MINERALOGIA POLONICA
Vol. 18, No 2 – 1987

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LITHIUM IN THE LOWER SILESIAN KAOLINS

Abstract. Lithium content in the Lower Silesian kaolins is fairly diversified. This phenomenon is evidently connected with the origin of their parent rocks. Lithium is most abundant in kaolins formed from weathered greisens and granitoids subjected to late- and post-magmatic alteration processes. In Lower Silesian kaolins, lithium is located not only in micas but also in kaolinite as diadochic admixture.

INTRODUCTION

The mean content of lithium in the commonest parent rocks of kaolins i.e. in granitoids is higher when compared with other igneous rocks. Depending on petrographic and genetic type of granitoid, the Li content usually ranges from 24 to 40 ppm (Wedepohl 1978). The role of alkali metals (such as potassium and, particularly, sodium) distinctly diminishes in kaolinization process. However, lithium behaves differently and its content may even increase. Consequently, the average Li content in clays amounts to 57 ppm (Turekian, Wedepohl 1961) and in some kaolins is even higher. According to Horstman (1957), the mean content of Li in 19 kaolin samples analyzed by him amounts to 120 ppm ($\sigma = 60$ ppm) whilst in 23 granites – 40 ppm ($\sigma = 30$ ppm). Very rich in lithium are some, purely kaolinitic, grain-size fractions below 2 μm, separated from kaolins of Montroux deposit in Massif Central Mts., France. The content of this element ranges from ca. 350 to ca. 600 ppm (Mosser 1982, 1983). This is, unquestionably, connected with high Li content in parent leucogranites containing sometimes more than 800 ppm Li (Aubert 1969, Burnol 1974 – *fide* Mosser 1982). Their kaolinization was often connected with hydrothermal processes.

The main lithium-bearing minerals is granitoids are micas – particularly biotite which can contain more than 90% of the total content of this element in a rock (Walenczak 1969). During metamorphic processes the concentration of Li in rocks tends to diminish. Consequently its content in biotites of metamorphic rocks is lower when compared with that of igneous ones (Gadomski 1968, Zawidzki 1976). Other rock-forming minerals of granitoids (quartz, feldspars) are distinctly lower in lithium. So e.g. in feldspars its content does not exceed 5 ppm (Wedepohl

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1978). Lithium content in plagioclases is higher than in potassium feldspars. This conclusion results from geochemical study of minerals contained in Lower Silesian granitoids from the environs of Strzegom and the Karkonosze Mts (Walenczak 1969).

As already mentioned, during kaolinization of granitoids the content of Li in these alteration products increases when compared with parent rocks. Lithium may be incorporated into the lattice of new-formed clay minerals (Wedepohl 1978) particularly into that of kaolinite (Horstman 1957, Mosser 1982, Mosser *et al.* 1985), what is due e.g. to small ionic radius and low valency of Li^{+}. In Horstman's (1957) opinion, heterovalent diadochy may have place in octahedral layers of kaolinite ($Al^{3+} \rightleftharpoons Mg^{2+}$). This hypothesis was later confirmed by ESR studies (Angel *et al.* 1974, Hall 1980). The above diadochy in connected with a deficit of charge. In elementary cell of kaolinite only four of six possible octahedral positions are occupied. Thus, the deficit of charge in connection with vacants in octahedral layers, could be the cause of entering monovalent Li^{+} into the lattice to reach its charge balance. Besides, the presence of this trace element in kaolinite lattice should also be related with the problem of so called structural iron. It is known that in kaolinite lattice the limited substitution of Fe^{3+} for Al^{3+} is possible. This phenomenon was confirmed e.g. by detailed studies of Lower Silesian kaolins containing, on the average 0.5% (Wiewióra 1970) and even 1% (Sikora 1974) structural Fe_2O_3. Moreover, Mössbauer (Malden, Meads 1967) and ESR (Angel *et al.* 1974) studies suggest the possibility of replacing Al^{3+} by Fe^{2+} in kaolinite lattice. Such replacement would need a compensation of charge what can be accomplished just by incorporation of small ions like Li^{+} into this lattice. Consequently, the opinion on the presence of trace amounts of lithium within kaolinite lattice is evidenced and well explained by heterovalent diadochy, first of all within octahedral layers of this mineral.

METHODS

The present study was carried out using some methods of chemical and phase analysis. Lithium and magnesium contents were determined by AAS using Perkin-Elmer spectrometer model 5000. The following parameters were applied:
air/acetylene flame

Li – wave length 670.8 nm, slit width – 1.4 nm
Mg – wave length 285.2 nm, slit width – 0.7 nm

The precision for Li and Mg is ±5% (1σ) and ±4% (1σ), respectively.

Phase analysis was carried out by means of *X*-ray and infrared spectroscopic methods. In *X*-ray studies the DRON-1,5 diffractometer (USSR production) with scintillation counter was used by applying CuKα radiation. Infrared spectroscopic analysis was carried out by means of C. Zeiss (Jena, GDR) UR-10 apparatus using KBr discs technique.

RESULTS

Lithium contents were determined in selected representative samples of both residual and sedimentary Lower Silesian kaolins. Their localization is presented in Fig. 1, whilst list of analyzed samples and the short characteristics – in Table 1.

The obtained results indicate considerably variable lithium contents in raw

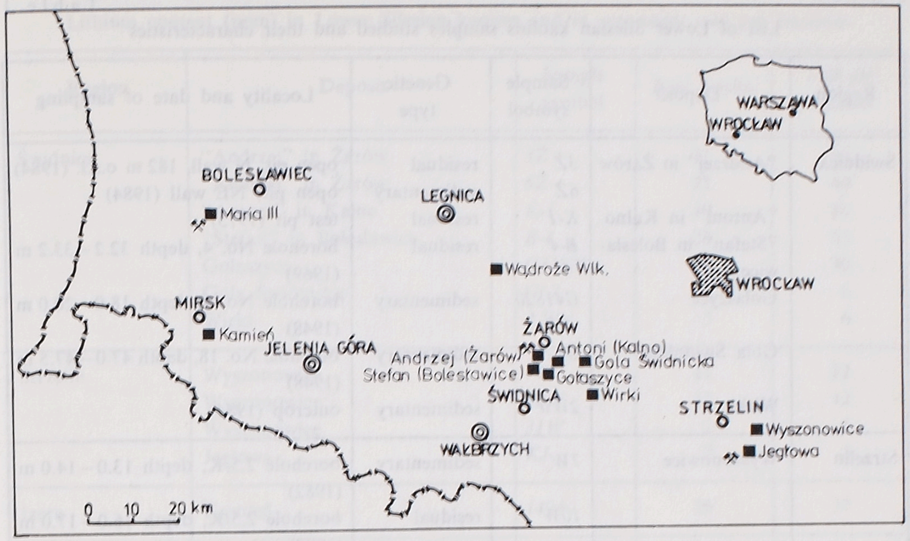

Fig. 1. Sampling sites of kaolins from studied Lower Silesian deposits

Lower Silesian kaolins (Tab. 2). The highest amounts of this trace alkali element were found in kaolin from Kamień near Mirsk (Izera region) and from the environs of Żarów near Świdnica („Andrzej" open pit in Żarów, „Antoni" deposit in Kalno, "Stefan" deposit in Bolesławice, Gołaszyce). Kaolin in Kamień was formed by weathering of leucogranites and gneisses intercalated with schists (Kościówko 1982). The rocks of this complex were subjected to greisenization (Karwowski 1977). Within the kaolin deposit in question a quartz-topaz vein was found (Budkiewicz 1971). Consequently, the increased Li content (29 ppm) in this kaolin should be related with greisenisation of its parent rocks. This process resulted, especially in the initial stage of greisenization, in concentration of Li in muscovite (Karwowski 1977). This mica, apart from dominant kaolinite and quartz, is one of the components of kaolin from Kamień.

Higher lithium contents in kaolins from the environs of Żarów are also, probably, connected with the origin of their parent granitoids of the Strzegom – Sobótka massif. In marginal parts it consists of more alkaline rocks subjected to pneumatolytic-hydrothermal processes (Kowalski 1967). In the environs of Żarów they are generally represented by two-mica granites, being products of late-magmatic alteration of different granites of Chwałków type which in their top and marginal parts of the massif were secondarily enriched in potassium and water. These processes resulted in microclinization of primary rocks, decalcification of plagioclases and crystallization of large muscovite flakes (Maciejewski 1973). Consequently, these two-mica granites represent a separate petrographic rock type (Gawroński 1982). According to recent opinions (Puziewicz 1985), based on the results of $^{87}Sr/^{86}Sr$ isotope studies, these two-mica granites and granodiorites of E part of the Strzegom – Sobótka massif were formed by partial melting of rocks dominated by upper mantle material and not, as was supposed previously (Majerowicz 1972), from the Sowie Góry Mts. gneisses. Among local differentiates and late- to post-magmatic products a two-mica Mrowiny granite was distinguished which is the parent rock for kaolin of Żarów (Gawroński 1982). Post-magmatic

Table 1

List of Lower Silesian kaolins samples studied and their characteristics

Region	Deposit	Sample symbol	Genetic type	Locality and date of sampling
Świdnica	"Andrzej" in Żarów	*3Ż*	residual	open pit, N wall, 182 m o.s.l. (1984)
		6Ż	sedimentary	open pit, NE wall (1984)
	"Antoni" in Kalno	*K-1*	residual	test pit (1973)
	"Stefan" in Bolesławice	*B-4*	residual	borehole No. 4, depth 32.2 – 33.2 m (1969)
	Gołaszyce	*G41820*	sedimentary	borehole No. 4, depth 18.0 – 20.0 m (1948)
	Gola Świdnicka	*Gl 2*	sedimentary	borehole No. 18, depth 47.0 – 47.5 m (1948)
	Wirki	*2Wir*	sedimentary	outcrop (1984)
Strzelin	Wyszonowice	*7W*	sedimentary	borehole 2,5/K, depth 13.0 – 14.0 m (1982)
		10W	residual	borehole 2,5/K, depth 16.0 – 17.0 m (1982)
		31W	residual	borehole 2,5/K, depth 37.0 – 38.0 m (1982)
	Jegłowa	*KJ*	residual	open pit K-4, SW wall, 200 m level (1985)
Izera	Kamień near Mirsk	*1494*	residual	open pit (1975), closed
Wądroże Wielkie	Wądroże Wielkie	*WW1*	residual	borehole 24/W, depth 21.4 – 28.9 m (1972)
		XV/4/1	residual	exploration ditch XV/4/1 (1984)
Bolesławiec	"Maria III" in Nowogrodziec	*III*	kaolinite sandstone	open pit, W wall, II level (1985)

processes may result e.g. in enrichment of micas in lithium what was evidenced for some granitoids of peripheral parts of the Strzegom – Sobótka massif (Zawidzki 1976). The role of pneumatolytic-hydrothermal processes is confirmed by increased concentration of Li in quartz from veins cutting residual kaolins in „Andrzej" open pit in Żarów. White quartz of these veins contains 10 – 20 ppm Li whilst its grey variety – up to 30 ppm of this element (Stenzel-Kolasa 1972).

Increased concentrations of Li are observed both in residual kaolins (samples *3Ż*, *K-1*, *B-4*) and in sedimentary ones (samples *6Ż*, *G41820*) from the environs of Żarnów (Tab. 2). The highest amounts of Li were discovered in residual kaolin from Kalno (39 ppm). Lithium is but slightly mobile during weathering of parent rock. This conclusion results from comparison of its contents in poorly kaolinized granite from „Andrzej" open pit in Żarów (22 ppm) and in both residual (sample *3Ż*) and sedimentary (sample *6Ż*) kaolins (Tab. 2). Besides, moderate mobility

Table 2

Lithium content (ppm) in Lower Silesian kaolins and in separated <40 μm fractions

Region	Deposit	Sample symbol	Raw kaolin	<40 μm fraction
Świdnica	"Andrzej" in Żarów	*3Ż*	27	38
	"Andrzej" in Żarów	*6Ż*	21	40
	"Antoni" in Kalno	*K-1*	39	88
	"Stefan" in Bolesławice	*B-4*	22	53
	Gołaszyce	*G41820*	26	30
	Gola Świdnicka	*Gl 2*	4	6
	Wirki	*2 Wir*	5	6
Strzelin	Wyszonowice	*7W*	11	12
	Wyszonowice	*10W*	12	12
	Wyszonowice	*31W*	2	5
	Jegłowa	*KJ*	7	7
Izera	Kamień	*1494*	29	27
Wądroże Wielkie	Wądroże Wielkie	*WW1*	21	28
	Wądroże Wielkie	*XV/4/1*	3	4
Bolesławiec	"Maria III" in Nowogrodziec*	*III*	9	41

* Kaolinite sandstone deposit.

of this trace alkali element is evidenced by higher Li contents in kaolins formed from granitoids containing lithium rich biotite. So e.g., the everage Li content in biotite from Gołaszyce granite calculated according to analyses of Zawidzki (1976) is very high and contains 675 ppm. Similarly, its content in kaolin from this locality is also relatively high – 26 ppm. On the other hand, biotite from Gola Świdnicka granodiorite contains less lithium i.e. 450 ppm (Zawidzki 1976), similarly as kaolin formed, most probably, from this rock (4 ppm). Contrary to kaolins originated from two-mica granites, sedimentary kaolin from Gola Świdnicka and from Wirki

Table 3

Lithium content (ppm) in selected Lower Silesian kaolins and in separated <40 μm fractions after preliminary 9% HCl treatment

Deposit	Sample symbol	Raw kaolin	<40 μm fraction
"Andrzej" in Żarów	*3Ż*	26	38
"Andrzej" in Żarów	*6Ż*	21	39
Gołaszyce	*G41820*	25	30
Wirki	*2 Wir*	5	5
Wyszonowice	*31W*	2	5
Jegłowa	*KJ*	7	6
Kamień	*1494*	28	27

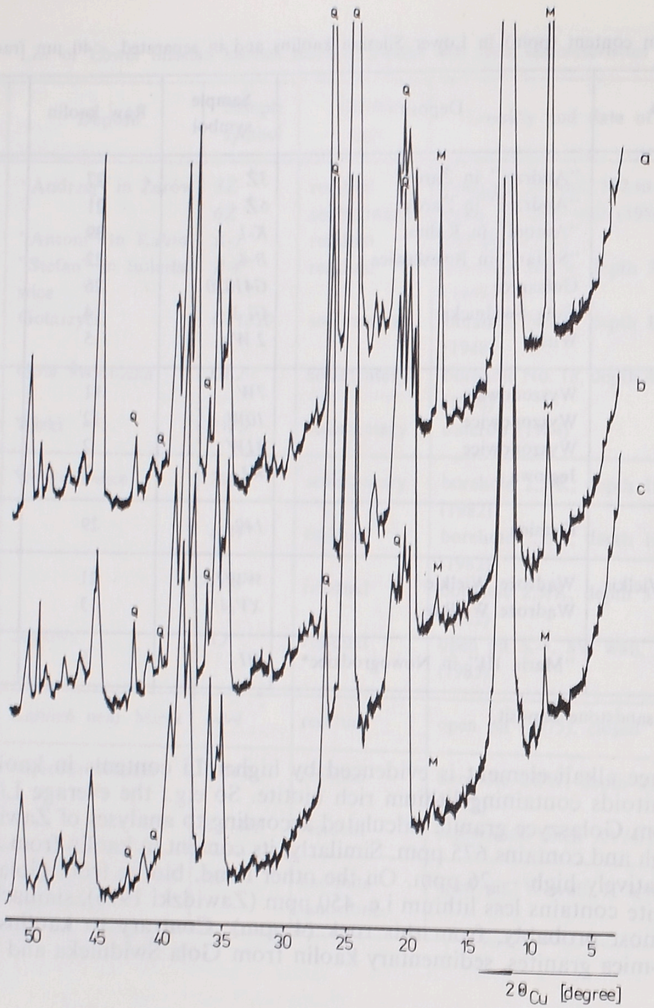

Fig. 2. Typical *X*-ray patterns of size fractions below 40 μm separated from: *a* – residual kaolin from "Antoni" deposit in Kalno (sample *K-1*), *b* – residual kaolin from Wyszonowice deposit (sample *31W*), *c* – sedimentary kaolin from Golaszyce deposit (sample *G41820*). Symbols: M – mica, Q – quartz, other not marked reflexions are of kaolinite

are very low in lithium (Tab. 2). This can be connected with the different type of parent rocks (so called Gola Świdnicka and Strzeblów granite type, respectively) (Gawroński 1982).

Kaolins of the Strzelin region (Wyszonowice, Jegłowa) contain small amounts of lithium (Tab. 2). In the former locality it is related with chemical composition of biotite, from which this kaolinite was mostly formed (Wyszomirski *et al.* 1987). These micas are considerably lower in Li when compared with those from two-mica granites of the Strzegom – Sobótka massif (Gadomski 1968). This is due to migration ability of this trace element during metamorphic processes. Variable amounts of lithium in kaolins from the borehole Wyszonowice 2.5/K (Tab. 2) are, probably, connected with changes in their mineral composition. So e.g., sample *31W*, containing no micas (according to *X*-ray data) is the lowest in lithium in comparison with the other samples.

Kaolins from Jegłowa, which are considered to be formed by hydrothermal alteration of sericite schists (Wiewióra 1973) are similarly low in Li (7 ppm) as their parent rocks (8 ppm).

Kaolins from the environs of Wądroże Wielkie are genetically related with granite-gneisses occurring there. Kaolinization of parent rocks is partical (sample *WW1*) what is confirmed by the presence of feldspar relicts or complete (sample *XV/4/1*). The content of Li in the both samples fairly diversified, whereby the lowest concentration are characteristic of the latter (Tab. 2).

Specific type of clay sediment occurs in Maria III open pit in Nowogrodziec near Bolesławiec. These are kaolinite sandstones, being actually the only raw material in Poland used for the production of washed kaolin. Lithium content in this deposit, determined for comparison purposes, amounts to 9 ppm.

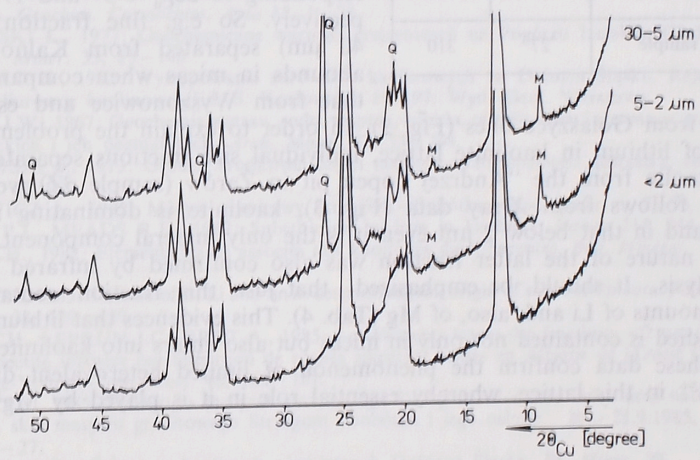

Fig. 3. *X*-ray patterns of size fractions separated from residual kaolin from "Andrzej" open pit in Żarów (sample *3Ż*)
Symbols: M – mica, Q – quartz, other not marked reflexions are of kaolinite

In the majority of analyzed samples we observe an increase of Li content in finer fractions of washed kaolins (Tab. 2). This phenomenon is much more distinct in typical residual kaolins (samples *3Ż*, *K-1*, *31W*) than in sedimentary ones (samples *G41820*, *Gl 2*, *2 Wir*). This is due, first of all, to removal of Li-poor quartz, accompanied by relative increase of kaolinite and fine-grained micas contents. So e.g. in washed kaolin from Maria III kaolinite sandstone, the amount of Li is 4.5 times higher than in primary rocks. On the other hand, no such enrichment is observed in finer fractions of washed kaolin from Kamień near Mirsk (Tab. 2). The raw kaolin from this deposit is relatively rich in light mica. Greisens, parent rocks of these kaolins, contain Li-enriched muscovite (Karwowski 1977). Consequently, we observe no increase of Li content in finer size fractions after washing of raw kaolin from Kamień.

Lithium contents were also determined in selected samples of raw and washed kaolins after preliminary treatment with boiling 9% HCl for 15 min. The obtained results (Tab. 3) do not differ – within analytical error – from those for primary material. This evidences that lithium in these rocks is not connected with easily soluble, poorly crystalline iron oxides and hydroxides, often sorbed onto the surfaces of clay minerals. It is, thus, supposed that Li occurs not only in micas but also in kaolinite lattice.

Table 4

Lithium and magnesium content (ppm) in size fractions separated from residual kaolin ("Andrzej" deposit in Żarów – sample *3Ż*)

Fraction	Content (ppm)	
	Li	Mg
30–5 μm	34	560
5–2 μm	39	760
<2 μm	56	780
Raw sample	27	310

The content of micas, considered to be the main Li-bearing minerals, is very variable in washed kaolins. It follows from *X*-ray data (Fig. 2), since their content can be evaluated on the ground of intensities of *(002)* and *(004)* reflexions, corresponding to $2\theta_{Cu} \approx 9°$ and 17.8°, respectively. So e.g. fine fraction (below 40 μm) separated from Kalno kaolin abounds in micas when compared with that from Wyszonowice and especially with that from Gołaszyce ones (Fig. 2). In order to explain the problem of the presence of lithium in kaolinite lattice, individual size fractions separated from residual kaolin from the "Andrzej" open pit in Żarów (sample *3Ż*) were analyzed. As follows from *X*-ray data (Fig. 3), kaolinite is dominating in finer fractions and in that below 2 μm even it is the only mineral component. Purely kaolinitic nature of the latter fraction was also confirmed by infrared absorption analysis. It should be emphasized that just this fraction contains the highest amounts of Li and, also, of Mg (Tab. 4). This evidences that lithium in the kaolin studied is contained not only in micas but also enters into kaolinite lattice. Besides, these data confirm the phenomenon of limited heterovalent diadochy $Me^{2+} \rightleftharpoons Al^{3+}$ in this lattice, whereby essential role in it is played by Mg^{2+} ions.

CONCLUSIONS

– Lithium content in Lower Silesian kaolins is fairly diversified. In raw materials it ranges from 2 ppm (residual kaolin from Wyszonowice) up to 39 ppm (residual kaolin from Kalno).

– Kaolins formed from greisens and granitoids subjected to late- and post-magmatic alteration are enriched in Li. This phenomenon is exemplified by kaolin from Kamień near Mirsk (Izera region) and by kaolins from the environs of Żarów (Świdnica region).

– Washing process of kaolins generally results in an increase of Li content in separated finer fractions below 40 μm. This is due to removal of quartz and relative increase of kaolinite and fine-grained micas content.

– Mineralogical and geochemical studies of very fine (below 2 μm) purely kaolinitic fraction of residual kaolin from Żarów confirmed the opinion on the presence of Li in kaolinite lattice. This is connected with heterovalent diadochy $Me^{2+} \rightleftharpoons Al^{3+}$ resulting in compensation of electrostatic charge of this lattice.

Acknowledgments. These studies were carried out partially within research programme MR-I-33. The present author would like to express his gratitude to Professor Dr. W. Narębski (Museum of the Earth, Polish Academy of Sciences) for critical reading the manuscript. He is also very obliged to Mrs. H. Kasprzak (Research Unit of the China Ware Factory, Wałbrzych) for carrying out chemical analyses.

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LIT W KAOLINACH DOLNEGO ŚLĄSKA

Streszczenie

Zawartość litu w kaolinach dolnośląskich jest zróżnicowana i waha się w materiale surowym od 2 ppm (kaolin pierwotny z Wyszonowic) do 39 ppm (kaolin pierwotny z Kalna). Najwyższą zawartością tego pierwiastka wyróżniają się kaoliny, które powstały w wyniku wietrzenia grejzenów (Kamień k/Mirska) a także granitoidów zmienionych na etapie późno- i pomagmowym (okolice Żarowa w rejonie świdnickim – Żarów, Kalno, Gołaszyce, Bolesławice).

Proces szlamowania kaolinów prowadzi najczęściej do wzrostu zawartości litu w wydzielonych w ten sposób frakcjach o uziarnieniu poniżej 40 µm. Wiąże się to z usunięciem ubogiego z reguły w ten pierwiastek kwarcu oraz względnym wzrostem zawartości kaolinitu i drobnoziarnistych mik.

Badania czysto kaolinitowej frakcji o uziarnieniu poniżej 2 µm – przeprowadzone na przykładzie kaolinu pierwotnego z Żarowa – potwierdziły pogląd, że lit może wchodzić do struktury kaolinitu. Związane jest to z heterowalentną diadochią $Me^{2+} \rightleftharpoons Al^{3+}$ i wynikającą stąd koniecznością kompensacji ładunku elektrostatycznego tego minerału.

OBJAŚNIENIA FIGUR

Fig. 1. Lokalizacja miejsc występowania badanych kaolinów dolnośląskich

Fig. 2. Przykładowe dyfraktogramy frakcji ziarnowych o uziarnieniu <40 µm wydzielonych z: *a* – kaolinu pierwotnego ze złoża "Antoni" w Kalnie (próbka *K-1*), *b* – kaolinu pierwotnego z Wyszonowic (próbka *31W*), *c* – kaolinu wtórnego z Gołaszyc (próbka *G41820*). Stosowane oznaczenia: M – mika, Q – kwarc (nieoznaczone refleksy pochodzą od kaolinitu)

Fig. 3. Dyfraktogramy frakcji ziarnowych wydzielonych z kaolinu pierwotnego z kop. „Andrzej" w Żarowie (próbka *3Ż*)
Stosowane oznaczenia: M – mika, Q – kwarc (nieoznaczone refleksy pochodzą od kaolinitu)

OBJAŚNIENIA TABEL

Tab. 1. Zestawienie badanych próbek kaolinów dolnośląskich i ich charakterystyka

Tab. 2. Zawartość litu (ppm) w kaolinach dolnośląskich i wydzielonych z nich frakcjach o uziarnieniu <40 µm

Tab. 3. Zawartość litu (ppm) w wybranych kaolinach dolnośląskich i wydzielonych z nich frakcjach o uziarnieniu <40 µm po obróbce chemicznej 9%-owym roztworem HCl

Tab. 4. Zawartość litu i magnezu we frakcjach ziarnowych wydzielonych z kaolinu pierwotnego z kop. „Andrzej" w Żarowie (próbka *3Ż*)

Петр ВЫШОМИРСКИ

ЛИТИЙ В КАОЛИНАХ НИЖНЕЙ СИЛЕЗИИ

Резюме

Содержание лития в нижнесилезских каолинах непостоянно и колеблется в сыром материале от 2 г/т (первичный каолин из Вышоновиц) до 39 г/т (первичный каолин из Кальна). Максимальным содержанием этого элемента отличаются каолины, которые образовались в результате выветривания грейзенов (Камень возле Мирска), а также гранитоидов, измененных в поздне- и послемагматических этапах (окрестности Жарова в свидницком районе — Жаров, Кально, Голашице, Болеславице).

Процесс отмучивания каолинов чаще всего ведет к увеличению содержания лития в выделенных фракциях зернистостью ниже 40 мкм. Это связано с удалением как правило бедного этим элементом кварца, а также относительным увеличением содержания каолинита и мелкозернистых слюд.

Исследования чисто каолинитовой фракции зернистостью ниже 2 мкм, проведенные на примере первичного каолина из Жарова, свидетельствует о возможном вхождении лития в структуру каолинита. Это связано с гетеровалентным изоморфизмом $Me^{2+} \rightleftharpoons Al^{3+}$, и следующей необходимости компенсации электростатического заряда структуры этого минерала.

ОБЪЯСНЕНИЯ К ФИГУРАМ

Фиг. 1. Местоположение изучаемых нижнесилезских каолинов

Фиг. 2. Некоторые дифрактограммы гранулометрических фракций <40 мкм, выделенных из: *а* — первичного каолина из месторождения „Антони" в Кальне (образец *K-1*), *b* — первичного каолина с Вышоновиц (образец *31W*), *c* — вторичного каолина из Голашиц (образец *G41820*). Применяемые обозначения: М — слюда, Q — кварц (не определенные отражения происходят от каолинита)

Фиг. 3. Дифрактограммы гранулометрических фракций, выделенных из первичного каолина из карвера „Анджей" в Жарове (образец *3Ż*)
Применяемые обозначения: М — слюда, Q — кварц (не определенные отражения происходят от каолинита).

ОГЛАВЛЕНИЕ ТАБЛИЦ

Табл. 1. Сводная характеристика изучаемых образцов нижнесилезских каолинов

Табл. 2. Содержание лития (г/т) в нижнесилезских каолинах и выделенных из них фракциях зернистости <40 мкм

Табл. 3. Содержание лития (г/т) в некоторых нижнесилезских каолинах и выделенных из них фракциях зернистости <40 мкм после химической обработки 9% раствором HCl

Табл. 4. Содержание лития и магния в гранулометрических фракциях, выделенных из первичного каолина из карьера „Анджей" в Жарове (образец *3Ż*)