

KAOLIN DEPOSITS OF LEUCOGIA, RHODOPE, GREECE: PROCESSES OF KAOLINIZATION

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ABSTRACT

Kaolin deposits of residual origin associated with metamorphic rocks (leucocratic gneisses and schist-gneisses) occur in Leucogia area, NE Greece. The chemical, mineralogical and textural characteristics of kaolin deposits were investigated by X-Ray diffraction (XRD), Scanning Electron Microscope (SEM) and chemical analyses. Attempts have been made to correlate the primary and secondary assemblages and to establish the factors controlling the step-wise character of the transition from the fresh gneisses to kaolinized rocks. The kaolinization process seems to follow two alteration pathways: (i) Sodic plagioclases ? Halloysite ? Kaolinite. (ii) K-Feldspars ? Kaolinite.

KEY WORDS: Leucogia; kaolinite; kaolinization; halloysite; weathering; residual kaolin; gneisses;

1. INTRODUCTION

The broader area in which the kaolin deposits occur belongs geotectonically to the Rhodope massif (Osswald, 1938; Dimitrov, 1959; Kronberg et al., 1970, Mposkos, 1989, Burg et al., 1996). It is mainly composed of metamorphic rocks, which according to their mineralogical composition, stratigraphy and metamorphism are distinguished into two tectonic units (Papanikolaou et al, 1982, Zachos and Dimadis, 1983, Mposkos and Liati, 1993, Wawzenitz and Mposkos, 1997).

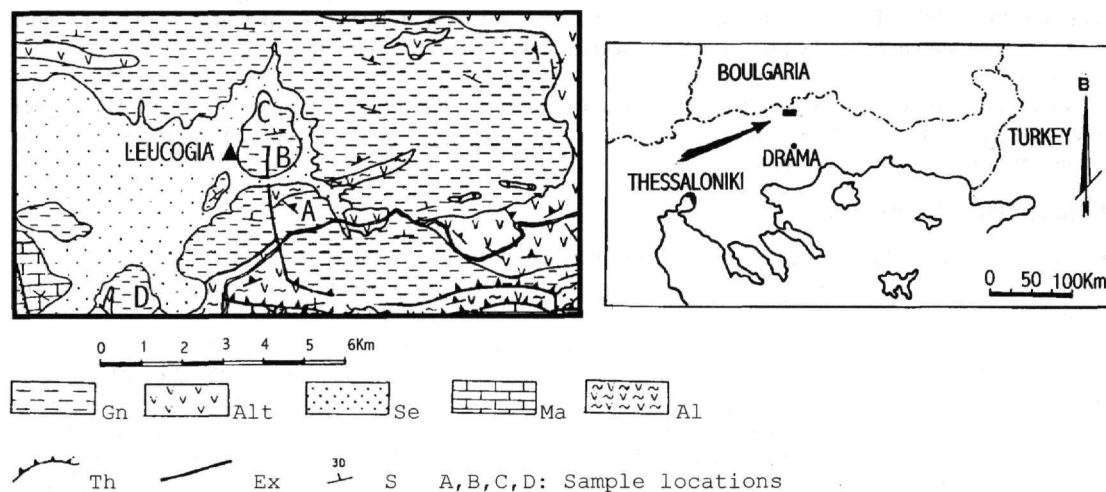


Figure 1. Geological map of Leucogia area (Chatzipanagis, 1991).

Gn: Gneisses, Alt: Alternation of Schist, Amphibolite, Gneisses and Marble, Se: Sediments, Ma: Marble, Al: Alternation of Schist, Gneisses, Marble, Th: Thrust, Ex: Extension fault, S: Inclination of Schistosity planes.

The lower tectonic unit characterized by metamorphism at conditions of the upper greenschist to lower amphibolite facies and the upper tectonic unit characterized by medium to upper amphibolite facies conditions (Mposkos and Liati, 1993). The Leucogia area is mainly composed of gneissic rocks of the upper tectonic unit.

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These rocks belong to the lower series of the metamorphic rocks of the western Rhodope (Osswald, 1938; Zachos and Dimadis, 1983).

The kaolin deposits lie on fine to coarsely crystalline and thinly bedded leucocratic gneisses and schist-gneisses. These rocks are mainly composed of quartz, feldspars (microcline, sodic plagioclases) and muscovite.

Michailidis and Tsirambides (1986); Tsirambides and Michailidis (1990), suggested a residual origin for the kaolins.

The kaolin deposits extend over an area of $\sim 20 \text{ km}^2$ and their thickness ranges from 1 to 50 m depending on the topographic relief (Ploumis and Chatzipanagis, 1993). They are grayish, yellowish to pale white in color and rarely stained by iron oxides. Sixty samples were collected from four different outcrops A,B,C and D (Fig. 1). At localities A and B the average thickness of the kaolin deposits is greater and the kaolinization is more advanced. At localities C and D the average thickness is about 10m, kaolins are less extended and show a lower degree of kaolinization.

2. ANALYTICAL TECHNIQUES

The studied samples were collected across several profiles, from the fresh gneiss to the upper parts of the profile which are intensively kaolinized.

Mineralogical analyses of bulk rock and clay fraction ($< 2\mu\text{m}$) samples were performed by XRD, using a Philips diffractometer PW1050/25, with Ni-filtered CuK α radiation. Powders from oriented specimens were scanned at $1^\circ 2\theta/\text{min}$ from 3 to $60^\circ 2\theta$. Clay minerals were identified from three XRD patterns (air-dried at 25°C , ethylene-glycolated, and heated at 490°C for 2h) of the clay-sized fraction ($< 2\mu\text{m}$) extracted by the standard sedimentation technique in deionized water.

Kaolin texture and kaolinite morphology were studied by a scanning electron microscope (SEM) equipped with EDS (Jeol SM 6300) operated at 20 kV.

The degree of kaolinite crystallinity or structural disorder was estimated by defining two crystallinity indices (i) R_2 Litard's test, determined on the (131) and (131) reflections, which is exclusively sensitive to the presence of stacking faults by translation of $\pm b/3$ (Litard, 1977) and (ii) test L_{002} related to the number of the layer-stacking faults along the c^* -axis and is determined by the enlargement at mid-height of the reflection L_{002} (Guinier, 1956). Hinckley index has not been determined because the reflections which are used for this index are forming a band and also the presence of plagioclase affects the reflection intensity at $20^\circ - 23^\circ 2\theta$ (Galan et al., 1994).

Chemical analyses of both major oxides and trace elements were carried out using I.C.P. on HClO_4/HF solutions, SiO_2 was determined using the X-Ray Fluorescence method after LiBO_2 fusion, Zr was measured by the xylenol orange colorimetric method.

3. RESULTS AND DISCUSSION

Five clay mineral phases were detected by XRD in the kaolins: kaolinite, halloysite, mixed-layer illite-smectite, smectite and discrete illite. Kaolinite increases significantly upwards from the unaltered parent rock to the upper horizons, whereas halloysite decreases.

The texture of Leucogia kaolin is typical of a residual kaolin produced by weathering (Keller, 1978); kaolin is replete with open interstices, the orientation of particles is random and its porosity is high. This texture would be developed where space for crystal growth is ample, due to leaching of material from the alteration of the parent rock. Morphologically kaolinite occurs as coarse, loosely packed "books" built of crystal plates. The orientation of plates is "face to face" (fig. 2). The results of the crystallinity indices are listed in table 1. It is evident from the data presented in table 1 that kaolinite show a high proportion of layer-stacking faults along the c^* -axis because $10 < L_{002} < 21$ and a few stacking faults by translation of $\pm b/3$, because $1,04 < R_2 < 1,26$ (Cases et al, 1982). SEM observations show c^* -axis disorder of kaolinite (fig. 3).

The structural formula of kaolinite from the Leucogia area, calculated from six representative electron microprobe analyses of kaolinite crystals on the basis of $\text{O}_{10}(\text{OH})_8$ is given in table 2 and show that kaolinite contains significant amounts of K^+ (0,10 p.f.u.) and Fe^{3+} (0,123 p.f.u.). Fe^{3+} substitutes for Al^{3+} in the octahedral sites and is responsible for the c^* -axis disorder of kaolinite (Mestdagh et al, 1980).

SEM observations show a mat of halloysite elongates lying on "books" and flakes of kaolinite (fig. 4). In the upper horizons of the profiles halloysite elongates are altered to kaolinite. In some cases remains of halloysite elongates appear in newly formed crystal plates of kaolinite (fig. 5). These observations indicate that the weathering process is not complete.

Table 1. Results of R_2 Litard's test and test L_{002} of kaolinite
n.d.: not determined because the presence of halloysite affects the intensity of the reflection 131 at 38° - $38,5^{\circ}$ 2θ .

| Sample | R_2 | L_{002} (mm) |
|--------|-------|----------------|
| L1 | 1,05 | 12 |
| L2 | 1,17 | 20 |
| L6 | 1,16 | 11 |
| L8 | 1,19 | 12 |
| L9 | 1,13 | 20 |
| L10 | 1,25 | 15 |
| L11 | 1,11 | 13 |
| L13 | n.d. | 18 |
| L14 | n.d. | 19 |
| L16b | n.d. | 14 |
| L18 | 1,15 | 20 |
| L22 | n.d. | 16 |

Table 2. Structural formula of kaolinite, on the basis of $O_{10}(OH)_8$ from Leucogia area.

| | Si | Al | Ti | Fe^{3+} | Mn | Mg | Ca | Na | K |
|---------|-------|------|-------|-----------|-------|-------|-------|------|------|
| Formula | 3,982 | 3,96 | 0,002 | 0,123 | 0,001 | 0,001 | 0,003 | 0,01 | 0,10 |

Major and trace element analyses of one gneissic rock and fifteen altered samples with different degree of kaolinization are given in table 3. The lowest potassium, calcium and sodium and the highest LOI contents are found in samples in which the alteration process is most advanced. The correlation coefficient of $CaO-Na_2O$ is +0,85, of Na_2O-LOI is -0,77, $CaO-LOI$ is -0,49 (table 4). The high loadings found between calcium and sodium shows that they decrease in the more altered samples following the same geochemical pathway. The high negative loadings found between LOI and sodium (fig. 6), of Na_2O and Al_2O_3 (fig. 7) and of SiO_2 and Al_2O_3 (fig. 8) show that LOI and Al_2O_3 increases in the more altered samples. These relations indicate that kaolinite is a product of the alteration of sodic plagioclases.

The correlation coefficient of Ba and K_2O , is +0,93 (fig. 9), suggesting that Ba content is related to K-feldspars. Electron microprobe analyses of representative K-feldspars showed high amount of BaO (~3%) and support the above suggestion. The potassium content in kaolin deposits is considered to be a good measure of the degree of alteration of the primary rocks. Tsolis-Katagas (1983) found that in kaolinitic samples with $K_2O < 1\%$ kaolinite was the dominant mineral constituent, while in samples with $K_2O > 2\%$ K-Feldspars were still present. The kaolinitic samples examined here gave K_2O values between 0,78%-4,73% suggesting that the alteration process is only completed in the upper horizons. The sodium content of the kaolinized rocks ranges between 0,03-2,63, suggesting that sodic plagioclases are practically completely altered.



Figure 2.

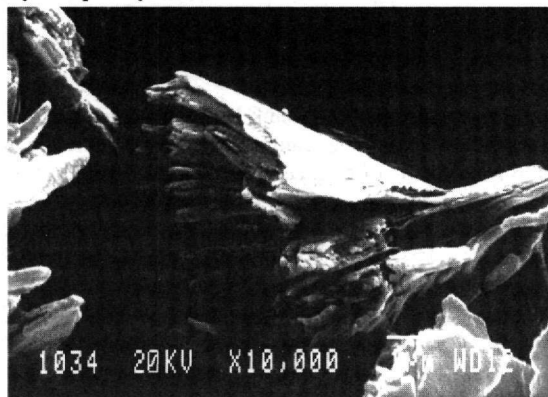


Figure 3.

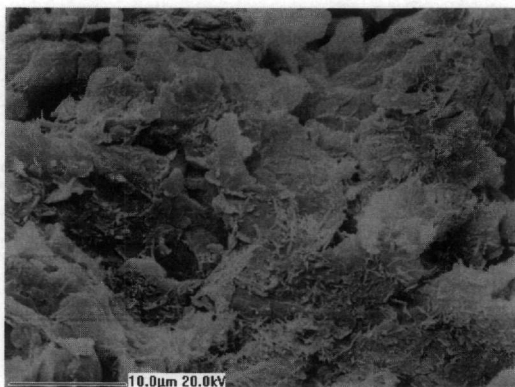


Figure 4.

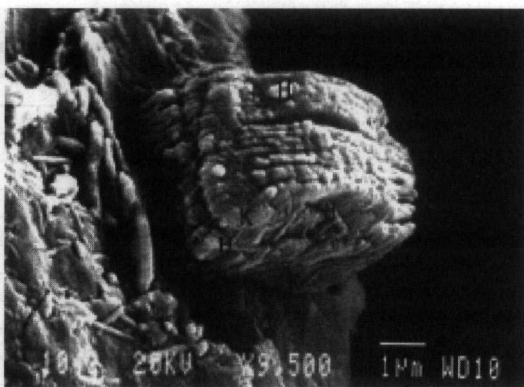


Figure 5.

Figure captions.

Figure 2. SEM micrograph of loosely packed books of kaolinite. Figure 3. SEM micrograph showing c-axis disorder of kaolinite. Figure 4. SEM micrograph showing mat of halloysite elongates lying on "books" and flakes of kaolinite. Figure 5. SEM micrograph showing remains of halloysite elongates in newly formed crystal plates of kaolinite, H=halloysite, K=kaolinite.*

4. CONCLUSIONS

1. The kaolinization process of Leucogia kaolin seems to follow two alteration pathways:
 - I Sodic plagioclases → Halloysite → Kaolinite.
 - II K-Feldspars → Kaolinite.
 In the first case sodic plagioclases are almost completely altered in the kaolins and the amount of kaolinite is higher than halloysite. In the second case K-feldspars are still present in the kaolins in considerable amount.
2. Kaolinite contains significant amounts of K^+ (0,10 p.f.u.) and Fe^{3+} (0,123 p.f.u.).
3. Kaolinite show a high proportion of layer-stacking faults along the c*-axis (c*-axis disordered) and limited stacking faults by translation of $\pm b/3$ caused by substitutions of Al^{3+} by Fe^{3+} in the octahedral sites.
4. a) The texture of kaolin is typical of residual kaolin produced by weathering, b) kaolinite increases significantly upwards from the unaltered gneissic parent rock to the upper horizons, whereas halloysite decreases, c) with increasing alteration Sr, an element which is related to hydrothermal fluids (Thorpe, 1982) and to kaolins of hydrothermal origin, is leaching and d) the kaolin formation has extensive surficial presence. The above observations indicate that kaolin deposits of Leucogia, are residual deposits formed in situ by the weathering of crystalline gneissic rocks.
5. Relative to the gneisses, kaolin is characterized by depletion of Si, K, Na, Ca, Ba and Sr and enrichment of Al and L.O.I.

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Table 3. Major and trace element analyses of parent rock* and kaolinized rocks from Leucogita. Major elements are in wt% and trace elements in ppm.

| Sample | SiO ₂ | Al ₂ O ₃ | Fe ₂ O ₃ | MgO | CaO | Na ₂ O | K ₂ O | TiO ₂ | P ₂ O ₅ | MnO | H ₂ O | L.O.I. |
|--------|------------------|--------------------------------|--------------------------------|------|------|-------------------|------------------|------------------|-------------------------------|-------|------------------|--------|
| L1 | 74,64 | 13,72 | 2,59 | 0,46 | 0,21 | 0,03 | 2,24 | 0,31 | 0,03 | 0,01 | 1,60 | 3,97 |
| L2 | 69,60 | 18,27 | 1,29 | 0,34 | 0,25 | 0,04 | 1,35 | 0,30 | 0,02 | 0,02 | 1,70 | 6,34 |
| L4 | 72,21 | 14,65 | 2,24 | 0,44 | 0,30 | 2,63 | 3,37 | 0,34 | 0,03 | <0,01 | 0,58 | 2,39 |
| L5 | 74,95 | 14,28 | 0,96 | 0,23 | 0,22 | 2,25 | 3,90 | 0,24 | 0,01 | <0,01 | 0,68 | 1,92 |
| L6 | 76,88 | 14,51 | 0,43 | 0,15 | 0,20 | 0,48 | 0,89 | 0,30 | 0,02 | 0,09 | 1,30 | 4,50 |
| L8 | 71,95 | 16,90 | 0,62 | 0,17 | 0,13 | 0,36 | 4,73 | 0,28 | 0,02 | 0,11 | 0,86 | 3,84 |
| L9 | 68,01 | 18,93 | 1,13 | 0,32 | 0,16 | 0,05 | 2,83 | 0,29 | 0,03 | 0,01 | 2,71 | 5,52 |
| L10 | 73,12 | 16,87 | 0,79 | 0,21 | 0,15 | 0,06 | 2,32 | 0,28 | 0,01 | <0,01 | 1,12 | 5,03 |
| L11 | 65,88 | 19,11 | 2,13 | 0,29 | 0,20 | 0,09 | 3,76 | 0,26 | 0,04 | 0,01 | 3,11 | 4,97 |
| L12 | 72,38 | 15,93 | 0,90 | 0,27 | 0,21 | 1,72 | 3,97 | 0,29 | 0,02 | <0,01 | 0,90 | 3,56 |
| L13 | 68,81 | 16,96 | 1,14 | 0,33 | 0,16 | 0,17 | 3,26 | 0,24 | 0,02 | <0,01 | 4,49 | 4,30 |
| L14 | 62,25 | 22,58 | 1,54 | 0,03 | 0,24 | 0,03 | 0,78 | 0,38 | 0,03 | <0,01 | 2,67 | 9,00 |
| L16b | 67,74 | 16,42 | 0,97 | 0,26 | 0,17 | 0,44 | 2,72 | 0,27 | 0,03 | 0,01 | 6,00 | 4,31 |
| L18 | 72,43 | 14,92 | 1,64 | 0,39 | 0,18 | 0,03 | 2,47 | 0,24 | 0,02 | <0,01 | 9,74 | 4,41 |
| L21* | 72,43 | 14,92 | 1,16 | 0,39 | 0,48 | 4,91 | 3,03 | 0,24 | 0,02 | <0,01 | 0,21 | 1,29 |
| L22 | 71,37 | 15,13 | 1,71 | 0,42 | 0,33 | 1,49 | 2,86 | 0,39 | 0,02 | <0,01 | 2,35 | 3,86 |

| Sample | Ba | Co | Cr | Cu | Li | Nb | Ni | Sc | Sr | V | Y | Zn | Zr | Ce | Nd | |
|--------|------|-----|----|----|----|----|----|----|-----|----|----|----|----|----|-----|----|
| L1 | 683 | 27 | 5 | 4 | 13 | 5 | 9 | 6 | 15 | 30 | 7 | 28 | 28 | 43 | 98 | 31 |
| L2 | 210 | 34 | 32 | 5 | 15 | 4 | 12 | 4 | 15 | 29 | 8 | 18 | 22 | 24 | 50 | 18 |
| L4 | 4711 | 50 | 6 | 2 | 10 | 7 | 9 | 2 | 288 | 27 | 21 | 34 | 20 | 58 | 100 | 40 |
| L5 | 1512 | 281 | 10 | 2 | 8 | 4 | 6 | 2 | 144 | 21 | 3 | 15 | 27 | 17 | 31 | 14 |
| L6 | 316 | 76 | 10 | 2 | 12 | 5 | 8 | 2 | 27 | 21 | 10 | 11 | 21 | 35 | 98 | 26 |
| L8 | 2245 | 31 | 6 | 2 | 8 | 5 | 5 | 3 | 106 | 21 | 28 | 13 | 30 | 46 | 87 | 32 |
| L9 | 720 | 16 | 8 | 2 | 10 | 5 | 7 | 6 | 33 | 25 | 7 | 21 | 27 | 44 | 118 | 28 |
| L10 | 817 | 54 | 8 | 2 | 8 | 5 | 7 | 5 | 43 | 17 | 2 | 12 | 24 | 14 | 30 | 12 |
| L11 | 1487 | 24 | 2 | 4 | 8 | 4 | 7 | 4 | 70 | 25 | 13 | 17 | 29 | 58 | 144 | 40 |
| L12 | 1377 | 33 | 8 | 1 | 10 | 6 | 6 | 2 | 131 | 22 | 10 | 17 | 33 | 40 | 83 | 26 |
| L13 | 867 | 23 | 6 | 2 | 11 | 4 | 6 | 4 | 43 | 23 | 10 | 23 | 27 | 44 | 84 | 34 |
| L14 | 157 | 52 | 14 | 4 | 9 | 7 | 11 | 9 | 18 | 30 | 6 | 23 | 28 | 50 | 107 | 32 |
| L16b | 1007 | 14 | 8 | 2 | 12 | 5 | 7 | 4 | 63 | 22 | 15 | 18 | 25 | 51 | 134 | 36 |
| L18 | 463 | 22 | 3 | 4 | 11 | 3 | 8 | 4 | 18 | 32 | 5 | 17 | 24 | 25 | 70 | 24 |
| L21* | 867 | 42 | 14 | 2 | 7 | 6 | 10 | 4 | 143 | 27 | 7 | 15 | 16 | 35 | 71 | 24 |
| L22 | 928 | 50 | 25 | 5 | 15 | 11 | 15 | 5 | 75 | 33 | 15 | 24 | 44 | 31 | 71 | 26 |

H₂O = loss at 110 °C, *LOI*=loss at 1000 °C

Table 4. The most significant correlation coefficients of major and trace elements from parent rock and kaolinized rocks of Leucogita.

| | SiO ₂ | Al ₂ O ₃ | TiO ₂ | Fe ₂ O ₃ | MnO | MgO | CaO | Na ₂ O | K ₂ O | P ₂ O ₅ | LOI | Ba | Sr | Zr | Zn | Cu | Ni | Cr | Co |
|--------------------------------|------------------|--------------------------------|------------------|--------------------------------|-------|-------|-------|-------------------|------------------|-------------------------------|-------|-------|-------|-------|-------|-------|-------|------|-------|
| Li | -0,05 | -0,16 | 0,51 | 0,22 | -0,01 | 0,41 | -0,19 | -0,43 | -0,47 | -0,01 | 0,32 | -0,53 | -0,59 | 0,32 | -0,14 | 0,57 | 0,56 | 0,55 | -0,22 |
| Co | 0,37 | 0,10 | -0,14 | -0,23 | -0,05 | -0,29 | 0,03 | 0,28 | 0,13 | -0,52 | -0,31 | 0,18 | 0,30 | -0,01 | -0,12 | -0,16 | -0,13 | 0,07 | |
| Cr | 0,07 | 0,03 | 0,51 | -0,05 | -0,08 | 0,17 | 0,35 | 0,13 | -0,40 | -0,25 | 0,21 | -0,39 | -0,11 | 0,13 | 0,21 | 0,50 | 0,79 | | |
| Ni | -0,12 | -0,02 | 0,68 | 0,40 | -0,26 | 0,53 | 0,49 | 0,12 | -0,53 | 0,06 | 0,24 | -0,53 | -0,22 | 0,27 | 0,20 | 0,75 | | | |
| Cu | -0,42 | 0,27 | 0,52 | 0,68 | -0,19 | 0,60 | 0,07 | -0,34 | -0,43 | 0,26 | 0,45 | -0,44 | -0,49 | 0,28 | 0,00 | | | | |
| Zn | -0,27 | 0,19 | 0,02 | -0,03 | -0,30 | -0,01 | 0,44 | 0,26 | 0,12 | 0,22 | -0,01 | 0,12 | 0,42 | 0,24 | | | | | |
| Zr | -0,05 | 0,00 | 0,58 | 0,27 | -0,10 | 0,14 | -0,13 | -0,18 | 0,31 | 0,09 | 0,08 | 0,30 | 0,05 | | | | | | |
| Sr | 0,45 | -0,37 | -0,40 | -0,18 | -0,04 | -0,26 | 0,62 | 0,81 | 0,66 | -0,08 | -0,76 | 0,73 | | | | | | | |
| Ba | 0,31 | -0,23 | -0,38 | -0,15 | 0,27 | -0,39 | 0,04 | 0,31 | 0,93 | 0,01 | -0,58 | | | | | | | | |
| LOI | -0,62 | 0,71 | 0,60 | 0,11 | 0,00 | 0,00 | -0,49 | -0,76 | -0,63 | 0,27 | | | | | | | | | |
| P ₂ O ₅ | -0,40 | 0,15 | 0,12 | 0,58 | -0,06 | 0,20 | 0,08 | -0,19 | -0,04 | | | | | | | | | | |
| K ₂ O | 0,18 | -0,20 | -0,48 | -0,07 | 0,04 | -0,14 | 0,02 | 0,34 | | | | | | | | | | | |
| Na ₂ O | 0,42 | -0,43 | -0,30 | -0,12 | -0,19 | 0,05 | 0,85 | | | | | | | | | | | | |
| CaO | 0,26 | -0,38 | -0,04 | 0,20 | -0,27 | 0,27 | | | | | | | | | | | | | |
| MgO | -0,37 | -0,04 | 0,14 | 0,75 | -0,58 | | | | | | | | | | | | | | |
| MnO | 0,33 | -0,20 | 0,03 | -0,47 | | | | | | | | | | | | | | | |
| Fe ₂ O ₃ | -0,30 | 0,03 | 0,20 | | | | | | | | | | | | | | | | |
| TiO ₂ | -0,13 | 0,18 | | | | | | | | | | | | | | | | | |
| Al ₂ O ₃ | -0,77 | | | | | | | | | | | | | | | | | | |

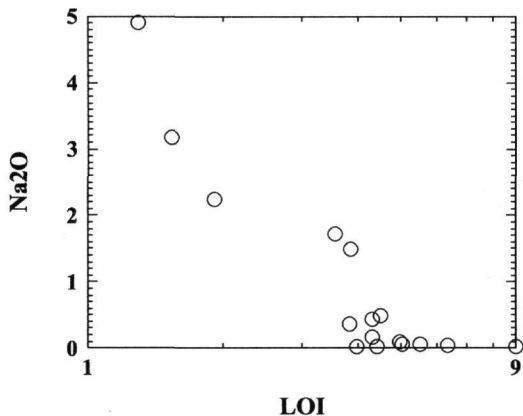


Figure 6.

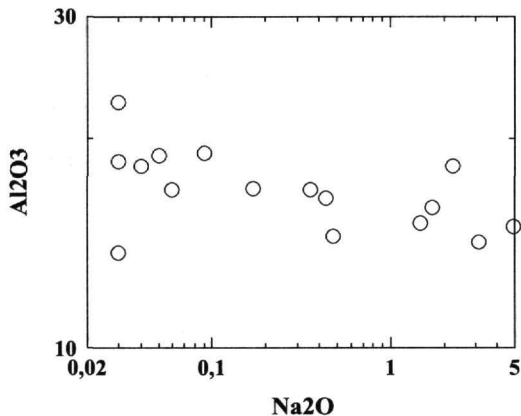


Figure 7.

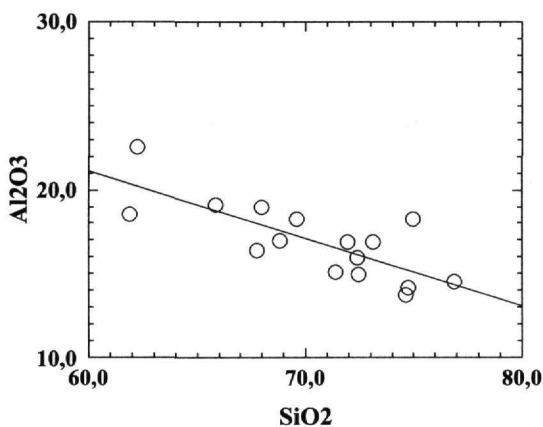


Figure 8. $y = -0,400x + 45,086$

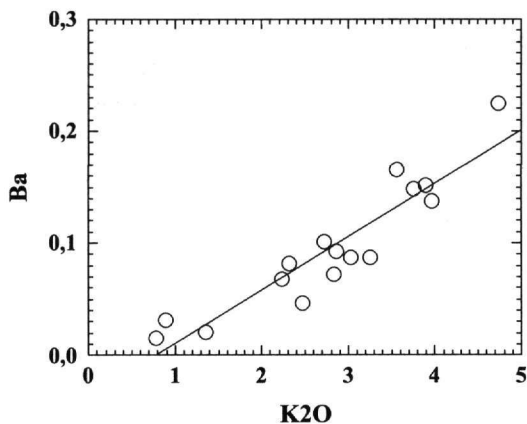


Figure 9. $y = 0,048x - 0,038$

Figures 6,7,8,9. Variation diagrams of selected major and trace elements of Leucogia kaolin and source rock.

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