Errata

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CARBON AND OXYGEN STABLE ISOTOPIC COMPOSITION IN THE MUSCHELKALK ROCKS OF THE CHRZANÓW AREA (UPPER SILESIA)

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Abstract. Fourteen determinations of C and O stable isotopic composition were made in limestones, dolomitic limestones and dolomites (dolostones) representing the upper members of the Gogolin Beds, Górażdża Beds, and the Lower Carboniferous carbonate rocks. A diagrammatic profile of these series, their brief petrographic description, and the results are presented in Table 2 and in Figures 1 and 2.

INTRODUCTION

Petrographic studies of the dolomites occurring in the Lower Muschelkalk of the Cracow-Silesian area were extended by determinations of the carbon and oxygen stable isotopes. This procedure was treated as a supplementary method for geological and petrographical investigations, giving new information on the conditions of origin of these rocks. This method is not commonly used in Poland. About twenty determinations of the C and O stable isotopes in Triassic limestones and dolomites (dolostones), together with their interpretation, were published by Pawłowska and Szuwarzyński (in press).

This paper is a contribution to the knowledge of the C and O isotopic composition in Triassic rocks. Determinations were made on samples derived from the continuous profile of the upper part of the Gogolin Beds and from the Górażdża Beds, which were compared with the dolomitized equivalents of the Górażdża Beds. Particularly valuable were the samples collected from a quarry in Płaza which permitted a characterization of limestones, dolomitic limestones, and early-diagenetic „primary" dolomi-

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tes (acc. to Siedlecki 1952). Ore-bearing dolomites were characterized by means of samples from the Trzebionka zinc and lead ore mine. Also samples of early-diagenetic dolomites occurring in the ore-bearing dolomites in the Trzebionka mine were investigated. To obtain comparative data, determinations were made for the Lower Carboniferous limestones and epigenetic dolomites from Czerna near Krzeszowice.

Several authors (e.g. Craig 1953, 1957; Clayton, Epstein 1958; Bathurst 1975) noticed differences in the isotopic composition of carbonate rocks depending on the mode of their formation and on the geologic processes that could have later affected them. The causes of this differentiation are not well known yet, but isotopic studies begin to gain ever increasing importance.

INVESTIGATION OF STABLE ISOTOPES

Fractionation effect of the carbon and oxygen stable isotopes is generally expressed in δ values (in permil ‰):

$$\delta^{13}C = \frac{[^{18}O/^{16}O]_{sample} - [^{18}O/^{16}O]_{standard}}{[^{13}C/^{12}C]_{standard}} \cdot 1000‰$$

$$\delta^{18}O = \frac{[^{18}O/^{16}O]_{sample} - [^{18}O/^{16}O]_{standard}}{[^{18}O/^{16}O]_{standard}} \cdot 1000‰$$

where: $[^{13}C/^{12}C]_{sample}$ — ratio of carbon isotopes in the samples;

$[^{18}O/^{16}O]_{standard}$ — ratio of oxygen isotopes in the measured standard.

For geochemical purposes, first measurements of $\delta^{13}C$ were referred to P.D.B. standard, whose isotopic composition had been determined by Craig (1957). Up till now, all results have been referred to this international standard which is a carbon dioxide obtained from $CaCO_3$ of the rostrum of Cretaceous belemnite from the Peede formation, South Carolina (P.D.B. — Peede Belemnite), by treatment with 100% phosphoric acid at 25.2°C. P.D.B. is materially non-existing standard, therefore other international standards for intercalibration versus P.D.B. are vailable, e.g.:

- NBS-20 Solenhofen limestone from the National Bureau of Standards (Washington, USA);
- Toilet Seat — $CaCO_3$ from the International Atomic Energy Agency (Vienna, Austria);
- Carrara Marble — marble from the University of Pisa (Italy).

A new universal $CaCO_3$ standard with a carbon and oxygen isotopic composition identical with that of P.D.B. will be prepared and distributed by the *International Atomic Energy Agency* in Vienna.

Table 1

Isotopic composition of standards

Sample	$\delta^{13}C$	$\delta^{18}O$
	versus PDB in ‰	
Marianna Biała	+1.68	−5.70
Toilet Seat	+1.88	−1.88
NBS	−1.06	−3.75
Carrara Marble *	+2.40	−1.41

* Results obtained in the Institute of Nuclear Geology in Piza, in the year 1969.

All the values discussed in this paper were measured vs. the Marianna Biała Marble (Kletno, Lower Silesia), which was calibrated versus P.D.B. using Toilet Seat, Carrara Marble and *NBS-20* standards (Table 1). The reproducibility of the presented values shows that there is a good correlation with the results obtained by other well established laboratories (Barański, 1976). Standard deviation of a single determination of $\delta^{13}C$ and $\delta^{18}O$ was ± 0.1‰.

Carbon and oxygen isotopes of investigated calcium carbonate samples can be introduced into the mass spectrometer for measurement only in the form of CO_2 gas. Carbon dioxide is generally obtained from calcium carbonate by treatment with 100% phosphoric acid. This reaction ensures liberation of CO_2 without changing its isotopic composition.

Preparation methods and measurement techniques applied to the samples discussed in this paper are described in detail by Barański (1975).

DESCRIPTION OF THE MATERIAL

The first detailed description of the Muschelkalk profile in Płaza near Chrzanów was given by Siedlecki (1952). A part of this profile was described again, basing on the recently exposed parts of the quarry (Fig. 1).

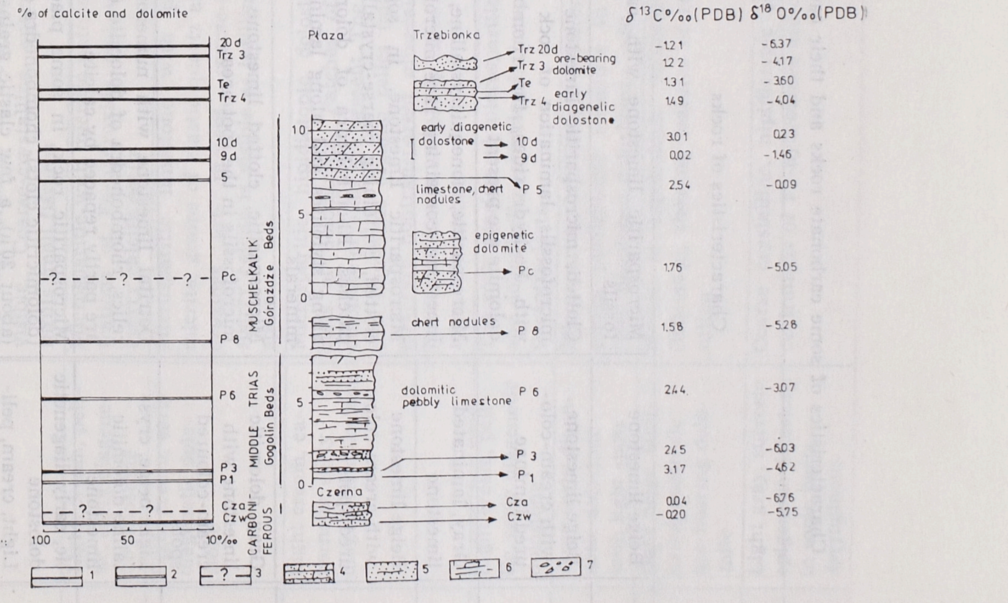

Fig. 1. The $\delta^{13}C$ and $\delta^{18}O$ along the simplified profile of Muschelkalk rocks and two samples of Lower Carboniferous rock. On the left-hand side diagram of percentage of calcite and dolomite in the examined samples

1 — dolomite, 2 — calcite, 3 — samples of rock with variable contents of dolomite and calcite in which quantitative measurements were aimless, 4 — dolostone, 5 — dolomitic limestone 6 — limestone, 7 — pebbly limestone, 8 — limestone with chert nodules

Table 2

Characteristics of some carbonate rocks and their carbon and oxygen isotopic composition

Sample		Characteristics of rocks	Isotopic composition			
			δ C¹³	δ O¹⁸	Range δ C¹³	Range δ O¹⁸
P_1 (Triassic)	Beige limestone	Microsparitic limestone with rare microfossils	3.17	−4.62	1.56—3.17	−4.62—−6.03
P_3 (Triassic)	Beige limestone with cream-coloured laminae	Clotted, microsparitic limestone with rare microfossils, lamination of rock underlined with Fe-hydroxides, the rhombohedra of dolomite are present	2.45	−6.03		
P_5 (Triassic)	Gray, laminated limestone	Microsparitic, nonequicrystalline, cavernous limestone, containing rare microfossils	1.56	−5.28		
P_6 (Triassic)	Beige limestone with cream-coloured spots	Microsparitic limestone, in some places clotted, containing coarse-crystalline calcite patches. Rhombohedra of dolomite occur along stylolites and along laminae of clay minerals	2.44	−3.07	1.76; 2.44	−3.07; −5.05
P_{C3} (Triassic)	Gray, dolomitic limestone with cream-coloured spots	Microsparitic, clotted limestone with rare microfossils in the spot area	1.76	−5.05		
Cz_{a2} (Carboniferous)	Light beige, crystaline dolomitic limestone	Detrital limestone with numerous organic relics, rhombohedra of dolomite (300—500 μ) are partly repaced by calcite	0.04	−6.76	0.04	−6.76
p_5 (Triassic)	tic, early diagenetic dolostone Light, cream, peli-	Microsparitic rock, in some places clotted (dolomicritic clots), rhombohedra of dolomite (about 20 μ), a few clastic grains of quartz and concentrations of Fe-hydroxides are visible. Coarse-crystalline sparry calcite infills small fissures	2.54	−0.08	0.02—3.01	−4.17—0.23
P_{9d} (Triassic)	Light, cream, pelitic, early diagenetic dolostone	Microsparitic with micritic patches, concentrations of Fe-hydroxide and veinlets of sparry calcite occur	0.02	−1.46		
P_{10d} (Triassic)	Light cream, early diagenetic dolostone	Microsparitic limestone with patches of micrite, grains of clastic quartz and Fe-hydroxides are present, coarse-crystalline sparry calcite is visible as infillings of free spaces	3.01	0.23		
Trz_4 (Triassic)	Light cream, cavernous, early diagenetic dolostone	Microsparitic, hipidiotopic dolostone, with clots of dolomicrite. Patches of idiotopic dolomite crystals are present. The rock contain numerous organic relics (microfossils) and rare nontransparent minerals (pyrite). Numerous cavers are partly filled with idiotopic dolorhombs	1.49	−4.04		
Te (Triassic)	Light cream, pelitic, early diagenetic dolostone	The rock consists of microsparitic dolomite and dolomicrite occurring in form similar to the intraclasts	1.31	−3.60		
Trz_3 (Triassic)	Gray early diagenetic dolostone with light spots	Microsparitic, hipidiotopic dolostone, insome places porfirotopic structure in observed (well developed dolorhombs − 60 μ)	1.22	−4.17		
Trz_{20d} (Triassic)	Dark beige, laminated, succrosic ore-bearing dolomite	Mosaic of hipidiotopic dolomite crystals (100—40 μ), showing features of recrystallization	−1.21	−6.37	−1.21	−6.37
Cz_{w1} (Carboniferous)	Light gray, sucrosic epigenetic dolostone in some parts ferruginous	Coarse crystalline, hipidiotopic rock, concentrations of Fe-hydroxides are common	−0.20	−5.75	0.20	−5.75

The lower part of the profile corresponds to the upper part of the Gogolin Beds (Siedlecki, 1952). The series in question are diversified, being represented by smooth limestones beige in colour, as well as by laminated, cavernous, spotted, nodular and conglomeratic limestones. The upper part of the profile, the Górażdża Beds, is less differentiated. It is made up of beige massive limestones, sometimes sugary grained, containing two intercalations of limestones with cherts (Fig. 1). About 10 m above the top of the Gogolin Beds is the bottom of a several-metre thick layer of early-diagenetic dolomites. In the Górażdża Beds, at the extension of limestones in another part of the quarry, dolomites have been found. The oblique boundary of their sole suggests epigenetic dolomites. According to Siedlecki (1952), they are ore-bearing dolomites which, in Bogacz's opinion (pers. comm.), owe their origin to surficial dolomitization.

Dolomites from Trzebionka were represented by four samples only. One was a typical ore-bearing dolomite, the remaining three were early-diagenetic dolomites, some of them partly altered intoore-bearing dolomites *.

Samples from Czerna represented Lower Carboniferous limestones and dolomites formed due to surface dolomitization **.

Table 2 gives a brief petrographic description of the samples whose isotopic composition was investigated. Figure 1 shows the lithologic profile and a diagram presenting the percentage of dolomite and calcite, whereby the degree of dolomitization of limestones was defined. The percentage of calcite in dolostones determines the content of calcite veinlets and inclusions in the rock.

RESULTS

The results presented in Figures 1, 2 and in Table 2 show homogenization of the isotopic composition of the rocks. For limestones not containing dolomite $\delta^{13}C$ values vary from +1.56‰ to +3.17‰, while for dolomitic limestones two values of ^{13}C determined vs. *PDB* are −1.76‰ and +2.44‰. $\delta^{18}O$ values for limestones range from −4.62‰ to −6.03‰, and those for dolomitic limestones are −3.07‰ and −5.05‰.

Early-diagenetic dolomites (Bogacz *et al.*, 1972) and similar, primary dolomites from Płaza (Siedlecki, 1952) had $\delta^{13}C$ values varying from +0.02 to +3.01‰, and $\delta^{18}O$ values ranging from −4.17‰ to +0.23‰. For epigenetic dolomites $\delta^{13}C$ was −0.21‰ and −0.20‰, and $\delta^{18}O$ — −6.37‰ and −5.75‰ (Table 2, Fig. 1).

A comparison of determinations of the oxygen isotopes for limestones, dolomitic limestones and early-diagenetic dolomites has revealed that the latter are enriched in heavy isotopes. This fact is consistent with the generally known regularities (Friedman, Sanders, 1967; O'Neil, Epstein, 1966). Worth noting is sample *P-10d* of early-diagenetic dolomite whose isotopic composition compared with that of limestones (e.g. sample *P-3*) shows an increase in the content of heavy isotopes by more than 6‰, therefore falling within the limits 6—10‰. This difference of 6—10‰ in the oxygen isotopic composition of dolomite and calcite suggests that the two carbonates and formed in the same sedimentary basin (O'Neil, Epstein 1966). In the profile studied, only one sample satisfies the above condition whereas another one, *P-5*, is close to it. However, they should not be left out of account because on other samples of early-diagenetic dolomites the content of heavy isotopes is expected to be underestimated. In samples *P-9d*, *P-5* and *P-10d* lower values can be due to the presence of calcite veinlets and infilling of caverns in the rocks; its content determined by point counts in polished sections amounts to a few per cent (Fig. 1). From Gross's studies (1964) it appears that the presence of secondary calcite is responsible for enrichment in both carbon and oxygen lighter isotopes.

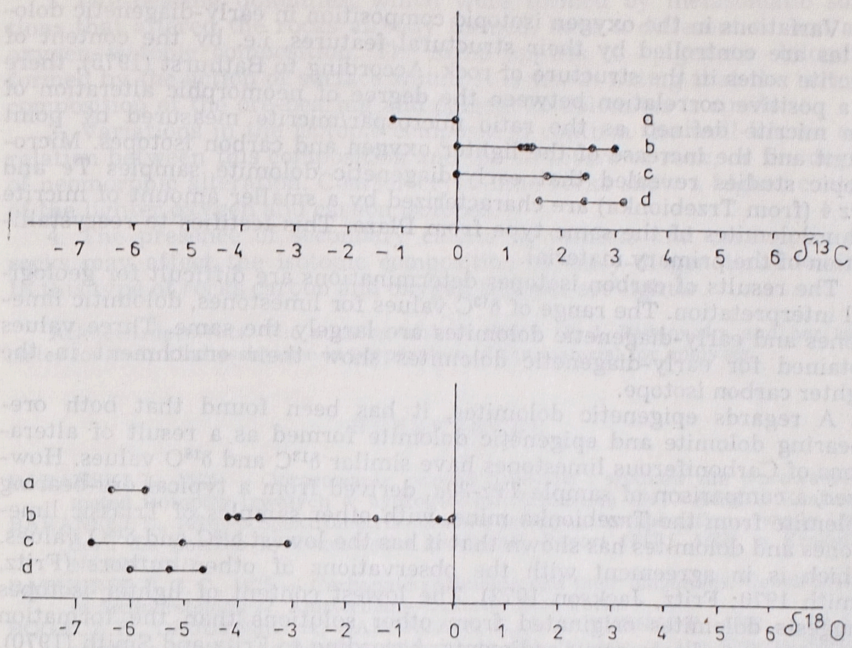

Fig. 2. Carbon and oxygen isotope rations in examined carbonate rocks
a — epigenetic dolostones, *b* — early-diagenetic dolostones, *c* — dolomitic limestones, *d* — limestones

Samples *Trz* 4, Te and *Trz* 3 also represent early-diagenetic dolomite from the same stratigraphic level as the dolomites in Płaza, originating however from the surroundings of ore-bearing dolomites. Lower values than those obtained for the Płaza dolomites may be due to subsequent dolomitization processes. Similarly, sample $P\text{-}C_3$ is a dolomitic limestone, but it differs from the other dolomitic limestones in its location in the border zone of limestones and epigenetic dolomites; therefore, it represents the front of epigenetic dolomitization observed in the Płaza profile

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* Samples obtained from Dr K. Bogacz and Prof. S. Dżułyński. The sample Trz_7 was classified by Bogacz *et al.* 1973 (Table XLIX, Fig. 2 — middle part) as ore-bearing dolomite. However, because of lack of typical petrographical features of ore-bearing dolomite, it was named now as only partly altered early diagenetic dolostone.

** Samples obtained from K. Bogacz.

Variations in the oxygen isotopic composition in early-diagenetic dolomites are controlled by their structural features, i.e. by the content of micrite zones in the structure of rock. According to Bathurst (1975), there is a positive correlation between the degree of neomorphic alteration of the micrite defined as the ratio microspar/micrite measured by point count and the increase of the lighter oxygen and carbon isotopes. Microscopic studies revealed that early-diagenetic dolomite samples *Te* and *Trz* 4 (from Trzebionka) are characterized by a smaller amount of micrite than dolomites of the same type from Płaza. This testifies to recrystallization of the primary material.

The results of carbon isotopes determinations are difficult for geological interpretation. The range of $\delta^{13}C$ values for limestones, dolomitic limestones and early-diagenetic dolomites are largely the same. Three values obtained for early-diagenetic dolomites show their enrichment in the lighter carbon isotope.

A regards epigenetic dolomites, it has been found that both ore-bearing dolomite and epigenetic dolomite formed as a result of alterations of Carboniferous limestones have similar $\delta^{13}C$ and $\delta^{18}O$ values. However, a comparison of sample *Trz-20a*, derived from a typical ore-bearing dolomite from the Trzebionka mine, with other samples of Triassic limestones and dolomites has shown that it has the lowest $\delta^{13}C$ and $\delta^{18}O$ values, which is in agreement with the observations of other authors (Fritz, Smith 1970; Fritz, Jackson 1972). The lowest content of lighter isotopes suggests dolomites originated from other solutions than the formation environment of calcareous sediments. According to Fritz and Smith (1970), secondary dolomites could have formed by the action of solutions containing lighter C and O isotopes compared with the solutions in the environment of precipitation of limestones, or the dolomitization temperature was 10—20°C higher.

Relatively insignificant differences in the isotopic composition of the ore-bearing dolomite and epigenetic dolomite from Czerna, being a dolomitized Carboniferous limestones, suggest that the nature of the dolomitizing solution was not so important. However, the samples discussed do not permit drawing any definite conclusions.

In the ore-bearing dolomites the correlation between the size of dolomite grains and the isotopic composition (Bathurst 1975) has been explicitly confirmed. The ore-bearing dolomites are coarsest-crystalline and simultaneously have the lowest content of lighter isotopes.

DISCUSSION

The foregoing analysis of the isotopic composition is to be treated as preliminary studies. More detailed petrographic investigations and a greater number of isotopic determinations will be able to extend our knowledge about the formation of the rocks. Worth noting, however, are the following regularities:

1. In the group of early-diagenetic dolomites both in Płaza and Trzebionka there are rocks with an isotopic composition suggesting their syngenetic origin with limestones in the same sedimentary basin. No later metasomatic action was necessary for their formation.

2. Ore-bearing dolomites, which were formed by metasomatic solutions that altered the rocks already formed, have a different carbon and oxygen isotopic composition. The same applies to epigenetic dolomites formed by the action of surface water. It is worth noting that the isotopic composition of the ore-bearing and epigenetic dolomites is similar.

3. Variations in the isotopic composition confirm the well-known correlation between this composition and the structure of rock, or the degree of neomorphic alteration. Coarser-crystalline rocks show a higher content of the lighter oxygen and carbon isotopes.

4. The presence of secondary calcite in veinlets and caverns in the rocks may affect the isotopic composition of early-diagenetic dolomites. In this type of rocks carbon and oxygen isotopes are lighter.

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OZNACZENIA SKŁADU IZOTOPOWEGO TLENU I WĘGLA W SKAŁACH WAPIENIA MUSZLOWEGO REJONU CHRZANOWSKIEGO

Streszczenie

Wykonano 14 oznaczeń składu izotopowego tlenu i węgla w wapieniach, wapieniach dolomitycznych i dolomitach, należących do górnych ogniw warstw gogolińskich, górażdżańskich oraz skał węglanowych wieku dolnokarbońskiego. Schematyczny profil badanych serii, skrócony opis petrograficzny oraz zestawienie wyników oznaczeń znajduje się w tabeli 2 oraz na figurach 1 i 2.

Z uwagi na niedużą liczbę oznaczeń artykuł ma charakter przyczynkowy i nie pozwala na wysnucie ogólniejszych wniosków. Nasuwają się jednak pewne prawidłowości w składzie izotopowym badanych skał:

1. Wśród dolomitów wczesno-diagenetycznych wyróżnić można skały, w których charakter izotopowy świadczy o tym, że powstały w tym samym zbiorniku co wapienie.

2. Odrębny charakter składu izotopowego wykazują dolomity kruszconośne, utworzone pod wpływem roztworów zmieniających skały już zdiagenezowane. Skład izotopowy dolomitów kruszconośnych i epigenetycznych utworzonych przez dolomityzację wodami powierzchniowymi jest zbliżony.

3. Zróżnicowanie w składzie izotopowym potwierdza znaną w literaturze zależność od stopnia zmian neomorficznych.

4. Czynnikami zaburzającymi skład izotopowy niektórych próbek dolomitów wczesno-diagenetycznych wydaje się być obecność wtórnego kalcytu, co powoduje że próbki stają się izotopowo lżejsze.

OBJAŚNIENIA FIGUR

Fig. 1. Skład izotopowy $\delta^{13}C$ i $\delta^{18}O$ próbek skał z profilu wapienia muszlowego oraz dwu próbek skał dolno-karbońskich. Z lewej strony wykres procentowej zawartości kalcytu i dolomitu w badanych próbkach
1 — dolomit, *2* — kalcyt, *3* — próbki, w których z powodu zmiennych zawartości kalcytu i dolomitu pomiary ilościowe były niecelowe, *4* — dolomit (skała), *5* — wapień dolomityczny, *6* — wapień, *7* — wapień zlepieńcowy, *3* — wapień z wkładkami krzemieni

Fig. 2. Skład izotopowy tlenu i węgla w badanych próbkach skał węglanowych
a — dolomity epigenetyczne, *b* — dolomity wczesno-diagenetyczne, *c* — wapienie dolomityczne, *d* — wapienie

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ОПРЕДЕЛЕНИЕ ИЗОТОПНОГО СОСТАВА КИСЛОРОДА И УГЛЕРОДА В СРЕДНЕТРЯСОВЫХ ПОРОЯХ РАЙОНА ХЖАНОВА

Резюме

Было сделано 14 определений изотопного состава кислорода и углерода в известняках, доломитных известняках и доломитах принадлежащих верхним звенам гоголинских слоёв, гураждканьских слоёв и карбонатным породам нижнекаменноугольной эпохи. Схематический профиль изучаемых ярусов, сокращённое петрографическое описание и сопоставление итогов определений находятся в таблице 2 и на фигурах 1 и 2.

Из-за небольшого количества определений эта статья имеет лишь характер приложения к вопросу о изотопном составе выше упомянутых элементов. Это является тоже причиной отсутствия более широких выводов. Но очевидны некоторые закономерности изотопного состава изучаемых пород:

1. Среди раннедиагенетических доломитов можно выделить породы, в которых изотопный характер является доказательством их образования в одном бассейне с известняками.

2. Другой характер изотопного состава имеют рудоносные доломиты образованые, образованных под воздействием растворов, изменяющих породы подверженные прежде диагенезису. Изотопный состав рулонсных и эпигенетических доломитов, образованных из-за доломитизации поверхностными водами, очень близкий.

3. Различия в изотопном составе подтверждают известную по литературным данным зависимость от степени неоморфических перевращений.

4. Фактором, который вызывает нарушение изотопного состава некоторых образцов раннедиагенетических доломитов, может быть присутствие вторичного кальцита, приводящего к тому, что образцы кажутся изотопно более лёгкими.

ОБЪЯСНЕНИЯ К ФИГУРАМ

Фиг. 1. Изотопный состав $\delta^{13}C$ и $\delta^{18}O$ образов пород из среднетрясового профиля и двух образцов нижнекаменноугольных пород. С левой стороны — график процентного содержания кальцита и доломита в изучаемых образцах
1 — доломит, *2* — кальцит, *3* — образцы, в которых из-за переменного содержания кальцита и доломита количественные измерения были нецелесообразными, *4* — доломит (порода), *5* — доломитовый известняк, *6* — известняк, *7* — конгломератный извесняк, *8* — известняк с включениями кремния

Фиг. 2. Изотопный состав кислорода и углерода в изучаемых образцах карбонатных пород
a — эпигенетические доломиты, *b* — раннедиагенетические доломиты, *c* — доломитные известняки, *d* — известняки