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Control of Oxygen Concentration by Using a Carbonaceous Substance

Mohanad Jadan, Jihad Said Addasi
Department of Basic Sciences, Tafila Applied University College
Al–Balqa' Applied University, P.O. Box 40, Al-Eys – 66141, Tafila, Jordan

Abstract: The control of oxygen concentration in gas flow may be used in chemical industry, heat power engineering, ecology, automobile construction and other industrial branches. This control is realized over a broad range of oxygen concentrations. The control of the oxygen concentration is based on passing of gas flow through a measuring cavity of radio spectrometer and measurement of a magnetic resonance signal. A change in the magnetic resonance signal of a dispersed carbonaceous substance, placed into the cavity, indicates to the changes in oxygen concentrations. The dispersed anthracite and thermal treatment cellulose substance in the oxygen-free medium are proposed to use as a carbonaceous substance.

Key words: Oxygen Concentration, Control, Gas Flow, Anthracite, Cellulose

INTRODUCTION

The control of oxygen concentration of gas flow relates to the technology of magnetic resonance. This technology may be widely used in chemical industry, heat power engineering, ecology, automobile construction and other industrial branches where control of oxygen concentration in gas flow is required. The concentration of oxygen can be determined by chemical methods, involving a sample of gas in measurement of the number of oxygen atoms in this sample. This sample of gas participates in the characteristic chemical reactions to measure the concentration of oxygen [1]. This method is unsuitable for the continuous control of oxygen concentration in gas flow. The optical methods of control over the concentration of gasses [2] are based on estimation of the optical characteristics (e.g. optical absorption) at a characteristic frequency of the Infrared Range (IR). However, these methods fail to solve the problem defined due to the fact that oxygen molecules have no specific IR-absorption lines.

The radio-spectroscopic method based on paramagnetic absorption of the radio-frequency energy directly by the molecules of oxygen, passing through the measuring cavity of radio spectrometer [3]. While having a rather high measuring rate, the last pointed method features fairly low sensitivity characteristic to gas radio-spectroscopy. The low sensitivity of radio-spectroscopic method is due to a limitation on the number of paramagnetic molecules in a gas. This gas, filling the measuring cavity, is characterized by low coefficients of magnetic resonance filling. The efforts to improve sensitivity by thus far proposed solutions of signal accumulation in time prevent the problem of control of oxygen concentration from being handled.

The principal object of this problem is to provide fast control over a broad range of oxygen concentrations. The control of oxygen concentration is realized by measurement of change in the magnetic resonance signal of a dispersed carbonaceous substance, putting into the measuring cavity of radio spectrometer. The change of this magnetic resonance signal is an indicative of the changes in the oxygen concentration in gas flow. The dispersed anthracite and thermal treatment cellulose in the oxygen-free medium can be used as a carbonaceous substance.

Theory: A magnetic resonance signal of a carbonaceous substance in oxygen-free medium is determined by the odd electrons localized at the bulk defects and near the surface of small-sized carbon clusters (micro crystallites). The magnetic resonance signal of non-dispersed anthracite with $g=2.0027\pm0.0003$ and $\Delta H=1\text{-}2$ Gauss (G) usually contains a component insensitive to the oxygen atmosphere, associated with the localized odd electrons within the micro crystallite bulk and the component sensitive to the content of oxygen due to electrons localized near the crystallite surface.

Dispersion of anthracite results in reduced number of the bulk-localized spins (decrease in the background signal component insensitive to the oxygen concentration). In addition dispersion of anthracite increases the oxygen-sensitive component due to the near-surface odd electrons and also increases the rate of oxygen sorption or desorption by the dispersed substance. This gives a considerable improvement in speed of control. Paramagnetic oxygen molecules reversibly sorbitized at the surface of small-sized carbonaceous clusters, owing to the magnetic effect of odd electrons on there surfaces, cause acceleration of relaxation processes of the paramagnetic energy and radical broadening of the initial magnetic-resonance line. This brings about a considerable lowering of the recorded signal intensity with g=2.0027. Each odd electron localized near the surface of a small-sized carbonaceous cluster is an effective spin mark responding to the presence of oxygen molecules. The increasing (decreasing) of the oxygen concentration in gaseous medium with a carbonaceous substance causes the decreasing (increasing) of intensity of a narrow-band signal for this mark. The efficiency of this process is improved due to the possibility of an optimal localization of the carbonaceous substance in the magnetic component loop of a microwave field in the cavity of radio spectrometer.

The interaction between the bulk-localized spins in the non-dispersed carbonaceous substance and the oxygen molecules is rather prolonged, cause this interaction can be neglected. In this process of oxygen sorption (desorption) the stabilization period for the intensity of Electron Paramagnetic Resonance (EPR) signal can be some minutes or some days. It means that the using of non-dispersed carbonaceous substance is unsuitable for the objective to be achieved fast oxygen control over a wide range of oxygen concentrations. The using of dispersed carbonaceous substance gives a fast response with stabilization period of several seconds for a wide range of oxygen concentrations (10^{16} - 10^{20} cm^{-2}).

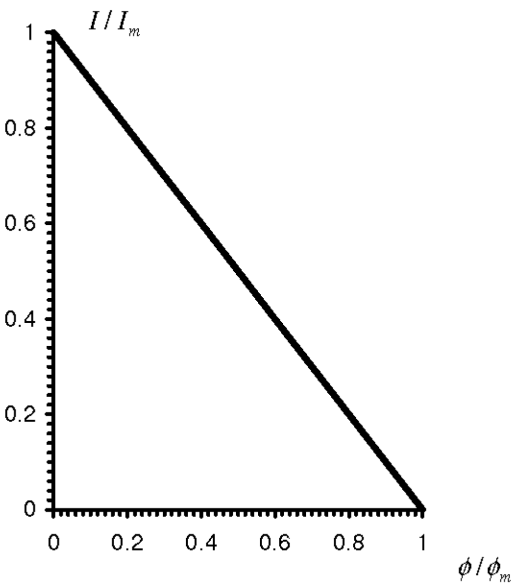

Fig. 1: Dependence of Intensity of Magnetic Resonance Signal I/I_m on Oxygen Flow ϕ/ϕ_m

Figure 1 represents a mechanism used in the control of the oxygen concentration. The intensity of signal of magnetic resonance for the dispersed substance in vacuum is I_m. For passing oxygen flow the signal intensity is decreasing and for a particular value Φ_m the signal of a sample is totally suppressed. The signal intensity I is related to the flow quantity Φ by the following equation:

$$I = I_m \cdot (1 - \Phi / \Phi_m) \qquad (1)$$

From this it is inferred that the quantity of oxygen flow Φ is related to the intensity of a magnetic resonance signal I by the following equation:

$$\Phi = \Phi_m \cdot (1 - I / I_m). \qquad (2)$$

To use eq. (2), the value of Φ_m is determined by using calibration method. The calibration gas flow Φ is used to find the intensity of a magnetic resonance signal I_c and then Φ_m is found from the following equation:

$$\Phi_m = \Phi_c \cdot I_c / (I_m - I_c). \qquad (3)$$

RESULTS AND DISCUSSION

Figure 2 represents a functional diagram of gas analyzer. The carbonaceous substance (2) can be placed into the measuring cavity (3) of electron paramagnetic resonance spectrometer with a recording unit (4). A portion of gas flow from the main gas line (9) is passing through the input coupler (7) and input block (5) to sorption chamber (1) located in the measuring cavity (3). From the sorption chamber (1) the gas flow is passing through the output block (6) and the output coupler (8) into the main gas line (9).

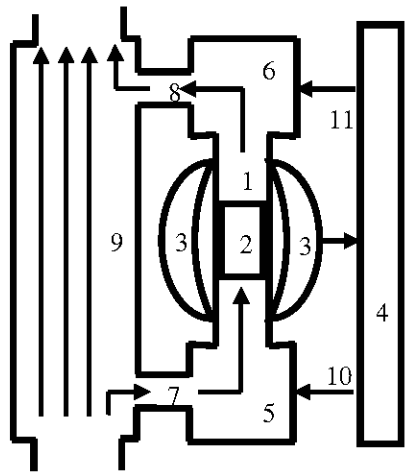

Fig. 2: Functional Diagram of Gas Analyzer, Where: 1-Sorption Chamber; 2-Carbonaceous Substance; 3-Measuring Cavity; 4-Recording Unit; 5-Input Block; 6-Output Block; 7-Input Coupler; 8-Output Coupler; 9-Main Line of Gas Flow; 10-Input Controller; 11-Output Controller

Molecular oxygen of the gas flow, passing through the carbonaceous substance (2), reduces the intensity of a magnetic resonance signal of this carbonaceous substance. This signal is further invariable provided the oxygen concentration in the gas flow remains unchanged. The intensity of this magnetic resonance signal is increasing (decreasing) with a decreasing (increasing) in the oxygen concentration in gas flow. Intensity variations of magnetic resonance signal of dispersed carbonaceous substance are determined by the reversible physical sorption of oxygen molecules at the surface paramagnetic centers of the carbonaceous substance.

Cellulose matter thermal treatment in oxygen-free medium is an efficient carbonaceous substance. The intensity of magnetic resonance signal of such this carbonaceous substance is a good indicator to the changes in the oxygen concentration in the surrounding atmosphere. This process (thermal treatment of cellulose matter) produces a stable extra fine-grained carbonaceous substance in oxygen-free medium has a relatively narrow-band (ΔH = 0.5-1.5 G) signal of magnetic resonance with g=2.0026 $\pm$0.0002 and intensity ~ 10^{22} sp g^{-1}.

In gaseous atmosphere with enhanced content of oxygen the signal intensity, for g=2.0026, is rapidly decreasing, producing a broadband signal, super-Lorentzian with form of oxygen-carbon complexes. This rapidly decreasing is due to reversible sorption of oxygen molecules at the localization site of the odd electron of carbonaceous substance. It should be noted that measurement of the signal intensity for the above-mentioned complexes provides an additional possibility to register an enhanced concentration of oxygen. This allows for the creation of particularly fast-response and sensitive objects for control of oxygen concentration over a broad range. These properties of cellulose are formed owing to a specific high-temperature modification of the carbonaceous substance in oxygen-free medium yielding extended low-dimensional structures, in which the majority of carbon atoms have an odd electron. As a consequence, access of the paramagnetic oxygen molecule to paramagnetic centers of the oxygen-sorbing substance is provided in case of their abnormally high local concentration.

The carbonaceous substance, with establish paramagnetic characteristics, is used as a source of information about the current oxygen concentration. In other cases its difficult to control the characteristics of the measured gaseous oxygen, which are greatly dependent on the external conditions (pressure, temperature, concentration, etc.). Last conditions decrease the accuracy of the measurement. This source of serious errors is excluded principally for proposed cellulose matter.

The anthracite and thermal treatment Cellulose can be used as a carbonaceous substance. Before dispersion of anthracite, the anthracite in the atmosphere of oxygen has exhibited the intensity of a magnetic resonance signal coming to $2{\cdot}10^{19}$ sp g^{-1} with factor g=2.0027. In vacuum the intensity after 30 sec. was growing up to value 4 $\cdot10^{19}$ sp g^{-1} due to oxygen desorption from the crystallite surface of solid anthracite. After dispersion down to the grain dimensions of 1-20 μm, the signal intensity in the oxygen atmosphere at g=2.0027 decreased to $2{\cdot}10^{17}$ sp g^{-1}, where as in vacuum it increased to $8{\cdot}10^{19}$ sp g^{-1} and setup time of a signal equilibrium value as a result of anthracite dispersion was reduced to 1 second. Further the sample of dispersed anthracite of 10 mg was used as a working substance for control of oxygen concentration in a gas flow. The gas flow is passing through a sorption chamber of the measuring cavity in magnetic resonance radio spectrometer of 3 cm range and any change in the magnetic resonance signal with g=2.00027 is displayed. The calibration has demonstrated that variation of the signal intensity from $2{\cdot}10^{15}$ sp to 10^{18} sp was associated with changes of the oxygen content in a gas flow from $3{\cdot}10^{18}$ to 10^{16} cm^{-2}.

The cellulose with thermal treatment at 800° C in oxygen-free medium is used as a carbonaceous substance. After this thermal treatment for cellulose the intensity of the magnetic resonance signal for 10 mg sample with g=2.0026 and ΔH = 1.4 G was correlated with $1.2{\cdot}10^{20}$ sp in vacuum and was lower by a factor of 10^5 when measured in air. The signal detection time in the processes of oxygen sorption (desorption) is not more than a fraction of second.

Execution of the operations in the proposed method provides control of oxygen concentration in a gas flow up to concentration 10^{20} cm^{-2} with fast response and high sensitivity.

CONCLUSION

Gas flow passing through the carbonaceous substance placed into the measuring cavity of radio spectrometer gives a magnetic resonance signal. The magnetic resonance signal of carbonaceous substance is an indicator of changes in the oxygen concentration. The change of intensity of magnetic resonance signal is used to control the concentration of oxygen. This control is realized for oxygen concentration in a gas flow in wide range 10^{16} - 10^{20} cm^{-2}. The anthracite dispersion to the grains of 1-20 μm dimensions and thermal treatment cellulose at 800°C in oxygen-free medium can be used as a carbonaceous substance.

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