

PARALAVA FROM COAL-DUMP COMBUSTION IN UPPER SILESIA, POLAND: MELT SEPARATION LEAVING A CORDIERITE-RICH RESTITE.

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Abstract

In this article, we present the results of the investigations into peculiar cordierite (indialite)-rich concentrations found in paralava from two coal dumps from the Upper Silesia Coal Basin, Poland. The temperature of the fusion process was in the range of 900–1100 °C and the main product is a ryolitic, peraluminous paralava. The crystallization and separation of cordierite (indialite)-rich restite from glassy paralava was probably controlled by changing oxygen fugacity and high viscosity of the fused rocks. The resorption of monazite and associated mobilization of phosphorus to form Fe–Mg–Ca phosphates and P-enrichment of the glassy paralava, pose questions about the behavior of REE and Th released from monazite during the fusion caused by coal combustion.

Keywords: paralava, cordierite, restite, oxygen fugacity

1 INTRODUCTION

Combustion pyrometamorphism is a common result of spontaneous and in many cases uncontrolled burning of coal-seams or coal-bearing waste-dumps at various scales. Backing and fusion of rocks occurs usually in the temperatures above 700 °C up to 1400 °C and at very low pressures, in near surface environments [1, 2], depending on the type of organic matter, presence of sulphides (especially pyrite), access of oxygen, and type of original rocks. During the coal combustion, the sedimentary rocks, associated with it, starts to melt and crystallize under a low pressure regime, corresponding to sanidynite to spurrite facies conditions [3, 4].

The term *paralava* is used to describe vesicular, glassy rock, with ropy structures, containing also the crystalline phases [5].

Upper Silesia (S-Poland) is one of the most intensively mined areas in Europe, and burning coal-dumps have created serious problems since several decades. In the Polish part of the Upper Silesia Coal Basin, 110 coal-fields have been documented, and currently, 45 coal-fields are under exploitation, with 15,284 M tonnes of coal produced overall. Different coal types are present, with so-called energetic coal being the most important [6]. Most of the coal seams are located in the Upper Silesian Sandstone Unit. Sedimentary rocks associated with thick coals include alluvial sandstones and conglomerates. Both „Sosnowiec” coal-mine and „Marcel” coal-mine exploited the Carboniferous coal-seams, located in the so-called Upper Silesian Sandstone Unit (Figure 1).

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In this article, we describe the case of cordierite-rich paralavas as the product of spontaneous coal-dumps combustion. The conditions of melting and processes of cordierite separation are discussed.

2 SAMPLING AND ANALYTICAL METHODS

The paralava samples showing dark and reddish segregation were found in two coal dumps that suffered spontaneous and uncontrolled combustion: near the inactive “Sosnowiec” Coal Mine (after 140 years, exploitation was stopped in 1997) and near currently active “Marcel” Coal Mine. The “Sosnowiec” dump covers an area of ~30 ha, while “Marcel” dump covers 56 ha, of which ~2 ha are continuously burning coal-bearing rocks. Both dumps mainly consist of silt- and sandstones originally associated with coal seams.

Microscope observations and micro-photographs as well as modal analyses were done on Olympus BX-51 microscope, while the morphological study was done using scanning electron microscopy (SE) using a FET Philips 30 electron microscope (15 kV and 1 nA), XRD analysis and lattice parameters calculation of mineral separates were carried out using Philips X’Pert diffractometer (CoK α , 10kV, 10mA, scan range 3.005–74.995° 2 θ) at the Faculty of Earth Sciences, University of Silesia, Sosnowiec, Poland. Mineral analyses were carried out in the Inter-Institution Laboratory of Microanalyses of Minerals and Synthetic Substances, Warsaw (CAMECA SX-100 electron microprobe; 15kV, 20nA). Whole rock analyses of two selected samples (reddish part and dark, cordierite-rich part) were completed by means of ICP-ES for major elements and ICP-MS for trace elements in ACME Analytical Laboratories, Canada. The calculations of paralava parameters (density and viscosity) as well as temperatures, were done using MAGMA software, by University of California (version 2.49.0122, 2008). Additionally, the procedure of magma temperature calculation by Harrison and Watson [7] was applied.

3 RESULTS

Both reddish and dark-grey parts of paralava show the oriented textures, as a result of flow. The vesicles content is greater in dark-grey segregations (28–45 vol%) while the reddish paralava is characterized by 18–30 vol% of vesicles. The mean vesicle size (used also for calculations) is 0.2 mm. The chemical compositions of both parts are similar, with exception of higher Fe₂O₃, MgO, and CaO contents and consequently, lower SiO₂, Al₂O₃ contents in reddish part of paralava in relation to dark one (Table 1, compare WR1 and WR2).

3.1 Red paralava

The glassy paralava is locally similar to pahoehoe structures, which could locally be developed as spherical droplets. Paralava portions show quite constant chemical composition, with SiO₂ content ranging from 64.2–75.2wt%, strongly peraluminous, with ASI in the range of 1.25–3.41. All EMPA paralava analyses show the high potassium content (4.86–8.03 wt%; Table 1) and could be classified as dacite-ryolite to alkali ryolite according to Cox et al. [8]), as well as according to Middlemost [9] classification schemes for volcanic rocks. In most of the spot analyses the enrichment in phosphorus was observed. The calculated liquidus temperature is in the range of 890–1078 °C (on the base of P₂O₅ content according to Harrison and Watson, 1984) or 1015–1141 °C (MAGMA software results). Calculated density of paralava is in the range of 2.32–2.42 g/cm³, while the viscosity values fall in the range of 586 to 4.69×10^7 Pa s (Table 1). Chemical changes in paralava composition could be observed within 50–100 μ m, visible as variation in normative components content. The whole-rock REE content is high and equals to 1613.94 ppm. The Chondrite (C1) normalized REE patterns are flat (Ce_N/Yb_N = 6.61), with negative Eu anomaly (Eu/Eu* = 0.61). Th prevails over U (Th/U = 2.66).

Paralava is poor in minerals, except for commonly found partly resorbed quartz grains. Skeletal forms of Fe–Ti–Al spinel, pseudobrookite, and cordierite (indialite) are components dispersed in glass.

3.2 Cordierite-rich paralava

Cordierite-rich paralava form lenticular and laminar accumulations, dark-grey in colour, ranging 1–30 mm in thickness. These paralava portions are relatively porous and rich in crystals, represented by (Fe–Mg)-cordierite (hexagonal variety called indialite), spinel, and ilmenite. Partly resorbed quartz grains are also found.

Cordierite (variety indialite according to XRD data) is predominant. It occurs as idiomorphic, hexagonal crystals (Figures 2a and b), often zoned, with Fe content growing from the core ($f_m = 0.247$ – 0.376) toward the rims ($f_m = 0.436$ – 0.556 ; Table 2). Locally, Fe-rich cordierite overgrows the poikilitic core with abundant inclusions of spinel and ilmenite. Calculated lattice parameters (assuming indialite $P6/mcc$ space group) are: $a_0 = 9.858(1)$ Å, $b_0 = 9.858(1)$ Å, $c_0 = 9.311(4)$ Å, while a distortion index $\Delta = 2\Theta_{131} - (2\Theta_{511} + 2\Theta_{421})/2$ equals to 0 and is similar to standard one for synthetic indialite

Both spinel and ilmenite show Al-enrichment. Spinel crystals form idiomorphic, locally skeletal crystals (Figure 3a), and non-stoichiometric compositional range: $Ti_{1.15}Fe^{+2}_{0.79}Fe^{+3}_{0.26}Al_{0.27}Mg_{0.02}V_{0.01}O_4 - Ti_{0.88}Fe^{+3}_{0.92}Al_{0.31}Fe^{+2}_{0.29}Mg_{0.06}Mn_{0.01}Si_{0.01}O_4$.

Locally, the monazite and xenotime crystals could be found, all showing resorption features. In the paralava matrix, micron-sized Fe–Mg–Ca phosphates occur adjacent to monazite.

The whole-rock REE content in cordierite-rich paralava is similar to red paralava portion (1746.81 ppm) as well as the Chondrite (C1) normalized REE patterns: $Ce_N/Yb_N = 6.36$), with negative Eu anomaly ($Eu/Eu^* = 0.62$). Th prevails over U ($Th/U = 2.76$).

4 DISCUSSION AND CONCLUSION

The high totals of chemical analyses of the paralava (~99–100 wt% oxides) suggest almost dry melting of the parent siltstone–sand–stone rocks that are supported by the high calculated liquidus temperatures. The chemographic projection (Figure 3) suggest the derivation of all components (glass, cordierite and spinel + ilmenite) from the parental rocks, which can be found as xenoliths in the paralava.

Possibly, the crystallization of cordierite (indialite) was restricted to those parts of viscous paralava, where the oxygen fugacity was relatively low. In most of the paralava the oxygen fugacity should be high, above the QMF buffer, as non-stoichiometric magnetite and pseudobrookite are the most common mineral components and oxidized iron (Fe^{3+}) prevail. On the other hand, formation of cordierite demands presence of Fe^{2+} , proved by the preliminary Mössbauer data. As the difference in chemistry of reddish and dark parts of paralava is meaningless, the oxygen fugacity variations are possibly explanation for the mineralogy and colour difference. One of the possible explanations is that the increased amount of cordierite (indialite) in selected portions of the paralava may be explained by the separation of early formed minerals from the melt during flow and steered mostly by low oxygen fugacity.

The resorption of monazite and associated mobilization of phosphorus to form Fe–Mg–Ca phosphates and P-enrichment of the glassy paralava, pose questions about the behaviour of REE and Th released from monazite during the fusion caused by coal combustion.

5 REFERENCES

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TABLE 1: Whole-rock compositions by ICP-ES (WR1 and WR2, in wt%), EMPA analyses of glass (#1–#6, in wt%) and calculated CIPW norms and other parameters of glassy paralava associated with cordierite segregations.									
Oxide component	LoD ⁽¹⁾	WR1 ⁽²⁾	WR2 ⁽²⁾	#1	#2	#3	#4	#5	#6
SiO ₂	0.01	61.55	60.02	74.84	74.15	64.16	75.15	74.31	69.19
TiO ₂	0.01	1.03	0.96	1.39	1.61	0.70	1.39	1.49	1.08
Al ₂ O ₃	0.01	20.96	19.31	12.03	12.69	20.69	12.44	12.82	15.21
Fe ₂ O ₃ -total	0.04	9.16	11.91	2.11	0.94	6.08	1.58	1.48	9.25
MnO	0.01	0.19	0.18	0.12	0.09	0.21	0.00	0.00	0.11
MgO	0.01	1.94	2.25	0.20	0.05	2.32	0.07	0.06	0.11
CaO	0.01	0.32	0.85	0.82	1.03	0.43	0.69	0.78	0.41
Na ₂ O	0.01	0.21	0.14	0.27	0.34	0.33	0.48	0.34	0.30
K ₂ O	0.01	3.39	3.07	6.24	7.65	4.74	8.03	7.66	4.86
P ₂ O ₅	0.01	0.14	0.12	0.20	0.32	0.11	0.26	0.18	0.11
Sum total	–	98.89	98.81	98.22	98.87	99.77	100.09	99.12	100.63
Mineral modal composition cf. CIPW, in vol%									
Qtz	–	44.21	42.66	47.91	41.56	40.10	40.81	41.83	47.86
C	–	16.71	14.50	3.80	2.74	14.51	2.34	2.99	8.92
Or	–	20.01	18.12	36.83	45.16	27.95	47.36	45.22	28.51
Ab	–	1.78	1.18	2.30	2.88	2.81	4.06	2.86	2.52
An	–	0.68	3.44	2.82	3.03	1.40	1.69	2.69	1.31
Hy	–	4.81	5.58	0.50	0.12	5.74	0.16	0.14	0.27
Ilm	–	0.41	0.39	0.26	0.19	0.46	–	–	0.23
Hm	–	9.18	11.93	2.11	0.94	6.09	1.59	1.48	9.21
Ap	–	0.31	0.26	0.43	0.71	0.24	0.58	0.39	0.24
Λ/CNK	–	4.918	4.013	1.465	1.276	3.341	1.230	1.303	2.439
Calculated parameters of glassy paralava associated with cordierite-rich restite									
T _{H&W} (°C)	–	920	905	1046	1078	890	1069	1009	896
T _{comp} (°C)	–	1149	1156	1015	1043	1141	1072	1048	1100
Density [g/cm ³]	–	2.418	2.436	2.327	2.326	2.418	2.325	2.323	2.360
Viscosity(10 ⁶ Pa s) ⁽³⁾	–	0.08	0.03	3.03	2.49	0.79	36.2	46.9	41.1
⁽¹⁾ LoD = Limit of Detection, in wt%. ⁽²⁾ WR – whole-rock analyses of dark(1) and red(2) paralava; #n – spot EMP analyses of glass. ⁽³⁾ Viscosity calculated after the method of Shaw (1972) assuming temperature 1000 °C.									

TABLE 2: Chemical compositions by EMPA in wt%, and calculated crystal-chemical formulae (in apfu) of selected cordierite (indialite) grains in paralava/restite.					
Oxide component	LoD	#1	#2	#3	#4
SiO ₂	0.03	48.04	48.28	46.45	44.91
TiO ₂	0.05	0.31	0.28	0.33	0.13
Al ₂ O ₃	0.02	33.57	32.72	33.88	38.45
FeO	0.17	9.93	13.79	12.45	10.21
MnO	0.09	0.16	0.37	0.26	0.16
NiO	0.03	0.05	0.00	0.00	0.00
ZnO	0.06	0.12	0.00	0.00	0.19
MgO	0.03	7.64	4.18	5.69	6.01
Na ₂ O	0.01	0.00	0.02	0.01	0.01
K ₂ O	0.01	0.21	1.21	0.09	0.68
P ₂ O ₅	0.10	0.36	0.00	0.00	0.00
Sum total	—	100.39	100.78	99.16	100.75
Calculated mineral compositions in apfu, based on 18O					
Si	—	5.021	5.191	4.847	4.711
Ti	—	0.024	0.023	0.026	0.010
Al	—	4.135	4.146	4.167	4.754
Σ(Si+Ti+Al)	—	9.180	9.360	9.040	9.48
Fe	—	0.391	0.558	1.087	0.403
Mn	—	0.013	0.031	0.023	0.013
Ni	—	0.004	0.000	0.000	0.000
Zn	—	0.009	0.000	0.000	0.015
Mg	—	1.190	0.670	0.885	0.940
Σ(Fe+Mg)	—	1.607	1.259	1.995	1.371
Na	—	0.000	0.004	0.002	0.002
K	—	0.028	0.166	0.012	0.091
P	—	0.032	0.000	0.000	0.000
Σ(Na+K)	—	0.060	0.170	0.014	0.093
fm=Fe/(Fe+Mg+Mn)	—	0.247	0.454	0.556	0.3000

LoD = Limit of Detection, in wt%.

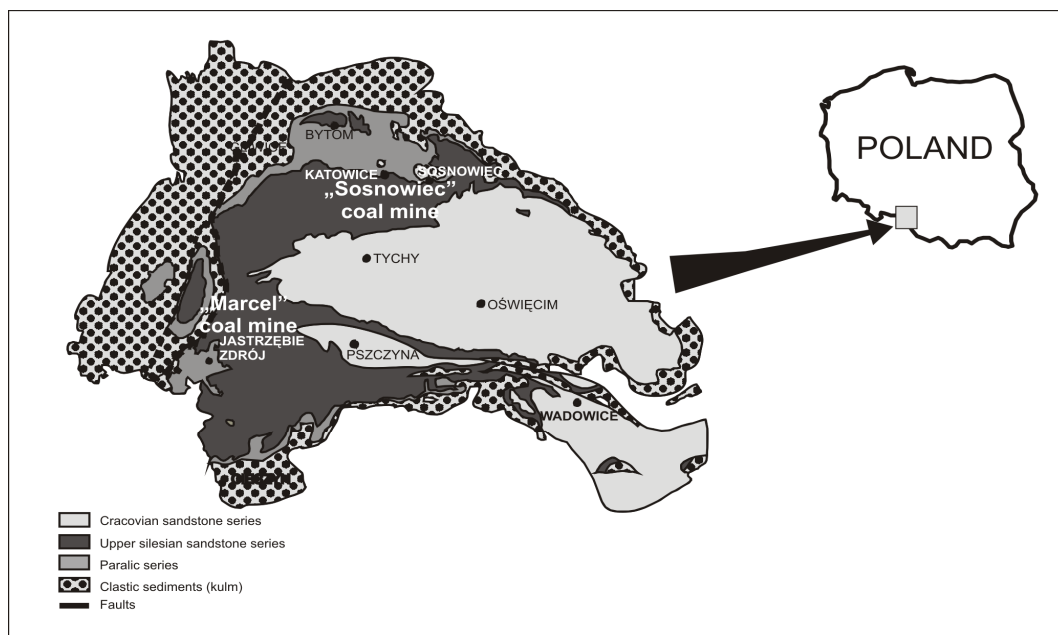


Figure 1: The simplified geological map of the Upper Silesia Coal Basin (USCB) with marked locations of the two sampled coal dumps: “Sosnowiec” and “Marcel” coal-mines dumps.

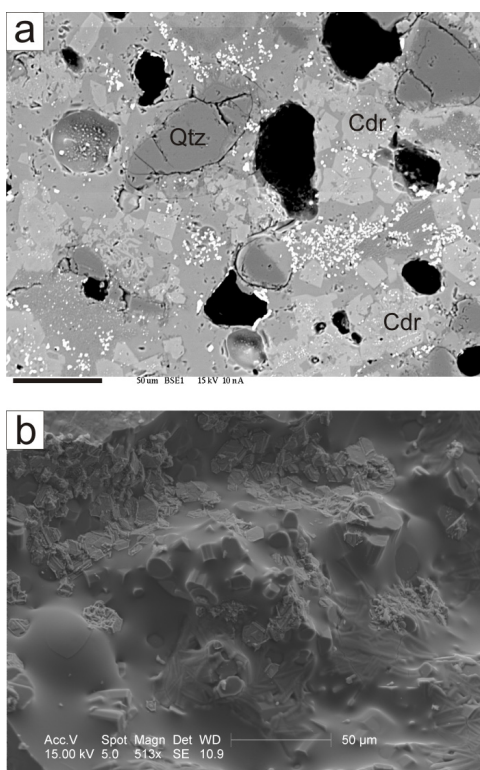


Figure 2: BSE (back-scattered electron (a) and SE (secondary electron (b) images of cordierite-rich paralavas showing relation of glassy paralava to cordierite crystals (Cdr) and resorbed quartz (Qtz) grains.

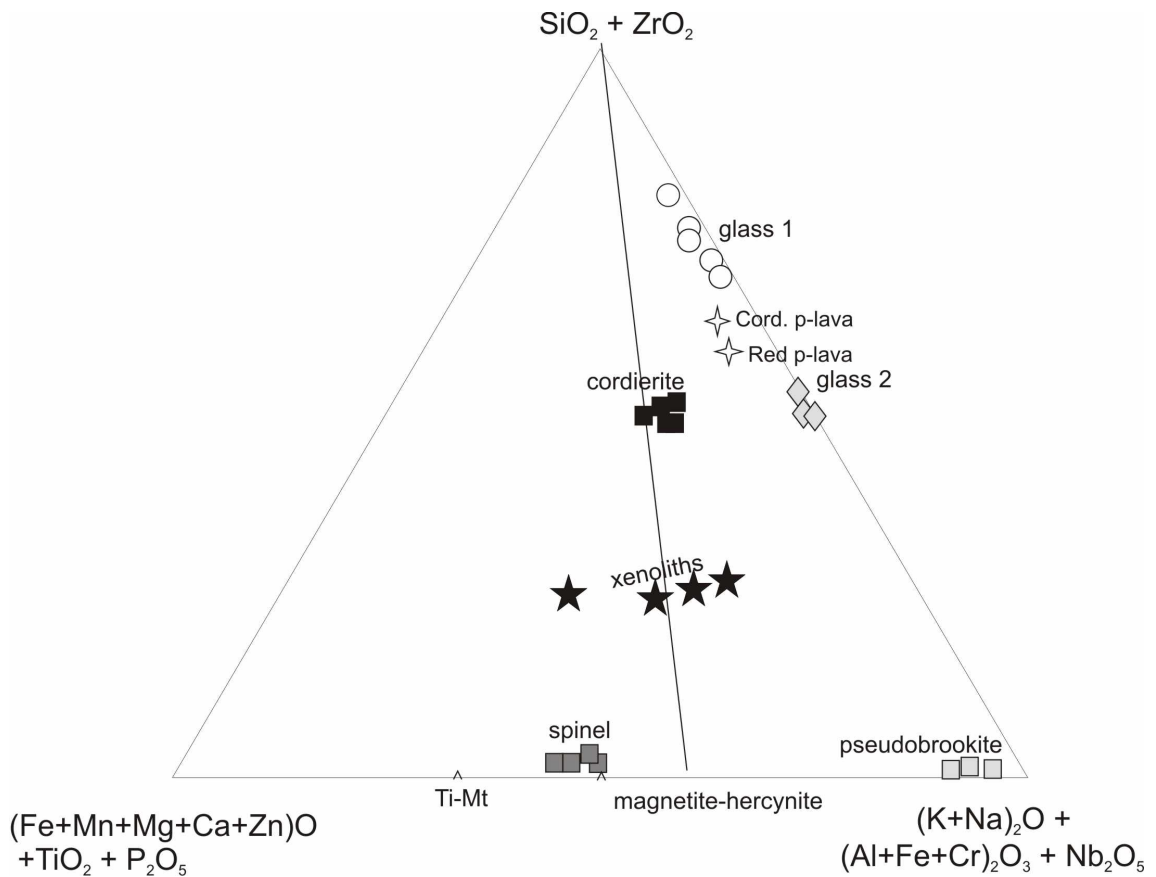


Figure 3: Chemographic projection (after [5]) of the whole-rock paralava samples, consisting minerals and micro-xenoliths.

