METROLOGY, QUALITY, STANDARDIZATION AND CERTIFICATION

EVALUATION OF NATURAL GAS QUALITY BY ITS CALORIFIC VALUE

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Abstract. The article develops and analyzes a method of the comprehensive evaluation of the quality of natural gas as an energy source. The method is based on establishing the calorific value of natural gas as a determinative index of its quality, taking into account all gas properties: both those that positively affect the gas calorific value and its energy content, and those that adversely affect. The generalized definition of natural gas quality as the degree to which the set of the own gas characteristics (component composition and physical properties) meet the requirements concerning energy content, safety, ecology, and other factors is given. The results of experimental researches on natural gas quality according to the developed procedure are also presented.

Key words: Natural Gas; Energy Source; Natural Gas Quality; Index of Natural Gas Quality; Properties of Natural Gas; Calorific Value of Natural Gas.

1. Introduction

Natural gas (NG), which is a high-energy and chemically valuable raw material, undoubtedly belongs to the strategic types of products, which has led to its widespread use both in everyday life and in many industries. NG is a vital energy resource of the world economy. In most European countries, the share of NG in the energy balance is 20–22 % and is likely to continue to grow. Therefore, the requirements for the quality of NG as an energy source, and, accordingly, the requirements for the methodology of gas quality evaluation are becoming more and more relevant, as evidenced by the increase in the number of publications on this issue. As stated in [1], the general performance, efficiency, emissions, and safety of most gas applications are affected by the quality of the gas and therefore a proper and stable gas quality must be guaranteed. Therefore, it has been tried for many years to reach a European standard that should define the quality ranges for cross-border gas. In particular, the calorific value variation range should be given in the new H-gas standard. The European Union on the political level strongly supports the development of a common European standard for the quality of H-gas within the internal market [2]. NG, consisting primarily of methane, has some advantages compared to liquid fuels such as gasoline and diesel fuel. However, to fully utilize the benefits of natural gas in-vehicle applications, many NG parameters regarding its quality as fuel need to be considered [3]. It is also important to harmonize the European standards for the quality of NG with the standards used in the United States [4].

The quality of NG is an important subject of contractual requirements between buyers and sellers. In some cases, national or state gas quality requirements are set. It should also be noted that NG has become an element of world politics today. On the other hand, NG is an expensive source of energy and also dangerous for the environment. This draws attention to the NG quality in terms of energy efficiency and environmental protection. In particular, the issue of the impact of the quality of hydrocarbon gases on the goals of sustainable development, that is economic development, which is carried out without depletion of natural resources [5]. However, as noted in ISO 13686:2013 [6], there is currently no generally accepted definition of NG quality. Thus, the relevance of the issue of NG quality evaluation has determined the subject of this article.

2. Analysis of the current state and main problems of natural gas quality evaluation

First of all, it should be noted that the interpretation of the concept of "*quality*" concerning NG is not clearly defined in the current normative documents in force. In particular, ISO 13686:2013 [6] and ISO 14532:2014 [7] provide a fairly broad definition of gas quality as an attribute of natural gas defined by its composition (main components, minor components, and trace elements), and its physical properties (calorific value, Wobbe index, compression ratio, relative density and dew points).

Important characteristics of NG as an energy carrier are its volume-basis gross (superior) $(H_V)_G$, MJ/m^3 and net (inferior) $(H_V)_N$, MJ/m^3 calorific value [5–7]. However, they are not directly used for the evaluation of gas quality and price. In most European countries, an important qualitative characteristic of natural

gas is Wobbe index W, MJ/m^3, namely volume-basis gross (superior) Wobbe index $\left(W_V\right)_G$, MJ/m^3, which is defined by the Eq. (1):

$$\left(W_V\right)_G = \frac{\left(H_V\right)_G}{\sqrt{d}},$$ (1)

where $\left(H_V\right)_G$ is the volume-basis gross (superior) calorific value at specified reference conditions, that is pressure $p_{ref} = 101.325$ kPa and temperature $t_{ref} = 20$ °C [8], MJ/m^3; d is the gas relative density at the same specified metering reference conditions.

As noted by K. Altfeld, and D. Pinchbeck in [9], the Wobbe index $\left(W_V\right)_G$ is by far the most important combustion parameter for gas appliances, and is also an indicator of the interchangeability of different fuel gases and is specified in all countries. Following the BS EN 437:2018 [10], respectively to the Wobbe index $\left(W_V\right)_G$ natural gas is divided into three groups: H-gas, L-gas, and E-gas. The Wobbe index $\left(W_V\right)_G$ limits for the H-gas are 45.7...54.7 MJ/m^3, for the L-gas – 39.1...44.8 MJ/m^3, and for the E-gas – 40.9...54.7 MJ/m^3. However, the using scope of BS EN 437:2018 is limited to test gases for appliances, and therefore it is not used as a characteristic of natural gas in the field of trade.

Mutual settlements between consumers and gas suppliers are based on energy E, MJ of the consumed gas. It is found according to ISO 15112: 2018 [11] as the product of the amount of gas Q (mass or volume) and the corresponding specific volume-basis calorific value of the gas at specified reference conditions.

Typically, the amount of gas consumed Q is expressed by the volume V, that is $Q = V$, m^3 and, accordingly, takes into account the volume-basis gross (superior) calorific value $\left(H_V\right)_G$, MJ/m^3 as shown in Eq. (2):

$$E = V \cdot \left(H_V\right)_G.$$ (2)

According to the generally accepted definition [7, 12], the volume-basis gross (superior) calorific value $\left(H_V\right)_G$, MJ/m^3 is an amount of heat that would be released by the complete combustion with the oxygen of a specified quantity of gas, in such a way that the pressure, p_1, at which the reaction takes place remains constant, and all the products of combustion are returned to the same specified temperature, t_1, like that of the reactants, all of these products being in the gaseous state except for water, which is condensed to the liquid state at t_1.

The value of the volume-basis gross (superior) calorific value $\left(H_V\right)_G$, MJ/m^3 according to ISO 15112: 2018 [11] is determined by two main methods:

• by direct measurement using a calorimeter;

• by the method of indirect measurement by calculation based on the component composition and volume fractions of combustible components of the gas mixture according to ISO 6976:2016 [12] using the method of gas chromatography [13].

Note. The procedure of determining the volume-basis gross (superior) calorific value $\left(H_V\right)_G$, MJ/m^3 and other its physicochemical properties necessary to find the actual calorific value of gas, and used in this article, is described in detail in ISO 6976:2016 [12].

Today, the method of indirect measurement is used to determine the value of $\left(H_V\right)_G$, MJ/m^3 of natural gas in real-time. As noted by Kenneth N. Marsh [14], the gross calorific value is difficult to measure accurately for a flowing stream whereas the analysis of the composition of the stream by gas chromatography can be done accurately by well-established techniques.

However, it should be noted that the value $\left(H_V\right)_G$, MJ/m^3, obtained by the calculation method, and, accordingly, the energy value E, MJ, do not reflect the real energy of the consumed gas. Because in this case the humidity of the gas and presence in the gas composition of non-combustible components, in particular, carbon dioxide, nitrogen, helium, etc., are not taken into account.

Thus, the methodology of natural gas quality evaluation as an energy source should be comprehensive and take into account the ratio of such important characteristics as caloric value, humidity, density, component composition, as well as the presence and number of non-combustible and harmful components. Therefore, to calculate the energy value E, MJ of the consumed natural gas, it is necessary to take into account its *real* (actual) volume-basis calorific value $\left(H_V\right)_R$, MJ/m^3. Resolving this issue is the main purpose of the present article.

3. The purpose of the article and the main objectives of the research

The *purpose* of the paper is to develop a method for determining the energy value of natural gas, taking into account such factors as the component composition, physicochemical properties, and humidity.

To achieve the stated objective, the following *tasks* have been identified:

• to develop a concept of the natural gas quality evaluation as an energy source;

• to analyze the impact of physicochemical properties of natural gas on its energy value and to establish a nomenclature of the quality indexes for determining the gas quality as an energy source;

• to establish the functional dependence between the determinative gas quality index as an energy source and its physicochemical properties;

• to carry out experimental research of natural gas to determine its quality as an energy source according to the developed methodology.

4. Principles of the method for evaluating natural gas quality by its calorific value

4.1. Definition of the notion of "*gas quality*"

According to generally accepted definitions, given in ISO 9000:2015 [15], *product quality* – the degree to which a set of own characteristics of a product satisfies the requirements. *The requirement* is a formulated need or expectation, and a *quality requirement* is a requirement related to quality. Accordingly, we formulate the basic notions and terms regarding the methodology for evaluating the quality of natural gas as an energy source.

Today, in the current standards ISO 13686:2013 [6] and ISO 14532:2014 [7], the *natural gas quality* is interpreted as an attribute of natural gas defined by its composition and physical properties. This definition does not fully meet the requirements of ISO 9000:2015 [15] on the definition of product quality. Consequently, it should be supplemented and clarified.

First of all, the *requirements* for the quality of NG should be formulated as the needs for energy content, safety, ecology, and other factors. The *own characteristics* of NG are its component composition (combustible, non-combustible, and harmful components) and physical properties (calorific value, Wobbe index, humidity, density, relative density, and compression factor).

Consequently, we obtain a generalized definition of the *natural gas quality* – that is the degree to which the set of the own gas characteristics (composition and physical properties) meet the requirements to energy intensity, safety, ecology, and other factors.

4.2. Concept of natural gas quality evaluation and establishment of the optimal structure of its quality indexes

Based on the theoretical foundations of qualimetry, the most effective method of evaluating product quality, in general, is a complex method by which a complex index of product quality P_c is found as a function of a number of its properties P_i, that is $P_c = f(P_i)$, $i = 1,2,\ldots,m$ (here m is the number of properties) [16, 17]. Thus, the complex quality index P_c is generally the determinative (main) index of product quality. This technique was used by the authors to assess the quality of natural gas as an energy source.

The authors have analyzed the main physico-chemical properties and component composition of natural gas as indexes of its quality as an energy source. The specific volume-basis gross (superior) calorific value of natural gas $(H_V)_G$, MJ/m^3 of natural gas is accepted as a determinative (main) index of its quality, and all its physical and chemical properties and components of the gas composition by the nature of the impact on its energy value are divided into two groups (see Table 1).

Table 1

Properties of natural gas as an energy source

Group 1 – properties, the ***increasing*** of which leads to ***increase*** of the gas quality level as an energy source	**Group 2** – properties, the ***increasing*** of which leads to ***decrease*** of the gas quality level as an energy source
• specific volume-basis gross (superior) calorific value $(H_V)_G$, MJ/m^3; • specific volumetric-basis net (inferior) calorific value $(H_V)_N$, MJ/m^3; • volume-basis gross (superior) Wobbe index $(W_V)_G$, MJ/m^3; • density ρ_S, kg/m^3 • relative density *d*	• gas moisture ϑ, g/m^3; • content of nitrogen C_{N_2}, %; • content of carbon dioxide C_{CO_2}, %; • mass concentration of hydrogen sulfide C_{H_2S}, g/m^3; • mass concentration of mercaptan sulfur C_{CH_4S}, g/m^3
Note. The compression factor Z_{mix} is used to calculate the gross (superior) $(H_V)_G$, MJ/m^3 and net (inferior) $(H_V)_N$, MJ/m^3 specific volume-basis calorific value of natural gas	

The *first group* includes natural gas properties, increasing values of which leads to the *increase* of the gas quality level as an energy source. The *second group* includes natural gas properties, increasing values of which leads to the *decrease* of the gas quality level as an energy source. Hydrogen sulfide, C_{H_2S}, although it is a combustible component, is a relatively insignificant energy factor and can be a significant cause of pollution and corrosion [18]. Therefore, it is classified in the second group of gas quality indexes.

The functional relationships between the natural gas calorific value and its properties are discussed below.

5. Methodology for determining the actual calorific value of natural gas

5.1. Determination of the actual volume-basis calorific value of natural gas taking into account its humidity and hydrogen content

Complete combustion of NG is observed when the products of its combustion do not contain combustible gas components or components of incomplete oxidation. Typically, the products of NG combustion contain water vapor, the mass of which consists of three parts [18]:

• vapor formed from moisture contained in the gas itself;

• vapor formed as a result of the gas components oxidation, in particular hydrogen;

• vapor, which is introduced together with the air used for gas combustion.

All the heat released during the flaring of gas is perceived by the combustion products and can be extracted from them and fully used only if they are cooled to a temperature below the water dew point t_ϑ, °C of the gas, in which there is the condensation of water vapor. However, in practice, such deep cooling of gas flaring products is usually not carried out and the latent heat of vaporization of water vapor contained in these products is not used.

Also, real NG, as mentioned above, contains non-combustible components. Therefore, it is advisable to correct the value of the higher volumetric heat of combustion of the gas to take into account the share of these components in the total volume of gas consumed and their impact on its calorific value.

Consequently, based on this analysis, the actual volume-basis calorific value $\left(H_V\right)_{act}$, MJ/m³ of NG, taking into account its humidity and the content of non-combustible components can be determined by the Eq. (3):

$$\left(H_V\right)_{act} = \left(H_V\right)_G \cdot k_\rho - q_{H_2O} \cdot$$
$$\cdot \left(m_{(H_2O)_\vartheta} + m_{(H_2O)_{H_2}} + m_{(H_2O)_{air}}\right), \qquad (3)$$

where $\left(H_V\right)_G$ is the volume-basis gross (superior) calorific value, MJ/m³; k_ρ is the correction factor to take into account the influence of non-flam components in the gas mixture on its volume-basis calorific value; q_{H_2O} is the amount of heat required for heating 1 kg of water to boiling and evaporation, MJ/kg; $m_{(H_2O)_\vartheta}$ is the mass of water in the gas, caused by its humidity ϑ, kg; $m_{(H_2O)_{H_2}}$ is the mass of water vapor formed by the oxidation of hydrogen H_2 in the gas, kg; $m_{(H_2O)_{ear}}$ is the mass of water vapor, which is introduced together with air in the process of gas combustion, kg.

Consider the quantities included in the formula (3), for its practical use. The volume-basis gross (superior) calorific value $\left(H_V\right)_G$, MJ/m³, as already mentioned above in paragraph 2, is advisable to determine by calculation according to the methodology described in ISO 6976:2016 [12]. The value of q_{H_2O} when heating 1 kg of water from 0 °C to 100 °C at a pressure $p = 0{,}101325$ MPa is equal to 2.51 MJ/kg. The mass of water $m_{(H_2O)_\vartheta}$, kg in the NG composition is determined depending on its humidity [19–21].

The concentration of water content in NG or gas absolute humidity ϑ, g/m³ at reference conditions, that is the pressure $p_{ref} = 101.325$ kPa and the temperature $t_{ref} = 20$ °C [8], is calculated depending on the measured water dew point temperature t_ϑ, °C of the gas according to Eq. (4) [20]:

$$\vartheta = 101.325 \cdot \frac{A}{p} + B, \qquad (4)$$

where A is the coefficient of the dependence of the water content on the water vapor pressure p at the measured water dew point t_ϑ, g/m³; B is the coefficient of the water content on the water dew point temperature t_ϑ and the component composition of the gas, g/m³; p is the absolute pressure of the test gas in the measuring chamber, kPa;

In general, the absolute humidity of the gas ϑ, g/m³ is equal to the mass of water vapor $m_{(H_2O)_\vartheta}$, g, which is contained in 1 m³ of the steam-gas mixture (g/m³ of wet gas), that is

$$\vartheta = \frac{m_{(H_2O)_\vartheta}}{V}. \qquad (5)$$

Then, provided that the volume of gas $V = 1$ m³, we obtain that $m_{(H_2O)_\vartheta} = \vartheta \cdot 10^{-3}$, kg.

As can be seen from the analysis [5], for most gas fields components $m_{(H_2O)_{H_2}}$ and $m_{(H_2O)_{air}}$ are negligibly small compared to $m_{(H_2O)_\vartheta}$ and formula (3) can be simplified, ie the actual volume-basis calorific value $\left(H_V\right)_{act}$, MJ/m³ of NG, taking into account its humidity and the content of non-combustible components can be calculated by equation (6):

$$\left(H_V\right)_{act} = \left(H_V\right)_G - q_{H_2O} \cdot m_{(H_2O)_\vartheta}. \qquad (6)$$

5.2. Influence of non-combustible components of natural gas composition on its volume-basis calorific value

It is advisable to introduce a correction to the measured value of the volume-basis gross (superior) calorific value $\left(H_V\right)_G$, MJ/m³ of real NG to take into account the share of non-combustible components in the total volume of gas consumed and their impact on its calorific value.

This correction will be determined by the following considerations. The amount of heat released during the combustion of any coolant depends on its mass, and for a known volume – on the density of this coolant [22]. Therefore, the actual volume-basis gross (superior) calorific value $\left(H_V\right)_{G,R}$, MJ/m³ of the real gas, that is the gas that contains both combustible and non-combustible components, can be determined by the following formula:

$$\left(H_V\right)_{G,\rho} = \left(H_V\right)_G \cdot \left(1 - \frac{\rho_{nc}}{\rho_c}\right), \text{MJ/m}^3, \qquad (7)$$

where $\left(H_V\right)_G$, MJ/m^3 is the specific volume-basis gross (superior) calorific value, determined by calculation according to the methodology described in ISO 6976:2016 [11]; ρ_{nc}, kg/m^3 is the density of a part of NG, which consists exclusively of non-combustible components; ρ_c, kg/m^3 is the total density of NG, which is determined by calculation according to the method described in ISO 6976:2016 [11] taking into account all gas components.

Therefore, based on equations (3) and (7), the calorific value $\left(H_V\right)_R$, MJ/m^3 of real NG, taking into account its humidity and the content of non-combustible components can be determined by the formula:

$$\left(H_V\right)_R = \left(H_V\right)_G \cdot \left(1 - \frac{\rho_{nc}}{\rho_c}\right) - q_{H_2O} \cdot \left(m_H + m_{H_2O}\right), \text{J/m}^3. \quad (8)$$

The actual volume-basis calorific value $\left(H_V\right)_R$, MJ/m^3 of NG determined by the formula (8), really reflects its energy value. It can be used to calculate the energy E, MJ of the NG following equation (2), and to set the price for gas consumed for both domestic and industrial consumers.

6. Determination of calorific value of natural gas

The authors have determined the calorific value of NG according to the developed method. Input information was taken from the results of experimental studies of NG samples from the deposit of Lviv region, Ukraine, carried out in the analytical laboratory of the Gas Production Division LvivGasVydobuvannya. The component composition of the gas was determined by the chromatographic method following ISO 6974-1:2012 [13]. Calculations of gas volume-basis gross (superior) $\left(H_V\right)_G$, MJ/m^3 and net (inferior) $\left(H_V\right)_N$, MJ/m^3 calorific value, total gas density ρ_c, kg/m^3, and the density of gas non-combustible components ρ_{nc}, kg/m^3 were performed based on the gas component composition according to ISO 6976:2016 [12]. The dew point temperature by humidity t_ϑ, °C was measured by the condensation method, and the absolute humidity of the gas ϑ, g/m^3 depending on the measured dew point of the gas by humidity t_ϑ, °C was determined according to the procedure by GOST 20060-83 [20].

According to the results of experimental studies of NG samples by formula (8), the values of the actual value of the volume-basis calorific value $\left(H_V\right)_R$, MJ/m^3 of gas on the 5th day of each month during the year are determined. The results of experimental studies of NG samples and calculations are given in Table 2.

The figure graphically shows the change in the volume-basis gross (superior) calorific value $\left(H_V\right)_G$, MJ/m^3, the theoretical net (inferior) calorific value $\left(H_V\right)_N$, MJ/m^3, and the actual value of the volume-basis calorific value $\left(H_V\right)_R$, MJ/m^3 of NG during the year on the 5th day of each month.

Table 2

**The results of experimental studies of natural gas samples
(the data for the 5th day of each month during the year)**

t, month	5.01	5.02	5.03	5.04	5.05	5.06	5.07	5.08	5.09	5.10	5.11	5.12
$\left(H_V\right)_G$, MJ/m^3	37.55	37.50	37.34	37.41	37.41	37.41	37.38	37.56	37.33	37.36	37.39	37.37
$\left(H_V\right)_N$, MJ/m^3	33.84	33.80	33.65	33.71	33.71	33.70	33.68	33.85	33.63	33.67	33.69	33.67
t_ϑ, °C	-5.7	-5.5	-4.4	-5.2	-4.9	-3.5	-3.8	-2.9	-3.0	-3.8	-3.4	-4.0
ϑ, g/m^3	0.097	0.098	0.105	0.099	0.110	0.119	0.115	0.124	0.123	0.113	0.114	0.112
ρ_c, kg/m^3	0.685	0.688	0.682	0.688	0.688	0.684	0.684	0.691	0.687	0.685	0.687	0.684
$\rho_{nc} \cdot 10^{-2}$, kg/m^3	0.567	0.627	0.554	0.651	0.627	0.573	0.573	0.551	0.603	0.559	0.581	0.573
$\left(H_V\right)_R$, MJ/m^3	32.81	32.77	32.01	29.69	31.46	31.27	27.59	30.32	26.97	29.63	31.41	33.28

Note 1. All symbols of the quantities given in the table correspond to the designation of these quantities in the text of the article.
Note 2. Dimensions of values, provided in the table, are taken for standard conditions, namely to $p_{ref} = 101.325$ kPa; $t_{ref} = 20$ °C.
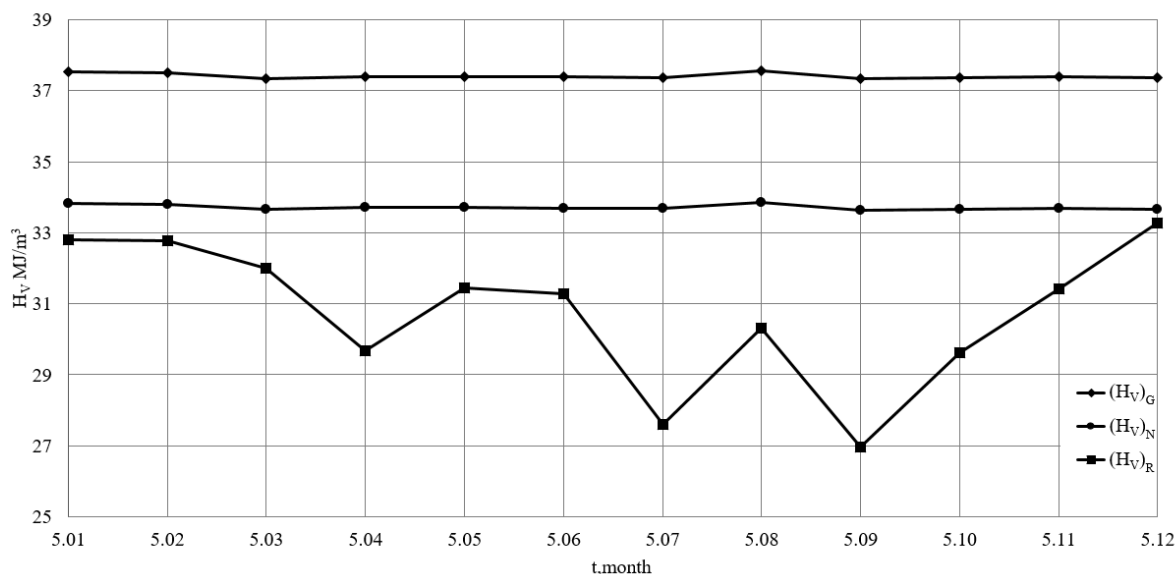
Fig. Change in the calorific value of natural gas during the year on the 5th day of each month

As can be seen from the data in Table 2, and the graphs are shown in the figure, the actual value of the volume-basis calorific value $(H_V)_R$, MJ/m^3, determined taking into account the humidity of the gas and the presence in its composition of non-combustible components, differs significantly from the theoretical net (inferior) calorific value $(H_V)_N$, MJ/m^3, determined according to the method according to ISO 6976:2016 [12], even if the concentration of carbon dioxide and nitrogen in the investigated gas field is low, and mercaptan sulfur is absent at all.

Thus, the experimental studies of NG samples taken from one gas field during the year fully confirmed the theoretical foundations of the method developed by the authors to assess the quality of NG as an energy source. The results of this work can be used in the practical activities of gas-industrial enterprises in assessing the quality of NG and setting the appropriate price for it, as well as in developing new regulations governing the requirements for NG quality.

7. Conclusions

The constant growth of consumption of natural gas, which is a high-energy and chemically valuable raw material, and the growth of world prices for natural gas leads to increased demands on its quality as an energy source and a way to determine its calorific value.

1. Growth in the consumption of natural gas, which is a high-energy and chemically valuable raw material, leads to increased demands for its quality as an energy source and to the development of the method of determining its calorific value.

2. The method for determining the calorific value of natural gas as a determinative gas quality index of its

energy value, taking into account the moisture content of the gas and the presence of non-combustible components facilitates the definition of the real energy value of gas.

3. Studies of natural gas samples confirmed the validity of the considered methodological foundation for assessing the quality of gas as an energy source and the possibility of its use in the gas industry practice and in developing regulations governing natural gas quality requirements.

8. Acknowledgments

9. Conflict of Interests

Conflict of interest while writing, preparing, and publishing the article as well as mutual claims by the co-authors is absent.

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