

ORIGIN OF MINE WATERS BASED ON THE ISOTOPIC AND CHEMICAL COMPOSITION (UPPER SILESIAN COAL BASIN, POLAND)

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ABSTRACT

The results of isotope investigations have shown that mine waters of different origin and residence time can be distinguished in the Upper Silesian Coal Basin (USCB) down to 1200 m depth. Stable isotope data allowed to assign the mine waters to the four main groups: 1) meteoric waters of the last infiltration period, 2) mixed infiltration and paleoinfiltration waters, 3) paleoinfiltration waters of different age, 4) Tertiary syndimentary waters. Meteoric waters of the last infiltration period squeeze out the relic waters and mix with them. Relic groundwaters occur in the deeper part of the Carboniferous strata. Mining activity is the fundamental factor modifying natural chemical and isotopic composition of groundwaters in the USCB. Deep penetration of low TDS waters from the overlying horizons and of technological mine waters gradually desalinate paleoinfiltration brines and change their isotopic composition. An interpretation using both the isotope and chemical data enabled to present a new identification of mine waters.

OUTLINE OF MINING

The USCB is now one of the biggest coal basins in Europe by its resources and output of hard coal (about 150 mln T/y). Mining has been active here since the second half of the 18th century. The coalfields cover an area of about 2,000 sq. km. The depth of mining works varies from 400 m to 1,200 m. The majority of coal mines lies in the area of shallow occurrence of productive Carboniferous series in the northeastern part of the USCB. The deeper coal deposits overlain by Tertiary clays are now intensely mined (Figure 1).

It should be stressed that the mean depth of mining works and mean mineralization (i.e. total dissolved solids - TDS) of mine waters markedly increase due to the construction of new, deeper exploitation levels in old mines and building of new deep mines in the southern part of the USCB. The mean depth of mining works was about 200-400 m in 1957 but 650 m in 1989 (Rózkowski, 1995). The mean mineralization of mine water discharged to rivers increased from 4.9 g/dm³ in 1970 to 10.9 g/dm³ in 1984 (Rózkowski, 1995), while the volume of water pumped

by coal mines increased only about 6% (Wilk et al., 1990). This indicates the growing inflow of high mineralized groundwater into mines in the last period.

Mine waters are pumped out from 56 coal mines in total quantity of 661.1 cub. m per min. TDS of natural mine waters ranges from 0.2 g/dm³ to 372 g/dm³ but TDS of the cumulative pumped out waters ranges from a few g/dm³ up to 110 g/dm³.

Mine waters are highly variable in their chemical composition and TDS. Pumped out mine waters belong to different chemical groups taking into account the following quantities:

- waters with TDS below 1 g/dm³ and Cl and SO₄ ion content below 0.6 g/dm³ (fresh waters) - 184.1 cub. m per min;
- waters with TDS from 1.0 g/dm³ to 3.0 g/dm³ and Cl and SO₄ ion content from 0.6 g/dm³ to 1.8 g/dm³ (industrial waters) - 266.6 cub. m per min;
- waters with TDS from 3.0 g/dm³ to 70 g/dm³ and Cl and SO₄ ion content from 1.8 g/dm³ to 42 g/dm³ (saline waters) - 157.7 cub. m per min;
- waters with TDS above 70 g/dm³ and Cl and SO₄ ion content above 42 g/dm³ (brines) - 52.7 cub. m per min.

GEOLOGICAL BACKGROUND

The USCB, 7,500 sq. km in area, is situated within the Upper Silesian Variscean depression. The basement of productive rocks of the USCB consists of the Precambrian, Cambrian, Devonian and Lower Carboniferous sequences.

The coal-bearing Upper Carboniferous formation of the USCB according to Jureczka and Kotas (1995) includes 4 lithostratigraphic series, 8,500 m in total thickness, developed within the zones of the greatest basal subsidence (Figure 1). These series are characterized by a gradual reduction of their thickness toward the east and southeast.

The Paralic Series embraces all paralic succession of sediments within the productive Carboniferous of the basin. The whole series belongs to the Upper Namurian A. The Paralic series is composed of conglomerates, sandstones, siltstones, claystones. The share of conglomerates and sandstones in the series profiles varies from 20% to 50%. The thickness of the Paralic series is variable: from 200 m to 3,780 m.

The Upper Silesian Sandstone Series (Namurian B-C) is characterized by a dominance of sandstones and conglomerates over claystones and siltstones. The share of coarse-grained clastic rocks in the series is high but variable, commonly exceeding 50%. The total thickness of the series approaches 1,100 m; it pinches out in the eastern part of the basin.

The Mudstone Series (Westphalian A-B) is rather monotonous with fine-grained clastic rocks being predominant. The share of sandstone in the whole series profile reaches to 16-23%. The thickness of the series ranges from 100 m in the eastern part of the basin up to 2,000 m in the central part.

The uppermost sequence of the productive Carboniferous consists of the Cracow Sandstone Series (Westphalian C, D). A predominance of coarse-grained sandstones (up to 90%) over siltstones and claystones is the characteristic feature of the series. The thickness of the series is up to 1,640 m.

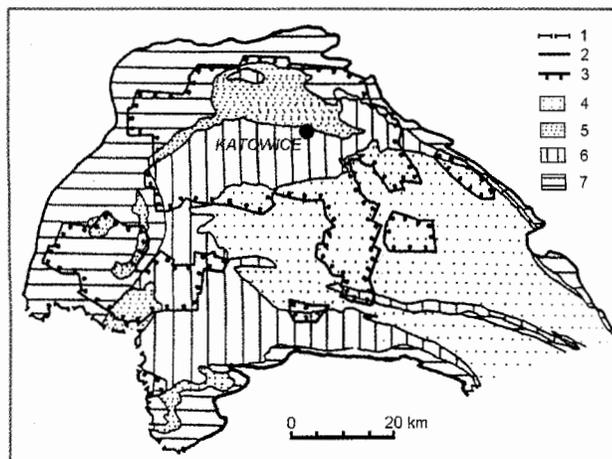


Figure 1. Permian, Mesozoic and Cainozoic subcrop map of the Upper Silesian Coal Basin (USCB), with outlined of coal mine fields. 1 - state boundary; 2 - extension of the USCB; 3 - mine areas; 4 - Cracow sandstone series; 5 - Upper Silesian sandstone series; 6 - Mudstone series; 7 - Paralic series.

The coal-bearing Carboniferous formations of the basin are covered by Permian, Triassic, Jurassic, Tertiary and Quaternary deposits. Permian and Jurassic deposits cover very small portions of the basin, stretching along the northeast limit of the Carboniferous subcrops. The Triassic, Quaternary, and most of all, the Tertiary cover of the basin is significant for the hydrogeological problems (Figure 2).

Triassic formations of the Upper Silesian Coal Basin reach up to 200 m in thickness. They are represented by claystones, sandstones (the Lower Triassic), dolomites and limestones, among which the ore-bearing dolomites are the most significant.

The Jurassic formations, ranging in thickness up to 100 m, are known to occur only in easternmost part of the basin.

Tertiary sediments of Miocene age bury the complex lying over the erosional land-surface of the basin (Figure 2). The formation consists of argillaceous sediments with subordinate fine-grained sand and sandstones intercalations. The thickness of that formation reaches up to 1,100 m in the southernmost area of the USCB.

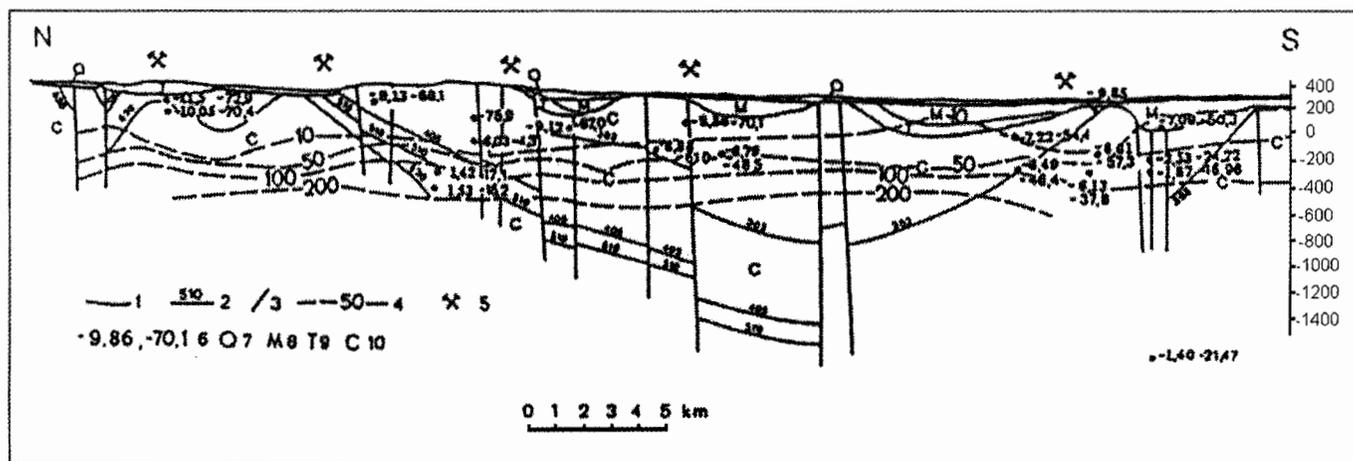


Figure 2. Hydrogeological cross-section. 1- boundaries of the stratigraphic series; 2- coal seams; 3- faults; 4- isolines of TDS (g/dm³); 5- coal mines; 6- d¹⁸O and dD values; 7- Quaternary; 8- Miocene; 9- Triassic; 10- Carboniferous.

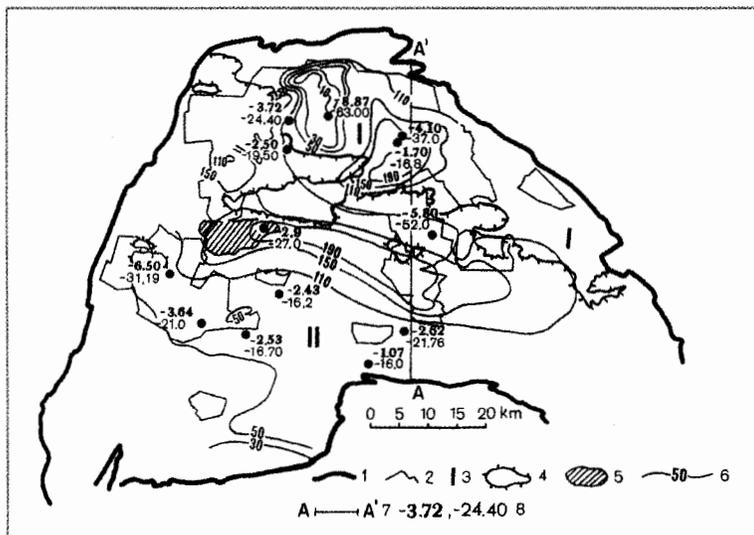


Figure 4. Sketch-map of the groundwater mineralization (USCB) at the depth of 500 m below sea level. 1- extension of the USCB; 2- extension of the coalfields; 3- hydrogeological regions; 4- extension of the isolating series of the Tertiary deposits; 5- extension of the salt deposits in the Tertiary formation; 6- isolines of TDS (g/dm³); 7- hydrogeological cross-section; 8- d¹⁸O and dD values. Hydrochemical setting according to Rózkowski et al., 1990.

Parameter	Depth intervals beginning from the surface (m)						
	0-200	200-400	400-600	600-800	800-1000	1000-1200	1200-1400
Open porosity (%)	18.0	12.5	11.0	10.5	9.5	7.1	6.7
Specific yield (%)	12.0	9.0	8.0	8.4	7.2	5.1	4.4
Permeability (mD)	300.0	80.0	20.0	19.0	1.6	0.17	0.16

Table 1. Arithmetic mean values of hydraulic properties of Carboniferous sandstones and mudstones within 200 m depth intervals.

Within the outcrops areas of the Carboniferous strata where the erosion and rock relaxation processes take place high permeability of Carboniferous sandstones independently on geological age of rocks is observed.

Field investigations carried out by Kleczkowski and Witczak (1967) in the eastern part of the USCB have shown very clearly the increase of hydraulic conductivity of sandstones until the depth of about 500 m due to rocks fissure. The values of K differ from 5.0×10^{-5} m/s to 3.0×10^{-7} m/s (mean value 1.0×10^{-6} m/s). Below this depth rocks fissure slowly disappeared and hydraulic conductivity is due to matrix porosity of sandstones. The results of test pumping and subsurface sampler tests carried out by Rózkowski and Wagner (1988) in the depth interval from 600 to 1600 m shown the declining of hydraulic conductivity values with the depth from 1.0×10^{-7} to 4.0×10^{-11} m/s.

Taking into account the results of laboratory and field investigations it should be accepted that Carboniferous sandstones and mudstones occurring below the depth of 700-800 m are practically impermeable. It does not concern the areas of mining where slides, cracks and unstraining of rocks accompanying mining excavation cause increase of rock permeability.

The USCB may be classified as a Variscan artesian basin. Three groundwater flow systems may be distinguished in this basin (Rózkowski, 1995). The travel distance and time of flow tend to increase from the upper flow system to the lower one. It was assumed that the local flow system is characterized by active groundwater flow through the rocks. The intermediate flow system is described by less active groundwater circulation, while the regional flow system is characterized by very sluggish groundwater flow, for length of time of millions years. The regional flow system occupies the zone of relict buried stagnant brines.

Topography and geology of the basin and hydraulic conductivity of the rocks, through which the groundwater moves, control the groundwater flow systems. Mining activity and, especially, mining drainage have recently become very important factors of flow system control.

A general trend of deepening and enlargement of the infiltration and intermediate zones is observed during the last 40 years due to deeper exploitation and intensive mining drainage.

HYDROCHEMISTRY

The groundwaters in the USCB vary in chemical composition and total mineralization (Rózkowski, 1995). Fresh waters occur in the Quaternary, Jurassic and Triassic formations of the cover and in the outcropping Carboniferous strata.

Mineralization of the groundwater in the Tertiary strata ranges from 0.5 to 220 g/dm³; the waters originally belong to the Cl-Na chemical type. The total mineralization of groundwaters in the coal-bearing Carboniferous strata ranges from 0.5 to 372 g/dm³.

Fresh waters (TDS < 1 g/dm³) are mainly of hydrochemical types: HCO₃-Ca, HCO₃-SO₄-Ca, SO₄-HCO₃-Ca-Mg. Their hydrochemical coefficients have values: $r(\text{Na}/\text{Cl}) > 1$, $r(\text{Ca}/\text{Mg}) > 1$, and $r(100 \times \text{SO}_4/\text{Cl}) > 1$. Nitrogen predominates in gaseous composition of these waters. The described waters occur in oxidation zone.

Saline waters, with TDS < 35 g/dm³, belong to multi-ion and Cl-Na hydrochemical types. The following values of coefficients are typical for them: $r(\text{Na}/\text{Cl}) = 1.3 - 0.87$ and $r(100 \times \text{SO}_4/\text{Cl})$ from 0.007 to 9.1. Nitrogen predominates in the upper part of their occurrence, and methane predominates in the lower one. This evidence allows one to assume that saline waters may occur in the oxidation as well as reduction zones.

Brackish mine waters from the oxidation zone are enriched in sulphate ions in the mining excavations, due to oxidation of pyrites and sulphur in coal seams.

The brines with TDS content above 35 g/dm³ belong to hydrochemical types Cl-Na and Cl-Na-Ca and have the following values of hydrochemical coefficients: $r(\text{Na}/\text{Cl}) = 0.72 - 0.96$ and $r(100 \times \text{SO}_4/\text{Cl}) < 1$. They occur in the reduction zone only. Highly mineralized waters in insulated structures are buried brines of the Cl-Na-Ca types. Methane from degazation of coal seams predominates in gaseous composition of those brines.

The presented hydrogeochemical data demonstrate a vertical succession of hydrochemical zones in the Carboniferous strata of the USCB. Three hydrochemical zones have been distinguished: the zone of infiltration waters, the intermediate zone of mixed waters, and the lower zone of buried brines (Palys, 1966; Rózkowski, 1995). These zones are defined on the basis of the values of hydrochemical coefficients and groundwater mineralization.

The zone of infiltration waters reaches the depth of about 300 m in the first hydrogeological region and 80 m in the second one. The lower boundary of the intermediate zone lies at depth of 450-650 m, 800 m at maximum, in the first hydrogeological region. The zone is about 200-300 m thick. In the second hydrogeological region, the lower boundary of this zone lies at depth of about 240 m. The zone of buried brines underlies the intermediate zone. Hydrochemical studies (Rózkowski, 1995) have shown that brines occur at the depth 650-850 m in the USCB, depending on the varying geological conditions and mining activity in the individual geological structures.

There is a general trend of increasing groundwater mineralization with depth, independently of the age of the rocks (Figure 2). This general trend is disturbed by hydrochemical anomalies. The anomalies have been observed, among others, in the uppermost part of the Carboniferous series in the Carpathian Foredeep, near the Tertiary salt deposit in the Zawada Graben, and along some regional fault zones (Kleczkowski, Witczak, 1967; Palys, 1966; Rózkowski, 1995). Antropogenic anomalies are due to mining activity (Rózkowski, 1995).

The variation of TDS zones in the USCB was studied on 1,233 chemical analyses of groundwater samples from mine works and boreholes.

Spatial variation of the groundwaters mineralization is clearly visible on the hydrochemical maps (Figures 3, 4) and on the cross-section (Figure 2).

ISOTOPE DATA

Isotope data of groundwaters gave an indication to the origin and residence time of groundwaters and to the depth of their extension within the USCB. The isotope studies (tritium and stable isotopes) permit also to recognise the effect of mining activity on the natural hydrochemical zonation and flow systems.

The results of isotope investigations have shown that within the USCB down to 1,200 m depth, it means to the maximal depth of coal mining, groundwaters of different origin and residence time can be distinguished (Rózkowski, Przewlocki, 1974; Rózkowski, 1995; Zuber, Pluta, 1989).

Stable isotope data (Figure 5) allowed to assign those groundwaters to the four main groups: 1) meteoric waters of the last infiltration period, 2) mixed infiltration and paleoinfiltration waters, 3) paleoinfiltration waters of different age and 4) Tertiary synsedimentary waters.

Groundwaters of the last infiltration period are of Holocene, Pleistocene and Prepleistocene age. Investigations carried out by Zuber and Grabczak (1985) and by Zuber and Pluta (1989) have shown that in the Carboniferous formation at the depth of a few hundred meters below the day surface Tertiary waters from the last stage of the last infiltration period have been found. Meteoric waters of the last infiltration period were sampled from the Quaternary, Jurassic, Triassic and the uppermost parts of the Tertiary and Carboniferous formations. In the diagram at the background of precipitation line described by the equation $\delta D = 8 \delta^{18}O + 10$ one can distinguish infiltration waters whose projection points are plotting at the precipitation line (Figure 5).

The contemporary infiltration waters belong to the Holocene and Pleistocene age. They have the $\delta^{18}O$ values varying from -12.1 to -9.0 per thousand and δD from -78.0 to -67.9 per thousand. In general they belong to the multi-ions hydrochemical type of groundwater and their TDS ranges from 0.2 to a few g/dm³. More detailed specification of contemporary infiltration groundwaters may be done by estimating tritium content. In the areas of outcrops of the Carboniferous formation within the first hydrogeological region tritium content was estimated in more than 200 mining water samples. Positive tritium presence was found till the depth of 150 to 250 m, locally even deeper. Tertiary infiltration waters are those which $\delta^{18}O$ vary from -6.2 to -8.1 per thousand and δD from -45.0 to -54.0 per thousand. TDS of these waters is rising up to 100 g/dm³. The depth of occurrence of these waters based on the isotopic investigations is up to 480 to 500 m.

Meteoric waters of the last infiltration period squeeze out the relic waters and mix with them. Mineralized groundwaters, which are a mixture of relic and meteoric waters of the last infiltration period, belong to the second group of waters distinguished on the precipitation line (Figure 5). They have been found in the Tertiary and mainly in the Carboniferous formations. In the opinion of Zuber and Pluta (1989), and Pluta (1988) to the mixed type of waters belong those which are the mixture of Tertiary waters of the last infiltration period with Quaternary waters as well as the Tertiary waters mixed with buried brines. According to these authors the $\delta^{18}O$ values range from -3.7 to -5.2 per thousand and δD from -27.0 to -36.0 per thousand. They are mainly brines of TDS up to 160 g/dm³. The depth of occurrence of tested waters is up to 480 m to 600 m. Position of the stable isotopes values of groundwaters from the Tertiary formation at the precipitation line (Figure 5) confirm the mixing process.

The buried paleo-infiltration brines in the Carboniferous formation belong to the fourth group of waters distinguished on the precipitation line. The tested brines are characterized by $\delta^{18}O$ values from -1.2 to -3.6 per thousand and δD from -16.9 to -26.0 per thousand. These brines were sampled at the depth below 480 m in the second hydrogeological region and at 600-700 m in the first one. The separated group of waters at the

precipitation line form the synsedimentary Cl - Na saline waters of Tertiary age in the sandy inserts of the Tertiary clays in the deep Alpine grabens, at the depth of about 600 m.

ISOTOPIC STRATIFICATION OF MINE WATERS

Mining activity is the fundamental factor modifying natural chemical and isotopic composition of groundwaters in the USCB down to the depth of 1,200 m. Deep penetration of low-TDS waters from the overlying horizons and of technological mine waters gradually desalinate paleoinfiltration brines and change their isotopic composition. The foregoing phenomena are intensified due to considerable recharge of Carboniferous horizons by atmospheric waters in the first hydrogeological region.

A close correlation exists between isotopic composition of groundwaters, groundwater mineralization in the Carboniferous strata, permeability of the overburden, and mining activity is shown on the hydrochemical cross-section (Figure 2).

The influence of geological and mining factors on the stable isotopic composition and groundwater mineralization in the USCB is shown on the sketch-maps of the groundwater mineralization and isotopic composition at the depth of 250 m (Figure 3) and 500 m (Figure 4) below sea level. A decrease in groundwater mineralization and stable isotope values within the coalmine fields is visible in these maps. This indicates mixing of groundwaters in these areas. As we can see in Figure 3, the TDS values in the first hydrogeological region, where exploitation proceeds below the depth of 500 m (250 m below sea level), usually do not exceed 10 g/dm³. Stable isotope values are typical for groundwaters of the last infiltration period, mainly of Quaternary age. An increase of TDS to 200 g/dm³ and stable isotope values typical for the mixed and paleoinfiltration types are observed in the second region. The highest TDS value is noted in the place where Tertiary salt deposits are present a top the Carboniferous strata. Unfortunately, no isotope data are available for these waters.

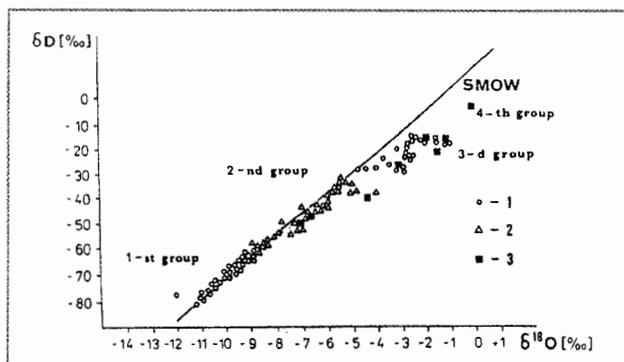


Figure 5. $\delta^{18}\text{O}$ plotted against δD for USCB groundwater. 1- waters from Carboniferous under permeable cover (I-st region); 2- waters from Carboniferous under permeable Tertiary (II-nd region); 3- waters from Tertiary formation.

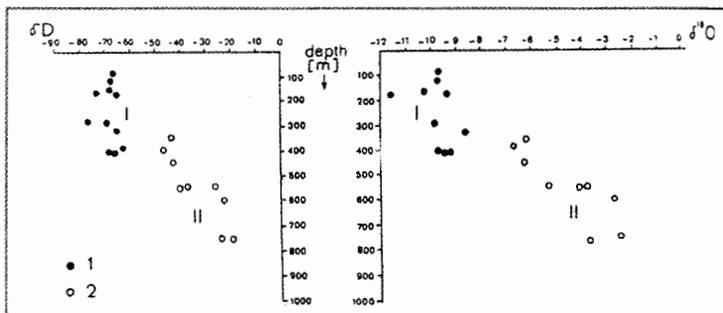


Figure 6. Isotopic composition of groundwaters from the Westphalian formation. I- 1-st hydrogeological region; II- nd hydrogeological region.

The sketch-map of groundwater mineralization and isotope composition at the depth of 780-800 m (500 m below sea level) shows different relations (Figure 4). The mineralized waters (below 10 g/dm³), showing stable isotope composition typical for infiltration waters, have been recognised only locally in the first hydrogeological region, where the mining activity has taken place to the depth of 900 m or more, and lasted for a long time. Brines with TDS values of 110-190 g/dm³, showing high values of stable isotopes ($\delta^{18}\text{O}$ -1.0 to -5.8 per mill and δD -16.0 to -52.0 per mill) occur in the remaining part of the USCB. As for their isotopic composition, these brines belong to the mixed waters and paleoinfiltration groups.

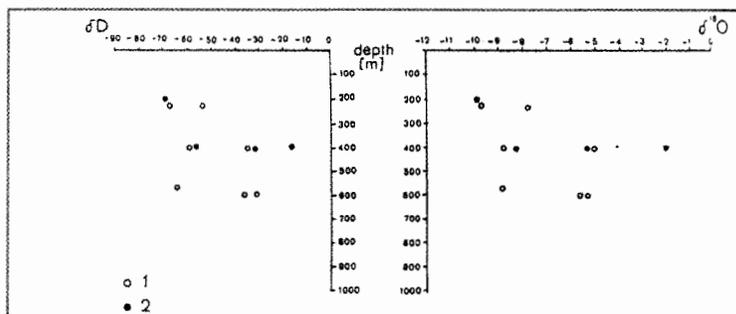


Figure 7. δD and $\delta^{18}\text{O}$ versus depth. 1- groundwaters from Jankowice mine; 2- groundwaters from Marcel mine.

Variation in stable isotope composition of waters in the Westphalian horizons, at the depth down to 800 m, is shown in Figure 6. The values of stable isotopes are low in the recharge area (the first hydrogeological region). There occur mine waters belonging to the recent infiltration period of Quaternary age. Less negative values of stable isotopes, typical of mixed and paleoinfiltration types of groundwaters, have been observed in the Westphalian horizons under the impermeable cover of the Tertiary clays, in the second hydrogeological region. A general trend of increasing of heavy isotopes and TDS content in waters with depth is observed in the second hydrogeological region. Isotope composition of waters sampled at the depth down to 420 m in the first region is much the same ($\delta^{18}\text{O}$ from -11.7 to -8.7 per mill and δD from -77.0 to -64.0 per mill). This is due to the replacement of waters in the mine area.

The chemical and isotope composition of groundwaters changes continuously during mine exploitation. This is clearly shown on the diagram of stable isotope values versus depth (Figure 7). The samples of brines were collected from the mines Jankowice and Marcel at the same time, the depth of 400 and 600 m. Waters sampled from the same depth have different isotope composition. So, e.g. the brine from the Jankowice mining level -400 m has isotope composition varying from $\delta^{18}\text{O}$ -2.0 to -8.3 per mill and δD from -18.0 to -62.0 per mill. It means that mixed and paleoinfiltration waters occur at the same depth. This is due to different timing of exploitation and drainage in separate mining fields at the same level.

Tritium analyses in the first hydrogeological region allow one to decipher the exchange process of mine water in more detail (Rózkowski, 1986). Tritium content of about 5 ± 2 TU is characteristic down to the depth of 150 m in the north-eastern part of the USCB, outside the mining areas. Recently, mine waters with such a tritium content have been recorded at the depth of about 300 m in the same area. This indicates that zone of young water distribution is about 100-150 m deeper. In the meantime, within the same area, mineralization of waters in one of the mines, at the depth of 300 m, decreased from 11.0 g/dm^3 in 1961 to 1.9 g/dm^3 in 1988, thus confirming active infiltration process of atmospheric waters into the mine workings.

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