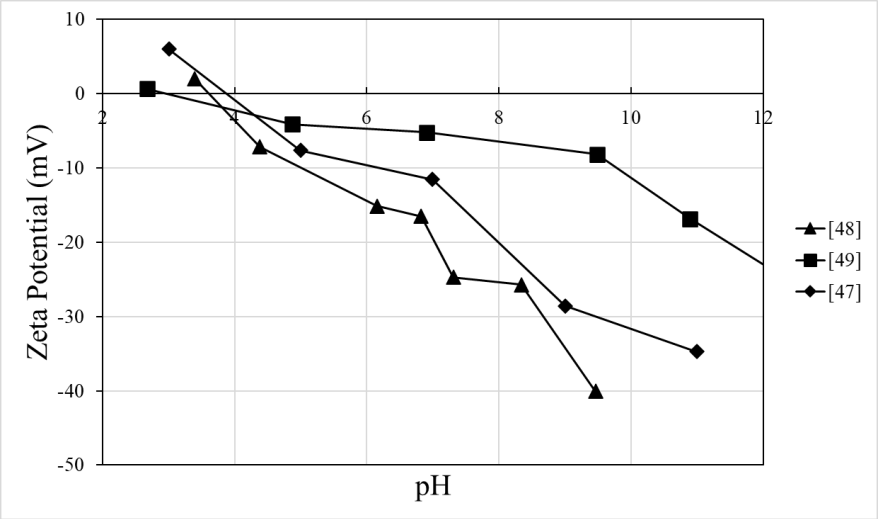

Fig. 7. Graphical representation of the variation of zeta potential with pH for pyrite [47-49].

Calcite is the last mineral reviewed with somewhat moderate contact angle values shown in Table 9. Figure 8 shows very inconsistent zeta potential curves in the literature for this mineral, questioning, for example, the measurement techniques and the nature of the calcite samples (could be variability in the composition).

Table 9. Calcite contact angles from the literature [50-53].

Contact Angle (°)	Source
55.6	[50]
42.1	[51]
40.3	[52]
~38	[53]

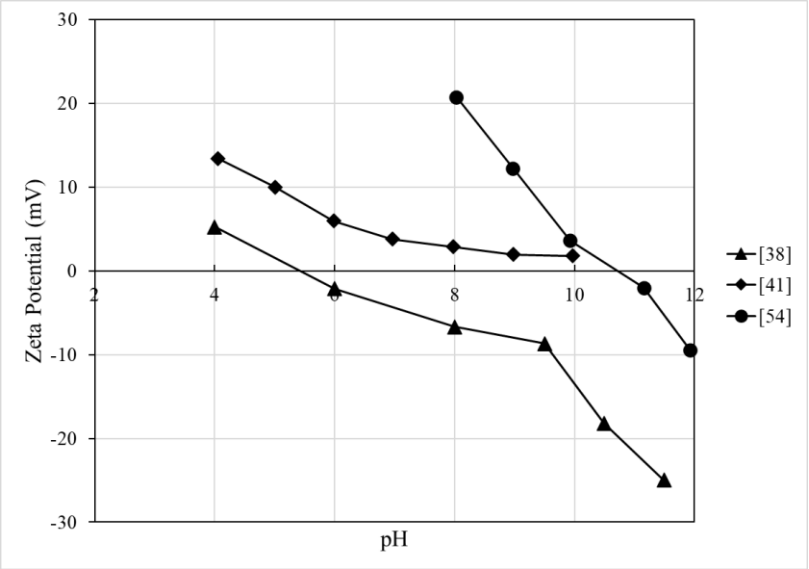

Fig. 8. Graphical representation of the variation of zeta potential with pH for calcite [38, 41, 54].

Values for the contact angle of coal are considerably higher than the coal-associated minerals described previously. Coal, especially higher rank coals, are quite hydrophobic and naturally floatable in terms related to froth flotation. Murata [55] provides the contact angle values for a variety of coals as is shown in Table 13. Table 14 further provides contact angle measurements for coals based on their rank as well as for macerals within these coals. Two measures for contact angle are also given. Captive bubble measurements are performed by placing an air bubble on the surface of a polished submerged specimen. Sessile drop measurements are conducted by placing a droplet of water on the surface of a polished specimen. The method for the previously presented contact angles was not identified so all values should be considered relative.

Table 10. Coal contact angles from the literature [55].

Coal	Contact Angle (°)	Coal	Contact Angle (°)
Taiheiyo upper seam lump	87.3	Miike 66 slack	86.6
Taiheiyo 62 slack	90.0	Takashima main seam lump	75.6
Ashibetsu No. 8 seam lump	87.4	Indonesia Kalimantan brown coal lump	113.6
Ashibetsu 70 slack	83.1	Australia Yallourn brown coal lump	89.1
Akabira No. 9 seam lump	74.9	Alaska Beluga brown coal lump	92.7
Horonai lump	85.7	Indonesia Bangko brown coal lump	97.9

As shown in Table 11, the higher rank medium volatile coals (US rank nomenclature) have higher contact angles. This relationship to coal rank can also be noted as higher contact angles in relation to higher vitrinite reflectance (% VRO mean max) values. The inertinite maceral fusinite also is notably less hydrophobic than associated vitrinite in these coals. Maceral content is often overlooked when considering coal flotation, dewatering, and dust suppression but should certainly be evaluated when processes do not behave as expected.

The zeta potential of several coals is found in Mori et al. [57]. As shown in Figure 9, most coals are negatively charged above ~pH 5.5 with some coals never exhibiting a positive charge even at very low pH values. Some of this behaviour can be explained by considering coal rank, as shown in Figure 10. The Lower Kittanning seam coal is the highest rank coal with the highest isoelectric point. The Upper Freeport seam coal is of slightly lower rank with the Pittsburgh seam coal being of the lowest rank in this group of coals from Pennsylvania, though it is still a high volatile A bituminous coal. The decrease in surface charge should also be noted in this figure when the coals are in tap water (presence of ions, notably Mg^{2+} and Ca^{2+}) as compared to distilled water. These ions tend to reduce the surface charge and promote agglomeration if the zeta potential values are reduced sufficiently.

Table 11. Coal contact angles from the literature [57].

Rank	Maceral	Contact Angle (°) for Captive Bubble Method	Contact Angle (°) for Sessile Drop Method	% C (dmmf)	% V_{RO} (mean max)
Medium Volatile	Vitrinite	61	63	90.4	1.44
	'Pseudovitrinite'	67	66		
High Volatile A	Vitrinite	62	69	85.5	0.94
	'Pseudovitrinite'	66	67		
High Volatile A	Vitrinite	53	62	84.3	0.84
	'Pseudovitrinite'	60	67		
Medium Volatile	Vitrinite	61	61	87.8	1.18
	Fusinite		58		
Medium Volatile	Vitrinite	60	66	87.7	1.11
	Fusinite	No contact	Soaks into sample		
High Volatile A	Vitrinite	60	62	85.5	0.94
	Fusinite	No contact	40		
High Volatile B	Vitrinite	56	57	82.7	0.62
	Fusinite	No contact	41		
Subbituminous A	Vitrinite	No contact	35	76	0.50
	Fusinite	No contact	25		

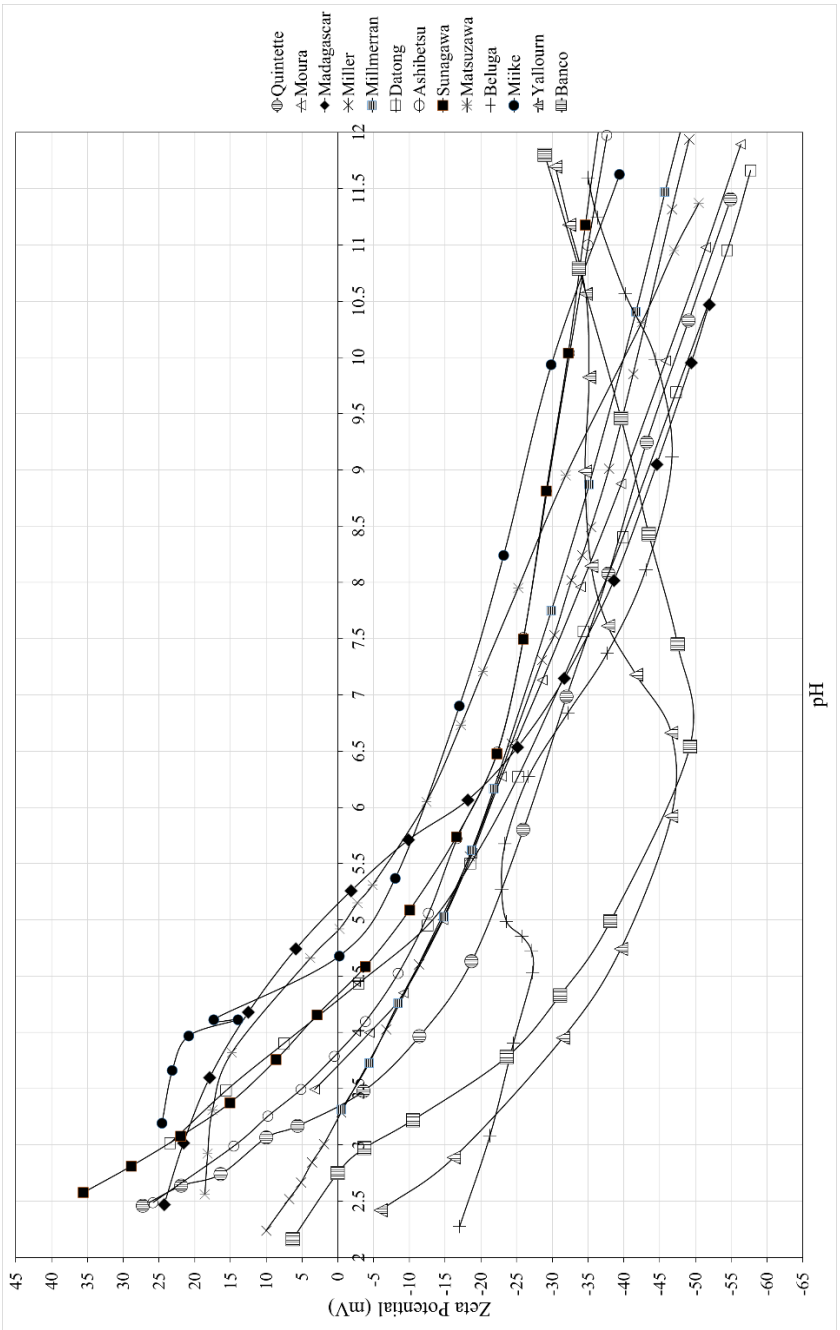

Fig. 9. Graphical representation of the variation of zeta potential with pH for different coal samples [57].

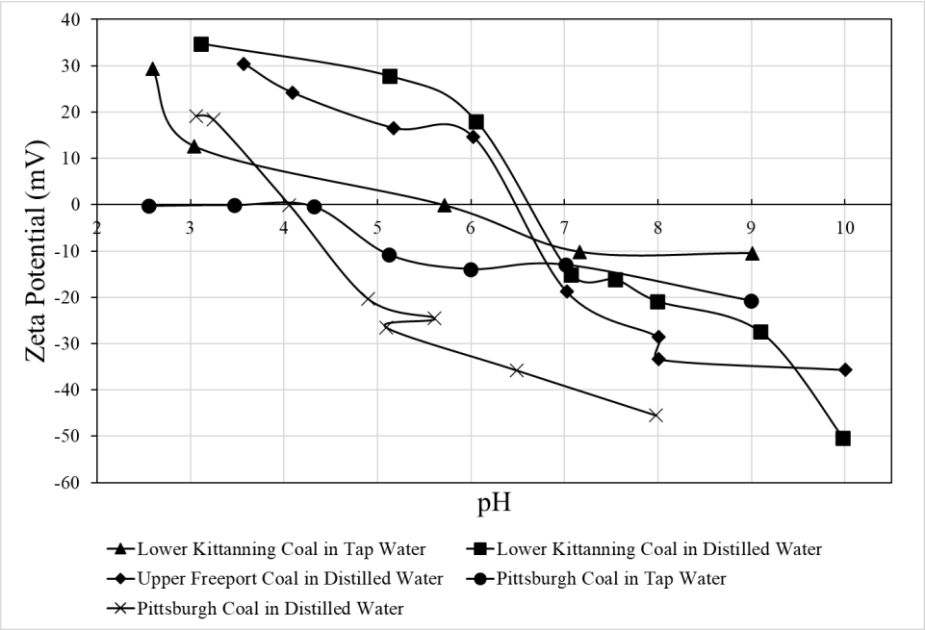

Fig. 10. Graphical representation of the variation of the zeta potential with pH for different Pennsylvania (USA) coal seams [3].

3 Discussion and summary

As a source of hydrophobicity and surface charge information, this paper serves many purposes. These data can inform engineers and operators about the effect of pH on surface charge that can lead to the selection of reagents for improving froth flotation, thickening, dewatering, and dust suppression as consideration should be given to both the properties of the coal as well as those of the minerals present. For example, flocculants often have different charges to assist in the flocculation process in thickeners. With clays being negatively charged under most conditions and coals sometimes being positively charged, the selection of a cationic (positively charged) flocculant may benefit the flocculation of the clays, while the addition of an anionic (negatively charged) flocculant may benefit the flocculation of misplaced coal particles in the thickener.

In dust suppression, it is important to select suppressants that will wet the coal and minerals. For higher rank coals, coal hydrophobicity may play a role in determining the best suppressant. However, if a significant amount of quartz is present in the dust, other suppressants may be utilized as quartz has limited hydrophobicity. If pyrite, a somewhat hydrophobic mineral is present, still other dust suppressants might be considered. The interaction with surface charge and pH should also be considered in these cases—complicated systems that have a direct bearing on the health of workers. Recent research has explored the use of reagents and dust suppressants to reduce the toxicity of these particles based on their surface chemistry [71].

4 Conclusions

As with all literature data, it is important to fully understand the conditions used in generating the data.

- For both contact angle and zeta potential measurements, were fresh surfaces always used? Oxidation of particles will disrupt the measured values and will certainly affect plant results.
- Was distilled or deionized water used in the experiments? Always consider water quality—the presence of various ions—and pH in any scenario. Run any plant related test work in the water that will be used in the plant. The use of distilled or deionized water will give good results in fundamental studies but not in real plant situations.
- Were the technicians skilled in operating the testing devices?
- Were coal maceral components recognized or quantified in these measurements? Macerals have varying properties as well.
- What impurities are found in the minerals tested? What affect will these impurities have on the results?

Therefore, it is important to use the data presented in relative terms or in a way that can guide the engineer, operator, or researcher to understand their own coal and coal-associated minerals in more detail. Do not overlook coal hydrophobicity and zeta potential, and that of the associated minerals, as well as oxidation, maceral content, and mineral phases in coal preparation plant design and operation.

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