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AN INVESTIGATION OF THE PERMEABILITY OF LARGE COAL SAMPLES TO THE FLOW OF GASES OF VARIOUS SORPTION ABILITIES

BADANIE PRZEPUSZCZALNOŚCI DUŻYCH PRÓBEK WĘGLA W PRZEPŁYWIE GAZÓW O ZRÓŻNICOWANYCH ZDOLNOŚCIACH SORPCYJNYCH

Experimental investigations of the flows of gases of various sorption abilities through large samples (volume over 1000 cm^3) cut from blocks of original rock material were performed. The authors measured the magnitudes and the rates of saturation of coal samples for pre-determined values of input gas pressure (greater than 0.05 MPa and less than 0.2 MPa). For the experiments the sorbing gases encountered in mines (methane, carbon dioxide) were used. For the purposes of comparison the flows of slightly sorbing gas (nitrogen) and non-sorbing gases (helium, argon) were also investigated. For the gases investigated the permeability of samples and the dependence of that permeability on gas pressure were determined. The authors have attempted to find a correlation between sample permeability and sample saturation rate.

Key words: coal, gases flow, swelling, sorption, permeability

Celem prowadzonych badań było rozpoznanie zjawisk zachodzących w wycinanych dużych próbkach węgla (ponad 1000 cm^3) w wyniku oddziaływania węgla z gazami różniącymi się zarówno wielkością cząsteczek, jak i charakterem chemicznym. Duże próbki węgla są bardziej zbliżone do węgla znajdującego się w pokładzie, oprócz mikroporów zawierają bowiem makropory, szczeliny, kawerny i płaszczyzny łupliwości. Taka struktura porów węgla odpowiada za zróżnicowany przebieg oddziaływania węgla z przepływającymi gazami.

Wycięta prostopadłościenna bryła węgla wklejona była do stalowego kształtownika pomiędzy równoległymi płytami czołowymi, w których znajdowały się zawory pozwalające na doprowadzenie stosowanego gazu do próbki oraz jego wypływ do atmosfery. Ciśnienie gazu mierzono przy użyciu czujników na wejściu do próbki oraz rozmieszczonych równomiernie na jej pobocznicy (rys. 1). W takich próbkach wycinanych z brył węgla pochodzącego z kopalni Śląsk prowadzono badania przepływu gazów o zróżnicowanych zdolnościach sorbowania: azotu (głównego składnika powietrza), niesorbujących się: helu i argonu, lecz przede wszystkim metanu i dwutlenku węgla (czyli gazów występujących w atmosferze kopalni i często stanowiących duże zagrożenie w trakcie prac prowadzo-

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nych w górnictwie podziemnym). W celu doboru odpowiednich próbek, przed przystąpieniem do właściwych badań, charakteryzowano wycięte próbki wyznaczając ich przepuszczalność standardową w przepływie azotu. Badania nasycania węgla i przepływu przez nie gazów prowadzono na próbkach różniących się porowatością (1,31 i 2,23%) i przepuszczalnością (43 i 247 mD). Na wejściu do próbki stosowano nadciśnienia gazu 0,05–0,2 MPa, przy których określano stopień i szybkość nasycania węgla tymi gazami. Łatwy dopływ gazu z wnętrza próbki do otworu pomiarowego (średnica otworu 2 mm, głębokość 5 cm) sprawiał, że ustalone ciśnienie gazu osiągano bardzo szybko. Narastanie ciśnienia gazu w poszczególnych otworach pobocznicy próbki rejestrowane w funkcji czasu jest więc najbardziej istotne na początku każdego eksperymentu (rys. 2 i 6). Za miarę nasycenia próbki gazem przyjęto stosunek nadciśnienia rejestrowanego aktualnie w danym otworze (na pobocznicy próbki) do nadciśnienia gazu zadanego aktualnie na jej wejściu. We wszystkich otworach próbki W01 (przepuszczalność 43 mD) czasy narastania ciśnienia są dla każdego gazu wyraźnie dłuższe niż dla próbki W02 (247 mD). Zaobserwowano też że wpływ rodzaju gazu, a zatem jego sorbowalności, jest tym większy, im mniejsza przepuszczalność badanej próbki (tabl. 1).

W każdym otworze pomiarowym nasycanie gazami przebiega w następującej kolejności: He, Ar, N_2, CH_4, CO_2, zgodnej z sorbowalnością stosowanych gazów. Już we wcześniejszych pracach (Żółcińska, Dyrga 1996; Żółcińska, Gudowska 1983) autorzy stwierdzili, że spośród przebadanych gazów dwutlenek węgla najlepiej sorbuje się na węglu kamiennym. Powoduje to wnikanie jego cząsteczek do wnętrza struktury węgla, jej rozluźnienie i pęcznienie, co znajduje swoje odbicie w przebiegu nasycania węgla gazem oraz jego przepuszczalności w przepływie danego gazu.

Rozkłady przepuszczalności wzdłuż badanych próbek wyznaczone przy ustalonym rozkładzie ciśnień wzdłuż próbki wskazują na dużą niejednorodność nie tylko pomiędzy poszczególnymi próbkami, lecz również wewnątrz każdej z nich (rys. 4 i 5). Pozwalają one określić dostępność poszczególnej części badanego węgla dla przepływającego gazu.

Zależność przepuszczalności badanych próbek od ciśnienia przepływającego gazu pokazuje dla obu badanych próbek, że ze wzrostem średniego ciśnienia gazu (zarówno sorbującego, jak i niesorbującego) przepływającego przez próbkę następuje spadek przepuszczalności, podobnie jak dla próbek małych (Żółcińska, Dyrga 1996). Wyznaczona przepuszczalność węgla jest tym wyższa, im mniej sorbuje się gaz przepływający przez niego. Szybkość zmiany przepuszczalności węgla rośnie bardzo wyraźnie z sorbowalnością użytych gazów i jest bardziej widoczna w próbce o niższej przepuszczalności.

Słowa kluczowe: węgiel, przepływy gazów, pęcznienie, sorpcja, przepuszczalność

1. Introduction

The purpose of the research was to study phenomena taking place as a result of the interaction of coal with gases differing in respect to both the size of their molecules and their chemical character in large samples cut from blocks of mined coal i.e. exceeding 1000 cm^3. Large coal samples resemble the coal as it actually exists in a coal-bed more closely as, apart from micro-pores, they contain macropores, fractures, cavities and cleavage planes. Such a coal pore structure is responsible for the diversity in the interaction between the coal and the flowing gases.

In order to examine the flows, the following gases have been used: nitrogen (the main component of air), non-sorbing gases (helium and argon) as well as methane and carbon dioxide, which closely resemble the gases that actually appear in a coal-mine atmosphere and are frequently a source of considerable danger during underground mining activities. The extent and the speed at which a stable level of gas-saturation of coal was reached were measured under pre-determined gas pressures.

Earlier tests of coal permeability only used small mined samples (Harpalani, Schraufnagel 1990; Gamson et al. 1993; Żółcińska, Dyrga 1996) or coal briquettes produced by compressing coal granules of various sizes under different pressures (e.g. Dyrga et al. 1992).

2. The method of measurement

A cuboidal coal lump cut from mined material was glued into a steel sleeve between parallel end plates containing valves, which made it possible both to introduce the gas to

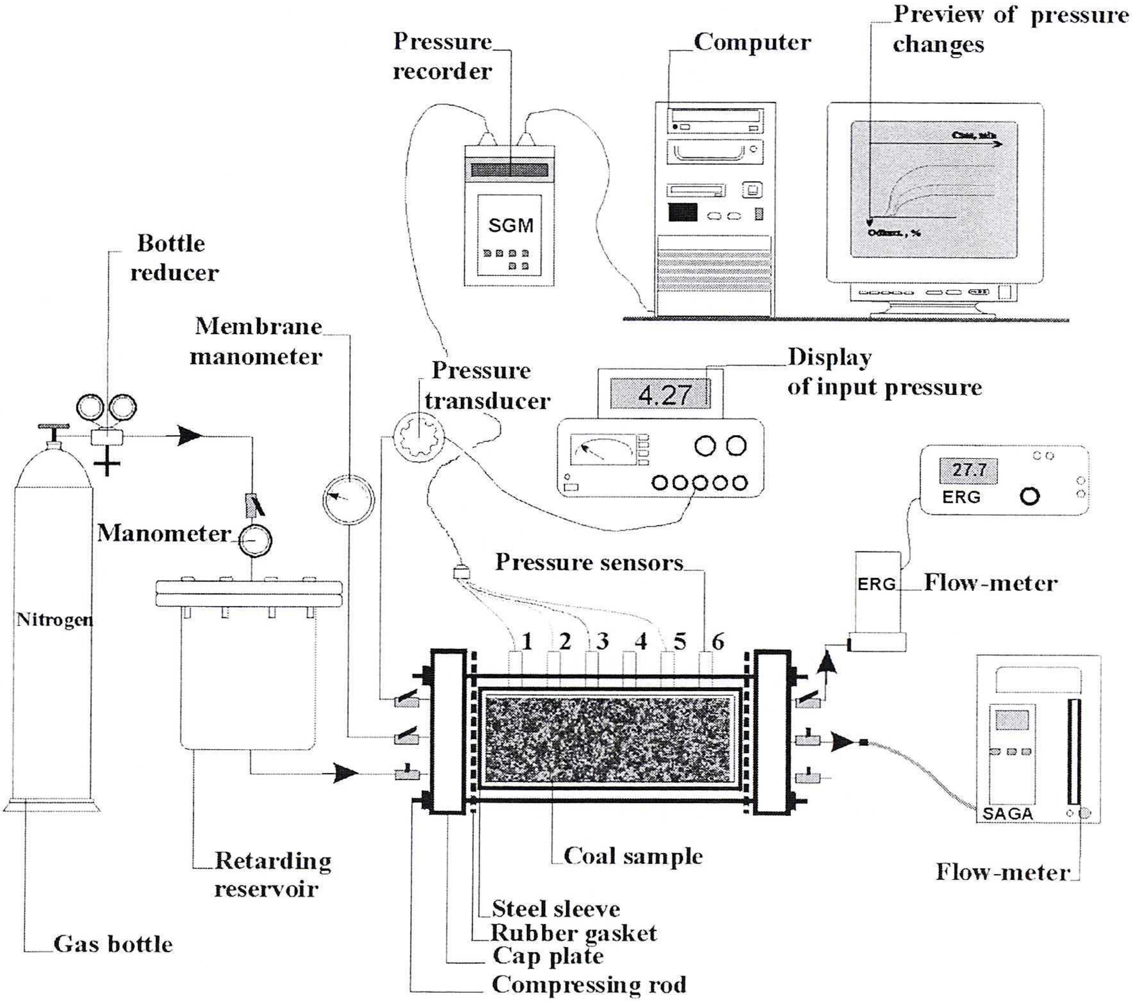

Fig. 1. The layout of the equipment for the flow investigations of gas flow

Rys. 1. Schemat aparatury do pomiaru przepływu gazów the sample and to release it to atmosphere. The source of gas was a bottle containing the gas being used at a particular experiment. The gas was passed through a reduction valve to the sample at a pre-determined pressure. The gas pressure at the inlet to the sample was measured with the use of a pressure transducer type P200C. Sensors placed equidistantly on the side surface of the rig transmitted the signals to a four-channel SGM recorder (Dyrga, Żółcińska 1999). The frequency with which sensor readings were recorded was adapted to an appropriate level for the experiment. The data obtained from the recorder were fed into a computer for their further processing. When the outlet was opened, gas pressure changes were measured on a time basis, together with quantitative measurement of the gases discharged with the aid of an appropriate instrument. The layout of the measuring equipment is illustrated in Fig. 1.

The coal that formed the subject of the examination had been obtained from the "Śląsk" mine, coal bed 502. In the dry air state such coal contains $C^a = 83\%$, $V^a = 28.2\%$, $A^a = 4.8\%$, $W^a = 0.8\%$. From the coal-mass cuboidal samples were cut for the purposes of the research. Each sample was about 16–17 cm long with a square cross-section of about 8 × 8 cm. On the sides of each sample in the steel section, sensors for measuring the pressure of the flowing gas were installed at equal intervals.

In order to select the most suitable samples, the tests themselves had been preceded by finding the characteristics of the samples, which was effected by determining their standard permeability to nitrogen flow.

3. The effect of gas-flow on coal

Coal is a fracture-porous medium of diversified structure as well as being an essentially porous medium. The impact of the type of pores upon the phenomena occurring in coal as a result of flowing medium is represented by Seewald's coal model(1985). Micro-pore areas are surrounded by fractures and cleavage planes, which serve as flow channels. The high pressure of the passing fluid leads to shrinkage of the micro-pores and the dilatation of the flow channels without damaging the coal's texture. The bag-like micro-pores and therefore the coal matrix, become filled by a process of diffusion, which results in their swelling. This in turn affects the macro-pore system, causing narrowing of the flow channels in the coal. Thus, due to the micro- and macro-pore systems existing in coal its permeability is closely connected with the type and amount of the fluid sorbed during the flow. The accumulation of gases in the coal is primarily dictated by the presence of the micro-pores. The gas flow, however, proceeds along various routes as it is dependent on the structure of the pores, the cavities and the fractures present in the coal (Gamson et al. 1993). Their dimensions vary from a few micrometres to as much as tens of centimetres. The penetration of gas into the coal depends on the interconnecting systems of pores and fractures within it. If the gas can penetrate the flow channels relatively easily, the coal-mass will become saturated over a short time-period.

4. The influence of gases of different sorbability on the speed of coal saturation

The research on coal saturation and gas flows in it was conducted on samples of varying porosity and permeability (W01 — 1.31%, $43 \cdot 10^{-15}m^2$, i.e. 43 mD and W02 — 2.23%, 247 mD). At the entrance to the sample gas, under pressures ranging from 0.05 to 0.2 MPa, was introduced. The relatively unimpeded flow through the sample to the first measuring aperture (2 mm in diameter, 5 cm in depth) was the reason why stable gas pressure was achieved almost immediately (in a few seconds) and simultaneously in all the measuring holes apertures. Thus, the increase of gas pressure in each of the side apertures, registered on a time function, is critical at the initial stage of every experiment. The stabilisation of the relationship between the inlet pressure and that registered at a particular side aperture was taken as an indication that the sample had become saturated with gas.

For example, the pattern of increase in pressure for the gases applied at the first, second and third apertures of sample W01, with an entrance pressure of 0.1 MPa, is shown in Fig. 2.

At each aperture saturation with gas takes place in the following order: He, Ar, N_2, CH_4, CO_2. The first gas to reach the interior of the sample was helium, easily penetrating the coal structure due to its small and chemically inert molecules; it was followed by the larger and virtually inactive argon molecules or the negligibly sorbing molecules of nitrogen. The slowest to arrive were methane and carbon dioxide molecules. At all apertures saturation was completed after about 30 seconds for all gases. This process is very quick (measurement interval 3 seconds), yet the results obtained in the experiment indicate a distinct dependence of the coal's saturation with gases on the sorbability of the gases applied.

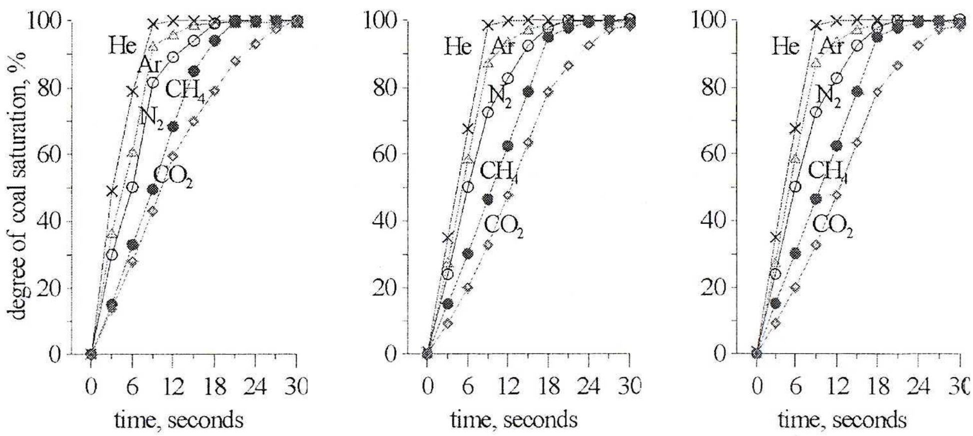

Fig. 2. The course pattern of pressure increase in the first, second and third measuring aperture of sample W01

Rys. 2. Przebieg narastania ciśnienia gazów w pierwszym, drugim i trzecim otworze próbki W01

Sorption of gases on identical sorbents increases in the series: He-Ar-N_2-CH_4-CO_2 (Kriwickaja et al. 1973) . The sorbing ability of coal does not correlate precisely with the molecular size of the gases applied since the chemical character of the sorbate is also an important factor. With approximately similar sized molecules, it is carbon dioxide that shows the greatest affinity to coal. Of all the gases applied, not only its quadrupolar moment, but also the polarizability of its particles is the highest.

In earlier studies (Żółcińska, Dyrga 1996; Żółcińska, Gudowska 1983) the authors had also observed that, of all the gases tested, it is carbon dioxide that is best sorbed in hard coal. Its molecules penetrate into coal structure, causing it to loosen and swell. In addition to the diffusive flow of gas dissolved in the coal substance, there also appears a diffusive flow in the adsorbed layers, which causes a diminution of the cross-section of the coal pores accessible to the flowing gas as well as the rate at which a sample becomes completely saturated. Because of the small differences between various gases only the results of saturating sample W01 in a flow of helium at a pressure of 0.1 MPa have been shown in Fig.3.

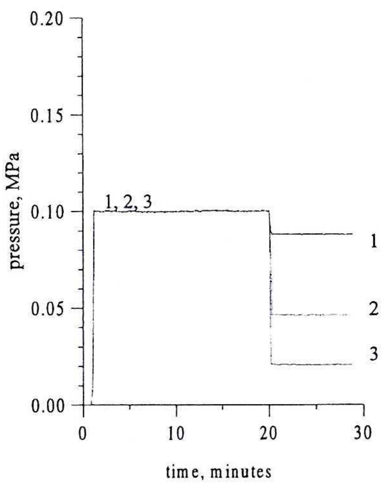

Fig. 3. The pressure variations in sample W01during its saturation with helium and its flow

Rys. 3. Zmiany ciśnienia w czasie nasycania próbki W01 helem i następnie przepływu przez nią gazu

The lack of marked differences is caused by the considerably greater permeability of sample W01 at its inlet than in the outlet area (Fig. 4). In the first part of this sample there are probably a greater number of transportation pores facilitating the flow of gas (Dyrga, Żółcińska 1999). On the horizontal axis of Figs. 4 and 5 the position of the measuring aperture has been marked and expressed as a percentage of the total length of the sample.

The distribution patterns of permeability along the samples under investigation indicate a considerable heterogeneity not only between the different samples, but also within each of them. They make it possible to determine the accessibility of the flowing gas for each part of the coal being tested.

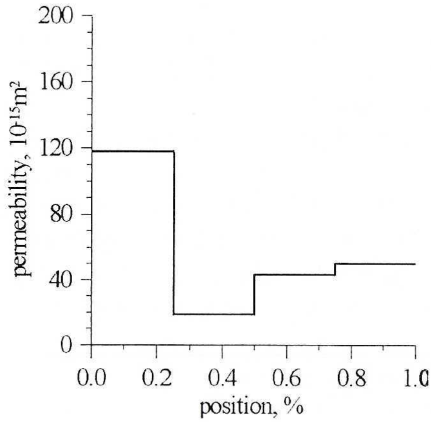

Fig. 4. The variations of permeability in different parts of the sample W01

Rys. 4. Zmiany przepuszczalności węgla wzdłuż próbki W01

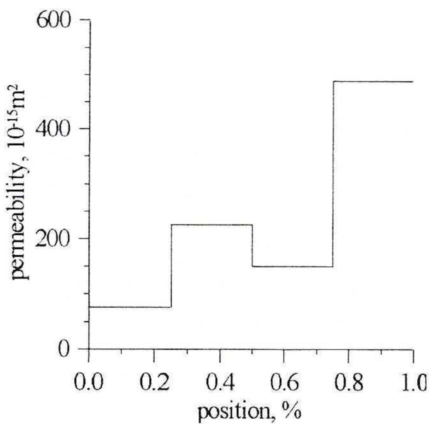

Fig. 5. The variations of permeability in different parts of the sample W02

Rys. 5. Zmiany przepuszczalności węgla wzdłuż próbki W02

Coal, treated as a molecular sieve, can interact with the flowing gas in many ways, in respect of both the size and the chemical character of its molecules (Żółcińska, Dyrga 1996; Kriwickaja et al. 1973). The effect of this interaction is reflected in the patterns gas flow changes in the samples tested and therefore in the changes of their permeability. This change is effected by, primarily, gas slippage on the coal pore walls (known and described as the Klinkenberg effect (Rose 1948; Harpalani, Chen 1993) as well as by changes occurring in the coal structure which are caused by the flowing gas (Harpalani, Chen 1993; Bustin 1997).The interaction of the coal with the fluid always produces further significant changes in the coal structure. It has been observed (Bustin 1997) that the shrinkage of coal during the desorption of gas from it (change in volume less than 2%) can result in as much as a twelve-fold increase in permeability whereas the effects of gas slippage can cause no more than a five-fold increase.

In the case of non-sorbing helium the whole process of permeability change may be ascribed to the slippage effect of its particles on the pore walls. The flow of a sorbing gas (methane, carbon dioxide) makes its possible to estimate the proportional contribution of structural changes in the coal structure to the observed changes of coal permeability.

Similarly, for sample W02, no distinct qualitative diversification was observed with respect to the type of gas applied in the course of the experiment. Nevertheless, at particular measurement apertures the changes in pressure during the saturation of the sample took place over a different time period — Fig. 6 (at saturation pressure of 0.1 MPa). A significant differentiation in the velocity of pressure increase for each gas can be observed only at the first aperture i.e. the one most proximal to the inlet. At this measuring site full saturation for all gases was effectively complete after ten minutes. Despite the high permeability of this sample, as determined by the flow of different gases, the gas flow in the initial area of the sample is clearly blocked and the permeability decreases (as can be seen in Fig. 5). This would seems to indicate the presence of less accessible pores in the area. On the other hand, the flow of gases through the more distant regions of the sample was very fast. At the second aperture complete saturation of the sample was achieved after about 12 seconds for helium and argon, slightly later for nitrogen and after about 24 seconds for methane and carbon dioxide. The time intervals to reach saturation at the third hole were even shorter. This indicates that the measuring stations were closely connected with the system of transport pores (fractures) appearing in this part of the coal.

Analysis of Fig. 2 (sample W01) and Fig. 6 (sample W02) leads to certain conclusions concerning the relationship between the results obtained for the flow of different gases in coal samples of variable permeability. The gas pressure in a given measuring

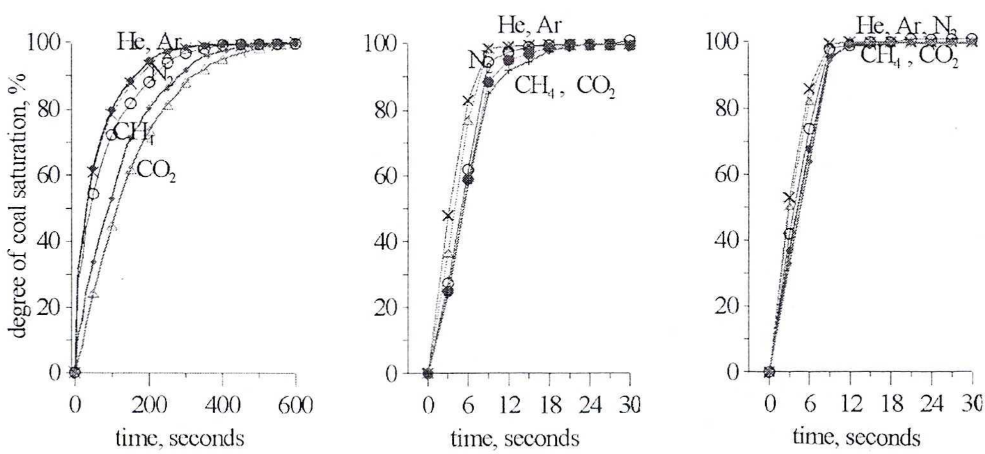

Fig. 6. The pattern of pressure increase in the first, second and third measuring aperture of sample W02

Rys. 6. Przebieg narastania ciśnienia gazów w pierwszym, drugim i trzecim otworze próbki W02

TABLE 1

The time for the samples to become saturated with various gases

TABLICA 1

Czas nasycania próbek poszczególnymi gazami

Gas	SampleW01	Sample W02
He	100	100
Ar	169	104
N_2	196	138
CH_4	221	169
CO_2	304	177

aperture was regarded as stable if the rate of increase in permeability between two subsequent measurements (at the interval of 3 seconds) did not exceed 1%.

Assuming the time of saturating the sample with helium as 100%, the time of pressure stabilisation for other gases was estimated at each of the three apertures. The values for each sample are compared in Table 1. The juxtaposition of the results clearly shows that the time periods of pressure increase for sample W01 (permeability 43 mD) are distinctly longer for all gases than is the case with sample W02 (permeability 247 mD). It confirms earlier observations which noted that the influence of the type of gas and, consequently its sorbability is greater when the permeability of the sample is lower. After the sample was completely saturated, the outlet was opened and the flow of released gases was studied. During the gas flow the pressure became stable almost immediately, although at different levels at particular measuring apertures.

5. The determination of the permeability of coal to the flow of different gases

The absolute permeability of samples was determined on the basis of the flow of helium, argon, nitrogen, methane and carbon dioxide from the measurements at the stabilised pressure differential between the inlet and outlet of the sample. The coal samples selected for investigation differ considerably, not only with respect to their porosity values, but also in their permeability, although they had been obtained from the same coal bed. The determination of permeability for sample W02 (high permeability) was accompanied by certain difficulties caused by the vast discharge of gas (e.g. at a pressure of 0.2 MPa at the sample inlet, the discharge of methane exceeded 14 $dm^3/min.$). The change in the permeability of both samples in the flow of different gases depending on the pressure of the gas applied are shown in Figs. 7 and 8.

For both samples tested, the increase in the mean pressure of the flowing gas (both sorbing and non-sorbing gases) is accompanied by a decrease in permeability, as was the case small samples were investigated (Żółcińska, Dyrga 1996). The permeability values

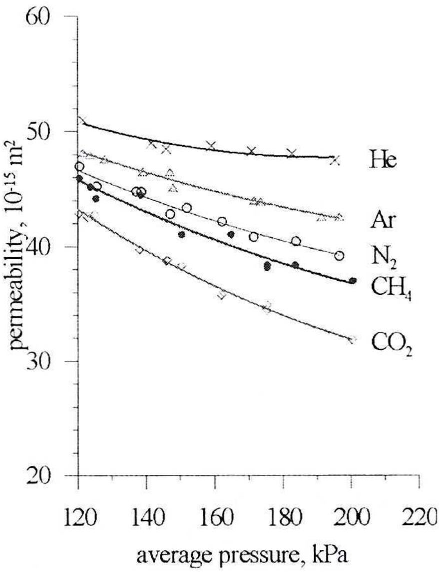

Fig. 7. The permeability variations in sample W01with the increase of gas pressure

Rys. 7. Zmiany przepuszczalności ze wzrostem ciśnienia gazu w próbce W01

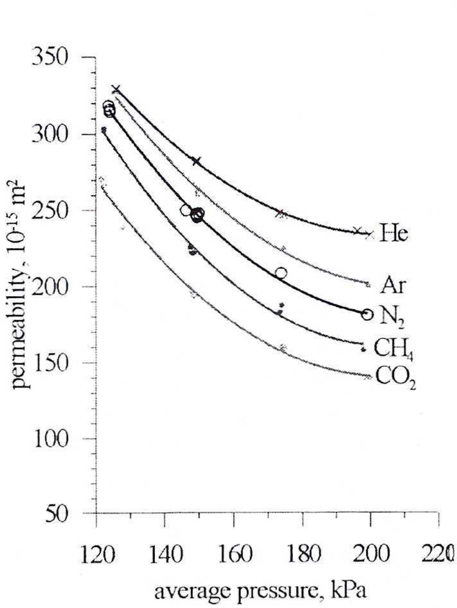

Fig. 8. The permeability variations in sample W02 with the increase of gas pressure

Rys. 8. Zmiany przepuszczalności ze wzrostem ciśnienia gazu w próbce W02 of the coal are also strongly dependent on the type of gas flowing; the less gas is sorbed, the higher the permeability of the coal is.

The rate of change in coal permeability (evaluated as the tangent of permeability change inclination in relation to the flowing gas pressure) grows very markedly with the sorbability of the gases applied. The changes in this rate are particularly evident in sample W01 (of lower permeability) since the sorption of gas diminishes the inside diameters of the transport channels to a greater extent than in the case of the other sample.

6. Conclusions

The research on gas flow conducted in large samples cut from mined coal-masses provides a closer approximation of the gas-flow in real mining conditions than can be simulated in tests carried out on small samples. Large samples contain not only micropores but macropores, fractures, cavities and laminar cleavage planes as well; therefore they more closely resemble the coal present in the actual coal-bed.

The saturation of coal and the flows of gases through it have been investigated with the use of gases appearing in the coal mine: nitrogen, methane and carbon dioxide (of different sorptive capacities) as well as helium and argon (inert gases, capable of penetrating the coal structure more easily).

On the basis of the results of the research, the following conclusions have been drawn:

- the lower the sorbability of a gas in coal, the higher the rate of coal saturation;
- the lower the sorbability of a gas, the higher the permeability of the coal to gas flow;
- the rate of changes in permeability with the increase in gas pressure grows very distinctly with the sorbability of the gases applied (due to the swelling effect). These changes in rate were more noticeable in the sample of lower permeability.

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