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1 **Assessment of surface heterogeneity of lime treated kaolinites: probed by low-**  
2 **pressure argon and nitrogen gas adsorption**

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14 **Abstract**

15 It is a common practice to alter the undesirable behavior of clayey soils (i.e. swelling-shrinkage  
16 and high plasticity) with lime addition. However, the reaction mechanism involved in the early  
17 stage (first minute up to 24 hr after mixing) of lime treatment is still not well explored. In the  
18 present work, the surface chemistry of lime ([Ca] = 22 mmol/l and pH=12.63) treated kaolinites  
19 (Kaol) was studied using argon and nitrogen gas adsorption. The effect of pH ([Ca] = 22 mmol/l  
20 and. pH=12.63 vs. [Ca] = 22 mmol/l and pH=7) was also examined. The derivative low-pressure  
21 adsorption isotherms were analyzed using derivative isotherm summation (DIS), focusing on the  
22 analysis of surface heterogeneity. After the treatment with lime, the specific surface area and  
23 adsorption energy distribution of Kaol were modified. The adsorption of the calcium cation at both

24 investigated pH (i.e. pH 7 and pH 12.63) takes place on the basal and lateral face of Kaol, however,  
25 the species of adsorbed calcium cation appeared to be different. The treatment also leads to a  
26 decrease in basal surface area due to basal face-basal face particle aggregation. The basal surface  
27 of Kaol treated with lime exhibited polar sites when probed with nitrogen molecule. These sites  
28 may have promoted linkage and flocculation of Kaol particles, which in turn play a role in the  
29 short-term modification of the macroscopic behavior of lime treated kaolins.

30 Keywords: Kaolinite, lime treated, low pressure, argon, nitrogen, surface heterogeneity

## 31 **1 Introduction**

32 The clayey soils usually exhibit high swelling-shrinkage behavior and compressibility and are  
33 generally unsuitable for construction works. These behaviors are mainly governed by the amount  
34 and type of clay minerals present in the soil. Hence, it is a common practice to use lime (CaO or  
35 Ca(OH)<sub>2</sub>) as a chemical additive to alter these undesirable properties and made the clayey soil  
36 suitable in construction activities (Bell, 1996; Dang et al., 2016; Emarah and Seleem, 2018, Das  
37 et al., 2021). When lime is added to the soil in presence of moisture, the swelling potential and  
38 plasticity decrease in the short-term, and mechanical strength increases in the long-term (Bell,  
39 1996).

40 The long-term stabilization mechanism (i.e. pozzolanic reaction) has been thoroughly studied and  
41 well understood (Bell, 1996; Al-Mukhtar et al., 2010; Pomakhina et al., 2012; Eisazadeh et al.,  
42 2012; Vitale et al., 2017). It is related to precipitation of cementitious hydrated gels. On the other  
43 hand, the short-term reaction mechanisms, which are mainly controlled by surface phenomena  
44 (Bell, 1996; Chemedi et al., 2015; Vitale et al., 2016), are less explored. The reaction at this stage  
45 is crucial in soil stabilization as it can affect the mixing ability of the soil with lime (Konan et al.,

46 2009) and retards long-term pozzolanic reaction (Chemedá et al., 2018). Hence, a better  
47 understanding of the reaction mechanism is central for the optimization of the technique.

48 The true surface interaction of clay minerals with a solution, which is difficult to look at directly,  
49 is usually inferred from adsorption at solid-liquid interface or characterization of surface chemistry  
50 of the treated sample. In the previous works (Konan et al., 2009; Chemedá et al., 2018), adsorption  
51 at solid-liquid interface were used to assess lime solution-Kaol interfacial phenomenon. Their  
52 findings revealed that adsorption of calcium cation mainly takes place on a permanent charge and  
53 Si-O<sup>-</sup> sites at neutral pH, whereas adsorption on pH-dependent charge site is dominant at alkaline  
54 pH. In addition, the adsorbed species of calcium cation at pH 7 and pH 12.63 found to be different.  
55 However, using this technic, it is impossible to identify the adsorption sites due to difficulty in  
56 isolation of the different faces (basal and lateral faces). Thus, a study on surface characteristics  
57 with techniques that can distinguish the different crystallographic surfaces is indispensable.

58 The different crystallographic surfaces of clay minerals can be probed using high-resolution gas  
59 adsorption coupled with DIS (Michot et al., 1990; Bardot et al., 1998). The technique, particularly  
60 low-pressure argon gas adsorption was successfully applied in previous works (Villiéras et al.,  
61 1992; 1997; Bardot et al., 1998; Hassan et al., 2005; Mbey et al., 2019) for a quantitative estimation  
62 of surface heterogeneity in terms of lateral and basal surfaces of different clay minerals.  
63 Furthermore, it has been demonstrated that the presence and nature of surface cation viz. size,  
64 electronegativity and polarizability influences the normal interaction energy between the probe  
65 gas (argon and nitrogen) and the probed surface (Villiéras et al., 1992; Bardot et al., 1998; Hassan  
66 et al., 2005). These advantages can be utilized to indirectly probe the nature (species) of the surface  
67 cations and adsorption sites.

68 In the present work, the surface chemistry of four Kaol samples treated with lime ( $\text{Ca}(\text{OH})_2$ ) was  
69 characterized using low-pressure quasi-equilibrium argon gas adsorption in order to identify  
70 calcium cation adsorption sites. For this purpose, the derivative gas adsorption isotherms of lime  
71 treated and Na-exchanged Kaol samples were analyzed using derivative isotherm summation  
72 (DIS), focusing on the analysis of surface heterogeneity. The effect of pH value was also  
73 investigated by comparing isotherms of lime ( $[\text{Ca}] = 22 \text{ mmol/l}$  and  $\text{pH}=12.63$ ) and calcium  
74 chloride ( $[\text{Ca}] = 22 \text{ mmol/l}$  and  $\text{pH}=7$ ) treated samples. Polar/nonpolar surface characteristics of  
75 Kaol samples were examined using nitrogen gas adsorption.

## 76 **2 Materials and experimental procedures**

### 77 **2.1 Materials**

78 Four Kaols of different properties and origins, namely KGa-2 (high defect), KGa-1b (low defect),  
79 SPwt (low defect) and B24 (high defect), were selected for this investigation. Kaol KGa-1b and  
80 KGa-2 were purchased from the Source Clay Repository of the Clay Minerals Society. Kaol SPwt  
81 and B24 were collected from Cornwall, UK and Charente basin, France, respectively. Chemical  
82 analysis and X-ray diffraction pattern showed the mineralogical purity of the Kaol samples is >  
83 95% ([Chemedda et al., 2018](#)). Minor phases in the samples include muscovite (in B24 and SPwt),  
84 quartz and anatase. Hydrated lime,  $\text{Ca}(\text{OH})_2$  (Merck, analytical grade, purity above 97%) and  
85  $\text{CaCl}_2 \cdot 6\text{H}_2\text{O}$  (analytical grade, Merck, Germany) were utilized for preparation of hydrated lime  
86 and calcium chloride solution, respectively. High purity argon gas (> 99.9996%) and nitrogen gas  
87 (> 99.999%) provided by Alphagas were used for gas adsorption experiment.

### 88 **2.2 Chemical treatment**

89 The Kaol samples were first purified (i.e. the samples were de-carbonated, organic matters were  
90 removed and clay sizes were fractionated) and then exchanged with sodium at pH 7. Na-Kaol was  
91 prepared by sodium exchange in 1M NaCl solution. The samples were then washed with distilled  
92 water repeatedly until the supernatant was chloride free and had lower conductivity ( $< 5 \mu\text{S}/\text{cm}$ ).  
93 Lime and calcium chloride treated Kaol were prepared by suspending Na-exchanged Kaol samples  
94 in freshly prepared calcium hydroxide ( $[\text{Ca}] = 22 \text{ mmol/l}$  and  $\text{pH}=12.63$ ) and calcium chloride  
95 ( $[\text{Ca}] = 22 \text{ mmol/l}$  and  $\text{pH}=7$ ) solution, respectively. For this purpose, 6 g sample were kept in  
96 contact with calcium chloride and lime solutions (30 ml) in centrifuge tubes and shaken  
97 mechanically. This was followed by centrifugation for 15 min at 8000 rpm to separate the solid  
98 from solution. The preparations (weighting and mixing) were performed under argon atmosphere  
99 in a nitrogen glove box.

### 100 **2.3 Low-pressure gas adsorption at 77K**

101 The low-pressure adsorption isotherms were recorded with lab-built quasi-equilibrium device (at  
102 LIEC (Laboratoire Interdisciplinaire des Environnements Continentaux), Nancy) equipped with  
103 three high accuracy differential pressure sensors (0 – 10 Pa, 0 – 103 Pa and 0 – 105 Pa). A full  
104 review of the technique and the setup of the apparatus could be found in previous works ([Michot  
105 et al., 1990; Bardot, 1998](#)). The principle of the measurement is based on introduction of adsorbate  
106 through a calibrated microleak continuously at a constant and slow flow rate ([Michot et al., 1990;  
107 Villieras et al., 1992; 1997](#)).

108 About 1 g of samples were put in the adsorption cell and outgassed for 18 hr under a residual  
109 pressure of  $10^{-4}$  Pa. The outgassing was carried out at three different temperatures in successive  
110 steps (110°C, 150°C and 200°C) on the same sample. More than 3000 data points were obtained  
111 from recording of quasi-equilibrium pressures as a function of time. To obtain derivative

112 adsorption isotherm, the data points were plotted as first derivative of adsorbed quantity vs.  
113 logarithm of relative pressure ( $\ln(P/P_0)$ ), with  $P_0$  being the saturated vapor pressure (Michot et al.,  
114 Villieras et al., 1992; 1997). The derivative adsorption isotherm represents the adsorption  
115 energy distribution for the used gaseous probe (Michot et al., 1990; Bardot, 1998). The  
116 experimental derivative adsorption isotherms are then modeled with the sum of “local” theoretical  
117 isotherms derived from Brunauer, Emmett and Teller (BET) formalism using DIS procedure  
118 (Villieras et al. 1992). The lateral interaction parameter ( $\omega$ ) was kept constant ( $\pm 0.2$ ) during the  
119 modeling of isotherms. For each “local” adsorption isotherm, three parameters were then obtained:  
120 the position of the peak ( $\ln P/P_0$ ), which depends on solid-gas and gas-gas interaction energies; the  
121 lateral interaction parameter between adjacent adsorbed gas molecules ( $\omega$ ), and a monolayer  
122 capacity ( $V_m$ ). The specific surface area was calculated from monolayer capacity of each domain  
123 of local isotherm by applying BET derived formalism.

### 124 **3. Results and discussion**

#### 125 **3.1. Effect of outgassing temperature**

126 The effect of outgassing temperature on the argon adsorption energy distribution were analysed  
127 first to identify cations on charged and neutral surface. An example of experimental derivative  
128 argon adsorption isotherm for different outgassing temperatures of Na-KGa-2 (high defect) and  
129 Na-KGa-1b (low defect) samples are presented in Figure 1. Irrespective of the outgassing  
130 temperature, all the derivative adsorption isotherms show the same shapes with a main peak  
131 located at low and medium energy ( $\ln(p/p_0) > -5$ ) representing the adsorption on the basal surfaces.  
132 While the shoulder and tail at high energy, located at  $\ln(p/p_0) < -6$ , attributed to the adsorption on  
133 lateral surfaces. At  $\ln(p/p_0) > -3$ , low energy domain represent adsorption on basal surface and  
134 multilayer adsorption (Villieras et al., 1992; 2002; Bardot et al., 1998; Hassan et al., 2005). With

135 increasing outgassing temperature, the major lower energy peak shifted to lower energy and the  
136 peak become narrow, i.e. the low energy surfaces become energetically less heterogeneous  
137 (Villieras et al., 1992; 2002).

138 The derivative argon adsorption isotherms were modelled using six local isotherms assigned to  
139 local derivative adsorption isotherms (Hassan et al., 2005) following DIS techniques. The three  
140 highest energy domains (1, 2 and 3) attributed to adsorption on lateral faces whereas the three low  
141 energy domains (4, 5 and 6) represent adsorption on basal faces (Bardot et al., 1998; Hassan et al.,  
142 2005). The obtained modelling parameters (i.e.  $\ln(P/P_0)$  which represents the normal interaction  
143 energy of the argon molecule with surface (Villieras et al., 1992;1997;2002), volume adsorbed  
144 ( $V_m$ ) and lateral interaction ( $\omega$ ) ) after different outgassing temperature are presented in Table 1,  
145 for Na-KGa-2 and Na-KGa-1b. Irrespective of the outgassing temperature, the total specific  
146 surface area remains nearly constant. It means that the modification of the derivative adsorption  
147 features is mainly related to variation in the argon adsorption energy distribution rather than the  
148 variation of amount of gas adsorbed. Concerning local isotherms, the position of domains 1, 2, 3  
149 and 4 appeared to be unaffected by outgassing temperature suggesting charged lateral (domain 1,  
150 2 and 3) and basal (domain 4) surfaces. On these sites, the surface cations bound tightly (Hassan,  
151 2005). On the other hand, domain 5 shows relatively greater sensitivity to outgassing temperature  
152 (i.e. the peak position shifts to lower energy after outgassing at 200°C) implying the elimination  
153 of physisorbed compounds and/or loosely bound cations from uncharged surface (Hassan, 2005).  
154 Since the purpose of using this technique is to identify the calcium adsorbed on charged surface  
155 (not neutral compound), domain 3 (charged lateral surface) and 4 (charged basal surface) were  
156 considered for further analysis (next section). The isotherms after outgassing at 200°C were also



157 considered to minimize the effect of physisorbed compounds on the gas adsorption energy  
158 distribution.

## 159 **3.2. Effect of chemical treatment**

### 160 **3.2.1. Derivative adsorption isotherms**

161 The derivative argon adsorption isotherms of the Na-exchanged, lime treated and calcium chloride  
162 treated samples are compared in [Figure 2](#), for an outgassing temperature at 200°C. After treatment,  
163 major differences are noted in the shape of the derivative adsorption isotherms, particularly for the  
164 low energy peak, due to change both in intensity and position of the peak. In both cases, the  
165 intensity of the major peak attributed to basal face decreases and becomes wider. However, the  
166 change is more pronounced for lime treated sample. This finding is consistent with more calcium  
167 adsorption and surface modification in the presence of lime than with calcium chloride solution  
168 ([Chemedá et al., 2015](#)). In the case of lateral faces, additional argon molecules are adsorbed at high  
169 energy range resulting in a well-developed high energy peak which could corresponds to a new  
170 adsorption site generated by flocculated structure ([Bardot et al., 1998](#)). The intensity of the peak  
171 is relatively higher for lime treated sample in accordance with higher flocculation in the presence  
172 of hydrated lime than calcium chloride solution ([Chemedá et al., 2015](#)). Generally, the major  
173 modification on the lower energy peak suggests the interactions of the two solutions (lime and  
174 calcium chloride solutions) with Kaol dominantly takes place on the basal surface. However, with  
175 this technique, it is impossible to know exactly whether the reaction is on silica or alumina basal  
176 faces.

177 An example of the derivative adsorption isotherms together with their decompositions before and  
178 after treatment with lime and calcium chloride solutions are presented in [Figure 3](#), for KGa-1b and  
179 KGa-2. The numerical DIS results for the four studied Kaol are summarized in [Table 2 & 3](#).

### 180 **3.2.2. Textural parameters**

181 The total specific surface area deduced from the sum of monolayer capacity of each domain of  
182 local isotherm by applying BET derived formalism decreased after lime and calcium chloride  
183 treatments as evidenced from the decrease in the intensity of low energy peak. Nevertheless, in  
184 some cases (KGa1b, KGa-2), the adsorption of additional argon in high energy region obscured  
185 this change ([Table 2](#)). The change is more evident in the low energy peak, particularly for domains  
186 4 and 5 ([Figure 4](#)). The extent of the change is different for the four studied Kaol samples, which  
187 appears to be depend on the degree of defect and cation exchange capacity. Indeed, there is an  
188 approximate linear relationship between the amounts of calcium adsorbed ([Chemedá et al., 2018](#))  
189 and the change (decreased) in basal specific surface area derived from domains 4 and 5 ([Figure 5](#)).  
190 The more the amount of calcium adsorbed, the more the basal specific surface area decreased. This  
191 phenomena can be interpreted in two ways: 1) involvement of exchangeable cation, in the present  
192 case calcium, in the first mono-layer adsorption site of argon molecule and/or 2) the aggregation  
193 of Kaol particles.

194 The first possibility can be checked by comparing the surface area occupied by the two cations  
195 (sodium and calcium) knowing their amount on the surface (from [Chemedá et al., 2018](#)). For this  
196 purpose, surface area occupied by Na and Ca-cations were compared for the raw (Na-Kaol) and  
197 calcium chloride treated samples ([Table 4](#)). The area occupied by Ca-cation is either equal or lower  
198 than the area occupied by Na-cation, which is not surprising as the size of the two cations are  
199 nearly the same ( $\text{Ca} = 0.99 \text{ \AA}$ ,  $\text{Na} = 0.97 \text{ \AA}$ ) and the former neutralizes two charge sites per cation.

200 It means that the area should increase when Na-cation is replaced by Ca-cation contrary to our  
201 observation. The other possibility is aggregation of Kaol particles. The replacement of monovalent  
202 cations by high order valency cations can change the particle arrangement from dispersed to  
203 flocculated structure (Mpofu et al., 2003, 2005; Chemedo et al., 2015). The decrease in basal  
204 specific surface area could be thus linked to the aggregation (probably basal face-basal face) of the  
205 clay particle that makes inaccessible a part of basal surface for argon adsorption. A similar  
206 phenomenon between monovalent and divalent cation (Na vs. Ca, Li vs. Mg and K vs. Ba) was  
207 observed for illite by Bardot (1998), although the change is more pronounced in the case of illite.  
208 However, the observed modification cannot solely accounted to change in texture (aggregation) of  
209 Kaol particles as there is also a shift in peak position in addition to decrease in the intensity. The  
210 observed difference could also be linked to the effect of the treatments on surface chemistry (this  
211 point addressed in next section).

#### 212 **3.2.4. Surface energetic parameters**

213 The energetic parameters ( $\ln(P/P_0)$ ) of domain 3 and 4 varied depending on the nature of the  
214 treatment (i.e. shifted to higher energy after lime treatment), revealing the modification of surface  
215 chemistry of lateral and basal surfaces (i.e. different surface cations) (Figure 6). This hypothesis  
216 is furthermore supported by the fact that the obtained peak position in the case of raw sample (Na-  
217 Kaol) and calcium chloride treated sample (Ca-Kaol) compared well with the peak position of Na-  
218 illite (for domain 4,  $\ln(P/P_0) = -4.53$  and for domain 3,  $\ln(P/P_0) = -6.95$ ) and Ca-illite (for domain  
219 4,  $\ln(P/P_0) = -4.56$  and for domain 3,  $\ln(P/P_0) = -7.05$ ), respectively (Bardot et al., 1998). On the  
220 other hand, the peak positions of both domains in the case of lime treated sample are higher than  
221 the calcium chloride treated samples. The only difference between the two solutions (i.e. calcium  
222 hydroxide and chloride solution) is the initial pH. The observed difference could be then attributed

223 to the effect of pH on surface charge density and the nature of calcium available for adsorption.  
224 However, as it has been demonstrated for the case of synthetic saponites with different layer charge  
225 (Michot and Villieras, 2002), the normal adsorption energy of argon on basal face is much more  
226 sensitive to the nature of surface cation than to the origin and the density of charge. One can then  
227 suppose that the nature of the charge compensating cations in the two cases are different. Indeed,  
228  $\text{CaOH}^+$  and  $\text{Ca}^{2+}$  are the dominant species of calcium available for adsorption at pH= 12.6 and 7,  
229 respectively (Mpofu et al., 2005; Chemedda et al., 2015). Hence, in the case of lime treated sample  
230 (i.e.  $\text{CaOH}^+$  as a surface cation), the argon molecule interacts with OH-ion in addition to calcium  
231 and the size is also larger than calcium cation. Therefore, the interaction energy is expected to be  
232 higher than the case of the calcium chloride treated sample (i.e.  $\text{Ca}^{2+}$  as surface cation).

### 233 **3.2.5. Argon vs. nitrogen isotherms**

234 Nitrogen and argon have different characteristics: argon molecule gives rise to only non-specific  
235 interactions, while due to its inducible quadrupolar momentum, nitrogen molecule interacts also  
236 with specific sites (Rouquerol et al., 1989; Villieras et al., 2002; Garnier et al., 2005), provided  
237 that the surface of the particle carries localized polar groups or positive charges. For the assessment  
238 of polar or non-polar surface characteristics before and after treatment, the adsorption energy  
239 distributions of the first monolayer as a function of natural logarithm of pressure ( $\ln P$ ) for nitrogen  
240 and argon were compared for selected samples (Figure 7). As usually observed for clay minerals,  
241 there is difference between the isotherms of the two gases at the higher energy range for the three  
242 samples irrespective of the treatment. It mainly corresponds to specific interaction of nitrogen with  
243 surface polar sites (Villieras et al., 1992; 2002; Bardot et al., 1998) and will not be further discussed  
244 here as it is not related to the effect of the treatment.

245 Concerning the basal face, the shape of the peak of the argon and nitrogen derivative adsorption  
246 isotherms appeared nearly identical (i.e. narrow and sharp) for Na-exchanged and calcium chloride  
247 treated sample. Furthermore, the main peak in both cases located at the same position (the values  
248 of  $\ln P$  around 6 to 7) suggesting non-polar nature of the basal surface. For the case of lime treated  
249 sample, the shape of the two gas isotherms are different as the peak of nitrogen isotherm is  
250 relatively wider due to additional shoulder at high energy side. The major peak, which located on  
251 the same position for the two gases ( $\ln P$  around 6) similar to Na-exchanged and calcium chloride  
252 treated samples, can be attributed to non-polar surface site. On this site, both nitrogen and argon  
253 adsorbed through low energy interaction (Villiéras et al., 2002; Garnier et al., 2005). A shoulder  
254 at higher energy side (indicated by arrow) in the case of nitrogen isotherm, could be attributed to  
255 the interaction of nitrogen molecules with some polar/ electropositive sites (Villiéras et al., 2002;  
256 Garnier et al., 2005) on the basal faces. The chemical nature of this site is not known exactly in  
257 the present study, hence, a deeper understanding of the surface chemistry of lime treated sample  
258 (at microscopic level) would be necessary to interpret this observation. It is tentatively attributed  
259 to  $\text{CaOH}^+$  or electropositive sites generated by adsorption of hydrolysis species of calcium as  
260 indicated by negative to positive surface charge reversal of Kaol particle in the presence of calcium  
261 rich alkaline ( $[\text{Ca}] = 22 \text{ mmol/l}$  and  $\text{pH}=12.63$ ) solution (Chemedda et al., 2015). These sites may  
262 promote linkage and aggregation of Kaol particles.

#### 263 **4. Conclusion**

264 The results of this work revealed that the adsorption of calcium cation from lime ( $[\text{Ca}] = 22 \text{ mmol/l}$   
265 and  $\text{pH}=12.63$ ) and calcium chloride ( $[\text{Ca}] = 22 \text{ mmol/l}$  and  $\text{pH}=7$ ) solution takes place both on  
266 the basal and lateral faces. These sites can be attributed to  $\text{Si-O}^-$  (lateral face) and local charge  
267 defect (i.e.  $\text{Al}^{+3}$  for  $\text{Si}^{+4}$  substitution) in tetrahedral sheet at pH 7, and additional charge sites

268 generated by dissociation of Al<sub>2</sub>OH and Al-OH functional groups on octahedral basal and lateral  
269 faces, respectively, at pH 12.6. The species of calcium cation available for adsorption at the two  
270 pH conditions are different as evidenced by the variation of the adsorption energy ( $\ln(P/P_0)$ )  
271 corresponds to charged surface on the basal and lateral faces depending on the source of the cation  
272 (calcium chloride or lime solution). The data indirectly validates the results obtained in previous  
273 works (i.e. Ca<sup>2+</sup> and CaOH<sup>+</sup> at pH 7 and 12.6, respectively) from adsorption at solid-liquid  
274 interface (Chemeda et al., 2015) and calculation of pH-dependent calcium cation hydrolysis  
275 (Mpofu et al., 2003; 2005). The face-to-face aggregation of Kaol particles, owing to compression  
276 of electrical double layer in the presence of calcium cation, mainly accounts for the decrease of  
277 basal specific surface area or adsorbed gas volume after lime and calcium chloride treatment. The  
278 basal surface of Na-exchanged and calcium chloride treated Kaol samples exhibits mainly non-  
279 polar characteristics, whereas lime treatment induces additional polar surface sites. These sites  
280 could be CaOH<sup>+</sup> or electropositive sites emerged on Kaol surface in the presence of calcium rich  
281 alkaline solution, which in turn induced particle linkage and short-term modification of  
282 macroscopic behavior of kaolins.

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