COMPARISON OF KINETIC EQUATIONS TO DESCRIBE POTASSIUM- CALCIUM EXCHANGE IN PURE AND IN MIXED SYSTEMS

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ABSTRACT

We used first-order, Elovich, parabolic diffusion, and zero-order equations to describe the kinetics of K-Ca exchange in kaolinite, montmorillonite, vermiculite, and soils of the Atlantic Coastal Plain Region. The first-order equation was the best of the various kinetic equations studied to describe the reaction rate of K adsorption in the clay minerals and soils, as evidenced by the highest simple correlation coefficients (r) and the lowest values of the standard error of the estimate (SE). The parabolic diffusion law described K adsorption best on vermiculite and soils dominated by vermiculitic clay minerals. The parabolic diffusion law did not describe K adsorption well on kaolinite and soils rich in kaolinite. These differences were related to the kinds of binding sites present in the pure and mixed systems. The Elovich and zero-order equations did not satisfactorily describe K adsorption in the soils and clays. Apparent potassium adsorption rate coefficients (k') were 0.23, 0.58, and 2.65 h⁻¹ for vermiculite, montmorillonite, and kaolinite, respectively. The k' values for the soils ranged from 0.94 to 1.66 h⁻¹, and their magnitude was related to the type and quantity of clay minerals present.

INTRODUCTION

A number of equations have been used to describe the kinetics of K exchange in clays and in soils (Kesy and Wild 1963; Burns and Barber 1961; Kloe and Gast 1967; Quirk and Chute 1968; Malcom and Kennedy 1969; Sparks et al. 1980a,b). However, little has appeared in the literature comparing these different equations for K exchange.

The Elovich equation was first developed to describe the kinetics of chemisorption of gases on solid surfaces (Low 1960). Paravano and Boudart (1955) criticized using the Elovich equation to describe a unique mechanism, because they found that it described a number of different processes, such as bulk or surface diffusion and activation and deactivation of catalytic surfaces. Recent theoretical studies with adsorption and desorption in oxide-aqueous solution systems showed that the applicability and method of fitting kinetic data to the Elovich equation require accurate data at short reaction times (Allen and Scaife 1966; Abahoni and Ungarish 1975, 1977). The Elovich equation described the kinetics of heterogeneous isotopic exchange reactions from goethite and gibbsite surfaces (Atkinson et al. 1970; Atkinson et al. 1972; Kyle et al. 1975). More recently, a modified form of the Elovich equation was used by Chien and Clayton (1968) to describe simultaneous first-order kinetics data for phosphate sorption and release in soils.

The Elovich equation may reveal irregularities in data ordinarily overlooked by other equations (Low 1960). It has been suggested that if n is characteristic of the nature of the sites involved in the adsorption process, then any "breaks" in the Elovich plot could indicate a changeover from one type of binding site to another (Low 1960; Atkinson et al. 1970; Chien and Clayton 1980). Such "breaks" may not be artifacts of kinetic treatments (Low 1960), but the nonlinear Elovich plots may indicate a different reactivity of sites for the adsorption of ions on an irregular surface (Atkinson et al. 1970). Hingston (1981) notes that the Elovich equation may be quite applicable to adsorption in soils and sediments where there is wide variation in activation energies because the mixture of adsorption surfaces is so complex.

The parabolic diffusion law has been used successfully in describing K release in clays (Bashard 1954; Chute and Quirk 1967), but it
does not always describe kinetics of exchange in soils adequately; (iv) Sisavabanjanian and Tali-
binden 1972; Evans and Jurinak 1976; Sparks et al. 1980a,b). The problem in using the para-
bolic equation for soil systems may be ascribed to the nebulous interpretation of the slope pa-
rameter. Sisavabanjanian and Talibinden (1972) obtained parabolic plots that had two slopes. They suggested this could be indicative of two simultaneous diffusion-controlled reac-
tions. Sparks et al. (1980b) noted a nonlinearity with the parabolic diffusion equation for the
initial minutes of K description in soils. They attributed this deviation to film diffusion-con-
trolled exchange in the early minutes of K ex-
change.
The first-order equation has been utilized by
many soil chemists to describe diffusion-con-
trolled kinetons in clays and in soils (Mortland
and Ellis 1951; Sawhney 1966; Sisavabanjan-
ian and Talibinden 1972; Sparks et al. 1980a,b).
Sawhney (1966) described the adsorption of Ca
on vermiculite as a pseudo first-order reaction.
Adsorption and desorption of K-exchange in soil
systems followed first-order kinetics (Sisavaban-
janian and Talibinden 1972; Sparks et al. 1980a,b; Sparks and Jardine 1981; and Rechcigl 1982). In a study examining the effects of temperate and moisture on K release from soils, Burns and Barber (1961) also noted first-
order kinetics.

The purpose of this study was to compare differ-
nent kinetic equations for K adsorption in the A and B horizons of Upper Atlantic Coastal
Plain Region soils and in standard clay minerals.

MATERIALS AND METHODS

Theoretical considerations

First-order equation

For a miscible displacement technique, a first-
order kinetic equation for K adsorption can be
derived as follows

\[
\frac{dK_*}{dt} = hK_* - K_e (1)
\]

where \(K_*\) = amount of K on the colloid at
time \(t\), and \(K_e\) = amount of K on the colloid at
equilibrium.

Separating variables

\[
\frac{dK_*}{K_*} = hK_* dt
\]

Integrating of the form \(\ln(y + x) = ln(1 + x)\)

\[
\log \frac{1 - K_*}{K_e} = hK_* t
\]

Because \(K_e\) is a constant, one can call this
product a constant \(k\). Thus

\[
\log \left(1 - \frac{K_*}{K_e}\right) = k t^{1/2}
\]

where \(k^{1/2}\) = apparent adsorption rate coefficient.

Elvish equation

The Elvish equation can be derived as fol-

\[
\frac{dq}{dt} = a^2 e^{-at}
\]

where \(q\) = amount of material adsorbed at time
\(t\), and \(a\) and \(\alpha\) = constants during any one
experiment. Assuming \(q = \text{constant}\) at \(t = 0\), Eq. (5) becomes,

\[
q = 2.3 \alpha \log(1 + a t)
\]

or

\[
q = 2.3 \alpha \frac{a}{1 + a t} = \frac{2.3}{a} \log a t
\]

where \(\alpha = 1/a\).

If a volume of gas, \(q_0\), is adsorbed instantan-
eously and before Eq. (5) begins to apply, the
integrated form of the equation becomes

\[
q = 2.3 \alpha \log(t + k) = 2.3 \alpha \log a t
\]

where \(k = t - \text{exp} a q_0\).

If \(k\) is negligible in comparison with \(t\), Eq. (8)
reduces to

\[
q = 2.3 \alpha \log t
\]

or

\[
q = 2.3 \alpha \log a t
\]
Equation (10) results directly from Eq. (6) if \( n = 1 \), as shown by Chien and Clayton (1988). Thus, for K adsorption one could evaluate the Elovich equation (Sparks et al. 1986), by plotting the amount of K adsorbed (\( \mu g \, K/g \) soil) versus \( \log t \) (min).

**Parabolic diffusion equation**

For K exchange, a radial diffusion law can be expressed as (Crane 1970)

\[
K_{c} = \frac{4}{r^{3/2}} \left( \frac{D t}{\alpha^{2}} \right)^{1/2} - \frac{D t}{\alpha^{2}} \tag{11}
\]

where

- \( K_{c} \) = quantity of K adsorbed at time \( t \)
- \( K_{e} \) = amount of K adsorbed at equilibrium
- \( \alpha \) = average radius of soil or clay particle
- \( D \) = diffusion coefficient and
- \( t \) = time

Equation (11) can also be written as

\[
\frac{1}{t} \left( \frac{K_{c}}{K_{e}} \right) = \frac{4}{\alpha^{3}} \left( \frac{D}{\alpha^{2}} \right)^{1/2} \ln \left( 1 + \frac{D t}{\alpha^{2}} \right) \tag{12}
\]

The parabolic diffusion equation (Laidler 1965) for K exchange can be expressed as

\[
K_{c} = \frac{D t}{\alpha^{2}} \times \text{constant} \tag{13}
\]

where \( R \) = the overall diffusion coefficient.

**Characterization analyses**

Soil samples from the A and B horizons of a Matapeake silt loam (Typic Haplustolls), a Kearnsville loamy sand (Arenic Hapludult), and a Downer sandy loam (Typic Hapludults) were taken, air-dried, and gently ground to pass a 2-mm sieve in preparation for laboratory analyses. These soils are prevalent in the Upper Atlantic Coastal Plain region. The standard clay minerals used in the study were Washington County, Texas, kaolinite; Conesville County, Texas, montmorillonite; and Llano vermiculite. These were obtained from the University of Missouri Soil Clay Minerals Depository. The clays were fractionated and prepared for laboratory analyses according to procedures outlined by Jackson (1976). The CEC of the clay minerals, as determined by Mg-Ca exchange (Rich 1992), was 1.15, 70.5, and 207.00 meq/100 g for kaolinite, montmorillonite, and vermiculite, respectively.

Soil pH was determined using a 1:1 water-to-soil ratio, and organic matter was quantified using a modified Walkley-Black procedure (Alison 1965). Cation exchange capacity was measured using an MgCl2 saturation with a subsequent displacement by CaCl2 (Rich 1992; Okazaki et al. 1985). Particle size analysis was determined by the hydrometer method (Day 1955).

Before mineralogical analysis, samples were treated with NaOCl-adjusted to pH 9.5 to remove organic matter (Andersen 1963). Iron oxides were removed using a Na-dithionite-citrate-bicarbonate procedure (Mehra and Jackson 1960). The clay and silt fractions were passed wet through a 300-mesh sieve and separated by centri- fugation. Oriented mounts of the clay fraction were prepared by depositing ~250 µg of clay on a ceramic tile, saturating with K or Mg under suction, washing free of salts, and glycolyzing the Mg-saturated samples. X-ray diffraction patterns of Mg glycerol-saturated samples at 25, 110, 300, and 500°C were obtained using a Diano XRD 8300 AD x-ray diffractometer equipped with a graphite monochromator, FEP-8 computer, and a printout. The samples were scanned at 2° (2θ) per min using CuKα radiation.

Prior to the kinetic studies, the soil horizons and clay minerals were made homogenic with respect to Ca by saturating with 0.5 M CaCl2. The excess Ca was removed with destilled water until a negative test for Ca was obtained using AgNO3.

**Kinetics of K adsorption using miscellaneous displacement**

The kinetics of K adsorption of the soils and clays was determined using a miscellaneous displacement technique (Sparks et al. 1988). Duplicate 1-µg samples of the basaltic soils and duplicate 0.5-µg samples of the clays were mixed with 50 µl of distilled water. The suspension was in- jected into a 47-mm Nuclepore filter column. The column was attached to a Florscent Al- pha fraction collector and a peristaltic pump. The pump delivered the K solution through the soils and clays at a uniform flow rate of 1.0 ml min⁻¹. All outputs were collected at various time intervals until the concentration of the leachate equaled that of the initial K solution. The K
concentration was measured in each aliquot using atomic absorption spectrophotometry. Mathematical equations, including the Eqvich, parabolic diffusion, first-order, and zero-order, were tested by least-square regression analysis for K adsorption in the soils and clays to determine which equation best described the data. The correlation coefficient (r) and the standard error of the estimate (SE) were calculated for each equation

\[ SE = \frac{1}{n-2} \left( \sum (x_i - \text{predicted})^2 \right) \]  

(14)

where \( k \) and \( k' \) are the measured and calculated concentration of K adsorbed at time \( t \), and \( n \) is the number of measurements.

RESULTS AND DISCUSSION

Selected chemical, mineralogical, and physical properties of the soils are given in Table 1. Of the three soils, the Matapexa contained the largest amount of clay. Clay content increased in all three soils with increasing depth. The <2-μm clay fraction of the A horizons of the Matapexa and Kennsawville soils were dominated by chloritized vermiculite with lesser quantities of kaolinite, mica, and quartz. The A horizon of the Dower soil contained substantial quantities of vermiculite, with lesser amounts of chloritized vermiculite, mica, and kaolinite. The <2-μm clay fraction of the B horizons of all three soils was dominated by kaolinite.

Kinetics of potassium adsorption

The kinetics of K adsorption in the clays and soils is shown in Figs. 1 and 2, respectively.

**TABLE 1**

<table>
<thead>
<tr>
<th>Horizon</th>
<th>Particle size analysis</th>
<th>Organic matter</th>
<th>CEC, meq/100 g</th>
<th>pH</th>
<th>Mineralogy of &lt;2-μm clay fractiona</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Sand</td>
<td>Sil %</td>
<td>Clay</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Matapexa silt loam</td>
<td>A</td>
<td>14.5</td>
<td>70.0</td>
<td>15.5</td>
<td>2.3</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>7.0</td>
<td>75.0</td>
<td>18.0</td>
<td>1.9</td>
</tr>
<tr>
<td>Kennsawville loamy sand</td>
<td>A</td>
<td>8.0</td>
<td>15.0</td>
<td>3.4</td>
<td>1.2</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>71.9</td>
<td>19.1</td>
<td>9.0</td>
<td>0.2</td>
</tr>
<tr>
<td>Dower sandy loam</td>
<td>A</td>
<td>78.5</td>
<td>15.0</td>
<td>8.5</td>
<td>1.3</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>70.0</td>
<td>16.0</td>
<td>14.0</td>
<td>0.5</td>
</tr>
</tbody>
</table>

*a C2+ = chloritized vermiculite, K+ = kaolinite, Mg = mica, Q4+ = quartz, Vr = vermiculite, Gi = gibbsite, 1 = most abundant, 5 = least abundant.
EXCHANGE IN CLAYS AND SOILS

The rate of K exchange on kaolinite was rapid, an apparent equilibrium being reached in 35 min. X-ray diffraction of the kaolinite we used in this study revealed only 7.12 Å total material, anal-

ysis in the clay (Bennas 1965) showed only small amounts of K, and the CEC was 0.012 meq g⁻¹.

Thus, the kaolinite was relatively pure. The rapid K adsorption on the kaolinite is due to exchange taking place only from planar external surface sites. The apparent equilibrium of K exchange on montmorillonite was attained in ~140 min. As would be expected, the rate of K adsorption was considerably slower on ver-

miculite than on kaolinite and montmorillonite, with apparent equilibrium being reached in ~250 min. The slow rate of K exchange on ver-

miculite was also observed by others (Kesy and White 1961; Malcom and Kennedy 1969). Mal-

colm and Kennedy (1969) found that 50 and 97% of Ba-K exchange reactions were complete after 10 and 720 s, respectively, using a batch equilibrium technique. With a miscible displace-

ment technique like ours, the rate of K exchange would be considerably lower (Sparks and Rech-

cigl 1982). The low rate of K exchange on ver-

miculite is due to interparticle transport and to tortuous exchange reactions (Barshad 1964; Sparks et al. 1990b).

More K was adsorbed in the B horizons of the soils than in the A horizons due to a higher percentage of clay and the type of clay found in the former horizons (Table 1, Fig. 2). The <2-

µm clay fraction of the B horizons was predomin-

antly 1:1 layer silicates, which do not have collapseable interlayer spaces. The larger quantity of this type of clay would afford more sites for K exchange. The Matapex soil adsorbed the largest quantity of K, and the Kennesawville soil adsorbed the least. This also conforms to the clay contents in the soils. Equilibrium in K adsorption averaged 105, 110, and 125 min in the two horizons of the Matapex, Kennes-

awville, and Downer soils, respectively. The longer equilibrium time in the Downer soil relative to the other soils is probably related to the higher amounts of vermiculite (Table 1).

Comparison of kinetic equations to describe K adsorption

The first-order equation was the best of the various kinetic equations studied to describe the reaction rate of K adsorption in the clay min-

erals, as evidenced by the overall highest value of r and the lowest value of SE (Table 2). That K adsorption in clays was described well by first-order kinetic's also shown in Fig. 3. This observation conforms to findings of others (Kesy and White 1961; Kloke and Gast 1967). The slope of the first-order plots was highest for kaolinite, indicating the highest rate of K ad-

sorption; the lowest rate of K adsorption oc-

urred in vermiculite.

The parabolic diffusion law described K adsorption best in vermiculite, as characterized by the highest r value and lowest SE (Table 2, Fig. 4). This observation shows intraparticle diffu-


tion.

| Table 2 | Correlation coefficient (r) and standard error of estimate (SE) of various kinetic equations for potassium adsorption in pure systems |
|---|---|---|---|---|
| Equation | Kaolinite | Montmorillonite | Vermiculite |
| SE* | r | SE | r | SE | r |
| 1. Elovich: | X(t) = a + b ln t | 7.27 | 0.999 | 11.50 | 0.999 | 24.79 | 0.985 |
| 2. Parabolic diffusion law: | X(t) = a + bt² | 8.40 | 0.942 | 3.10 | 0.968 | 2.36 | 0.997 |
| 3. First-order: | ln(1 - X(t)/X∞) = a - bt | 1.19 | -0.997 | 1.25 | -0.994 | 1.06 | -0.992 |
| 4. Zero-order: | (1 - X(t)/X∞) = a - bt | 15.40 | -0.682 | 10.85 | -0.962 | 12.14 | 0.977 |

* SE = in micrograms per gram.
sion-controlled exchange in vermiculite. This was also found by others (Bardad 1954; Mortland and Ellis 1959; Mortland 1963; Reed and Scott 1962; Chute and Quirk 1967; Feigenbaum et al. 1981). The parabolic diffusion law did not describe K adsorption well in kaolinite, as evidenced by the relatively low r values and high SE values (Table 2, Fig. 4). Relatively pure kaolinite, which we worked with, should have only external planar surface areas exposed for K exchange, and thus only film diffusion-controlled exchange should occur. This type of exchange is usually not described well by the parabolic diffusion law (Chute and Quirk 1967; Sparks et al. 1980b), because the latter best describes intraparticle transport (Boyd et al. 1947).

The Klovich equation is not suitable to describe K adsorption in pure clays, as can be seen from the large values of SE, even though the r values are quite high. The zero-order equation also did not describe K adsorption well, as shown by the larger SE values and lower r values than for the other equations (Table 2). The kinetics of K adsorption in the soils was also best described by the first-order equation (Table 3, Fig. 5), as shown by abnormally high r values and low SE values. The parabolic diffusion equation also described the kinetics of K adsorption well, although conformity was better in the A than in the B horizons (Table 3). This can probably be related to clay mineralogical differences in the two horizons of the three soils. Kaolinite predominated in the B horizons of all three soils (Table 3), and vermiculitic clay minerals were most prevalent in the A horizons. It was previously shown (Table 2, Fig. 4) that the

![Figure 3: First-order plots of K adsorption in pure systems.](image)

![Figure 4: Parabolic diffusion plots for K adsorption in pure systems.](image)

### Table 3

<table>
<thead>
<tr>
<th>Position</th>
<th>Horizon</th>
<th>Matapexes silt loam</th>
<th>Kaolinite loamy sand</th>
<th>Donner sandy loam</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>SE r</td>
<td>SE r</td>
<td>SE r</td>
</tr>
<tr>
<td>1. Klovich</td>
<td>A</td>
<td>3.93 0.58</td>
<td>2.58 0.59</td>
<td>8.27 0.64</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>1.13 0.49</td>
<td>1.41 0.49</td>
<td>1.50 0.68</td>
</tr>
<tr>
<td>2. Parabolic</td>
<td>A</td>
<td>1.20 0.68</td>
<td>1.29 0.67</td>
<td>1.40 0.69</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>1.77 0.97</td>
<td>1.79 0.97</td>
<td>1.60 0.99</td>
</tr>
<tr>
<td>3. First-order:</td>
<td>A</td>
<td>1.14 -0.95</td>
<td>1.41 -0.95</td>
<td>1.33 -0.96</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>1.39 -0.98</td>
<td>1.42 -0.99</td>
<td>2.18 -0.98</td>
</tr>
<tr>
<td>4. Zero-order:</td>
<td>A</td>
<td>15.80 -0.96</td>
<td>12.50 -0.99</td>
<td>15.50 -0.96</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>14.32 -0.92</td>
<td>14.60 -0.90</td>
<td>15.42 -0.92</td>
</tr>
</tbody>
</table>

* SE is in micrograms per gram.
The $k_h$ values were higher and the $t_{1/2}$ values were lower in the A horizons of the three soils than in the B horizons. Even though the A horizons had more chlortalidone vermiculite relative to the B horizons, the A horizons contained a much larger percentage of organo-matter, causing the $k_h$ and $t_{1/2}$ values of these horizons to be higher and lower, respectively. The Mispapea and Kannasville soils had average $k_h$ values for the two horizons, which were similar, as would be expected, due to similar mineral suites in the two soils (Table 1). However, the $k_h$ values of the Downer soil were lower than for the other two soils. This can be ascribed to the large amount of vermiculite in the Downer soil (Table 4).

### Table 4

<table>
<thead>
<tr>
<th>System</th>
<th>Horizon</th>
<th>$k_h$</th>
<th>$t_{1/2}$ h</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mispapea</td>
<td>A</td>
<td>1.86</td>
<td>0.57</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>1.50</td>
<td>0.46</td>
</tr>
<tr>
<td>Kannasville</td>
<td>A</td>
<td>0.96</td>
<td>0.72</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>0.96</td>
<td>0.72</td>
</tr>
<tr>
<td>Downer sandy</td>
<td>A</td>
<td>0.96</td>
<td>0.44</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>2.65</td>
<td>0.38</td>
</tr>
<tr>
<td>Mispapea</td>
<td>B</td>
<td>0.68</td>
<td>1.20</td>
</tr>
<tr>
<td>Vermiculite</td>
<td></td>
<td>0.23</td>
<td>3.01</td>
</tr>
</tbody>
</table>

$*k_h$ values were calculated from the first-order equation.

parabolic diffusion law did not describe $K$ adsorption well on kaolinite, though it did on vermiculite. As was observed for the clay minerals, the Elvico and zero-order equations did not describe K adsorption kinetics well in the soils, producing lower r values and higher SE values than were found with first-order and parabolic diffusion equations.

**Apparent adsorption rate coefficients ($k_h$)**

<table>
<thead>
<tr>
<th>System</th>
<th>$k_h$</th>
<th>$t_{1/2}$ h</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mispapea</td>
<td>A</td>
<td>1.86</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>1.50</td>
</tr>
<tr>
<td>Kannasville</td>
<td>A</td>
<td>0.96</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>0.96</td>
</tr>
<tr>
<td>Downer sandy</td>
<td>A</td>
<td>0.96</td>
</tr>
<tr>
<td></td>
<td>B</td>
<td>2.65</td>
</tr>
<tr>
<td>Mispapea</td>
<td>B</td>
<td>0.68</td>
</tr>
<tr>
<td>Vermiculite</td>
<td></td>
<td>0.23</td>
</tr>
</tbody>
</table>

* $k_h$ values were calculated from the first-order equation.
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