



# Surface Electrical Behaviors of Apatite, Dolomite, Quartz, and Phosphate Ore

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The phosphate ore flotation is achieved through the surface electrical behavior difference among apatite, dolomite and quartz, but it is usually affected by the dissolved ions from pulp, especially  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ . The zeta potentials of apatite, dolomite, quartz, and phosphate ore in different solutions were measured to discuss the surface electrical behaviors of them in this study. It was found that the apatite, dolomite and phosphate ore supernatants have limited impact on the zeta potentials of the respective surfaces themselves, but the surface electrical behaviors of apatite and dolomite in the supernatant of the other is similar to the supernatant mineral source. This will increase the separation difficulty of apatite from dolomite in the phosphate ore flotation process. The negative zeta potentials of apatite, dolomite, quartz and phosphate ore increase in the presence of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ . Moreover, a more zeta potential increase in presence of  $\text{Ca}^{2+}$  than  $\text{Mg}^{2+}$  was investigated, suggesting a stronger influence of  $\text{Ca}^{2+}$  than  $\text{Mg}^{2+}$  on the surface electrical behaviors of ores.

**Keywords:** apatite, dolomite, quartz, phosphate ore, zeta potential

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## INTRODUCTION

Phosphorus is an important chemical raw material, and is also a primary constituent of plants. The industrial phosphorus is extracted from the phosphate ore to produce elemental phosphorus, phosphoric acid, phosphatic fertilizer and phosphate (Santana et al., 2011; Xiao et al., 2013). Phosphate ore is a significant mineral source of phosphorus, which is general floated to recover the phosphorus in the apatite minerals ( $\text{Ca}_5(\text{PO}_4\text{CO}_3)_3(\text{F, OH, Cl})$ ) (Zhou et al., 2017). However, most of the phosphate ore industry is currently focused on recovery of apatite from low-grade ores due to the decrease of high-grade phosphate ore. Flotation as the most important method is applied in phosphate ore processing to separate the valuable mineral of apatite from the gangue minerals, such as dolomite and quartz (Zhou et al., 2015). Mineral type and electrolyte environment will result in different surface electrical behaviors of different minerals, and even the same mineral (Alroudhan et al., 2016). Therefore, the investigation of those surface electrical behaviors, which influences interfacial process such as flotation, has obvious research significance. Apatite and dolomite belong to the group of salt-type minerals. It has thus been assumed that they have similar surface electrical behaviors due to the dissolved metal ions (Mishra, 1978; Hu and Wang, 1992). Metal ions in the bulk solution which can be adsorbed on the mineral surface by physical and/or chemical interaction exist in various forms, such as bonding to mineral lattice, generating metal

hydroxides, or carbonate hydroxides precipitated on mineral surface, producing new active center of beneficiation reagent, transferring mineral surface to improve or weaken the role of mineral processing reagents (Demir et al., 2003).

Dolomite can produce  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ions into the pulp during the ore grinding and pulp-mixing processes (Amankonah et al., 1985; dos Santos et al., 2010; Li et al., 2017). The valuable mineral, apatite, in phosphate ore has the same lattice ion,  $\text{Ca}^{2+}$ , as dolomite, which results in a significant effect on the selective flotation of apatite from dolomite. In the real phosphate ore flotation process, different ions in the pulp change the surface electrical behavior of minerals and hence change the selective adsorption of beneficiation reagent on the mineral surface, which enlarges the difficulties in the selective separation of apatite from dolomite and quartz (Amankonah et al., 1985).

The surface electrical behaviors of apatite, dolomite, and quartz in mineral supernatants were discussed in this study. The surface electrical behaviors of apatite, dolomite and quartz in solutions with different  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  concentrations have also been studied to further confirm the effects of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ . The comparison of surface electrical behaviors for phosphate ore in phosphate ore supernatants and in solutions containing

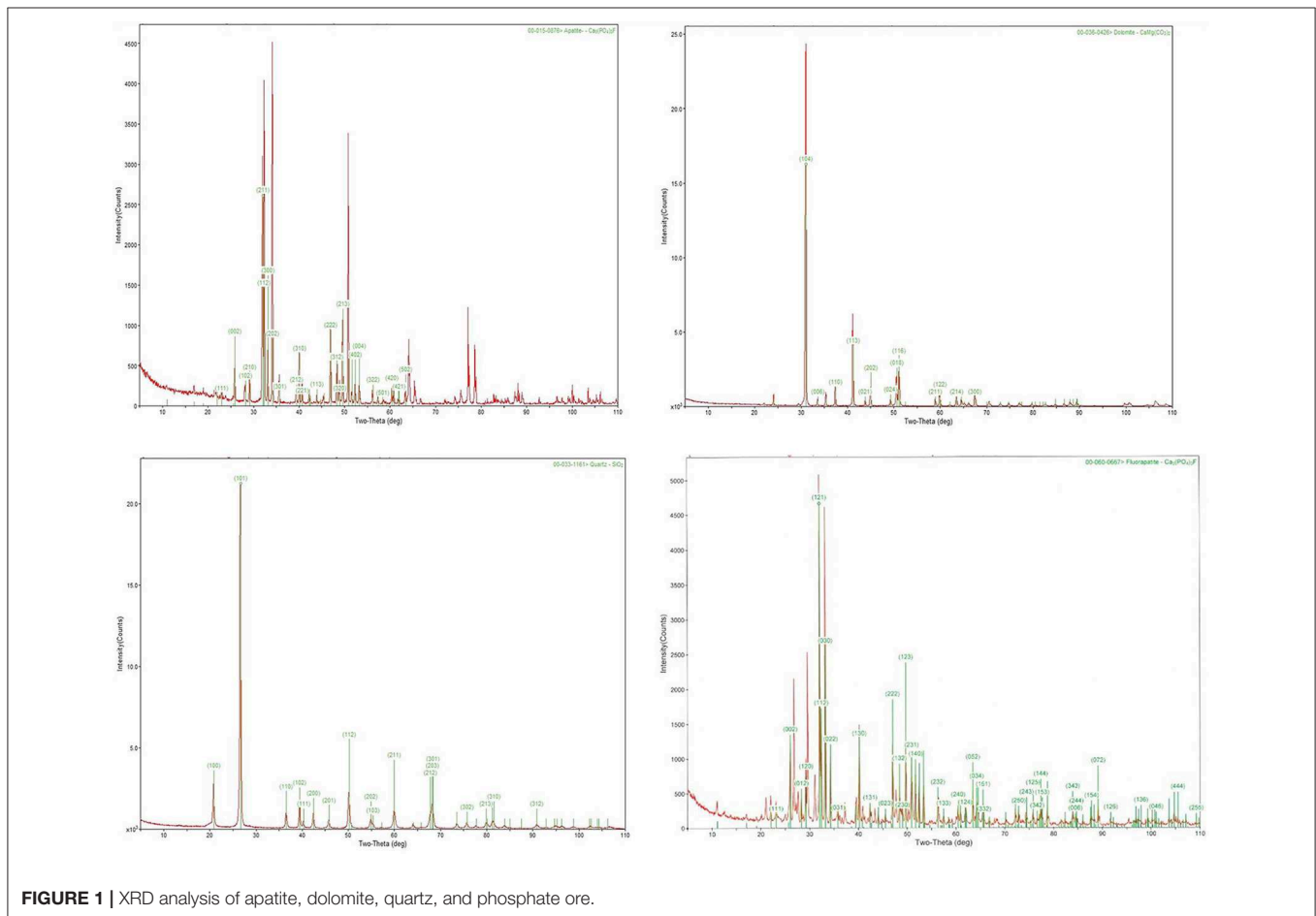
$\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  are conducted to explore the surface chemistry changes in phosphate ore flotation.

## MATERIALS AND METHODS

### Materials

Potassium chloride, calcium chloride, and magnesium chloride were used in reagent grade. Reagent grade hydrochloric acid and sodium hydroxide were used as pH modifiers.

The single minerals of apatite, dolomite and quartz were purchased from Canada Wards Natural Science Establishment Company, hand-crushed by a hammer, ground, and sieved to obtain particles with diameter smaller than  $20\ \mu\text{m}$  for zeta potential measurement. The phosphate ore from Yichang City, China, was ground by a Mortar Grinder (Fisher Scientific) to obtain the same size fractions as the single minerals for zeta potential measurement. All mineral samples prepared were analyzed by XRD shown in **Figure 1**. As seen in **Figure 1**, the XRD peaks of the tested apatite dolomite and quartz matched closely with that of the standard samples, confirming that the samples is mainly apatite, dolomite, and quartz, respectively. The phosphate ore sample is mainly fluorapatite.



## Methods

The mineral supernatants of apatite, dolomite, quartz, and phosphate ore were prepared by immersing the single mineral, respectively, in 10 mM KCl solutions for 48 h followed by suction filtration. The absence of mineral particles in the mineral supernatants was determined by an optical microscope. The ore samples were placed in the test solutions to prepare ore suspensions, which were hand-shaken for 2 min. After 10 min standing for mineral particles to reach equilibrium, the supernatant of the suspension was diluted by the test solutions, and then placed under an ultrasonic equipment HS2060A (Fisher Scientific) for 10 min. The detailed description can be found in the literature (Zhou et al., 2015). The obtained solutions were used for zeta potential measurement by Zetasize R-Malvern Nano-ZS. The measurements were repeated six times for the same suspension, and the average values as well as the standard deviations of a given ore sample were reported. All the experimental set-up was usually flushed by nitrogen gas to avoid the possible precipitation of calcium carbonate in the system and the formation of carbonate species on the ore surface (Amankonah et al., 1985). The measurement was conducted at ambient temperature of  $22 \pm 0.5^\circ\text{C}$ .

## RESULTS AND DISCUSSION

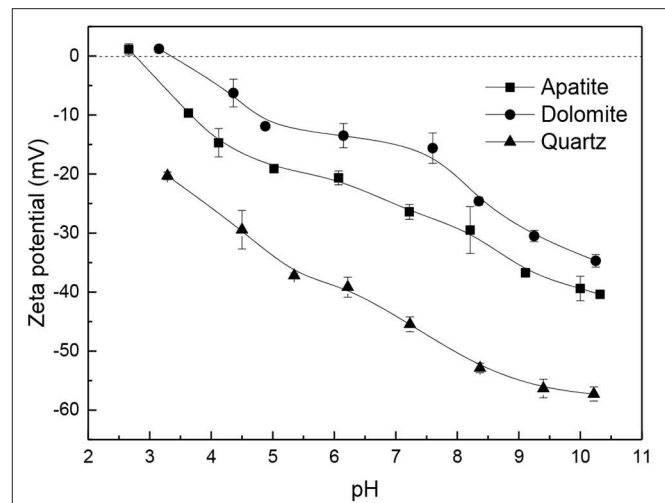
### Zeta Potentials of Apatite, Dolomite, Quartz

Results obtained for the zeta potentials of apatite, dolomite, and quartz in 10 mM KCl solutions are given in **Figure 2**. The Zeta potentials of apatite, dolomite, and quartz decrease with the pH increasing among the tested pH range. Some literatures report that isoelectric point (IEP) value of apatite from different deposits changes from 2.0 to 8.0, and the dolomite is around 3.0–6.0, but they can shift in the presence of electrolyte (Yu et al., 1985; Merma et al., 2013; Kasha et al., 2015). Therefore, isoelectric points of apatite and dolomite in 10 mM KCl solutions are observed, but no isoelectric points of quartz are determined around the tested pH range from 3.1 to 10.2. In general, the zeta potential of apatite is larger than that of dolomite, and that of quartz is the most electrostatically charged among the three ores.

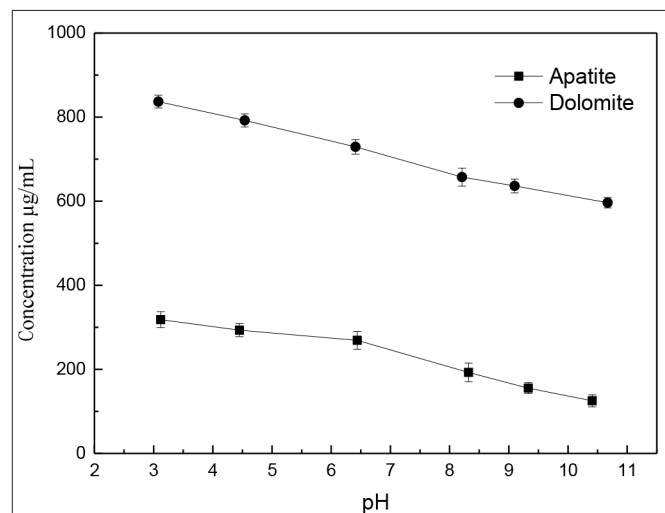
### Dissolution of Apatite and Dolomite

Apatite and dolomite are slightly soluble minerals, which can release the dissolved  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  to affect the surface electrical behaviors of minerals. Since the dissolution of apatite and dolomite can achieve equilibrium in 2 h according to the previous studies, the dissolved  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  contents as a function of pH were investigated with the two minerals soaked in different pH solutions for 2 h, respectively.

From **Figure 3**, the dissolved  $\text{Ca}^{2+}$  content in acid solutions is relatively high due to the carbonate and phosphate minerals reacting with hydrogen ions. But it decreases in alkaline environment, which is due to the precipitation of  $\text{Ca}(\text{OH})_2$ . This phenomenon is similar to that of  $\text{Mg}^{2+}$  in **Figure 4**, where the dissolved  $\text{Mg}^{2+}$  from dolomite decreases with the solution pH increase. Furthermore, the dissolved  $\text{Ca}^{2+}$  from dolomite is more than that of apatite, which is consistent with literature



**FIGURE 2** | Zeta potentials of apatite, dolomite, and quartz in 10 mM KCl solutions.

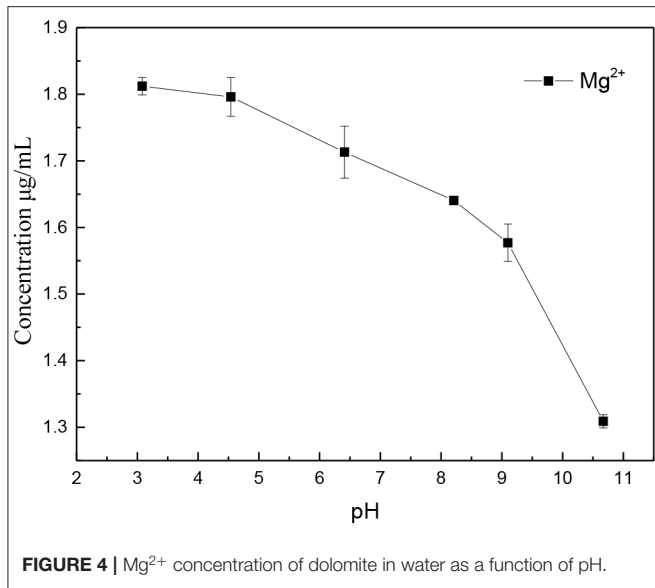


**FIGURE 3** |  $\text{Ca}^{2+}$  concentration of dolomite and apatite in water as a function of pH.

(Amankonah et al., 1985). Amankonah et al. (1985) found that the dissolved  $\text{Ca}^{2+}$  from calcite is more than that of apatite when calcite and apatite are soaked in open water, closed water and 2 mM  $\text{KNO}_3$  solution. Generally, the dissolved  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  will affect the surface electrical behavior of minerals in solution.

### Surface Electrical Behavior of Apatite, Dolomite, and Quartz in Apatite Supernatants

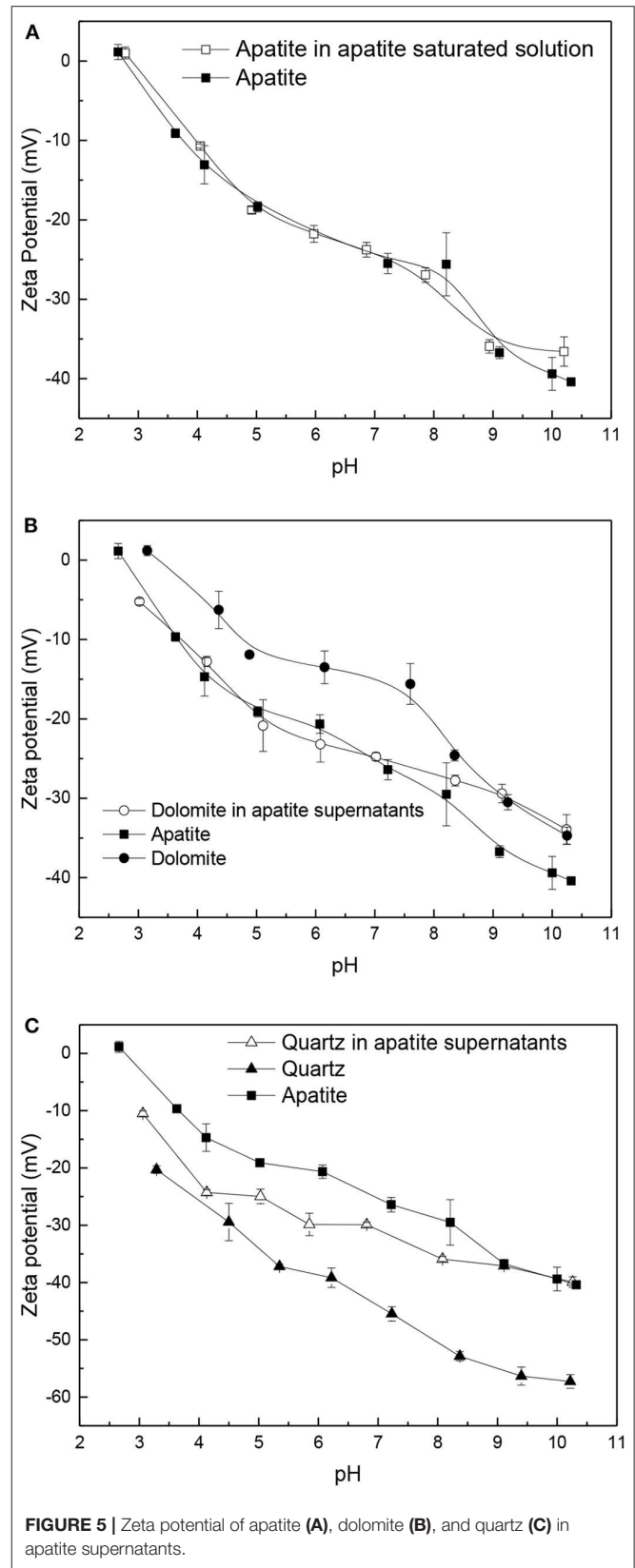
Results obtained for the zeta potentials of apatite in apatite supernatants was compared to that of apatite in 10 mM KCl shown in **Figure 5A**. It can be seen that there was no marked difference in the zeta potential values because of the similar surfaces of apatite in the two solutions. The experimental dates

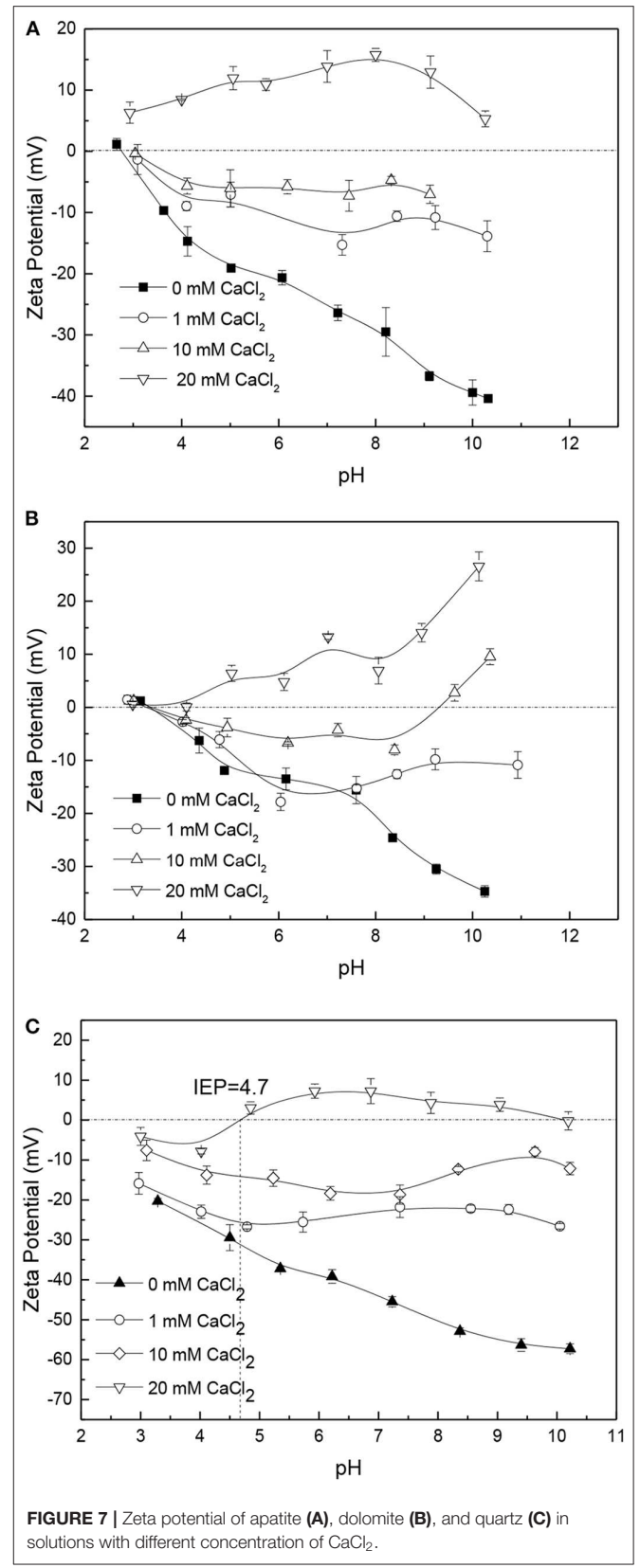
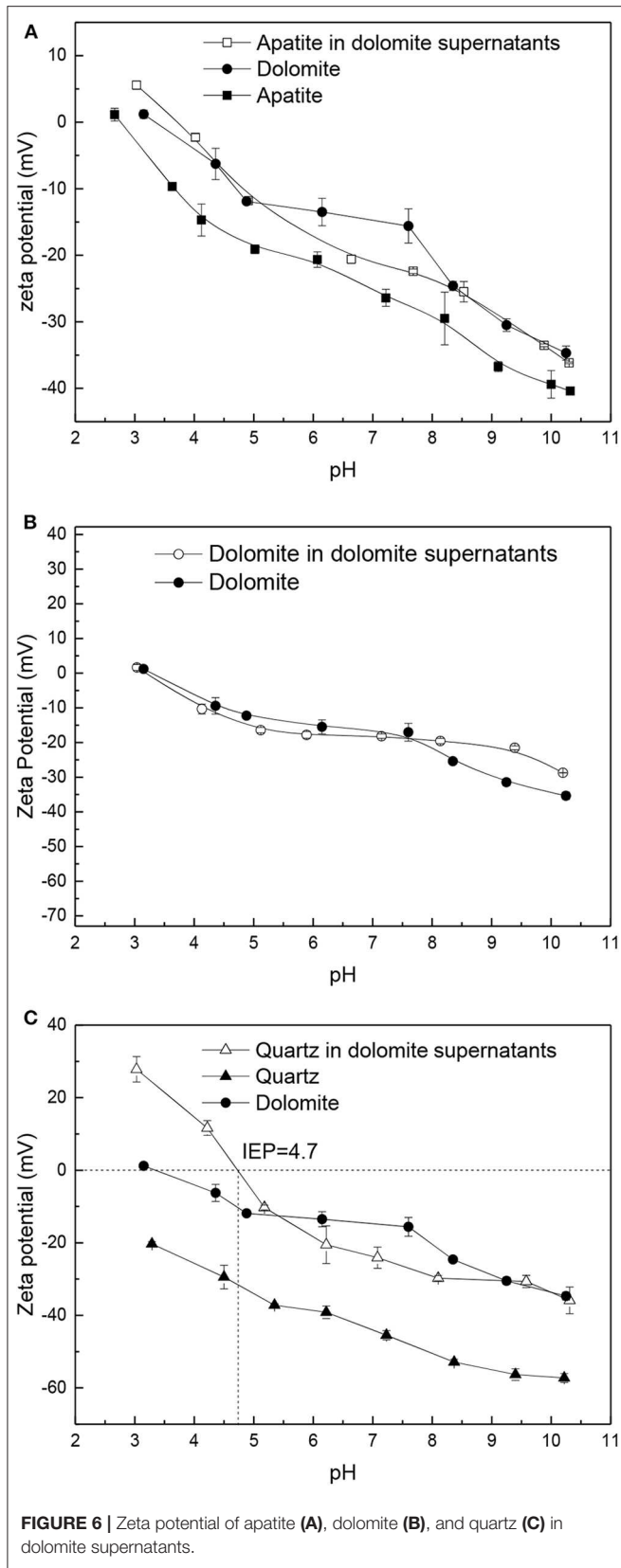


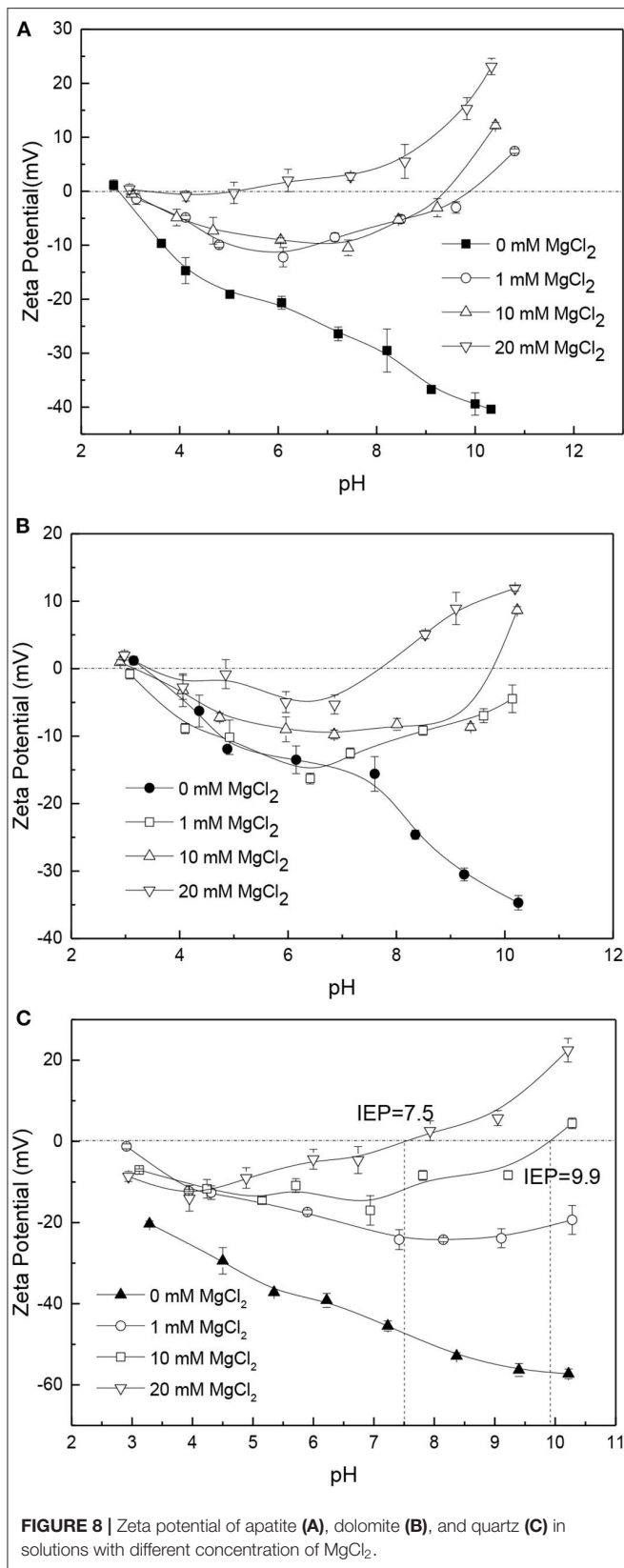
suggest that the influence of dissolved ions from apatite on the degree of variation in zeta potential values of apatite can be ignored. No changes can be deduced, because the new surface formed by the dissolved ions precipitated and/or adsorbed on apatite surface is similar to the apatite surface in the absence of apatite supernatants. The dissolved ions just change the electrolyte solutions, and further the influence is tiny because of the low concentration.

For dolomite in apatite supernatants, it is interesting to find that there is no marked difference between the zeta potential-curve of dolomite in apatite supernatants and the zeta potential-curve of apatite, but both of them are distinct to the dolomite. This result is similar to the interaction of apatite and calcite reported by Somasundaran (Amankonah et al., 1985). The observations indicate that the dissolved ions from apatite change the surface of dolomite to become similar to the apatite surface due to the precipitation and adsorption of dissolved ions on the dolomite surface.

In **Figure 5C**, the zeta potential-curve of quartz in apatite supernatants shifts toward less negative zeta potential compared to the original values of quartz, suggesting that the zeta potentials of quartz increase at the same pH because of the dissolved positive ions from apatite. There may be two reasons. The one is that the dissolved ions change the electrolyte solutions due to the zeta potential depending on the electrolyte and the ionic concentration (Sze et al., 2003). The second is that the dissolved ions are adsorbed or precipitated on the quartz surface to form calcium phosphate or hydroxide so as to change its surface electrical behavior. However, the zeta potential for quartz in apatite supernatants is still distinct to that of apatite, indicating that the dissolved ions cannot transfer the quartz surface to the apatite surface, very different from its effects on the dolomite.







## Surface Electrical Behavior of Apatite, Dolomite, and Quartz in Dolomite Supernatants

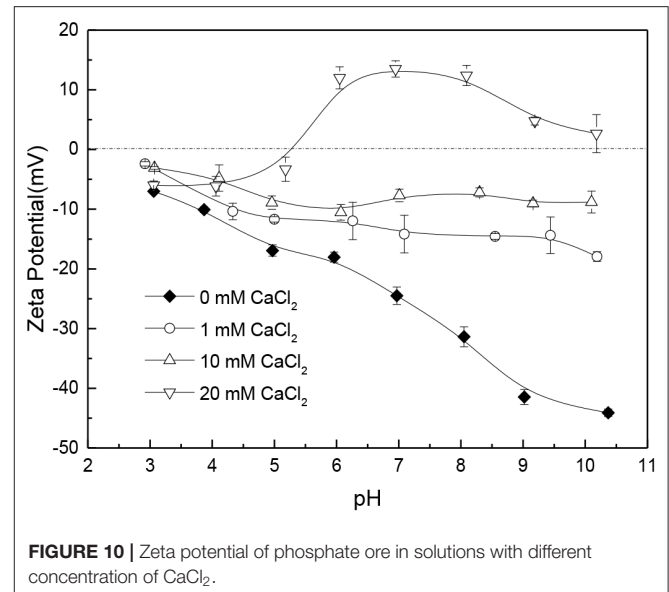
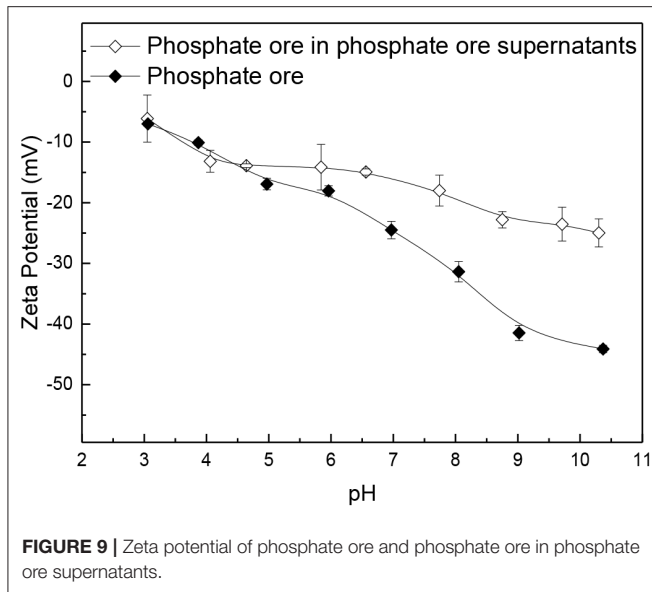
The results given in **Figure 6A** show the zeta potentials of apatite are increased by the dolomite supernatants, and move to the zeta potential-curve of dolomite, indicating the surface conversion of apatite to dolomite with the impact of dissolved ions from dolomite because the zeta potential increased with increase in ionic concentration of cations (Ozeki et al., 1989). This surface conversion from apatite to dolomite in dolomite supernatants is similar to the surface change from dolomite to apatite in apatite supernatants shown in **Figure 5B**. The zeta potentials of dolomite in dolomite supernatants are similar to that of dolomite over the entire pH range investigated shown in **Figure 6B**, indicating a less impact of the dissolved ions from dolomite on the zeta potential variation of dolomite. The results are similar to the effects of apatite supernatants on the surface electrical behavior of apatite in **Figure 5A**. The reason of the surface conversion for apatite and dolomite in the other supernatants may be the similar lattice cations of  $Ca^{2+}$  and  $Mg^{2+}$  for apatite and dolomite, which will adsorb on the surface of the minerals or form hydroxide precipitations. For quartz in dolomite supernatants shown in **Figure 6C**, the original negative zeta potential of quartz is increased and IEP at pH 4.7 is observed because of the increasing of dissolved ions from dolomite, which can be deduced they are divalent  $Ca^{2+}$  and  $Mg^{2+}$  ions due to the chemical component of dolomite,  $CaMg(CO_3)_2$  (Pokrovsky et al., 1999).

## Effects of $Ca^{2+}$ on the Surface Electrical Behavior of Apatite, Dolomite, and Quartz

The effect of  $Ca^{2+}$  on the surface electrical behavior of apatite, dolomite and quartz is shown in **Figure 7**, respectively. Examination of the results shows that the zeta potentials of the three ores are more positively charged in solutions containing  $Ca^{2+}$  than in 10 mM KCl among the tested pH range, and the  $Ca^{2+}$  can alter the zeta potentials of these ores from negative to positive. The affinity between the  $Ca^{2+}$  and the zeta potentials of apatite, dolomite, and quartz is affected by the concentration of  $Ca^{2+}$ . The greater the concentration of  $Ca^{2+}$  is, the higher the zeta potentials of these ores in calcium chloride solution is. The increase in the zeta potential values resulting from the concentration of calcium ions could be attributed to the compression of the double layer under high ionic strength conditions (Amankonah et al., 1985). Furthermore, a high concentration of  $CaCl_2$  can be expected to cause the precipitation such as calcium hydroxide on the surface of ores to produce more positive zeta-potential values.

## Effects of $Mg^{2+}$ on the Surface Electrical Behavior of Apatite, Dolomite, and Quartz

The effect of  $Mg^{2+}$  at different concentration on the surface electrical behaviors of apatite, dolomite, and quartz is shown in **Figure 8**. The  $Mg^{2+}$  causes the zeta potentials of these ores to be increased similar to the effect of  $Ca^{2+}$  among the tested pH



range. A higher concentration of  $\text{Mg}^{2+}$  has a greater impact on the surface electrical behaviors of apatite, dolomite and quartz. At a relative high concentration, 20 mM  $\text{MgCl}_2$ , the original negative zeta potentials of apatite, dolomite, and quartz change to be positive. The isoelectric points of these ores shift to a relatively small values. In the acidic solution,  $\text{Mg}^{2+}$  is in the form of positioning ion adsorbed on the surface of apatite and dolomite in the form of characteristic adion. Under the condition of high pH value, the adsorption of  $\text{Mg}^{2+}$  and  $\text{OH}^-$  may create precipitation on the surface of ores to produce more positive zeta potential values (Tang et al., 2010).

### Electrokinetic Behavior of Phosphate Ore Surface Electrical Behavior of Phosphate Ore in Phosphate Ore Supernatants

To understand the surface electrical behavior of phosphate ore, a phosphate rock from China was chosen to investigate the effects of its supernatants,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , on its zeta potentials which can affects interfacial process such as flotation. **Figure 9** shows the comparison of zeta potential-curves of phosphate ore and phosphate ore in phosphate ore supernatants. The two curves are basically coincident when  $\text{pH} < 6$ , but as pH increases, they separate gradually. In general, the zeta potential of phosphate ore in phosphate ore supernatants is greater than that of phosphate ore. The phosphate ore supernatants contain metal cations dissolved from phosphate ore. When  $\text{pH} > 8$ , these cations can attach and cause precipitation to the surface of phosphate ore to produce more positive zeta potential values.

### Effects of $\text{Ca}^{2+}$ on the Surface Electrical Behavior of Phosphate Ore

The effects of  $\text{Ca}^{2+}$  with different concentrations on the zeta potential of phosphate ore are shown in **Figure 10**. The results show that the zeta potentials of phosphate ore in 0 mM  $\text{CaCl}_2$ , 1 mM  $\text{CaCl}_2$ , and 10 mM  $\text{CaCl}_2$  are negative over the entire pH range investigated. The isoelectric point of phosphate ore in 20 mM  $\text{CaCl}_2$  is about 5.2. Data given in **Figure 10** for the

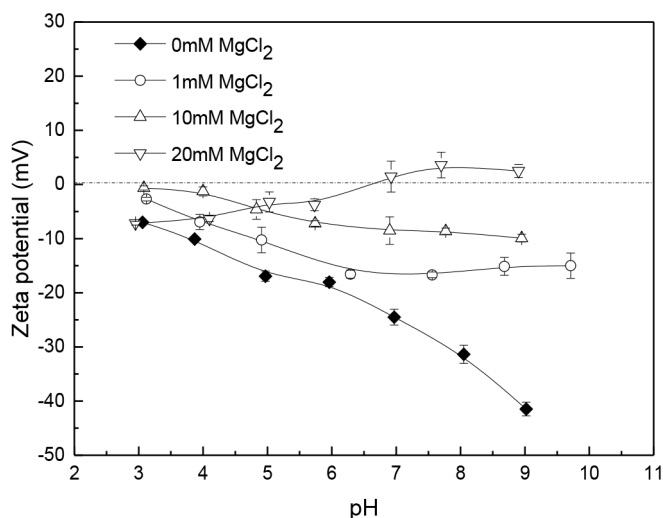
zeta potential of phosphate ore in calcium chloride of several concentrations show that the  $\text{Ca}^{2+}$  has the same impact on the zeta potential of phosphate ore to the other three ores, apatite, dolomite, and quartz. The greater the concentration of  $\text{Ca}^{2+}$  is, the higher the zeta potential of phosphate ore in  $\text{CaCl}_2$  solution is.

### Effects of $\text{Mg}^{2+}$ on the Surface Electrical Behavior of Phosphate Ore

**Figure 11** shows the effects of  $\text{Mg}^{2+}$  on the zeta potential of phosphate ore. The original negative zeta potential of phosphate ore is increased in the presence of  $\text{Mg}^{2+}$ . The results suggest that the  $\text{Mg}^{2+}$  reveals the similar influence on the zeta potential of phosphate ore as  $\text{Ca}^{2+}$ . However, the  $\text{Ca}^{2+}$  shows a greater impact than the  $\text{Mg}^{2+}$  on the zeta potential of phosphate ore among the tested pH range according to the comparison of **Figures 10, 11**. The isoelectric points of phosphate ore in 20 mM  $\text{MgCl}_2$  solutions, is about 6.6, but the isoelectric points in 20 mM  $\text{CaCl}_2$  solutions is about 5.2. A relatively higher influence of  $\text{CaCl}_2$  solutions than  $\text{MgCl}_2$  solutions are also investigated for the zeta potentials of apatite, dolomite, and quartz according to the **Figures 7, 8**, which is consistent with the results on silica reported in literature (Datta et al., 2009). Mammen et al. (1997) used a simple model of a silica surface to present that a higher degree of association of  $\text{Ca}^{2+}$  to the silica surface are consistent with the experimental zeta potentials tested by Datta et al. (2009). The higher zeta potentials of  $\text{Ca}^{2+}$  observed for comparison to  $\text{Mg}^{2+}$  may result from the increase in association of  $\text{Ca}^{2+}$  to the mineral surface and the larger ion radius of  $\text{Ca}^{2+}$  leading to lower polarizability than  $\text{Mg}^{2+}$  as well as its smaller influence to the electrons on the outer surface of minerals.

## CONCLUSION

No impact or limited impact of the apatite, dolomite and phosphate ore supernatants are found on the surface electrical



**FIGURE 11** | Zeta potential of phosphate ore in solutions with different concentration of MgCl<sub>2</sub>.

behaviors of the respective surfaces. The zeta potentials of quartz in apatite or dolomite supernatants increase compared to the original negative zeta potential of quartz. When apatite and dolomite are conditioned in the supernatants of each other, they interchange their zeta potential-curves over the entire pH range investigated. The zeta potential-curve of apatite in dolomite supernatants moves from that of apatite to dolomite, while the zeta potential-curve of dolomite in apatite supernatants moves from that of dolomite to apatite similarly. At high Ca<sup>2+</sup> and Mg<sup>2+</sup> concentration, especially 20 mM, the zeta potentials of apatite, dolomite, quartz and phosphate ore increase with IEP investigated, which is distinctly different from the original negative charge. Furthermore, it is interesting to find that the effects of Ca<sup>2+</sup> is greater than that of Mg<sup>2+</sup> on the surface electrical behaviors of apatite, dolomite, quartz, and phosphate ore.

These results are significant for the phosphate ore flotation. In the flotation circuit, the calcium and magnesium species from dissolved ores, not only increase the consumption of flotation reagents, but also change the surface properties of apatite,

dolomite and quartz. Especially for apatite and dolomite, the similar variation on surface charge will enlarge the difficulty of selective flotation of phosphate ore.

## DATA AVAILABILITY STATEMENT

All datasets generated for this study are included in the article/supplementary material.

## AUTHOR CONTRIBUTIONS

FZ designed experiments and wrote the manuscript. QL, XL, WL, and JF carried out experiments. RC analyzed experimental results.

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**Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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