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WATER-ROCK INTERACTION IN SOME MINERAL WATERS IN THE SUDETES, POLAND: IMPLICATIONS FOR CHEMICAL GEOTHERMOMETRY

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Abstract: So-called chemical geothermometers influenced greatly estimation of groundwater temperatures. However, it has been shown that in low enthalpy waters in the Sudetes geothermometers have a limited application. We found that for the available chemical analyses with field pH and the carefully chosen set of thermodynamical data practically all mineral waters (thermal and carbonated) are far from overall equilibrium with minerals at any temperature. Only exceptionally could a particular mineral pair be candidate for a geothermometer in the Sudetes. We found that the reasonable range of the expected water temperatures in the Sudetes is 50° - 95°C in waters in Duszniki (Pieniawa Chopina) and 60° - 70°C in Cieplice (Basenowe springs and C-2 borehole, which is quite close to the present output temperature of the waters.

Key words: geothermometry, thermal waters, equilibration, Sudetes

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INTRODUCTION

Estimation of the temperature of the ground waters that could be made available at the surface was always an important asset in geothermal prospecting. The so-called chemical geothermometers are the formulas expressing functional temperature dependency versus chemical composition. They are widely known since the classical work of Fournier & Truesdell (1973). Chemical geothermometer results from either calibration of a mineral pair within the range of temperatures or simply the best fit of specific cation ratios versus temperatures. In geothermal areas their use is quite successful, however in low enthalpy waters their estimations are often dubious.

The usefulness of chemical geothermometers can be simply tested by comparison of the solubility product with the actual activity product resulting from chemical analyses. Then construction of saturation diagrams over the range of temperatures allows to estimate the temperature of an underground reservoir. The method was originally proposed by Michard & Roekens (1983) and Reed & Spycher (1984).

Table 1

Log K(T) of the minerals used for the calculations

Temperature	25°	60°	80°	100°	125°	150°
Minerals						
Low albite	-20.21	-18.19	-17.27	-16.48	-15.69	-15.07
Microcline	-23.3	-20.78	-19.60	-18.60	-17.56	-16.73
Muscovite	-27.51	-2409	-22.48	-21.15	-19.88	-18.99
Anorthite	20.08	15.13	12.73	10.60	8.23	6.15
Kaolinite	-36.92	-33.14	-31.31	-29.68	-27.87	-26.28
Sepiolite	-40.08	-38.04	-37.05	-36.17	-35.19	-34.33
Analcime	-16.00	-14.48	-13.78	-13.20	-12.61	-12.17
Laumontite	-32,30	-2949	-28.23	-27.18	-26.15	-25.40
Adularia	-21.3	-19.09	-18.08	-17.22	-16.35	-15.66
Ca-montm.	-111.46	-97.64	-91.00	-85.56	-80.39	-76.83
Mg-montm.	-112.1	-98.56	-92.05	-86.71	-81.64	-78.17
Na-montm.	-55.61	-48.17	-44.60	-41.65	-38.81	-36.81
K-montm.	-56.06	-48.82	-45.34	-42.46	-39.70	-37.75
Mg-chlorite	-85.2	-91.85	-92.05	-86.71	-81.64	-78.18
Chalcedony	-3.73	-3.19	-3.01	-2.84	-2.65	-2.49
Prehnite	-41.2	-38.63	-37.57	-36.78	-36.13	-35.83
Illite	-40.3	-36.10	-39.78	-39.63	-39.45	-39.30
Gypsum	-4.60	-4.58	-4.57	-4.56	-4.55	-4.54
Diopside	-36.22	-34.60	-33.81	-33.11	-32.34	-31.65
Calcite	-8.48	-8.76	-8.99	-9.27	-9.68	-10.16
Fluorite	-10.96	-10.70	-10.58	-10.53	-10.52	-10.55
Quartz	-3.99	-3.48	-3.25	-3.06	-2.84	-2.66

Dissolution of minerals is written in terms of H_4SiO_4, H^+, H_2O, Na^+, K^+, Ca^{2+}, Mg^{2+}, $Al(OH)_4^-$ species. Temperature is given in degree centigrades. Abbreviation "montm" designates montmorillonite

Theoretically, the temperature of the underground reservoir is unequivocally determined if several curves of the saturation indices of the minerals cross SI = 0 axis (SI = IAP/K, where: SI – saturation index, IAP – ion activity product, K – thermodynamic stability constant for mineral as specified in

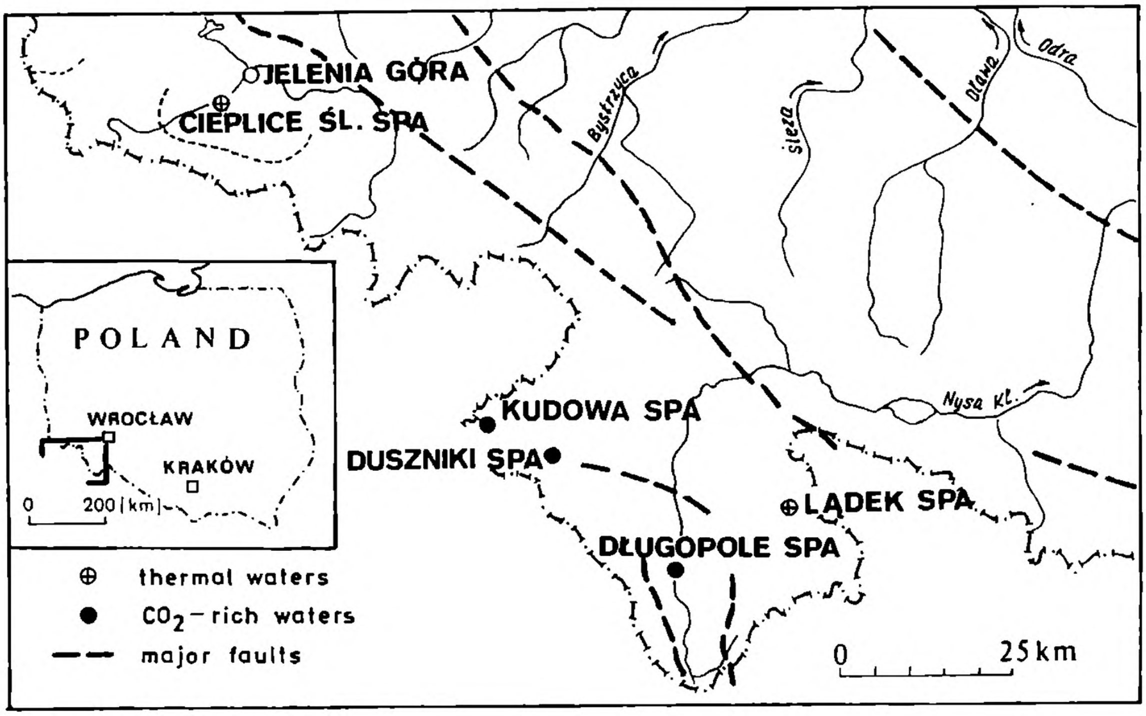


Fig. 1 Locations of mineral waters of interest in Sudetes

Tab. 1.) at the same temperature. This is the case of the high temperature waters, when nearly all kinetic barriers to the equilibrium are overcome. The main advantage of this method over the conventional geothermometers is that no assumption of particular mineral equilibrium is required, while for example in applying Na/K geothermometer an assumption about equilibrium of phases containing Na and K with water is made, sometimes implicitly. In the former case, however, the pH values and chemical analyses of water with reliable total ionic aluminium concentrations are needed.

OCCURRENCE OF MINERAL WATERS

Thermal waters in the Sudetes Mountains are known at Cieplice (Karkonosze-Izera Block) and Lądek (Lądek-Śnieżnik Massif). At Cieplice, thermal waters occur within fissured Karkonosze granite (Upper Carboniferous) covered by several meters thick Quaternary deposits. The rock-forming minerals of the Karkonosze granite are: plagioclase, orthoclase and quartz (Borkowska, 1966). In the search for thermal waters in the Cieplice bore-hole C-2 (Fig. 1) penetrating granite until the final depth of about 750 m, muscovite, microcline, chalcedony and calcite have been reported. The main zones of water circulation seem to be tectonically disturbed areas of NW-SE and NE-SW directions, though the problem is still not solved (Fistek & Tęsiorowska, 1973; Ciężkowski *et al.*, 1992).

In this region thermal waters discharged by natural springs and boreholes are characterised by a low TDS content and by a considerable amounts of fluoride and silica. They are of SO_4-HCO_3-Na type (Tab.2) and fall in the temperature range 22°C and 58°C (borehole C-2).

Thermal waters at Lądek circulate in the fissured Lower Palaeozoic Gierałtów gneisses. Natural springs are located in the NW part of the Gieraltów synclinorium in an intensively disturbed zone by numerous faults of NW-SE and NE-SW directions. The temperature of spring waters ranges around 20-29°C, while the temperature at the outflow of the borehole L-2 was 45°C. They are slightly mineralised waters of HCO_3-(SO_4)-Na type (Tab. 2).

Table 2

Chemical analyses, pH temperatures and calculated PCO_2 of Sudetic thermal waters

	Cieplice Spa							Lądek Spa	
	Sobieski	C-2	Basen-5	Basen-6	Nowe	Antoni	Mary-sieńka	L-2	Jerzy
T (°C)	23.6	57.9	40.6	39.6	34	17.3	21.5	41.2	28.2
pH	7.72	8.15	8.1	8.2	8.02	7.45	8.4	9.0	9.0
Ca^{+2}	69.9	9.14	10.8	8.03	11.1	30.1	9.94	2.73	4.3
Mg^{+2}	10.7	0.97	1.02	1.02	0.49	4.08	0.73	0.24	0.73
Na^+	120	160	165	165	150	145	163	48.5	45.0
K^+	42.0	6.6	4.6	5	4.7	13.5	4.7	0.75	0.65
Al^{+3}	0.02	0.002	0.003	0.003	0.003	0.003	0.002	0.003	0.003
H_4SiO_4	70.4	134	134	137	128	102	44.8	76.8	60.8
Cl^-	131	42.2	42.2	42.2	40.8	56.4	43.6	3.55	8.86
Alk.	262	166	166	154	154	195	154	72	51.4
SO_4^{-2}	113	164	165	167	154	156	160	22	17.5
F^-	2.1	12.4	12.4	12.4	12	9.8	11	10.8	10.2
PCO_2	$4.4\ 10^{-3}$	$1.8\ 10^{-3}$	$1.6\ 10^{-3}$	$1.7\ 10^{-3}$	$1.6\ 10^{-3}$	$6\ 10^{-3}$	$5.2\ 10^{-4}$	$6.7\ 10^{-5}$	$4.3\ 10^{-5}$

Alk = alkalinity i.e. = $HCO_3^- + 2CO_3^{2-} + OH^- - H^+$
PH values were measured in the field. All concentrations are given in mg/dm^3

Possible existence of thermal waters within other Sudetic crystalline formations, e.g. Duszniki and Kudowa areas has been suggested by Dowgiałło (1987a, b). According to this author, these waters may reach relatively high temperatures during their circulation in the underground, though at the surface they are cold. Low temperature of spring waters is due to their cooling during ascension to the surface and/or to mixing with the cold water of a shallow circulation.

CO_2-rich waters at Duszniki circulate within strongly fissured metamorphic formation of Early Palaeozoic age, consisting of micaschists and paragneisses. The occurrence of waters under consideration is connected with tectonically disturbed zone where a major fault bordering the Intra-Sudetic synclinorium upon Bystrzyckie and Orlickie Mts., is crossed by younger transversal faults. Carbonated waters are discharged by several boreholes at a maximum depth of 165 m. From the chemical point of view the waters belong to the HCO_3-Ca-Na(Mg) type (Tab.3) with a relatively high total mineralization (about 2.5 g/dm^3) and a pressure of CO_2 about 0.1 MPa at the surface.

Cold carbonated waters within the Kudowa syncline occur within the Upper Cretaceous sediments underlain by Permian deposits and Lower Palaeozoic metamorphic formations. The syncline is framed to the east by a granitoid massif of the Carboniferous age. The TDS content of the Kudowa mineral waters varies from 1.3 to 3.5 g/dm^3 with a carbon dioxide pressure of about 0.1 MPa at the surface. They are of HCO_3-Na-Ca and HCO_3-Ca-Na types (Tab.3).

Table 3

Chemical analyses, pH, temperatures of cold, carbonated Sudetic mineral waters

	Kudowa Spa		Duszniki Spa			Długopole Spa	
	K-200	Pieniawa Chopina	Jan. K	B-4	B-3	Kazimierz	Emilia
T (°C)	13.0	18.0	17.1	17.2	13.6	10.0	10.3
pH	5.9	5.75	5.6	5.85	5.75	5.3	5.3
Ca^{+2}	211	228	165	263	189	75.3	31.8
Mg^{+2}	85.7	66.9	50.2	86.0	21.7	60.9	53.3
Na^{+}	530	169	146	205	47	54	67
K^{+}	58.0	80.0	63.5	95.0	26.0	9.5	11.0
Al^{+3}	0.003	0.05	0.04	0.001	0.04	0.08	0.06
H_4SiO_4	92.8	83.1	51.2	89.6	40.0	42.9	49.6
Cl^-	77.2	12.3	11.4	11.4	10.5	15.8	15.8
Alk.	2191	1555	1159	1875	1052	665	841
SO_4^{-2}	243	55.1	64.0	56.8	32.3	25.5	1.77
F^-	0.56	0.42	0.45	0.29	0.27	0.38	0.25

Alk = alkalinity i.e. $+ HCO_3^- + 2CO_3^{2-} + OH^- - H^+$

PH values were measured in the field. All concentrations are given in mg/dm^3

METHODS

Calculations of saturation indices were performed with the computer program PHREEQE (Parkhurst *et al.*, 1980) with a carefully compiled thermodynamic data set: for carbonate species of Plummer & Busenberg (1982), for silicates and $Al(OH)_4^-$ of Arnorsson *et al.* (1982). Other thermodynamic data for minerals are taken from Robie *et al.* (1978). Table 1 contains the stability constants for minerals from 25°C to 150°C. Apart of the thermodynamic data, the most critical parameter involved in the calculations is the total concentration of $Al^{3+}{}_T$. As a free ion Al^{3+} takes part in equilibration of solutions with aluminium silicates. However, available chemical analyses of the waters suffered from the invalid laboratory procedure that consisted of the acidification of the samples without previous filtration. Therefore, all suspended matter was dissolved by acid action and additional Al^{3+} entered the solutions. The difference among some analyses is reaching even four orders of magnitude. In the recent borehole C-2, the reported $Al^{3+}{}_T$ concentration was 3.5 mg/dm^3. If such concentration is used in the calculation the estimated temperatures would reach 200°C. To avoid serious overestimation of the temperatures, the minimum Al^{3+} concentrations reported ever since were used for calculations. Al^{3+} concentration of 0.1mg/dm^3 (Fig. 2 a) results in temperature of equilibration of about 100°-110°C. With Al^{3+} minimum reported concentration (0.002), while other components are constant and the estimated temperature is in the range of 50° to 70°C (Fig. 2 b).

CIEPLICE SPA WATER SYSTEM

Calculated partial pressures of CO_2 in Cieplice are less than in the typical groundwaters in equilibrium with soil CO_2, and in Lądek are even less than in the atmosphere (Tab. 2), which suggests that all waters belong to the chemical water-rock systems closed to the external reservoir of CO_2 (TDC = const). No supply of CO_2 is evident. At the emergence temperature the waters are supersaturated with respect to albite, microcline, muscovite, and several other minerals that perhaps are not present in the thermal water reservoir. Taking into account secondary minerals, smectites represent the group in which saturation indices are apparently exceeded (Figs. 2 to 5).

According to the chemical and isotopic data, the borehole C-2 and the Sobieski thermal spring represent the end-members in Cieplice waters (Ciężkowski *et al.*, 1992). The Sobieski thermal spring is the water mostly mixed by a shallow circulation while the C-2 represents water from deep reservoir, possibly cooled on the route to the surface. It is apparent from Fig. 2B, that the majority of the SI-curves for C-2 cut the SI=O axis between 50° and 75°C. Small difference between temperatures (60°-70°C) at which albite and microcline cut SI=0 axis suggests that the Na/K geotermometer calibrated by Arnorsson *et al.* (1983) could be used with confidence for estimation of the underground reservoir temperature. However, this low albite - K-feldspar geothermometer written as

$$t = 933/(0.933 + \log(Na^{+}/K^{+}) - 273.15$$

gives an equilibration temperature of 104°C. Position of other SI-curves suggests that the above temperature could be regarded as the upper limit.

A spread in the water-silicate equilibration temperature of about 30°C span was found in the low temperature spring waters of Cieplice (Fig. 3 a b, 4 a b, 5 a b). This is probably a consequence of dilution by shallow waters or the effect of differing rates of re-equilibration of minerals with waters that are cooled during their ascent.

The study of SI diagram for the Sobieski spring (Fig. 4 b) indicates that SI-curves are dispersed within the temperature range of 40°-110°C. In this case, contrary to the borehole C-2, albite and K-feldspar do not equilibrate with each other at any temperature. This is the reason why geothermometer based on Na/K ratio gives a meaningless result of 400°C. For the Antoni-Wacław and the Nowe boreholes we obtain very similar SI's versus temperature characteristics, which can be compared to the Basenowe springs and the C-2 borehole, indicating that water equilibrates with potassium and sodium aluminosilicates close to 65°C. The Marysieńka water (Fig. 4a) shows lower temperature though the equilibration curves are still much more consistent with each other than in the Sobieski (Fig. 4 b) spring. Silica phases, quartz and chalcedony are supersatured but show inconsistent temperatures. It is known (Arnorsson, 1975), that quartz does not equilibrate at temperatures lower than 180°C and chalcedony is a much better indicator of lower temperatures. However, in the Cieplice springs temperature of equilibration of chalcedony with water is also inconsistent with temperature displayed by equilibrium of water with alkali silicate phases.

LĄDEK SPA WATER SYSTEM

Lądek thermal waters are generally undersaturated with respect to most minerals (Fig. 6 a b). Any particular mineral pair does not reach equilibrium. Only chalcedony show equilibration temperature of about 95°C, but lack of equilibrium with other silicate phases suggest that chalcedony in Lądek thermal waters is not a good temperature indicator. The non-equilibrium state of waters under consideration suggests that all chemical thermometers, in this case, are likely to fail. Other thermal waters in Lądek have similar SI versus temperature's dependencies and were not included in the discussion.

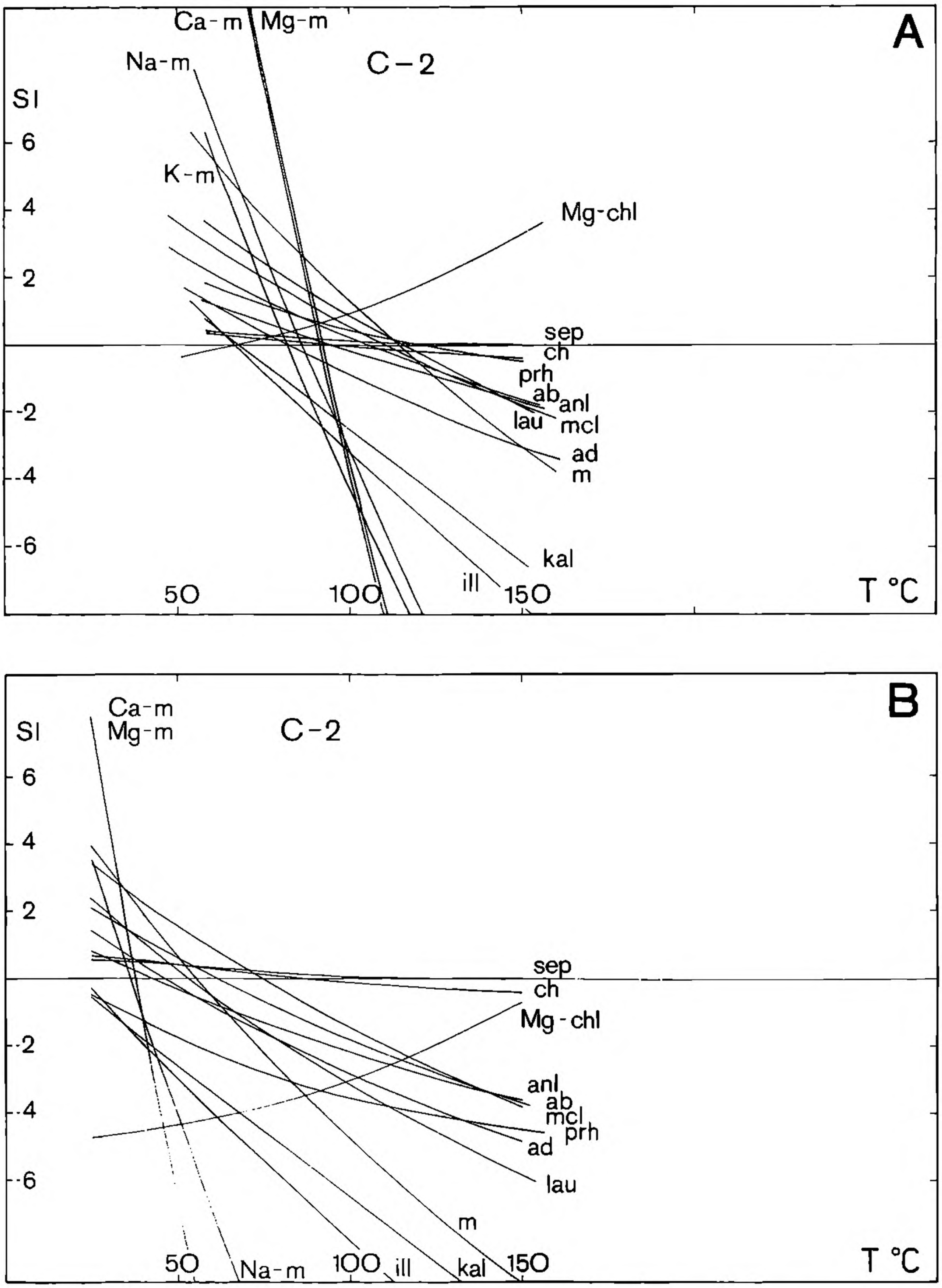


Fig. 2 SI-curves versus temperature for Cieplice waters (C-2), temperature in °C. *ab* – albite; *mcl* – microcline; *m* – muscovite; *an* – anorthite; *kal* – kaolinite; *sep* – sepiolite; *anl* – analcime; *lau* – laumontite; *ad* – adularia; *Ca-m* – Ca-montmorillonite; *Mg-m* – Mg-montmorillonite; *Na-m* – Na-montmorillonite; *K-m* – K-montmorillonite; *Mg-ch* – Mg-chlorite; *ch* – chalcedony; *prh* – prehnite; *ill* – illite; *gyp* – gypsum; *d* – diopside; *c* – calcite; *f* – fluorite; *q* – quartz

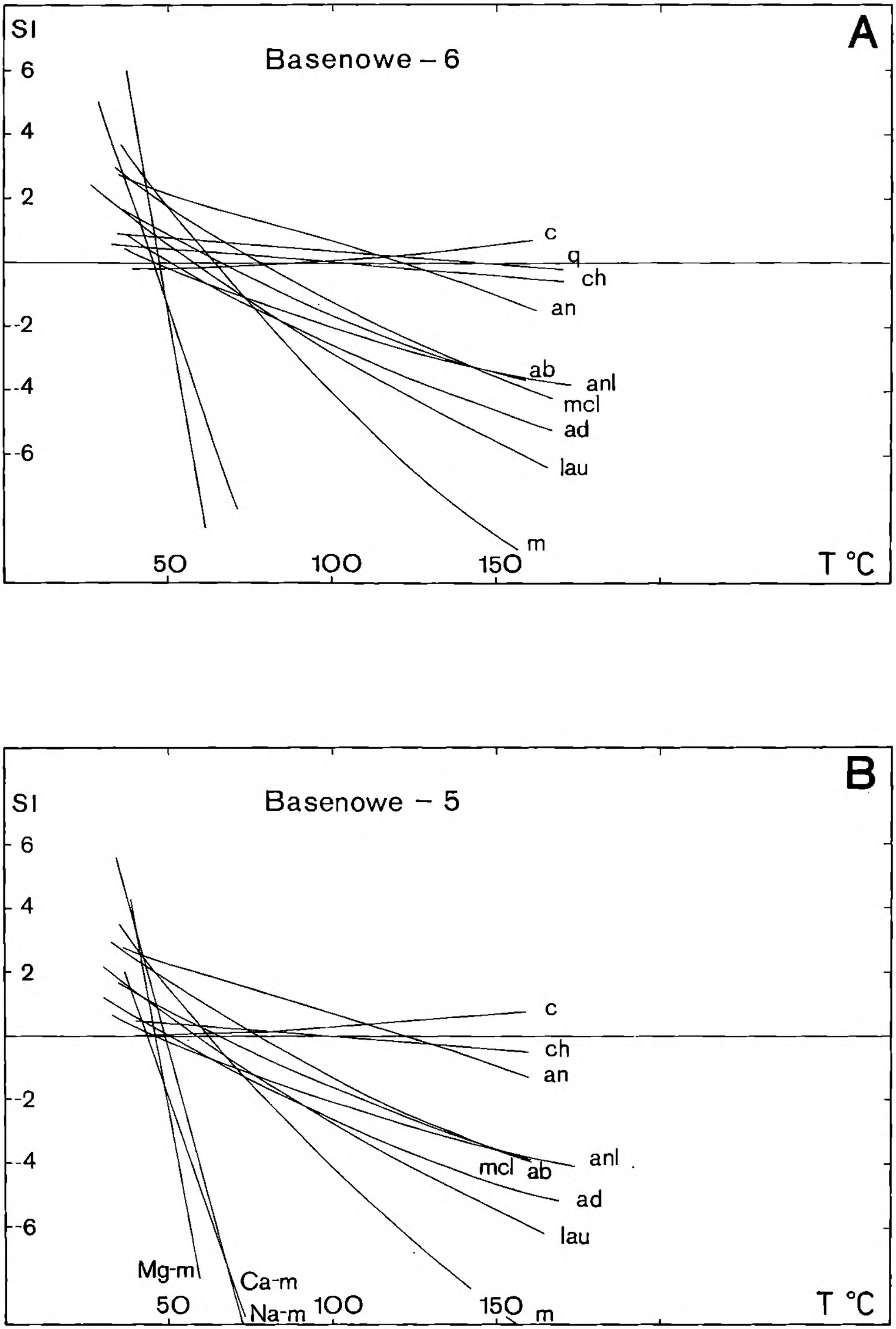
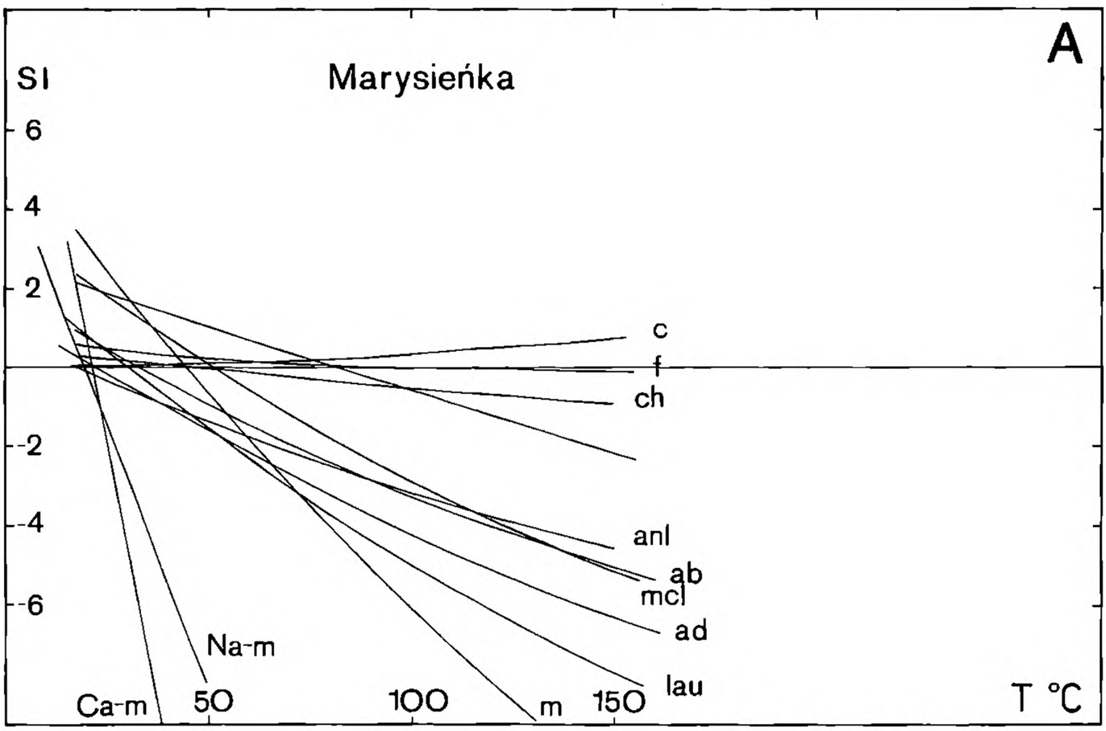


Fig. 3 SI-curves versus temperature for Cieplice waters (Basenowe-6, Basenowe-5), temperature in °C. Abbreviations as in Fig. 2



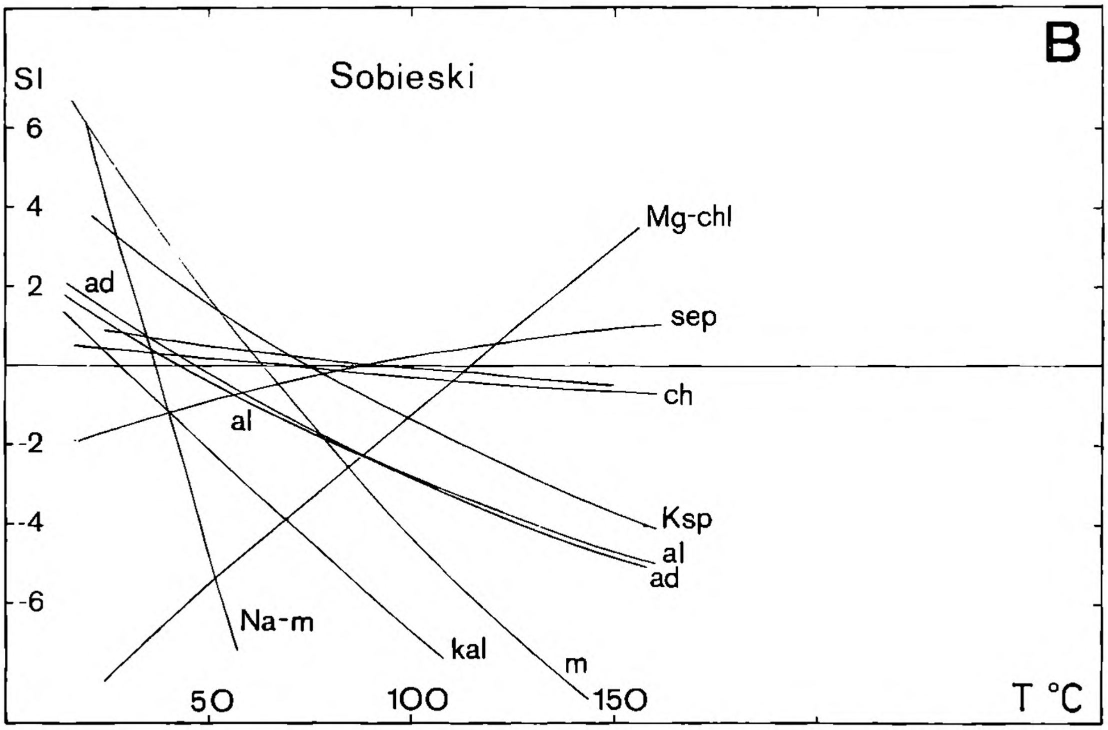
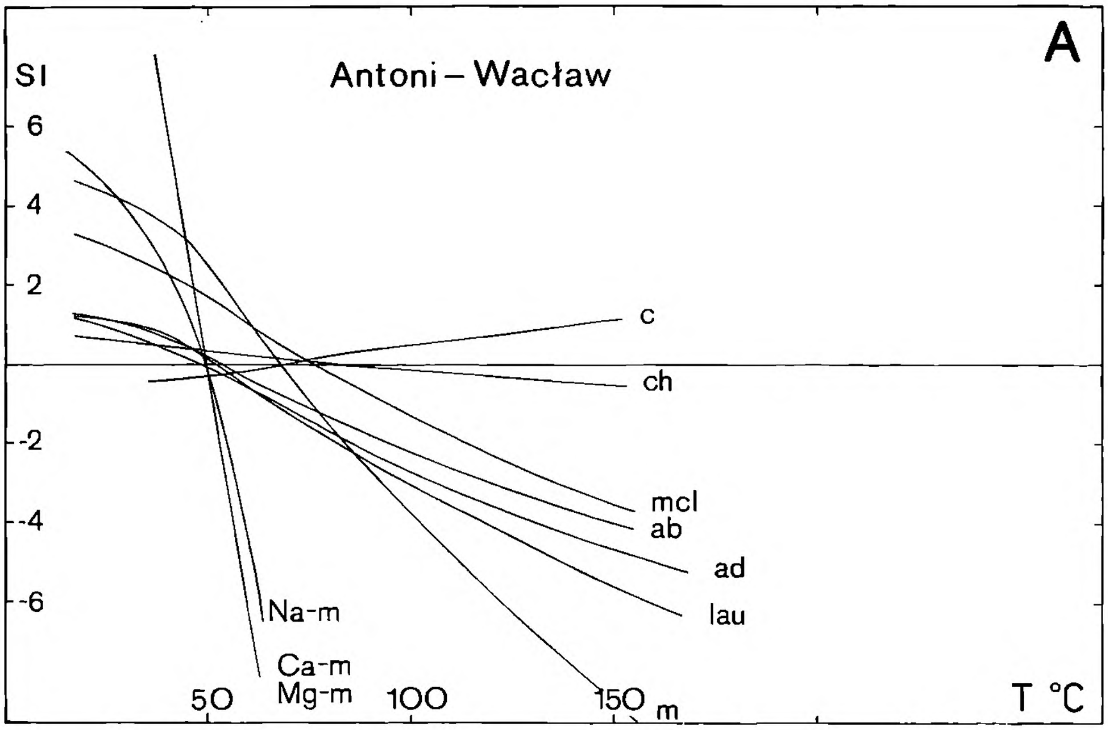


Fig. 4 SI-curves versus temperature for Cieplice waters (Marysieńka, Sobieski), temperature in °C. Abbreviations as in Fig. 2



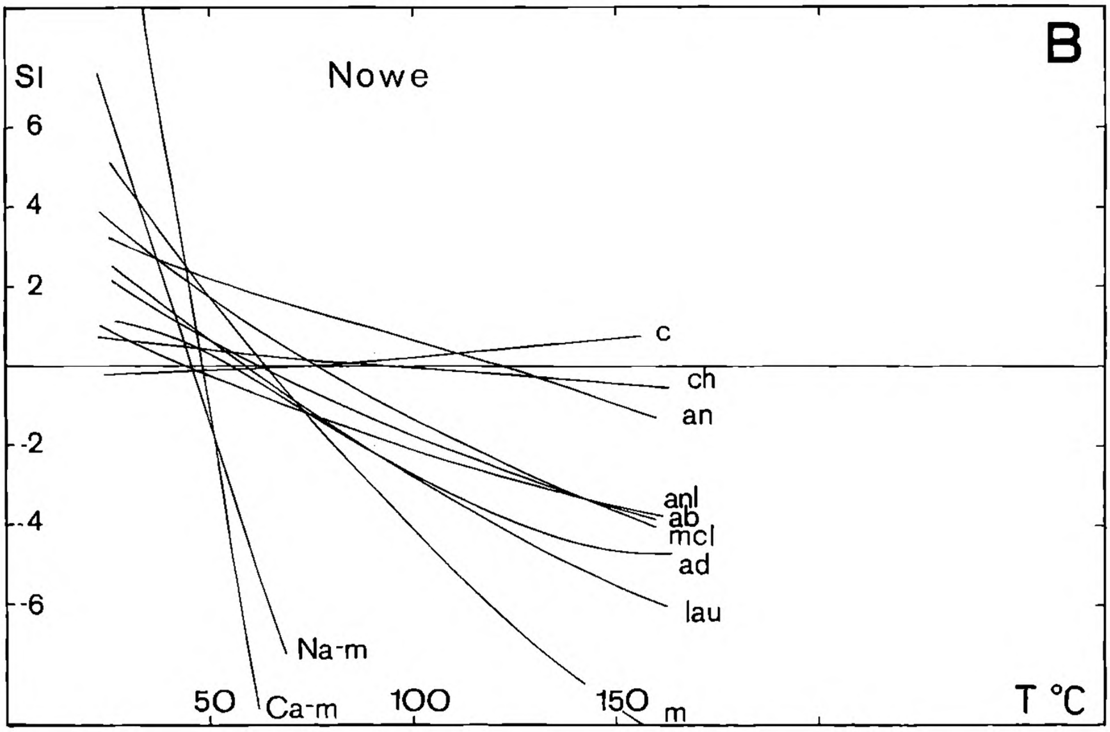
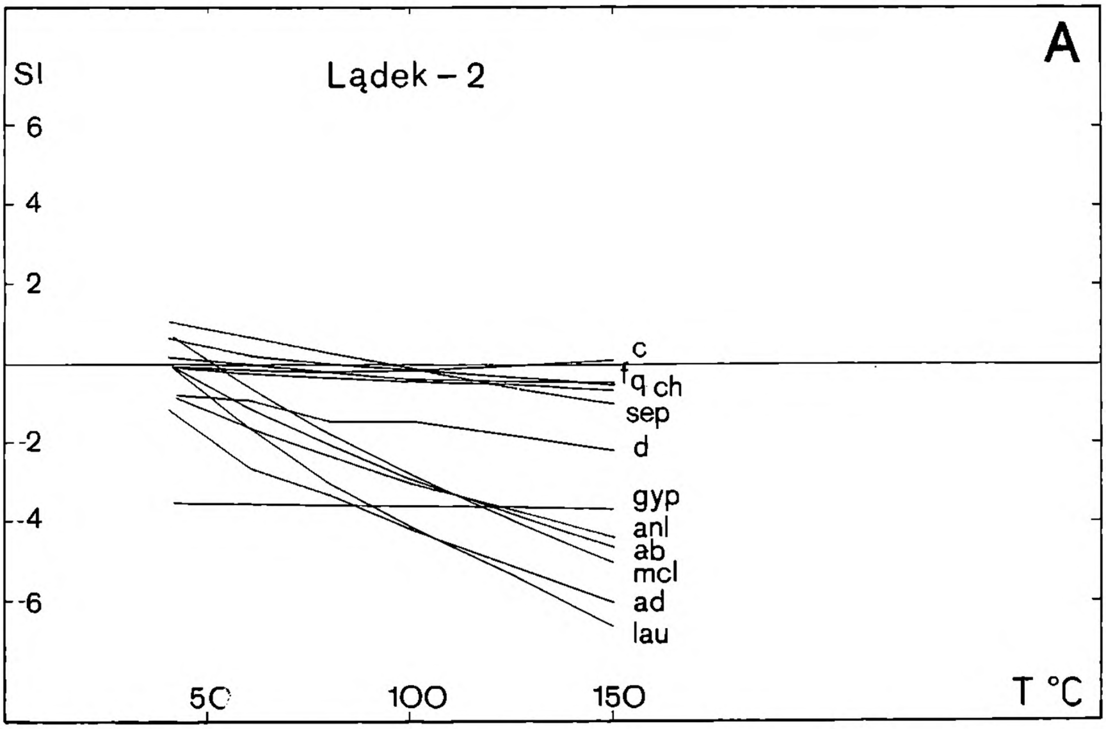


Fig. 5 SI-curves versus temperature for Cieplice waters (Antoni-Wacław, Nowe), temperature in °C. Abbreviations as in Fig. 2



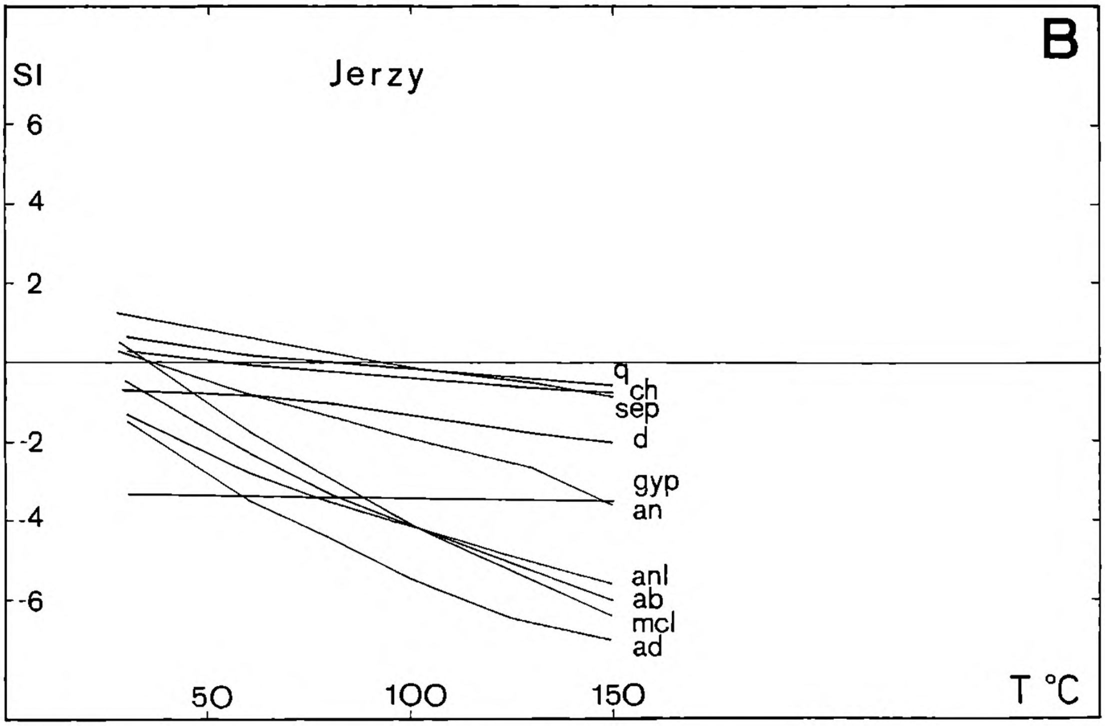


Fig. 6 SI-curves versus temperature for Lądek waters (L-2, Jerzy), temperature in °C. Abbreviations as in Fig. 2

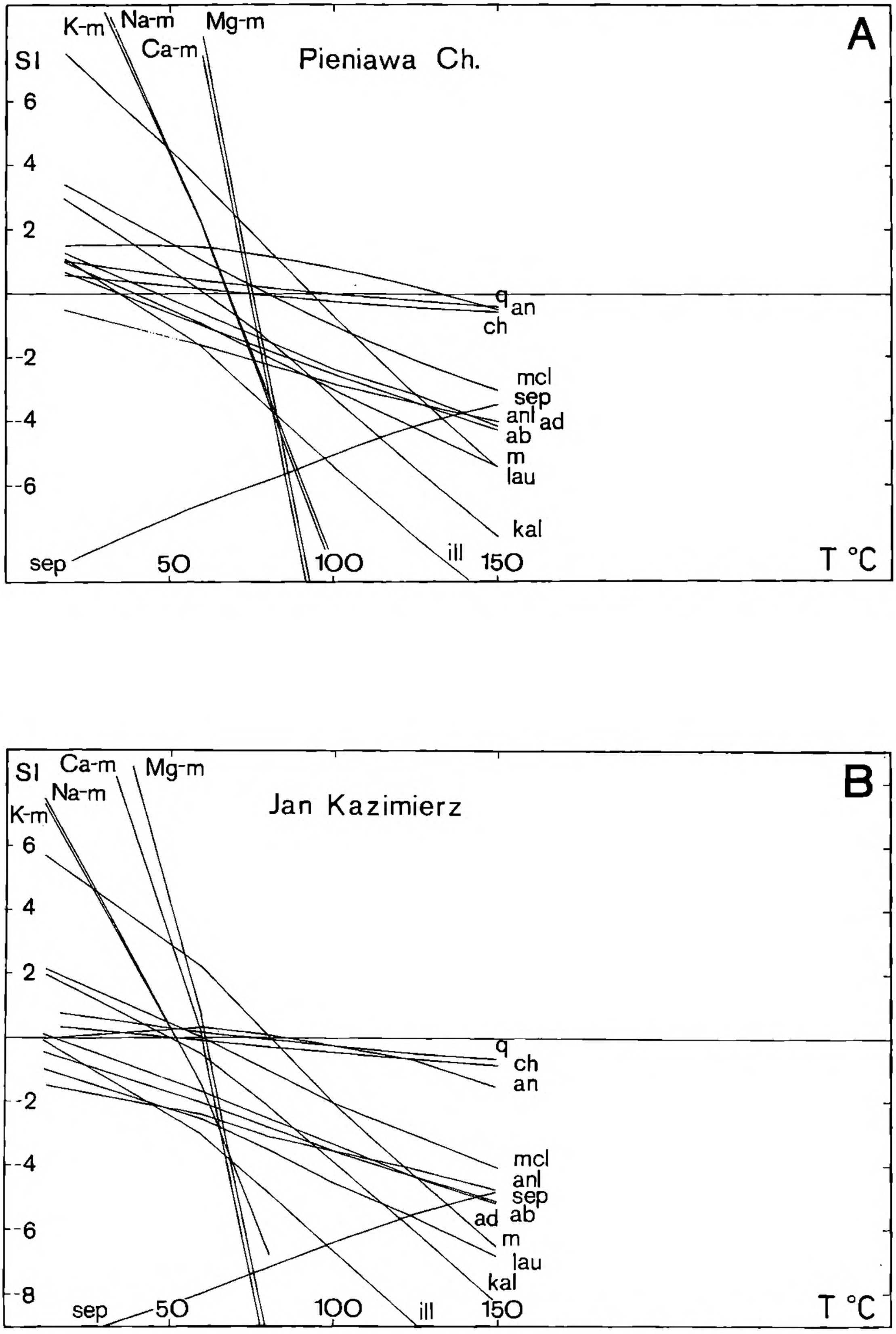


Fig. 7 SI-curves versus temperature for Duszniki waters (Pieniawa Chopina, Kazimierz) temperature in °C. Abbreviations as in Fig. 2

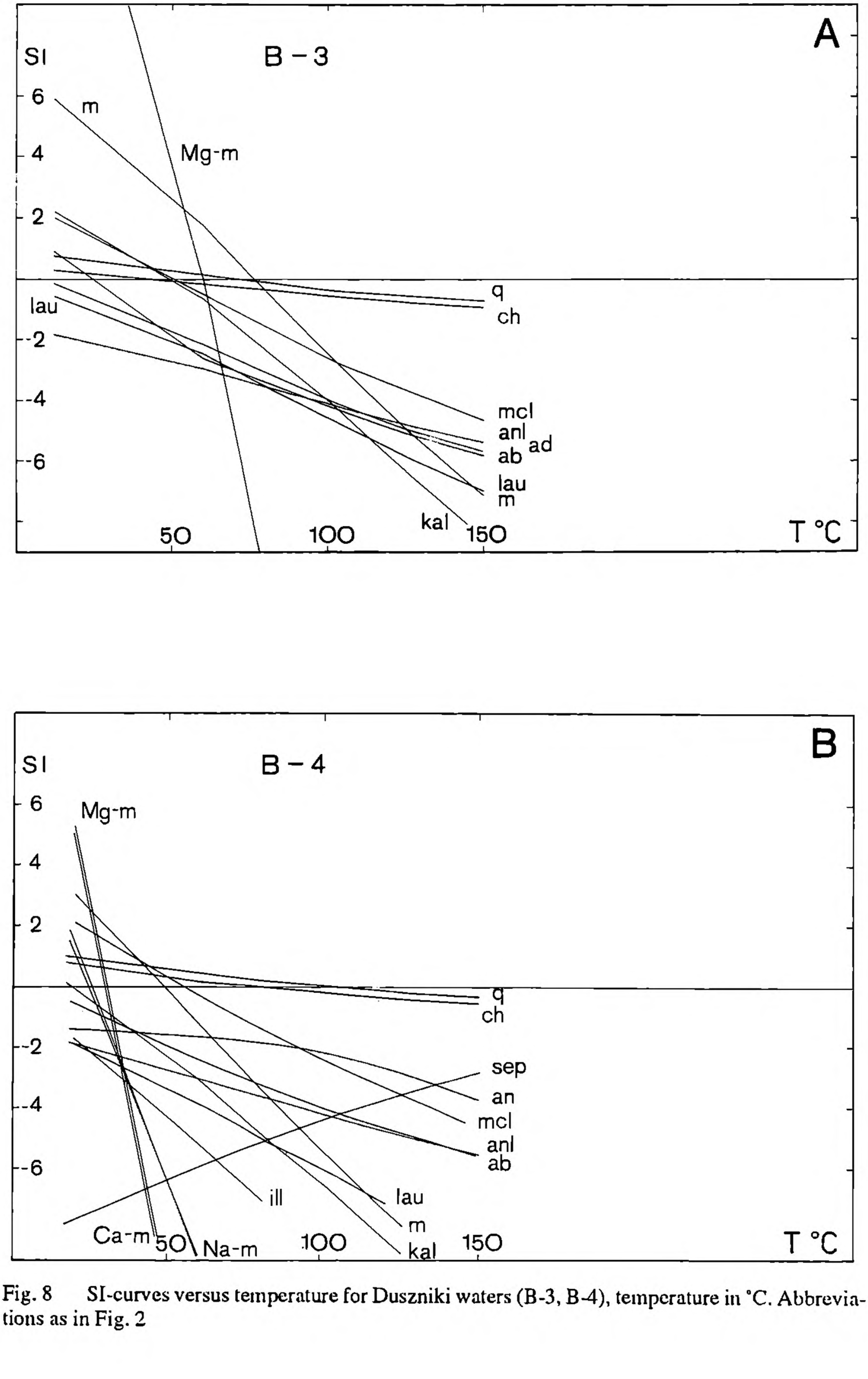


Fig. 8 SI-curves versus temperature for Duszniki waters (B-3, B-4), temperature in °C. Abbreviations as in Fig. 2

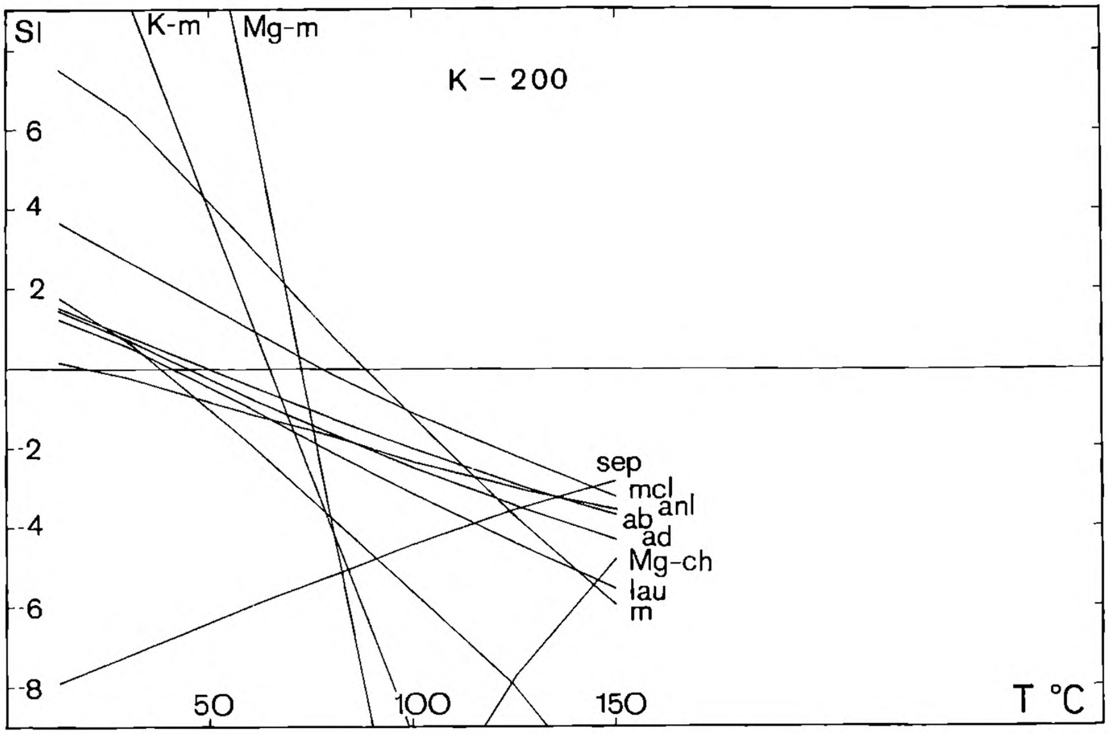


Fig. 9 SI-curves versus temperature for Kudowa waters (K-200), temperature in °C. Abbreviations as in Fig. 2

DUSZNIKI AND KUDOWA SPA WATER SYSTEM(S)

The Duszniki and Kudowa carbonated waters have rather low temperatures at the orifices though in Duszniki the constant temperatures of about 17°C have been reported in several wells. The important difference between the thermal waters of Cieplice and Lądek and CO_2-rich ones as in Duszniki and Kudowa is also related to water-mineral equilibrium. Figs. 7, 8 and 9 show the evolution of saturation indices of some minerals versus temperatures for Duszniki and Kudowa mineral waters. It is apparent from the diagrams that the highest temperatures of equilibration (50°-95°C) are displayed in Pieniawa Chopina. However, even here sodium and potassium silicates phases show temperatures of equilibration below 60°C which indicates the lack of overall equilibrium conditions between water and reservoir minerals in the system. Secondary phases show higher temperature but due to their instability they are not good temperature indicators. The only exception is K-feldspar and Mg-montmorillonite mineral pair that, except for B-4, "equilibrates" in a narrow temperature range. This seems to be rather fortuitous because primary and secondary minerals do not form a reasonable mineral-pair geothermometer. As K^+ and Mg^{2+} ions are concerned K-feldspar-clinochlore geothermometer was proposed by Giggenbach *et al.* (1983) and used again in Giggenbach (1988).

However, in our plots SI-curves fall far from each other at any reasonable temperature range. Therefore any application of this geothermometer makes no sense.

DŁUGOPOLE SPA WATER SYSTEM

These are CO_2-rich waters at a low pH of about 5-5.5 and apparently undersaturated with respect to most of the minerals. They seem to represent the first stage of evolution of CO_2-rich waters, where the only supersaturated phase is chalcedony. Chalcedony crosses SI=0 axis at about 56°C indicating the upper limit of reservoir temperature. These springs belong to the low pH category of springs with shallow circulation as evidenced by tritium data (Ciężkowski *et al.*,1986), which has never equilibrated with silicate minerals. Therefore geothermetry is hardly applicable to these waters. Consequently no figures are shown for Długopole.

CONCLUSIONS

Plots of saturation indices versus temperature impose several constraints on the use of geothermometers in the Sudetic thermal waters. In case they equilibrate with several minerals, they are also subject to mixing processes or re-equilibration at lower temperature with secondary minerals. Therefore, overall equilibrium with many mineral phases cannot be expected. The potential use of chemical geothermometers for the Sudetic thermal waters needs a detailed study on particular mineral pairs as for instance K-feldspar and clinochlore.

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Streszczenie

ODDZIAŁYWANIE WODA-SKAŁA W NIEKTÓRYCH WODACH MINERALNYCH SUDETÓW: ZNACZENIE DLA GEOTERMOMETRII CHEMICZNEJ

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Tak zwane geotermometry wywarły duży wpływ na szacowanie temperatur wód podziemnych. Jednakże jak pokazaliśmy, w wodach o niskiej entalpii występujących w Sudetach, geotermometry mają ograniczoną stosowalność. Stwierdzono, na podstawie dostępnych analiz i pomiarów pH wykonanych w terenie, wraz ze starannie wyselekcjonowanymi danymi termodynamicznymi, że praktycznie wszystkie wody są dalekie od stanu całkowitej równowagi z minerałami w każdej temperaturze. Zupełnie wyjątkowo para mineralna może być kandydatem na geotermometr w wodach mineralnych w Sudetach. Stwierdzono, że zasadnym zakresem spodziewanych temperatur jest 50°-95°C w wodach węglanowych w Dusznikach (Pieniawa Chopina) i 60°-70°C w

Cieplicach (ujścia Basenowe i odwiert C-2), który to zakres jest bliski aktualnej temperaturze na wypływie (C-2).