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Quality Requirements of the Application of Biogases in Natural Gas Public Utility Services in Hungary

Theses of PhD Dissertation

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1. The aim of the research work

Among the long term energy goals of the European Union the furtherance of the role of renewable energy resources in a wider and wider area can unambiguously be shown. Hungary, as a member state committed itself to take similar steps. The agricultural history of the country and its geographical conditions represent a biogas exploitation potential that is much higher than the present rate of utilisation. The domestic political, economic and technical regulations environment does not yet favour the transport and trade of biogases through the natural gas network. It is mainly the heat and electric power generation that is favoured. In the field of technical regulations of injecting biogas into natural gas networks some basic questions have to be clarified and laid down.

The main goal of the research work was to set up a *gas quality requirement system* that is suitable to assess the *injection of renewable gases,* originating in several sources, *into the natural gas network* in Hungary. Over and above quality requirements in case of certain gases not prepared for non-biomethane quality the maximum injection quantities were also determined as a function of the natural gas quality supplied through the given network section. A further element of the requirement scheme is the comprehensive analysis of the effect the gas (prepared and supplied in this way) quality has on consumer gas equipment. A further goal of the research work was to investigate interchangeability methods prepared for natural gas and applied in the international practice from the point whether they can be applied to the simultaneous modelling of heating technology parameters of natural gases and biogases. For the complexity of the subject a further objective of the paper was to determine the theoretical biogas potential of Hungary based on summarising the available resources.

2. Scientific preliminaries

Biogas (marsh gas) itself was discovered by Shirley in 1677 already. The first operating biogas equipment was put in operation in Bombay in 1857. Fermentation of biogases from animal manure through an anaerobic process started in Hungary in the 1950s that targeted the energy supply of animal breeding farms without gas supply. Currently 7 biogas plants operate in the country, 5 plants are under construction and there are project plans for a further 20 biogas plants.

The domestic technical literature about the injection of biogases into natural gas networks is very poor. The only current professional book (*Bai, 2007*) presents the sample injection project implemented in Pucking, Austria. However, the very wide technical literature database of the web is available where I could review the results of several unabridged research reports and technical publications at Hungarian, English and German language sites. (*Kilinski et al, 2006; Theißig et al., 2006; Hornbachner et al., 2005; Ramesohl et al, 2005; Biocomm 2002-2005*). My literature research on the web was mainly by sites organised on professional basis (EurObserv'ER, IEA-Biogas, Biogas Netzeinspeisung, Marcogaz, British Petrol, European Commission, etc.).

International organisations of the gas industry recently realised the importance and future role of the connection of biogas and natural gas systems. Upon this influence the Brussels based Marcogaz created its recommendation *WG-Biogas-06-18* in 2006 with the title "Injection of Gases from Non-Conventional Sources into Gas Networks". Similar motivations led the Dutch Kiwa N.V. to prepare its report No. *GT-070127* where it shared its experience gained through already operating injections with the professional audience.

The relevant chapter of the Energy User Manual (*Barótfi, 1994*), published in 1994, designates the specifications of the then already existing standard *MSZ 1648: 1990* 'Natural gas for public utility services' for the quality requirements of "bio natural gas". These requirements are more or less identical with the specifications of standard *MSZ 1648:2000* currently in force which is the basic document of the domestic natural gas industry regarding quality, and accepted by all parties in the sector. In the international practice when investigating the quality requirements of biogas injection the relevant parts of the German worksheet No. *DVGW G 262* are typically referred to. In fact it was this regulation that provided the principal technical connection conditions by which the injection of biogas into natural gas networks became manageable for both parties in Germany.

A serious limitation of utilising biogases in public utility services is the heating technology behaviour of gas when burnt in the consumers' equipment. The so-called „interchangeability methods" can be applied to investigate this issue. Several researchers and institutes studied replacement procedures since the '20s (e.g. Wobbe, 1946; Weaver, 1951; Delbourg, 1953; Holmquist, 1957; Schustre, 1957; Van der Linden, 1970; Sommers, 1973; Dutton, 1978). The question of replacement of combustible gases was already raised in the domestic gas industry in the '70s. The latest related domestic technical literature is a gas industrial technical book by two German authors, translated into Hungarian, that sets forth the most important procedures applied in the international practice. (*Joos, 2005; Cerbe, 2007*)

3. Investigations performed, data collection methods

Because of the complexity of the research my investigations had three main directions:

- exploring the role of biogas in the domestic energy balance and its potentials;
- determining the quantitative and qualitative terms and conditions derived from the injection of biogases into the Hungarian natural gas network from the point of the cooperating natural gas system; and
- the analysis of effects of the injected biogases and their natural gas mixtures on consumers' equipment using the interchangeability methods worked out for natural gases and mostly used in Europe.

I prepared a calculation scheme for determining the domestic theoretical biogas potential that is based on summarising the available resources (cultivation, animal breeding, municipal waste). A novelty of the procedure is that it only uses publicly accessible statistical data (e.g. National Statistical Agency, Eurostat).

For thoroughly investigating the injection of biogases into the Hungarian natural gas network it should be laid down in the first step what extreme qualities may occur in the domestic practice both on the side of natural gas and on the side of biogas. Hungary basically relies on two natural gas resources. One is the imported Russian gas typically with a higher than 96 mol% methane content arriving from the direction of Beregdaróc and Győr. The other is the H and S quality, respectively, natural gas of domestic production. Well distinguishable supply regions can be marked off in the country upon these resources and their mixtures. Five natural gas compositions derived from real data and typical to the given supply area were determined for calculating injection possibilities which unanimously characterize the extreme and average values related to the domestic natural gas qualities.

Regarding the composition of biogases produced by fermentation processes no such quite typical composition values can be given as the quality of the produced biogases varies in time not only plant by plant, but also within the plant, depending on the base material quality. When selecting the 5 pcs biogas sample compositions used for the calculations I was aiming that the methane content of the sample gases covers the compositions found in practice (from the approx. 40 mol% methane content of gas from household waste to the approx. 85 mol% methane content of hazardous animal wastes). Sample gas compositions typical to the individual biogas groups were taken based on table B.1 of the German specification *DVGW G 262.* Only main components of the sample gases were tested when determining the maximum injection values.

Pursuant to the principle applied the extreme biogas values, and average compositions, resp., that should be taken into account from the point of Hungarian gas network injection can be clearly characterized that make the investigation of the terms and conditions of injection possible.

The calculation of material and heating technology features (heat value, molecular weight, density, Wobbe-index, compression factor, specific heat, inert content, dynamic viscosity, methane number, water dew point, hydrocarbon dew point, theoretical oxygen and air requirement, flame propagation velocity, inflammation concentration limit values, flue gas composition, combustion product dew point) of the natural gas types supplied to each area as well as that of the biogas types injected into the network in different proportions (sample compositions) was performed using the correlations found in different standards and in the technical literature, and using professional software. (*e.g.: MSZ ISO 6976: 1997; Meszléry, 1978; Farkas-Nagy, 1984; N.V. Nederlandse Gasunie, 1988; Joos, 2005; Program SUPERTRAPP, 2003*). The reference base was the gas technology normal state in all cases.

For the examination of the effect of the individual gas mixtures on gas consuming equipment i.e. their replacement potential harmonized standard *MSZ ISO 13686: 1999* was used. I performed a detailed analysis of the five test methods described in the standard using other technical literature (*Bobok, 1997; Joos, 2005; Poellnitz, 2009*) to determine which method can be used directly for the sample gas compositions. Detailed calculations and analysis was performed with the two selected methods (Wobbe-number and Weaver-index methods) that were found suitable.

4. New scientific results

The new scientific results of the paper can be summarised as follows.

Thesis No. 1

I prepared a calculation method suitable to determine the annually and theoretically available biogas quantity, and using this method I performed the calculations for Hungary. I determined that the theoretical biogas potential of Hungary is approximately an average value of 138 PJ annually.

Presentation of the thesis:
Pursuant to the working hypothesis established when developing the calculation method, only biogas quantities that can be generated from by-products were taken into account for each raw material (cultivation and animal farming by-products, municipal waste). Accordingly the value received does not include the biogas quantities that can be generated from plants cultivated expressly for biogas generation and from organic industrial waste. Furthermore, it does not include the quantity of bio-synthesis gases that can be generated by thermal gasification of solid biomass either. Biogas quantities that can be generated from the individual base materials are based on the specific biogas yield indices described in the technical literature. As these indices cover a value band in every case, therefore a minimum and maximum theoretic potential value was determined for every possible base material. The final potential value can be derived from the arithmetic average of these values. The energy content of the individual biogas types was taken into account based on the recommendation of the German stipulation DVGW G 262 for the methane content of individual biogas types.

Thesis No. 2

I have demonstrated by calculations that hydrocarbon condensation must not be considered at all when preparing a mixture of biogases containing carbon dioxide (or nitrogen) and commercial propane gas at 4°C hydrocarbon dew point temperature used at natural gases – pursuant to standard MSZ 1648:2000 – up to 17.5 bar overpressure. Hydrocarbon condensation might occur between 17.5 bar and 24.3 bar overpressure according to the nitrogen and carbon dioxide proportion of the biogas; above 24.3 bar overpressure hydrocarbon condensation can be predicted.

Presentation of the thesis:
Hydrocarbon condensation is not an allowable phenomenon in the natural gas system. The correlation between pressure and temperature of the hydrocarbon condensation is given by the condensation curve for the given gas composition. Problems typically occur when higher carbon atomic number components are present. When injecting biogases into the natural gas network special attention should be paid to this phenomenon if the preparation of the biogas to network quality is performed using commercial propane (or with high-grade hydrocarbons). The calculation of the related thermodynamic properties of gas mixtures was prepared using the SUPERTRAPP software based on the Peng-Robinson equation of state.

In addition it was determined that the preparation of biogases for natural gas quality, by separating their inert components (the preparation of biomethane), or mixing them with natural gas, would not result hydrocarbon condensation at the pressure and temperature conditions of the Hungarian natural gas system.

Thesis No. 3

Based on my calculation I found that – taking into account the quality provisions in force for gases transported and supplied in the co-operating Hungarian natural gas system – only biogases (having a significant carbon dioxide content) with a methane content reaching 82.1 mol% can be injected into the natural gas system as full value replacement gases.

Presentation of the thesis:
In order that the replacement gas to be injected reaches the lower limit of the 2H quality specified in the standard its methane content should be at least 95.5 mol%. If the methane content is more than 89.3 mol% but less than 95.5 mol% the gas would meet the heat value provisions, however, it cannot be classified neither into the 2S nor the 2H Hungarian gas quality category.

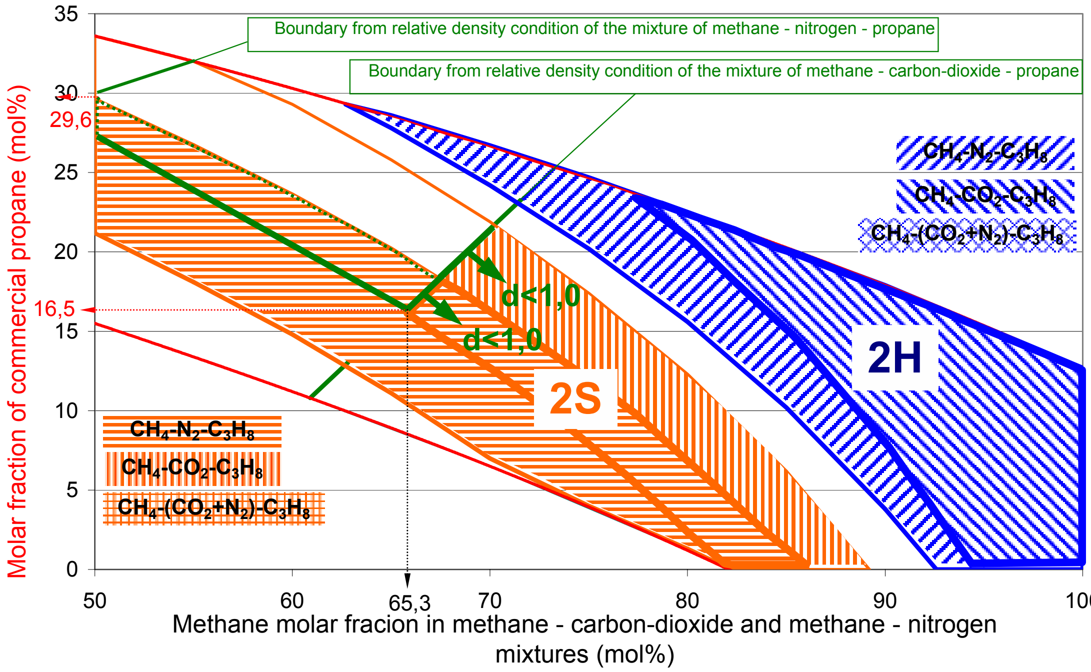
If, besides carbon dioxide, the biogas contains also nitrogen in significant quantity (several tens of percentages), as a consequence of the upper Wobbe number range the minimal methane content cannot be less than 92.6 mol% for 2H gas quality. For 2S quality – although as a consequence of the Wobbe number a mixture containing 77.4 mol% methane would satisfy the requirements of standard MSZ 1648 – the provision for the upper heat value does not permit to inject gas with a methane content less than 82.1 mol% into the network.

Certainly it could be necessary on areas supplied with domestic natural gas to mix commercial propane to the biogas already processed to improve quality when preparing biogas to become full value replacement gas so that the heat value of the gas supplied in the given network section reaches the lower limit value of natural gas.

Thesis No. 4

I prepared a method to graphically determine the maximum commercial propane quantities to be mixed into biogases and required to inject biogases as full value replacement gases into the natural gas network (the biogas in question is reduced to the main components: methane, carbon dioxide and nitrogen, and still contains inert substances).

Presentation of the thesis:
The methane - carbon dioxide – nitrogen, as well as commercial propane mixtures inside the shaded areas (see legend on the diagram area) satisfy the requirements of Hungarian standard MSZ 1648:2000 for the Wobbe number and heat value.



Mixtures containing carbon dioxide and nitrogen (and oxygen, respectively), as well as methane and commercial propane always satisfy the condition of a maximum relative density of 1.0, which is a determinant condition for the safety of the natural gas industry (green lines) in case of 2H gas quality (areas confined with blue lines). In case of 2S gas quality (areas confined with orange lines) the examination of methane and carbon dioxide, methane and nitrogen, and methane and carbon-dioxide+nitrogen mixtures must be separated. If the mixture under examination is to the left of the thick green line (which is the relative density limiting condition) on the shaded orange area (the area confined with a thick and dashed green line) the fulfilment of the relative density of the mixture can be ascertained only by another test. In the case of such mixtures within the limit value range the compliance to the condition of relative density can be decided by another *chart forming a three-dimensional surface*.

Thesis No. 5

I have demonstrated that a biogas-propane mixture produced by quality improvement using commercial propane, and having the same Wobbe number than the natural gas supplied in the network can only be used in limited quantity i.e. as additive gas ('off-spec' injection), for injection into the Hungarian natural gas system.

Presentation of the thesis:
The calculations performed have clearly demonstrated that – taking the typical Hungarian natural gas compositions as the basis – the individual biogas types cannot fulfil the lower heat value requirement (with a maximum deviation of ±5%) characteristic to the given network by adding commercial propane only, when correlated with identical Wobbe number natural gas and biogas-propane mixtures.

Thesis No. 6

My investigations demonstrated that the maximum quantity of biogases of fermentation origin, to be injected into the Hungarian natural gas network as additive gases without improving their quality, can be determined in every case upon the composition of the gas supplied through the natural gas network and that of the given biogas type (first of all its methane content). In the area of Hungary the quantity to be injected into the network could even be 1-100 mol%, depending on the composition of natural gas and biogas.

Presentation of the thesis:
When investigating the limited ('off-spec') injection possibilities of the sample biogas compositions, representing the extreme quality range of biogases, into the sample natural gas compositions, representing the extreme qualities of the natural gas supplied in Hungary, it was demonstrated that the biogas having the highest methane content in Hungary (85 mol%) can be mixed with 2H natural gases with a higher than 97 mol% methane content even in proportion close to 40 mol%, in case it meets the requirements of the standard for gas mixtures. In case of other natural gases that contain less than 90 mol% methane but contain a few mol% high-grade hydrocarbons the mixture ratio is significantly lower (10 – 17 mol%); for biogases that are of the „poorest quality" (containing less than 50 mol% methane) the maximum mixture proportion is a few (2 - 4) mol% only.

In case of natural gas marked 2S the maximum mixing proportion derived is more than 20 mol% (biogas from sewage treatment plant). In case of biogas containing 85 mol% methane the lower heat value of the gas is quite close to the lower heat value of the Hungarian 2S gas quality. Consequently a situation might occur when an unlimited quantity of biogas even without quality improvement can be injected into the given network marked 2S.

Thesis No. 7

My calculations performed for the multiplying factors of the spill equation of the Wobbe number method clearly demonstrated that in case of applying the natural gas compositions characteristic for Hungary, and the representative biogas compositions originating from fermentation processes as replacement gases, or – without improving the quality – as additive gas mixed into the natural gas network, the examination (identity) of the Wobbe numbers derived from the upper heat value is sufficient from the point of interchangeability.

Presentation of the thesis:
The calculations supported that it is sufficient to investigate the identity of the Wobbe numbers of the basic gas (network natural gas) and the replacement gases (biogas or the mixture of biogas and natural gas) when investigating the interchangeability conditions of biogas qualities to be injected into the natural gas network when the quality of the biogas was not improved, or when it was improved using commercial propane, or when quality improving gas (commercial propane) was not added (only the inert content of the biogas was reduced). Other factors than the Wobbe number that affect the amount of heat output of the burner (isentropic and compression factor) influence the amount of chemically bonded energy flowing out of the burner only to a negligible extent (maximum ±0.15%) in the same device, at the same gas temperature, gas pressure and ambient pressure.

5. Utilisation of the results

The results to be transmitted by this paper are suitable to determine the quantitative and qualitative limit values of biogases coming from specific sources to be injected into the network (as replacement gas or additive gas) as a function of natural gas qualities supplied in certain areas of Hungary that are of different qualities but comply with the stipulations of the Hungarian standard. By these limit values the operation of the gas network and consumers' equipment can smoothly continue.

Furthermore, the research results presented in the paper are suitable for the preparation of the presently not yet available technical and regulation provisions of the Hungarian natural gas sector to be made on biogases. The preparation of these regulations is mandatory according to clause (9) § 132 of Act XL of 2008 about natural gas supply, entering into force on 1st July, 2009.

The results of determining the theoretical Hungarian biogas potential can clarify the actual role of renewable gases in the Hungarian primary energy balance for all players of the gas sector.

The results can well be utilised in the university and postgraduate training of gas industry professionals because the knowledge of biogas injection into natural gas networks and transportation are very incomplete. Our undertakings as an EU member state regarding the proportion of renewable energy can be fulfilled only if these energy resources - including biogas that has a determinant potential from the point of the country - are known, accepted and utilised in an increasingly wide area.

One possible direction of further developing the scientific results is to prepare a computer programme based on the results drafted in this paper. The computer programme should be able to determine the terms and conditions of injection as function of the given biogas resource, the quality supplied through the natural gas network used for injection, and the economical feasibility, by providing several alternatives (replacement gas, additive gas, quality improvement).

I tried to draw up the paper so that it is easy to understand, I also tried to put down the chain of thoughts in detail and support with samples if possible. I also had the goal that with the help of the results presented and the related theoretical samples, samples from the technical literature and from already operating European systems the issue is made easy to understand and transparent to all interested professionals. I hope that with the results presented I could raise the interest of certain players of both the natural gas and the biogas industry.

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