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DEVELOPMENT OF NEW INSTANT TECHNOLOGY OF NATURAL GAS QUALITY DETERMINATION

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ABSTRACT

World experience shows that important factor in the calculations for natural gas consumption between suppliers and consumers is not only the volume of natural gas, but the quality indicators. With gas market liberalization, gas properties are expected to vary more frequently and strongly (composition, heating value etc.). Quality of natural gas is currently a topical issue, considering the steady increase of gas consumption in the world in recent decades. Existent chromatographs and calorimeters are very accurate in gas quality determination, but general expenditure and maintenance costs are still considerable. Market demands alternative lower cost methods of natural gas quality determination for transparent energy billing and technological process control.

Investigation results indicate that heating value (HV) is a nonlinear function of such parameters as sound velocity in gas, N_2 and CO_2 concentration. Those parameters show strong correlation with natural gas properties of interest (HV, density, Wobbe index), during analysis conducted on natural gas sample database. For solving nonlinear multivariable approximation task of HV determination, artificial neural networks were used. Proposed approach allowed excluding N_2 concentration from input parameters with maintenance of sufficient accuracy of HV determination equal to 3.7% (with consideration of N_2 concentration – 2.4%) on sample database. For validating of received results corresponding experimental investigation was conducted with reference analysis of physical and chemical parameters of natural gas samples by gas chromatography and followed superior HV calculation according to ISO 6976:1995. Developed experimental setup consist of measuring chamber with ultrasonic transducer, reflector, pressure, temperature and

humidity sensors, ultrasonic inspection equipment for sound velocity measurements and CO_2 concentration sensor with relevant instrument. The experimental setup allows measurement of sound velocity at 1MHz frequency and CO_2 concentration in natural gas sample along with parameters control (temperature, humidity, pressure). The HV calculation algorithm was based on specially designed and trained artificial neural networks.

Experimental investigation of proposed approach was conducted on 40 real samples of locally distributed natural gas. Obtained results, in comparison to reference values, showed absolute error in Lower HV (net calorific value) determination equal 166 kJ/m³, while relative error was equal 4.66%. Developed technology allows construction of autonomous instrument for instant natural gas quality determination, which can be combined with volume meters in order to provide transparent energy flow measurement and billing for gas consumers. Additionally it can be used for gas sensitive technological process control.

INTRODUCTION

With gas market liberalization in the EU, relevant gas properties expect to vary more frequently and strongly (composition, heating value, Wobbe index etc.), which affects consumers respectively, resulting in gas quality variations [1]. Main and most used quality characteristic of natural gas is its heating (calorific) value – indicator that determines its energy content. An average difference between stated in existing regulations minimum and maximum values of heating value (HV) constitutes 10.17 MJ/m³ in Europe (25%) [2], while in

Ukraine it can reach up to 15.0 MJ/m^3 (38%) [3]. Difference in HV equal 10.17 MJ/m^3 , for annual consumption of average household (up to $2500 \text{ m}^3/\text{p.m.}$) could lead to 1000 EUR annual overpayment, for small power plant (up to $100\,000 \text{ m}^3/\text{p.m.}$) – 80 000 EUR annual overpayment (according to average 2011'EU gas price).

In energy billing, the HV of the natural gas and other gas property, such as normal density and CO_2 mole fractions data are required to determine gas law deviation factor according to ISO 12213-3:2006, and together with measured volume for consumed energy calculation on the basis of ISO 15112:2001. Therefore independent HV determination is the key for transparent energy billing considering gas price rises last years. In Ukraine the excessive and not regulated content of CO_2 and N_2 are the reasons of incomplete gas combustion, creating a ballast mixture, which lowers gas calorific value [3]. Gas energy properties variations can also cause reduction of received energy and increasing volume of combusted gas, and its express determination is of great importance for technological process control in gas sensitive industrial applications (chemical, power producing etc).

Gas chromatographs and calorimeters have now reached a stage where gas content and energy properties measurement uncertainty and susceptibility to failure are very low [4-6]. However, capital expenditure and maintenance costs are still considerable, while analysis can be time-consuming. Alternative lower cost methods are under intense development over last years. These new measurement methods are usually physical properties (correlative) methods are considered to be very perspective in terms of cost, ease of use and short analysis time [7]. The aim of this paper was development of new technology for instant natural gas quality determination that would conform to the requirements of scientific validity, precision and usability, remain inexpensive enough for wide end user usage.

BACKGROUND

Regular response of various physical properties as a function of carbon atom number is the reason why hydrocarbon mixtures, involve good correlative relations between some of their properties [7, 8]. Therefore hydrocarbons (alkanes) contained in natural gas can be characterized by a single or several physical properties.

In previous research possibility to use artificial neural network (ANN) for solving nonlinear, multi-parametric approximations of HV as a function of sound velocity in gas, nitrogen (N_2) and carbon dioxide (CO_2) concentration was showed [10]. Considering that sound velocity corresponds with hydrocarbons concentration and N_2 concentration is equal to the difference between gas total composition and sum of CO_2 and hydrocarbons concentration, N_2 concentration indirectly depends both of this parameters and can be characterized by them implicitly. Towards development of previously proposed approach, it was proposed to exclude N_2 concentration from input parameters for HV determination and compare received results with previous one on gas sample database and real gas

sample. Excluding N_2 concentration from input parameters will be highly important and obvious since there are no inexpensive sensors to measure it on the market, and it allows decreasing measuring instrument producing cost.

In order to check the proposed approach, we performed modelling of natural gas HV determination by means of Matlabs's ANN Fitting Tool. It provides choice of architecture, training algorithm and network parameters for specific needs. ANN architecture is the number of layers and neurons in them as well as the shape of performance function in neurons. It must be mentioned that selection of architecture in each case is carried out individually based on the assumption about the task complexity, available computing power and input data amount. During set of preliminary investigation, several ANN architectures and activation functions have been examined. Chosen two-layered feed-forward ANN architecture, which demonstrated the best performance results, is shown in Fig. 1. The activation functions for neurons in hidden layer were *tansig*, and for neurons in output level *logsig*. As a training algorithm, the Levenberg–Marquardt algorithm of error back-propagation has been used, since it is recommended for cases when there is no large quantity of training pairs in a data set.

Fig. 1. Designed ANN architecture

The modeling was based on gas sample database provided in [11]. This database is array of 95 natural gas mixtures parameters determined by chromatography analysis. Gas samples superior HV was calculated according to [6] and was between 36.8151 MJ/m^3 and 42.9323 MJ/m^3 . Gas samples CO_2 and N_2 concentration ranges from 0.44% to 6% and from 0.97% to 7.40% respectively. Sound velocity was estimated from gas composition in accordance with [12].

Gas properties database has been divided into two data sets consisting of 86 and 9 entries. First 86 entries were used for ANN training and performance testing according to Matlab's ANN Fitting Tool training procedure. Rest 9 unknown for ANN entries were used for its simulation and error estimation. According to ANN performance practice, all the data were normalized, dividing them by the difference between maximum and minimum values. On the two inputs of ANN there were feed sound velocity and CO_2 concentration parameters, and one output parameter was obtained – superior HV of gas sample.

The results of ANN simulation were accurate enough. The HV, obtained by ANN coincides with actual values (chromatographic) and relative to the measurement range error was 3.7% (with consideration of N_2 concentration – 2.4%).

In respect that natural gas supplied to consumers in Ukraine differ in gas composition and parameters spread range, additional testing on designed and trained ANN on 8 real natural gas samples was performed afterwards, in order to prove its reliability and adequacy in superior HV determination in real conditions. Gas samples properties were obtained from gas mixture quality certificates prepared by local gas distributing company in Ukraine. The results of ANN simulation showed in table 1. The maximum error of HV determination by ANN without N₂ concentration consideration reached 4.4%, while for ANN with N₂ concentration consideration it was 1.2%.

Table 1. Results of ANN simulation on real gas data

Gas sample	Real HV, kJ/m ³	ANN HV (with %N ₂), kJ/m ³	ANN HV (without %N ₂), kJ/m ³
1	35702.3	35913.2	37287.3
2	36914.9	36642.1	36842.4
3	39253.6	39762.6	39385.3
4	36623.5	36404.3	37173.1
5	36870.3	36476.1	36651.3
6	37245.6	37306.3	37271.3
7	37256.5	37483.7	37953.2
8	37203.1	37364.5	37442.5

The gained results of ANN simulation on sample database and real gas samples confirmed possibility of for reducing measured input parameters (excluding N₂ concentration) without significant accuracy drop and created a good background for further approach development and its experimental validation.

It should be emphasized that for proper HV determination ANN should be trained on gas sample with properties that are common for local distributed natural gas, to reduce gas composition and parameters spread range, in order to achieve better accuracy and reliability.

APPARATUS & PROCEDURE

For validating of received theoretical results, corresponding experimental investigation were conducted, with for the purpose of establish the possibility of practical application of the proposed approach directly to the consumers of natural gas.

The essence of experimental investigation was in the following: two identical samples of natural gas were selected simultaneously from gas network. Thereafter one of the samples was analyzed by means of specially developed experimental setup for HV determination according with proposed method. Another gas sample was analyzed by the means of gas chromatograph LHM (industrial No.3875), for component composition determination and following natural gas HV calculation [6]. After the above-mentioned investigations were performed, their results were compared. To reduce the influence of ambient temperature readings for research measuring results,

the investigations were conducted simultaneously in same room and gas sample parameters were controlled correspondingly.

The experimental setup for gas HV determination consist of the following main functional blocks (Figure 1): gas preparation unit, measuring chamber, sound velocity measuring unit, CO₂ concentration measuring unit, temperature sensor, pressure and humidity sensors, processing and visualization unit (personal computer).

1 – gas preparation unit; 2 – measuring chamber; 3 – sound velocity measuring unit; 4 – CO₂ concentration sensor; 5 – temperature sensor; 6 – pressure sensor; 7 – humidity sensor; 8 – processing and visualization unit
Figure 1. Experimental setup block diagram

The actual experimental setup design is showed in Figure 2. Gas preparation unit aimed to clear gas sample from mechanical impurities. It includes water segregator, and flow-metering tube, designed to clean the samples of dust and moisture. Sound velocity measuring unit consists of ultrasonic transducer, reflector and ultrasonic inspection instrument DIO 562 (serial No. 138).

1 – gas preparation block; 2 – measuring chamber; 3 – CO₂ sensor; 4 – gas analyzer; 5 – pressure gauge; 6 – thermal hygrometer; 7 – ultrasonic inspection instrument
Figure 2. Experimental setup for natural gas HV determination

CO₂ concentration measuring unit consists of CO₂ infrared sensor Dynamet MSH-P-CO₂ and gas analyzer DOZOR-C (serial No. 3002). The experimental setup was also equipped with gas physical parameters sensors (pressure, temperature, humidity): pressure gauge of type MT-2H (TR 33.2-33884768.001-2006), thermal hygrometer of type OVT-6-7302 (serial No. 08.082.341).

The specifically designed and manufactured measuring chamber is of particular interest (Figure 3). Measuring chamber is a tight construction made of stainless steel in cylindrical form, into which the natural gas was supplied. The specific transducer of own production was set up in the block. Its frequency is at up to 1 MHz, which works in couple mode as transmitter-receiver. The reflector was made of stainless steel with diameter of 22 mm and a surface roughness $R_z = 20$ was positioned at the calculated and precisely set distance of 57.7 mm for receiving the maximum energy value of reflected signal.

1 – tap; 2 – fitting; 3 – reflector; 4 – measuring chamber;
5 – ultrasonic transducer; 6 – pressure gauge; 7 – connector;
8 – captivating plate; 9 – temperature and humidity sensor

Figure 3. Measuring chamber dismantled

First series of experiments showed that experimental setup required improvement, because it was necessary to take into account the effect of humidity and temperature on gas samples. For this purpose the block of sound dispersion velocity in gas velocity was updated by the including humidity and temperature sensors directly into the cylindrical measuring chamber unit.

In addition, it was also defined that accuracy and reliability of sound velocity measurement should be improved. For this purpose there have been improved the internal surface of measuring chamber by introducing scattering coverage to avoid unwanted ultrasonic beams multiple reflections (Figure 4).

1 – ultrasonic transducer; 2, 3 – temperature and humidity sensor; 4 – scattering surface; 5 – concave reflector

Figure 4. Measuring chamber

The reflector was replaced by the new one – concave shaped reflector, made of stainless steel with the diameter 20 mm and the curvature surface of 520 mm, allowing to focus acoustic signal from the primary converter and to increase the energy value of received acoustic oscillations. In addition, the block construction of sound dispersion velocity in gas provides the possibility of distance regulation from the primary reflector to the transducer, that allows get the maximum energy value of the reflected signal due to the radiated frequency.

Procedure for natural gas heating value determination by means of the experimental setup was the next. Sample gas enters the measuring chamber, where sound velocity is determined. CO₂ concentration was measured by respective sensor. For carrying out correct CO₂ concentration measurement in natural gas, gas sample needs to be additionally cleaned from mechanical impurities and dried with the preparing samples block. In addition, gas sample parameters, such as the pressure, temperature and humidity were measured and controlled. During measurement procedure, the temperature and pressure inside measuring chamber were stabilized at values of (20 ± 1) °C and (2.2 ± 0.2) kPa correspondingly.

For the verification of the new approach of HV determination on the basis of experimental setup 40 samples of typical local distributed natural gas were selected at separate sample points. By means of experimental setup, the following informative parameters were determined: sound velocity in gas and CO₂ concentration. Actual natural gas sample HV value was determined by calculation from gas composition obtained from gas chromatograph [6]. In addition, information about actual CO₂ concentration and sound velocity were prepared for experimental setup single properties measurement accuracy estimation.

RESULTS

Considering the fact that in Ukraine only Lower HV (LHV) property of distributed natural gas is specified in normative documents, it is the main quality characteristic of interest for gas consumers and energy billing. The experimental setup was configured to determine LHV by the way of training ANN to calculate it from measured input parameters. Alternatively other gas quality characteristics, such as higher HV and WI can be used for ANN training and further determination.

For the experiment, there have been used gas samples with next main physical properties variations range, typical for locally distributed natural gas: methane content – not lower 96%; CO₂ concentration – 0-2%; N₂ concentration – 0-2%, sulfur content – lower 0.5%.

All the gas samples were analyzed on the experimental setup and reference gas chromatograph in order to measure informative parameters and determine the accuracy of single parameter measurement. The received results were compared with data obtained from gas chromatograph. Sound velocity was calculated from gas composition in accordance with [12].

Overall, measured by experimental setup single properties (sound velocity and CO₂ concentration) coincide to real values with minor error (Figure 5 and Figure 6).

Figure 5. Response of measured and real sound velocity in gas samples

Figure 6. Response of measured and real CO₂ concentration in gas samples

After applying calibration correction coefficients in measured by experimental setup informative parameters, they was used for ANN training and simulation. In order to calculate the local distributed natural gas LHV newly created ANN was used. The same as described above architecture and neurons activation functions were utilized.

First 30 sets of 40 samples were selected to train the ANN. Simulation of ANN operation has been carried out with the help of other 10 sets, unknown for ANN, which differ from training sets. They have been used for testing performance of ANN and determine its accuracy.

As it can be seen from the Table 3, the LHV of testing samples, defined by ANN on the basis of informational parameters measured by experimental setup, coincides with actual LHV calculated from gas sample composition in accordance with [6]. Maximal absolute error was equal 166

kJ/m³ and reduced to the range relative error 4.66 %. These results can be considered as acceptable for energy billing calculations purposes.

Table 2. Results of the natural gas LHV determination

Sample No.	HV by ANN, kJ/m ³	HV from composition, kJ/m ³	Absolute error, kJ/m ³
1	33109.57	33219.49	109.92
2	33759.95	33613.54	146.41
3	33917.17	34005.1	87.93
4	33558.87	33683.08	124.21
5	34110.02	34276.09	166.07
6	33313.54	33159.05	154.49
7	34797.17	34925.1	127.93
8	35601.08	35698.87	97.79
9	35345.49	35209.57	135.92
10	34513.54	34404.73	108.81

DISCUSSION

It should be mentioned, that designed and trained ANN has its operational limits in LHV calculation and gas mixture concentration. These limits consider to be the next gas mixture properties variations ranges: methane content – not lower 96%; CO₂ concentration – 0-2%; N₂ concentration – 0-2%, sulfur content – lower 0.5%, humidity – not more 30%.

Within the operational limits of natural gas components concentrations, designed and trained ANN showed good performance and acceptable error. Good performance in HV determination within operation limits can be explained by ANN exclusive ability to interpolate multiparametric nonlinear functions. But ANN can not be used outside the range of the provided for training data, since the result of it simulation in the case will be unpredictable and groundless. The ANN simulation approximation solving can be showed as the surface of calculated HV as function of input parameters, velocity of sound and CO₂ concentration (Figure 7). Operational range of designed ANN for HV determination is from 30100 kJ/m³ to 36650 kJ/m³.

Operational limits and accuracy of HV determination by proposed approach feasibly can be improved, while performing ANN training on the bigger amount of reference data with wider physical properties variations range. In connection with this additional research should be performed.

However, this have been a laboratory based study, and the actual composition of natural gas samples was additionally defined by reference gas chromatograph. Therefore, it was certainly know that the concentrations of separate components do not exceed operational limits. One main issue arise when transferring the approach to the field. The actual amount of main energy content harmful components, such as CO₂ and N₂, could possibly exceed predefined proper operations limits. The actual CO₂ concentration can be measured by CO₂ sensor, and if it will exceed predefined limits it will signalized about it and no

HV calculation performed. More difficulties will arise when N_2 concentration will exceed predefined limit of 2% of gas volume fraction. However, this issue can be overcome with periodic reference analysis of actual gas composition by gas chromatograph to ensure that N_2 concentration lie in operation limits.

Figure 7. Approximation surface generated by ANN

CONCLUSION

A new approach of natural gas HV determination has been proposed, which is based on the following: nonlinear multiparametric approximation of HV as a function of sound velocity and CO_2 concentration using ANN. It will allow to decrease final cost of final measuring instrument, since only two input parameter are measured.

Proposed approach was checked on the reference values of natural gas qualitative data and showed acceptable performance. Experimental validation of proposed approach on developed experimental setup showed absolute error 166 kJ/m³ in HV determination in predefined operational limits.

This study demonstrates the following advantages of the proposed approach for natural gas quality evaluation, based on ANN:

- possibility of fast calibration on respectively small amount of reference data, on account of neural network core of the approach;
- possibility of the continuous improvement of the ANN using additional new reference data, which will result in increased accuracy;
- alternative natural gas quality properties determination (Higher and Lower HV, WI), since ANN can be trained to approximate needed property.

Currently, there are performed activities on development of commercial prototype of the LHV measuring instrument. Furthering innovation can be the creating of an autonomic

device for measuring LHV of natural gas, which can be combined with gas volume meter, to provide complete energy flow calculation. The dimensions of the measuring instrument should not exceed dimension of current gas volume meters.

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