

**EXTENDED OPTIONS FOR THE CONDITIONING OF PROCESSED
FERMENTATION GAS USING LIQUEFIED PETROLEUM GAS (LPG)**

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1. ABSTRACT

The basic conditions for the gas grid access in Germany are specified by the Gas Network Access Ordinance (GasNZV). The GasNZV regulates the conditions, on which the gas grid operators must grant connected customers access to the gas grids. The amended version came into force on 09.09.2010. A goal of the new regulation is the promotion of the feed of fermentation gas, by the simplification of the grid access for the fermentation gas of plant operators in Germany. So approximately 6 billion m³ is to be fed and up to the year 2030 10 billion m³ bio methane up to the year 2020.

The feeding and the transport of processed fermentation gas depend both on technical and on economic criteria. For the efficient development of the use of gas from renewable sources the answer to the questions, whether the technical-physical and economic possibilities are given in particular to the LPG addition in the background of the relevant regulations and which options result from it are of great importance.

The paper at hand addresses the determination and argumentation of the technical-physically LPG addition boundary conditions and economic aspects with consideration of the following points:

- The GasNZV sets the following boundary conditions: The gas has to comply DVGW code of practice G 260 and G 262 (date of issue 2007) as well as aspects by intermediate revised versions and possibly higher requirements of the processing, if the condition of the grid gas cannot be achieved by LPG conditioning.
- Composition of the market available LPG (influence of components beside propane and butane). Determination of LPG compositions which allow achievement of higher calorific values.
- Determination of the maximum LPG quantities with consideration of the phase behaviour in comparison to the distributed natural gases on basis of a detailed analysis of distributed natural gas up to C₄₀ hydrocarbons.
- Determination of the methane numbers and K-number-deviations for selected pressure and temperature ranges.
- Influence of other admixtures (sulfur and oil residues) on the condensation behavior.
- View of further technical options: Using ethane. Background is the consideration of all conceivable options and their evaluation (especially for higher calorific values).

Subsequently data on LPG and distributed natural gas has been collected by analytical gas probe examination. Using this data base and the characteristic data of the processing routes the largest calorific values are indicated by consideration of the boundary conditions. Additionally conditioning with ethane has been treated and evaluated as option. Finally economic aspects are discussed to the individual options of conditioning. The summary supplies the attainable calorific values under the given conditions.

In this work funded by DVGWG the max. attainable calorific values is addressed to. Starting point is thereby the formulation in the GasNZV that a connection inquiry can be granted negatively, if it is technically and economically not possible and/or unreasonable (§ 17 - 2 of German Energy Industry Act EnWG). This could happen if the grid operator cannot achieve the calorific value of the distributed gas by the technically limited LPG addition due to a not sufficient methane concentration in the processed fermentation gas, although the processed gas is supplied complying G 260/G 262. The permissible quantities of LPG (DIN 51622) are limited through:

1. The DVGW code of practice G 486 (measurement of volume, compressibility, applicability SGERG, AGA8)
2. The DVGW code of practice G 260 (density, condensation of higher hydrocarbons)
3. The methane number (automotive applications, DIN 51624, MZ > 70, max. 6% propane)
4. Condensation higher hydrocarbons (e.g. Natural gas vehicle refueling procedure, processing, expansion)

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2. INTRODUCTONAL EXAMPLES FOR L- AND H-GAS

For a short introduction to the problem addressed two examples for L- and H-gas are outlined.

Example: L-gas: A rule-compliant L-gas is not producible from this basic composition without conditioning. It has to be admitted at least air, since the Wobbe index is too large (theoretically a gas with 10%, prepared on 93% methane with air addition is G 260/262-complying).

Referring to the plant operator the following aspect has to be considered: The necessary LPG addition by the network operator for calorific value rise must be co-ordinated with the processing degrees and the air addition (G 260/262 date of issue 2007). The regulations for the air addition (plant operator) and the liquid gas addition (network operator) must be co-ordinated. The figure 1 shows the range of the rule-conforming mixtures (green marked) with a processing to 93 % methane with propane/air addition for a goal calorific value of 10,3 kWh/m³. The range is restricted by the CO₂-concentration, maximum LPG quantity and the O₂ concentration. LPG addition yields a smaller methane number. For the marked calorific value volume 4-10% LPG addition are necessary.

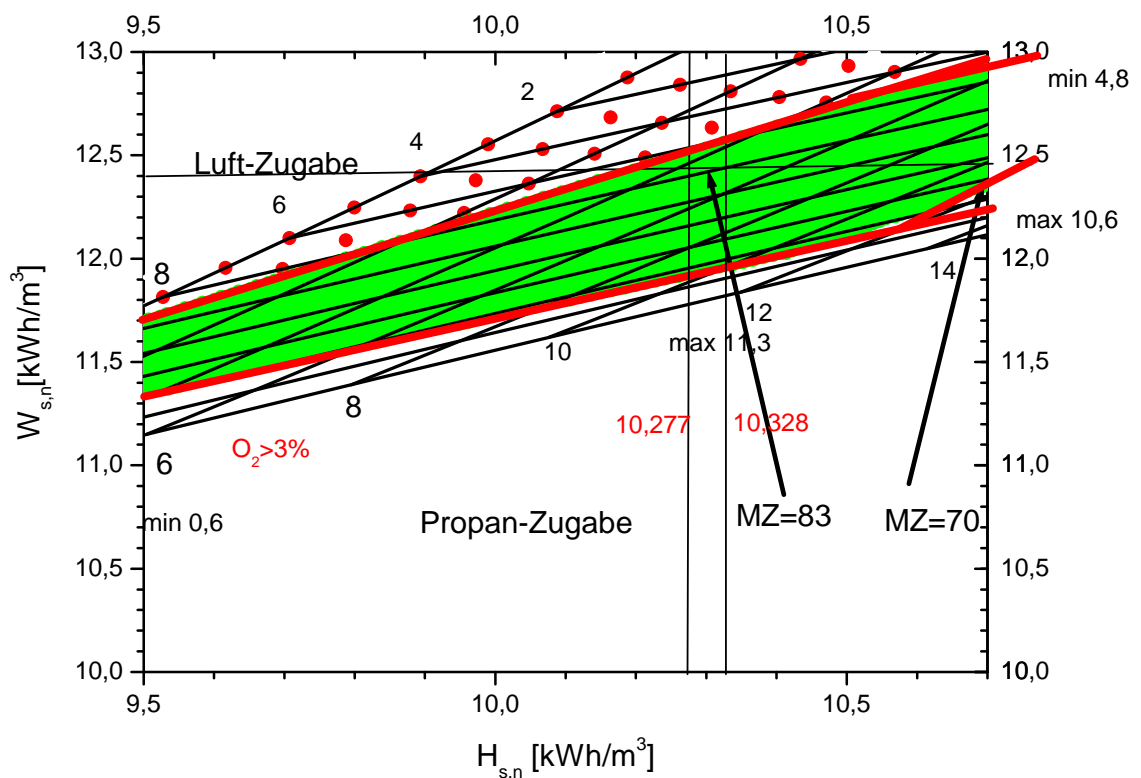


Figure 1: Detailed view: 93 Vol.-% methane, addition of propane/air

Summarized this means that by the 6 VOL. - % boundary condition from G 262 in particular this is a quite complex condition for L-gases to be fulfilled and large LPG quantities are required.

Example: H-gas:

The following figure 2 points the necessary LPG addition to rise the calorific value of the processed fermentation gas to natural gas H - values as function of the preparation degree by the concrete example and within the set of rules restrictions (amount of propane/butane and max. calorific value). With a minimum processing after G 260/G 262 (6 VOL.-% CO₂) there are in any case more than 6% propane/butane necessary in order to reach the calorific value of a H-gas with calorific values exceeding 11.4 kWh/m³ - with a calorific value of 12 kWh/m³ this yields approx. 10%. In figure 2 the boundary conditions from G 486 are drawn in: 3.5 % propane (AGA8 p > 100 bar), 5 % (SGERG) and 6 % (AGA8, CNG). With small preparation degrees furthermore the range of the relative density leaves the range specified in G 260. With higher preparation degrees the methane number falls below a value of 70 starting from approx. 8% LPG addition.

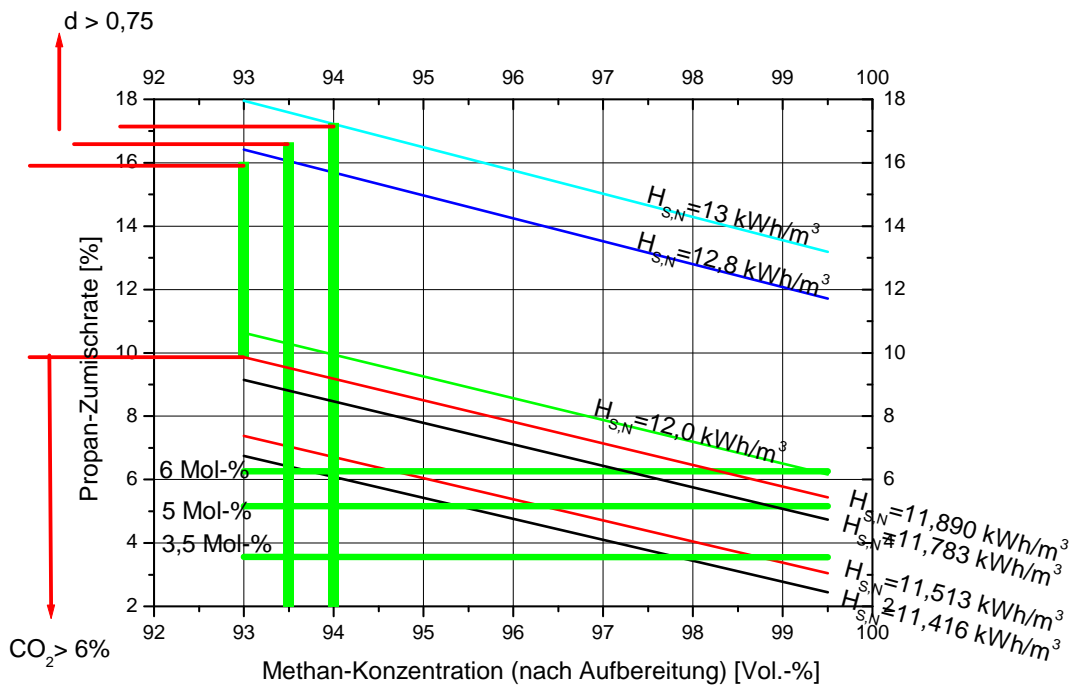


Figure 2: Propane admixing as function of processing degree

Conditioning and/or admixing LPG due to the calorific value adjustment - particularly on values of higher calorific H-gases depends thus directly on the preparation degree. This is clarified in Figure 3: With processing on 94 % methane approx. 10 % LPG is necessary, but the methane numbers are smaller than 71 for a calorific value of 12 kWh/m³.

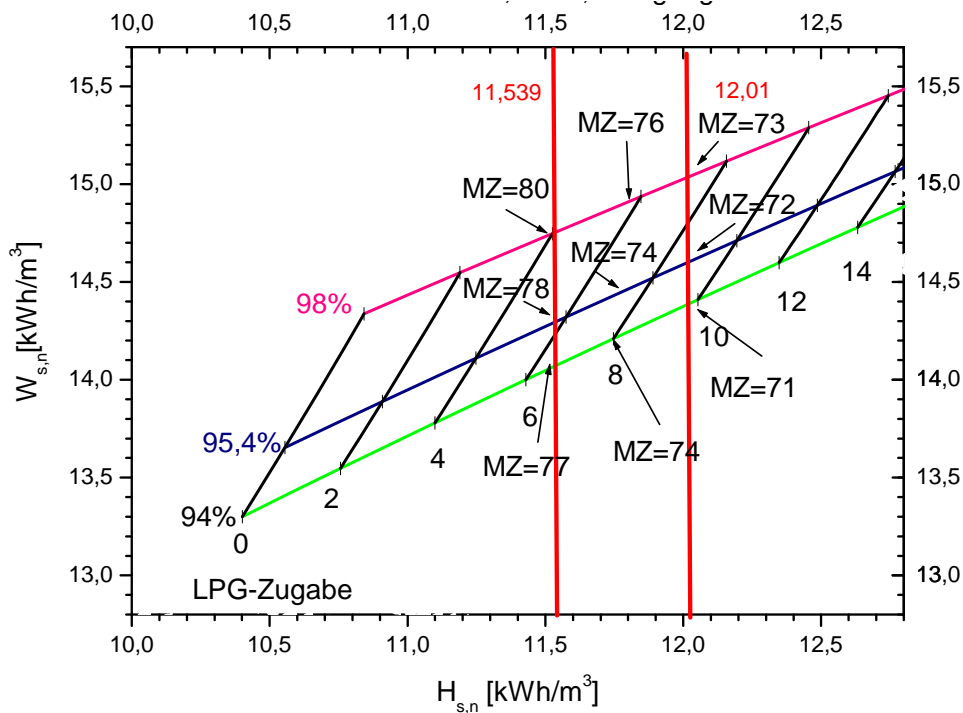


Figure 3: LPG-admixing as parameter

A processing on 98 VOL.-% methane requires a LPG addition (propane) of 7,4 % to the prepared gas. The methane number is 73, however conflicts with code of practice G 486 result.

3. NEW ASPECTS

The consideration to define a minimum content of methane from the processing for L and H-gases will be introduced by the revised version of the G 262. The limit of 6 VOL. - % CO₂ will not be applicable any more, if for L-gas a processing up to 90 and for H-gas 95 VOL. - % will be prescribed 8. Hence it follows that with carbon dioxide concentrations in the prepared fermentation gas, which result from the Wobbe index limits of G 260 it is possible to achieve the rule-conforming Wobbe index range (10.5-13 kWh/m³) for L-gases without an air addition. The processing and conditioning of fermentation gas can thus be arranged theoretically simpler and more economical, since the sophisticated case of the liquid gas and air mixing is no more necessary. The range of the rule-conforming mixtures is increased and thus the access for prepared fermentation gas to the natural gas grid is simplified.

To verify this approach a study was accomplished for „the determination of the CO₂-limit for operation of domestic appliances “by the DVGW 9.

In the case of H-gases it is considered by the range of values of larger/equivalent 95 VOL. - % that a smaller processing requires such a huge quantity of liquid gas to achieve calorific values of 11,5 kWh/m³ that technical limits are reached. The Gas Network Access Ordinance (GasNZV) declared that even higher requirements than specified in G 260/G 262 can be prescribed, if the condition in the grid cannot be achieved 11. Main focus of the study at hand is to supply information, which facilitates an argumentation and makes also grids with higher calorific values attainable for fermentation gas.

The framework of the technical boundary conditions of the feed of prepared fermentation gas is thus set by the GasNZV and the current rule and standardization work. The main statements are:

1. A minimum preparation of the fermentation gas is demanded by a plant operator in accordance with the DVGW codes of practice G 260 and G 262 (GasNZV).
2. The conditioning of the prepared fermentation gas is to be accomplished by the grid operator who is responsible for the adherence to the standards in accordance with code of practice G 685 at the point of delivery.

Furthermore from the rule and/or standardization work the DVGW code of practice G 486 „deviation from ideal gas factors and compressibility figures of natural gases “and the DIN 51622 (liquid gases) as well as DIN 51624 (natural gas as vehicle fuel) have to be taken into account. Some of them are at present in the revision.

For a closed evaluation of the context detailed information is necessary over liquid gases for conditioning, distributed natural gases and the usual upgrading technologies, which are arranged shortly in the following.

4. LIQUEFIED PETROLEUM GAS

In the context of the gas net entrance regulation conditioning with liquid gas (LPG places: Liquefied petroleum gas) is a standard solution for the rule-conforming feed of fermentation gas into the natural gas grid. Liquid gas results as by-product during oil refining and exploration of oil and natural gas. The fuel LPG is gaseous at ambient temperature on atmosphere conditions, can already be with low pressures liquefied. In the liquid condition its specific volume is approx. 260-mal smaller than in the gaseous condition. Therefore large energy quantities in relatively small containers can be transported and stored.

It is differentiated between field and refinery commodity. The first one is delivered by LPG tankers in Brunsbüttel, continued to transport and processed usually as propulsion gas. Further applications are given by the use in the chemical industry than starting point for plastics, heating gas and automotive fuel („autogas “). In Germany LPG means „autogas “that consists in the summer/winters of 40/60 and/or 60/40 mass. - % propane/butane. Liquid gas according to DIN 51622 consists of C₃ and C₄-hydrocarbons and/or mixtures of these.

The transport of LPG is made world-wide with sea-tankers, river boats, by pipelines, with railway tankers, road tank car or in liquid gas bottles. The German liquid gas market is based on altogether approx. 14 refineries, and about 140 fuel depots. The composition of these gases takes place behind the fuel depot. Delivery is according to DIN 51622, DIN 589 13, or optimized (alkenes limited) or as mixture from DIN 51622 and butane.

For calorific value adjustment and/or - increase liquid gas is added to the prepared fermentation gas, in order to fulfill the defaults given by G 685 14. Commercial propane consists of at least 95 mass per cent propane and propene, whereby the propane portion has to be higher. The rest may be composed of ethene (C₂H₄) ethane (C₂H₆), butane (C₄H₁₀) - and buten (C₄H₈) - isomers. The classification for commercial Propen, butane and butene is equivalent. The degrees of purity has to be considered according to DIN 51622. There limit values of sulfur and/or sulfur compounds are specified.

Further limits for a liquid gas admixing are specified in other sets of rules. Dependent on the applying boundary conditions, e.g. CNG stations, these limit values are to be consulted likewise. In DIN 51624 „fuels for motor vehicles - natural gas requirements and testing methods “ upper limits for the propane/butane mol fractions in the natural gas of 6%/2% in the total mixture and a methane number of 70 are demanded. The EASEE gas CBP 15 indicates a hydrocarbon dew point of -2 °C at 1-70 bar.

In principle it is possible to refer other liquid gas compositions and to thus convert the limits mentioned for propane and butane concentrations with appropriate mixed liquid gases more effectively. In past studies almost exclusively propane in the composition of 95/5 mass. - % for the computations was uses. Here additionally an „ vehicle gas“ with 60/40 and an adapted mixture with 75/25 mass. - % is uses. Furthermore influences are considered from deviations to these theoretically pure mixtures by propene /butane-fractions.

To get an overview of the variance of the mixtures within the DIN specs at different locations samples were arranged and analyzed by the DVGW research centre at the Engler Bunte institute Karlsruhe of the institute for technology (KIT). This permits a qualitative evaluation of the different compositions.

The calorific values varied within a range from 27,457 to 28.388 kWh/m³. This results from larger propane/propen volume fractions and at the same time smaller butane percentages by volume. In practice this means that conditioning must be designed for slightly larger liquid gas mixing. For the conversion it is to be considered further that the methane concentration from the processing can be subject to fluctuations likewise. The regulation elements of the mixing plant as well as the measuring instruments exhibit tolerances, so that the error propagation by the system with the range to be achieved has to be considered.

Table 1 shows the influence on the mixing quantity during a theoretically pure composition of 95/5 mass. - % propane/butane, a mixture according to DIN 51622. Basis are the natural gas compositions in the next chapter and the fermentation gas upgrading with Pressure Swing absorption, Pressure water washing (DWW), Genosorb- (GW) and DEA-technology (DEA).

The addition of liquid gas is to be understood as mixing to the prepared fermentation gas. That means that the portion of the total mixture (100% + X) is smaller than the mixing value. The liquid gas mixing used here represents the admitted portion. The values for the Wobbe index, calorific value and the delimitations of the propane and butane portions result from the total mixture. The mixing degree is computed as follows:

$$Z = \frac{\dot{V}_{LPG}}{\dot{V}_{Biogas}}$$

Table 1 Admixing for different upgrading processes conditioned with 95/5 Mass.-% Propan/Butan

Flüssiggas 95/5		PSA	DWW	GW	DEA
Vol.-% Methan		97	98,5	98,5	99,7
Brennwert in kWh/m ³		Flüssiggas- zugabe in % zum Biogas	Flüssiggas- zugabe in % zum Biogas	Flüssiggas- zugabe in % zum Biogas	Flüssiggas- zugabe in % zum Biogas
Waidhaus/Russland	11,226	2,8	1,9	1,9	1,1
RWE Süd	11,337	3,5	2,6	2,6	1,8
Verbund E.On/gegen Russland	11,404	4,0	3,0	3,0	2,2
Nordsee/Dornum	11,492	4,5	3,5	3,5	2,7
Hünxe	11,551	4,8	3,9	3,9	3,1
Phillips Emden	12,028	7,9	6,9	6,9	6,1
Dong Gas Dänemark	12,076	8,2	7,2	7,2	6,4

For the technical conversion of the liquid gas admixing the measuring technique and the equipment technology are extremely important. A calorimeter/PGC takes up the calorific value of the grid gas, the regulation can determine and admit the correct quantity of LPG on the basis of the desired value and the current methane content of the prepared fermentation gas. Dependent on the differences caloric values can be adjusted in a timeframe from 3 to 5 minutes.

5. DISTRIBUTED NATURAL GAS

For an overview of natural gas distributed in Germany samples were taken and analysed (table 2). Due to the fact that natural gas is „a natural product“, it may just be interpreted as candid photograph, which shows however the main differences. For the calculations of the liquid gas admixing quantities as well as the K-numbers, hydrocarbon-dew-points and amount of hydrocarbon condensate these were taken as basic values. The table 2 shows burn technical values values. The calorific values of the analyzed samples cover a range from 11,226-12,086 kWh/m³, higher values may be possible.

Table 2: Burn-technical values of natural gas probes

	Bezeichnung	Nordsee/ Dornum	Verbund E.ON / gegen Russland	Dong Gas Dänemark	RWE Süd	Waidhaus/ Russland	Philips Emden	Hünxe
	Probenahmeort	Wardenburg	Werne von Paffrath	Schuby	Stolberg von Bocholtz	Waidhaus	Rysum	GVH
Bezeichnung	Kurzbez. / Phys. Einheit							
Molare Masse	M [kg/kmol]	18,071	18,043	18,097	18,331	16,686	18,544	18,108
Realgasfaktor	Z _n [-]	0,9971	0,9971	0,9969	0,9971	0,9974	0,9968	0,9971
Normdichte	ρ _n [kg/m ³]	0,8086	0,8073	0,8099	0,8202	0,7464	0,8300	0,8103
Relative Dichte	d [-]	0,6254	0,6244	0,6264	0,6344	0,5773	0,6419	0,6267
Brennwert (molar)	H _{sm} [KJ/mol]	924,536	917,577	971,407	912,124	903,514	967,471	929,328
Brennwert (Masse)	H _s [MJ/kg]	51,160	50,856	53,678	49,758	54,147	52,172	51,320
Brennwert (Volumen)	H _{sv} [MJ/m ³]	41,370	41,056	43,475	40,813	40,413	43,302	41,584
Brennwert (Volumen)	H _{sv} [kWh/m ³]	11,492	11,404	12,076	11,337	11,226	12,028	11,551
Heizwert (molar)	H _{lm} [KJ/mol]	835,187	828,830	878,286	824,157	814,957	874,988	839,669
Heizwert (Masse)	H _i [MJ/kg]	46,216	45,937	48,532	44,959	48,840	47,185	46,369
Heizwert (Volumen)	H _{lv} [MJ/m ³]	37,372	37,085	39,307	36,877	36,452	39,163	37,572
Heizwert (Volumen)	H _{lv} [kWh/m ³]	10,381	10,301	10,919	10,244	10,126	10,878	10,437
Wobbeindex	W _s [MJ/m ³]	52,311	51,957	54,929	51,240	53,191	54,045	52,529
Wobbeindex	W _s [kWh/m ³]	14,531	14,433	15,258	14,234	14,775	15,013	14,591

Combined with the data of the liquified petroleum gas probe analyse this represents the basis data for the calculation.

6. RESULTS AND CONCLUSION

The figure 4 presents the results in summary. The calorific value is shown as function of the liquid gas admixing for 3 liquid gas compositions (95/5, 75/25 and 60/40 propane/butane) and 3 upgrading degrees (97, 98, 99.5 % methane in the prepared fermentation gas, conditioned with the respective liquid gas compositions). The methane numbers (MZ) are noted at the points of mole limits (3.5, 5 and 6 mol %) from G 486 (small symbols). Additionally limits are shown for methane numbers butane mol portions of 1,5 %, K-number-deviations and condensation lines (KKT). As a limit for the condensation lines a krikondenthermtemperature of approximately -17°C was drawn in, in order to achieve a sufficient distance to the 1 mol quality lines most natural gas:

Using a composition of 95/5 propane/butane the limits based on mole fractions referring to G 486 (3.5/1.5 mol % propane/butane (AGA8 p > 100 bar), 5 mol % propane (SGERG), 6/1.5 mol % propane/butane (AGA8 p < 100 bar) are approx. 3.7, 5.4 and 6.5% admixing. The methane number of 70 with approx. 7.6% (99.5% processing), and with approx. 9.8 % the phase limit line with approx. -17°C will be reached.

Using a composition of 75/25 propane/butane the limits based on mole fractions referring to G 486 (3.5/1.5 mol % propane/butane (AGA8 p > 100 bar), 5 mol % propane (SGERG), 6/1.5 mol % propane/butane (AGA8 p < 100 bar) are approx. 4.5, 6.6 und 8 % admixing. The 1,5 Mol-% butane-limit is reached at approx.

7.7 %. The methane number of 70 with approx. 6.6% (99.5% processing), and with approx. 6.7 % the phase limit line with approx. -17°C will be reached.

Using a composition of 60/40 propane/butane the limits based on mole fractions referring to G 486 (3.5/1.5 mol % propane/butane (AGA8 p > 100 bar), 5 mol % propane (SGERG), 6/1.5 mol % propane/butane (AGA8 p < 100 bar) are approx. 5.5, 8 und 9,8 % admixing. The 1,5 Mol-% butane-limit is reached at approx. 4.4 %. The methane number of 70 with approx. 6.6% (99.5% processing), and with approx. 5.6 % the phase limit line with approx. -17°C will be reached.

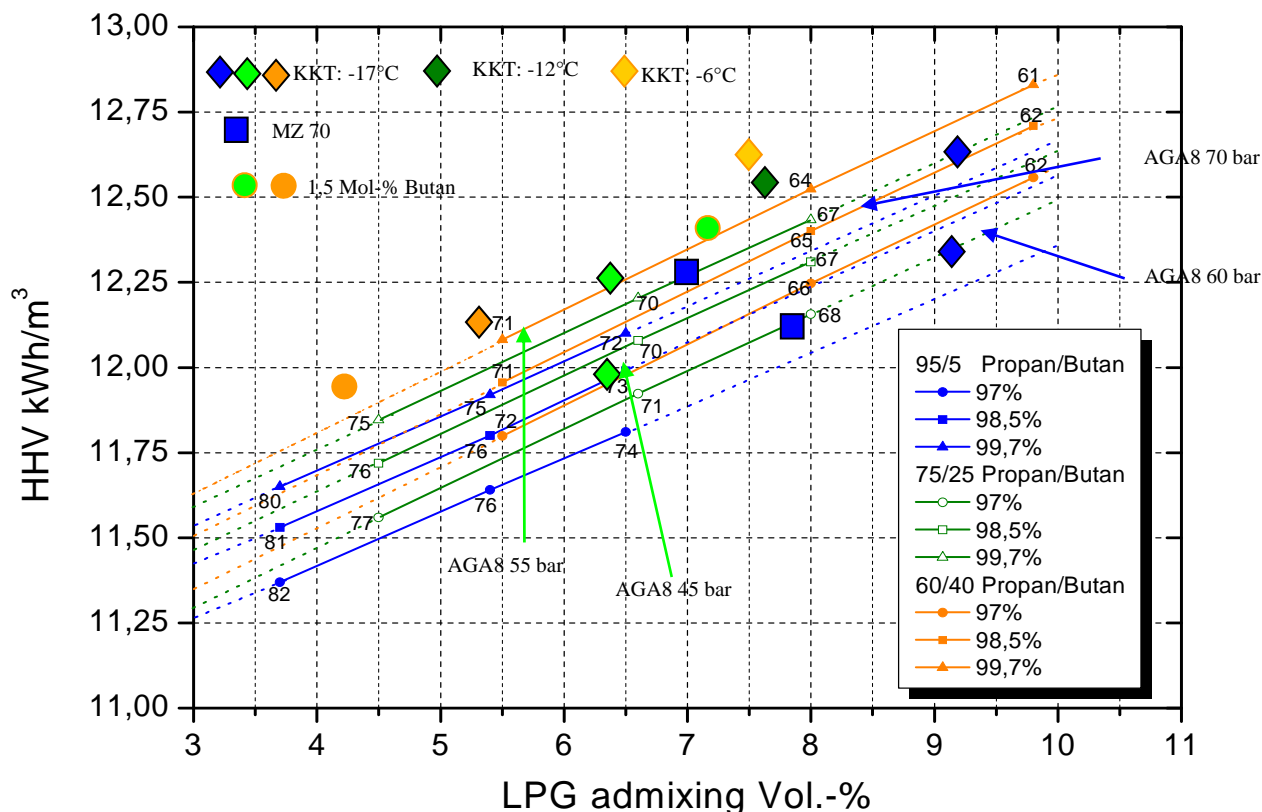


Figure 4: HHV as a function of LPG-Admixing for 95/5, 75/25 und 60/40 propane/butane with mole-fraction limits an methane numers marked

With smaller butane portions the attainable calorific values are limited first by the propane mol fraction, the methane number of 70 goes below the limit at higher mixing rates. The sequence reads: Mole fractions (applicability of the equations of state (SGERG, AGA8)). Methane number (MZ=70), condensation of higher hydrocarbons, K-number-deviation. With 75/25 propane/butane there is another order of limits: Mole fractions (applicability of the equations of state (SGERG, AGA8)) to 5 mol % propane. Methane number (MZ=70), condensation of higher hydrocarbons, mole fraction of butane 1.5%, K-number-deviation (AGA8 50 bar), mole fraction limit of 6 mol % propane.

With 60/40 propane/butane the sequence of limits reads: Mole fraction limit butane 1.5%, methane number (MZ=70), K-number-deviation (AGA8 50 bar), condensation of higher hydrocarbons, mole fractions (applicability of the equations of state (SGERG, AGA8)) up to 5 mol % propane, mole fraction of 6 mol % propane.

With the 3 liquid gas mixture variants and 99.5% methane from the processing approximately 11.63 can be attained 11.8 and 11.89 kWh/m³ in the calorific value below the first restriction (3.5/1.5 Mol% propane/butane). In the case of feed up to 25 bar, without consideration of downstream grids calorific values are attainable until 12.2 kWh/m³ under adherence to a methane number border of 70 with propane/butane 75/25, with a processing on 97% methane approx. 11.9 kWh/m³ is possible. In each case the measurability of the liquid gas portions is to be considered.

The cost framework for the conditioning gas depends on the preparation degree of the fermentation gas and the calorific value which can be obtained and lies with current liquid gas prices under assumptions made between 2 and 14 ct/m³ (product gas). With calorific values higher than 12 kWh/m³ the costs are approx. 11 ct/m³. A difference in the preparation degree from 99,5 to 97% methane results in 3 ct/m³ higher costs of the product gas.

Using ethane instead of propane/butane mixtures calorific values are possible up to 11.8 kWh/m³ within the limits of G 486. But there is no market for ethane in Europe at present contrary to the American continent. Due to the larger necessary quantities the ethane can cost approx. 580 €/t inclusive transport and taxes, in order to draw even with propane/butane mixtures.

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