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Study on the Physiochemical Properties of Liquefied Petroleum Gas Available by Cylinders in Bangladesh

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Abstract: The work analyzed the composition and other physiochemical properties of liquefied petroleum gas (LPG mixture available in Bangladesh by cylinder. LPG Composition (C_2-C_6), Butadiene, Total Sulfur, Cu Corrosion, Relative Density and Vapor Pressure were determined by respective ASTM methods. Free water content was measured visually and odour tested by smell detection. Data from nine samples were used to compare with ASTM and Indian Standards. The results of this analysis revealed that the LPG contains propane and butane with a certain range of ratio. But that has no free water content. The total Sulfur content is within the permissible limit according to ASTM Standards. The samples do not have corrosive hydrogen sulfide. The vapor pressure is lower than the maximum limit and have very low linear relationship with relative density. The calorific value is highly dependent on the propane and butane content and moderately related to the relative density. Compared to ASTM and Indian standards, the examined LPG mixtures are found to be safe for the tropical country.

Keywords: LPG mixture, physiochemical properties, propane-butane ratio, calorific value of LPG.

I. Introduction

Liquefied petroleum gas (LPG) is a valuable energy source used worldwide for numerous business applications in industry and transportation. The largest market for LPG is the residential/commercial market, followed by the chemical industry, where it is used as a petrochemical feedstock and agriculture [1]. LPG is a pressurized fuel gas that contains a mixture of hydrocarbon gases, most commonly propane (C_3H_8) and butane (C_4H_{10}). It is pressurized in the form of a liquid and stored in a steel container or cylinder for sale or commercial use. The types of LPG sold and purchased worldwide mainly include propane (C_3H_8) or butane (C_4H_{10}). In the polar climate region of Northern Hemisphere, the mixture contains more propane in winter, while it contains more butane in summer since propane has a lower boiling point of -42°C compared to -0.4°C for butane [2].

The demand for liquefied petroleum gas (LPG) in Bangladesh is very high. The JICA survey team made various assumptions, such as all the new housing except existing natural gas pipeline connections will be forced to use LPG, the demand for gasoline will grow faster, and the demand will be partially met by LPG. The growth rate in 2041 is increasing dramatically and is 15 times higher than in 2016 [3]. In the public sector, 12,361 tonnes have been produced in the financial year 2021-22, while 1,531,231 tonnes imported by private companies. Therefore, the public and private sectors together market 1.54 million tonnes of LPG, which meets a certain part of the country's LPG demand [4]. But according to the LPG Operators Association of Bangladesh (LOAB), the country's annual LPG consumption has been reached about 1.8 million tonnes. To meet this demand, about 27 LPG operators are presently catering in Bangladesh [5]. Different operators distribute LPG with different limits of certain specification. In this study, the LPG specifications of different operators have been compared with ASTM Standards and the respective calorific values and propane and butane ratios were examined to acquire knowledge regarding the LPG available in Bangladeshi market. The ASTM Standard Specification covers the products commonly referred to as liquefied gases, consisting of propane, propene (propylene), butane and mixtures of these materials. To cover common areas of application, four basic types of liquid gases are available. This specification applies to products intended for use as residential, commercial and industrial heating and fuels [2].

This article examines the composition and other physiochemical properties of LPG available in Bangladesh and compares with the ASTM and Indian Standards. The aim is to collect information about the LPG available in Bangladeshi market as there is no or very few studies on this topic. The study was also conducted for the validation and future reference.

II. Literature Review

LPG production did not begin until around 1904, almost 40 years after oil and gas production began in North America around 1860. By 1900, natural gas (methane) cooking and lighting appliances were ubiquitous, but the gaseous fuel was difficult to transport [2]. Compressed natural gas (CNG) could neither be transported nor stored in the large pressure vessels of the time. For mobile and remote gas applications, LPG is the best fuel. At average ambient temperatures and mild pressures (250 psig), it is a fluid of high calorific value (BTU) that is easy to store and transport. Once vaporized, it can be used in natural gas appliances that convert into gaseous LPG mixtures with minor adjustments to the air/fuel ratio. Without any standardization or regulatory

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restrictions, the industry's early years were characterized by the need to address immediate problems. One of the first consumer goods to be transported and sold in pressure vessels, was liquefied natural gas. Manufacturing, transporting and ultimately selling to the general population presented numerous economic and technical obstacles. The industry has been a leader in several areas of research and development, from manufacturing techniques, equipment and devices to analytical testing methods for composition and properties. The results were (in hindsight) predictable and there were many excesses. However, marketing and regulatory constraints ensured that the new business flourished in a short period of time [2].

LPG is produced by refining petroleum (crude oil) or extracting streams of petroleum or natural gas that emerge from the ground. Walter O. Snelling created it in 1910 and the first commercial goods hit the market in 1912. It currently produces about 3% of total energy consumption and burns relatively cleanly, with no soot and very little sulfur. Because it is a gas, it does not pollute the soil or water, but it can pollute the air. LPG (propane) has a specific calorific value of 46.1 MJ/kg, compared to 42.5 MJ/kg for heating oil and 43.5 MJ/kg for premium grade gasoline [6].

However, its energy density per unit volume is lower than that of gasoline or heating oil at 26 MJ/L due to its lower relative density (approximately 0.50 - 58 kg/L compared to 0.71 - 0.77 kg/L for gasoline/gasoline). Since the density and vapor pressure of LPG (or its components) change significantly with temperature, this fact must be taken into account whenever the application involves safety or custody transfer operations [7].

LPG is an excellent fuel for cooking due to its higher calorific value, good stove efficiency, low cost and environmentally friendly properties. The simple and precise control of the liquid gas stove not only makes cooking easier, but also saves time. Due to increasing urbanization, both urban and rural communities are increasingly reliant on LPG. Since it is very costly to extend gas distribution networks to remote areas, LPG is the perfect choice for household cooking [8]. A comparison of the calorific values of LPG and various cooking fuels [9] is shown in table -1.

Table 1 Calorific values of various fuels [9].

Name of the Fuel	Calorific value (KJ/kg)
Wood	14400-17400
Charcoal	29600
Kerosene Oil	41000
LPG	45750

III. Sampling and Analytical Procedures

A random sampling method was used for laboratory testing and primary data collection. Three LPG cylinders of different brands/companies (available in Bangladesh) were collected from the local market on the same day. All of the three samples (named S1, S2 and S3) were 12kg LPG cylinders. The samples were prepared in the laboratory on the same day and went through the specification/method (table 2) for testing and data collection. In this study, secondary data for six samples (named S4, S5, S6, S7, S8 and S9) were used which were collected from different LPG companies available in Bangladesh and their supplier. They have supplied their analytical reports which were also done by the same analytical procedures.

Table 2 Testing methods for data collection.

Serial no.	Description of Test	Method
01	LPG Composition (C_2-C_6)	ASTM D2163
02	Butadiene	ASTM D2163
03	Total Sulfur	ASTM D6667
04	Free Water	Visual
05	Cu Corrosion	ASTM D1838
06	Relative Density	ASTM D1657
07	Vapor Pressure	ASTM D1267
08	Odour	Smell

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Gas chromatography was used to determine hydrocarbons in liquid gases and propane/butane mixtures according to ASTM D2163 (the official designation for this standard). This method applies to the determination of the number of individual hydrocarbons in liquefied petroleum gas and propane-propene/butane mixtures, with the exception of high-purity propene in the range C_2 to C_5. The concentrations of the components are measured in the range from 0.01 to 100% [10]. To properly characterize the LPG sample, additional testing like infrared (IR) spectroscopy, gravimetry, gas chromatography with flame ionization detection etc. are required as the test conducted is unable to identify hydrocarbons heavier than C_5 as well as non-hydrocarbon components.

ASTM D1657 is a standard test method for measuring the density or specific gravity of light hydrocarbons using a pressure hydrometer. This test method is used to determine the density or specific gravity of light hydrocarbons such as liquefied petroleum gases (LPG) with the vapor pressures greater than 101.325 kPa (14,696 psi). At the test temperature, materials with a vapor pressure greater than 1.4 MPa (200 psi) must not be used with the approved device. Higher pressures can apply to other equipment designs for measuring the density of Castor oil, silicon oil, propylene glycol, ethylene glycol, and ethanol etc. [11]. The initial readings from the pressure hydrometer are not density measurements, but rather uncorrected readings from the hydrometer. Readings are taken with a hydrometer at a reference temperature of 15°C and then corrected to the reference temperature using calculations and the addition to the D1250 Guide to Petroleum Measurement Tables (API MPMS) for the meniscus effect, thermal glass expansion effect, alternative calibration temperature effects and Chapter 11.1 or API MPMS Chapter 11.2.4 (GPA TP-27), if applicable [12]. ASTM D1267 applies to the determination of gauge vapor pressure of LPG products at temperatures from 37.8°C (100°F) to 70°C (158°F) [13]. In this case, 37.8 °C (100 °F) was considered as a reference for measuring vapor pressure. In addition, the internal pressure volume and net weight were determined by laboratory instruments.

ASTM D1838 is the standard method for copper corrosion testing to determine compounds in liquid gases with a vapor pressure greater than 124 kPa at 37.8 °C that may be corrosive to copper. Copper corrosion limits provide assurance that problems will not occur with deterioration of the copper and copper alloy fittings and connections of equipment commonly used in production, treatment, processing, storage and transportation operations in the oil industry. For the ASTM D1838 copper corrosion test method, a polished copper strip is immersed in approximately 100 mL of the sample and exposed to a temperature of 37.8 °C (100 °F) for 1 hour in a cylinder capable of withstanding a hydrostatic pressure of at least 6900 kPa or 1000 psig [14]. The copper strip is cleaned and inspected for signs of deterioration. The results are evaluated by comparing the spots on the copper strip to the ASTM color match scale of 1A to 4C. A rating of 1A is given for the appearance of freshly polished copper coupons with slight but barely noticeable discoloration. The 1B rating indicates a slight haze, and the ratings go further down the scale as the corrosion discoloration of the test piece increases, with 4C being the worst and typically appearing as a heavily corroded, blackened and pitted specimen.

Propane has a lower boiling point of -42°C compared to -0.4°C for butane. Therefore, propane will continue to evaporate in colder climates [15]. The calorific value of LPG refers to the amount of thermal energy released from a given volume of fuel. Liquefied gas consists of the main components propane and butane, which have a calorific value of around 46 MJ/kg and 49 MJ/kg respectively [16]. Hence the calorific values of LPG mixtures of samples were calculated as $CV = (46 \times Propane\ \%) + (49 \times Butane\ \%)$ MJ/kg (1)

Since constituents other than C_3 and C_4 contribute a very small amount in the composition of LPG (less than 1%), they are omitted in the calculation of calorific values for the samples.

IV. Results and Discussion

The results from laboratory test and secondary data sheets are depicted here in the following tables. Table -3 shows the composition of LPG of the analyzed samples. Table-4 shows other physiochemical properties of the analyzed samples. Table 3 shows that LPG is mostly composed of propane and butane. These two components contribute more than 99% of the total mixture. The propane and butane ratio of the analyzed samples ranges from 29.63:69.67 to 40.511:59.147. The primary data reflects that the propane content ranges from 32.771 to 40.511 mol percent. Sulphur content ranges from 13 to 15 ppm. The minimum vapor pressure is 544.69 and maximum vapor pressure is 599.84 KPa. Relative density ranges from 0.549 to 0.551 kg/L. Calorific value ranges from 47.62 to 47.89 MJ/kg. The secondary data reflects that the propane content ranges from 29.63 to 34.69 mol percent. Sulphur content ranges from 5 to 16.97 ppm. The minimum vapor pressure is 583.49 and maximum vapor pressure is 667.8 KPa. Relative density ranges from 0.5506 to 0.5535 kg/L. Calorific value ranges from 47.6928 to 47.7681 MJ/kg. The free water content is nill for both primary and secondary samples. All the primary and secondary data show that the butadiene is less than 0.01 of mol percent, residue on evaporation is less than 0.05 ml/100 ml. The cu corrosion reflects 1A for all the samples.

There are ASTM D1835 and Gas Processors Association GPA 2140 specifications for commercial propane-butane blends, but these are rarely used for consumer applications in North America. There is no current Canadian General Standards Board (CGSB) specification for propane-butane blends because winter temperatures are too cold and butane demand for winter gasoline production is high. In polar climates, propane must be used year-round to ensure low temperature operation. Tropical climates (no winter temperatures, no winter gasoline) tend to use propane-butane blends year-round to utilize the butane. In temperate climates with large seasonal temperature fluctuations, propane could be used in winter and propane-butane mixtures in summer [17].

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Some countries have either100% propane (Australia & USA). An LPG gas mixture of 60:40 propane: butane is used in New Zealand & Belgium. The percentage of propane and butane (propane : butane) in LPG around 35:65 is used in India, Spain & Hungary [15]

Table 3 Composition of LPG of analyzed samples.

Sl. No.	Test Parameter	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6	Sample 7	Sample 8	Sample 9
1	C_2, Ethane (mol%)	0.152	0.141	0.139	0.2	0.2	0.28	0.37	0.37	0.29
2	C_3, Propane (mol%)	40.511	40.33	32.771	34.68	34.67	34.59	29.64	29.63	34.69
3	C_4, Butane (mol%)	59.147	59.328	66.967	64.83	64.84	64.86	69.66	69.67	64.89
4	Pentane (mol%)	0.128	0.156	0.103	0.29	0.29	0.27	0.33	0.33	0.13
5	Propane : Butane	40.511 : 59.147	40.33 : 59.328	32.771 : 66.967	34.68 : 64.83	34.67 : 64.84	34.59 : 64.86	29.64 : 69.66	29.63 : 69.67	34.69 : 64.89
6	Total Sulfur (ppm)	14	15	13	15	15	5	8	8	16.97
7	Butadiene (mol%)	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
8	Residue on Evap. @ 38 Deg C (ml/100ml)	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05

Table 4 Physio Chemical Properties of Analyzed Samples.

Sl. No.	Test Parameter	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6	Sample 7	Sample 8	Sample 9
1	Vapor Pressure @37.8 Deg C (KPA)	579.16	599.84	544.69	620.02	620.02	620.85	583.49	583.49	667.8
2	Relative Density @37.8 Deg C (kg/L)	0.55	0.549	0.551	0.5524	0.5526	0.5506	0.5552	0.5535	0.5507
3	Free water	Nill	Nill	Nill	Nill	Nill	Nill	Nill	Nill	Nill
4	Cu Corrosion, 1H@37.8	1A	1A	1A	1A	1A	1A	1A	1A	1A

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	Deg C									
5	Odour	Detectable	Detectable	Detectable	Detectable	Detectable	Detectable	Detectable	Detectable	Detectable
6	Calorific value MJ/kg	47.61709	47.62252	47.88849	47.7195	47.7198	47.6928	47.7678	47.7681	47.7535

In comparison with butane (-0.4 ºC), propane's boiling point (-42ºC) is lower. This property makes propane a suitable choice for colder climate. Only propane as LPG is not suitable for Bangladesh considering heating value, price and safety issue and climatic condition. Being a tropical country, LPG mixture containing more butane is preferred in Bangladesh. A simple comparison among propane and butane ratio of analyzed samples and commonly used ratio in India, Spain and Hungary is depicted in figure 1. Figure 1 reflects that propane and butane ratio of the samples named S4, S5, S6 and S9 are very close to the commonly used ratio in India, Spain and Hungary. The samples S1 and S2 contain more propane but samples S3, S7 and S8 contain more butane.

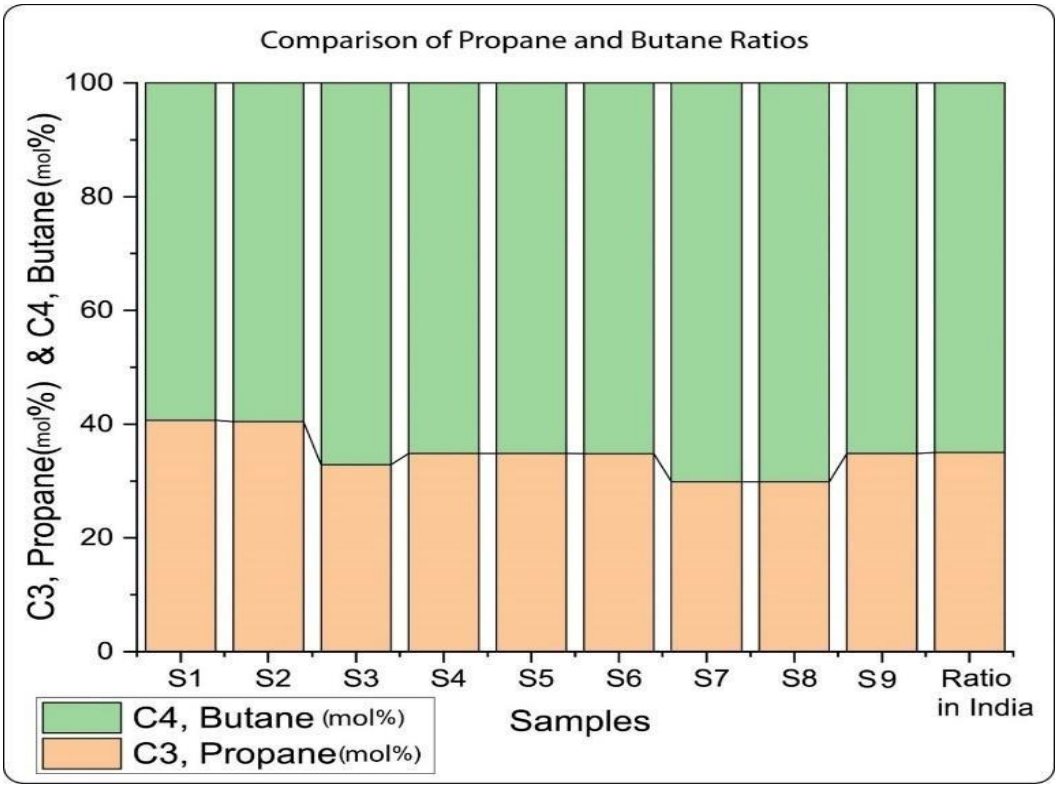


Fig. 1 A simple comparison of propane and butane ratios among the analyzed samples and referenced ratio used in India.

Sulfur can be present in liquid gas in the form of hydrogen sulfide, carbonyl sulfide, carbon disulfide and mercaptan. All forms can be present in the same liquid. Sulfur contamination not only leads to odour problems, but can also form unwanted oxides during combustion and pollute the environment [1], [18]. As per the ASTM standards the permissible sulfur content in LPG mixture is 140 ppm [19] and The Indian Standard limits the sulfur content up to 150 ppm [20]. But Automotive LPG fuel standards in Korea permit maximum 40 ppm which was 100 ppm before 2009 [21]. Table-3 shows that sample 9 contains maximum level of sulfur as 16.97 ppm. The sulfur contents of LPG mixtures available in Bangladesh compared to ASTM, Korea and Indian standards are shown in figure-2. Figure-2 shows there are very low sulfur content in the LPG mixtures available in Bangladesh.

Pentane and heavier hydrocarbons (C_5 + , condensate) are liquids at ambient temperature and pressure [22]. According to ASTM standards the maximum permissible limit of pentane and heavier hydrocarbons in LPG mixture is 2.0% [23]. But in Indian standards it is about 2.5% [20]. Pentane and heavier hydrocarbons content ranges from 0.103 to 0.33 percent (figure-3) which is far less than the ASTM, Korean and Indian standards.

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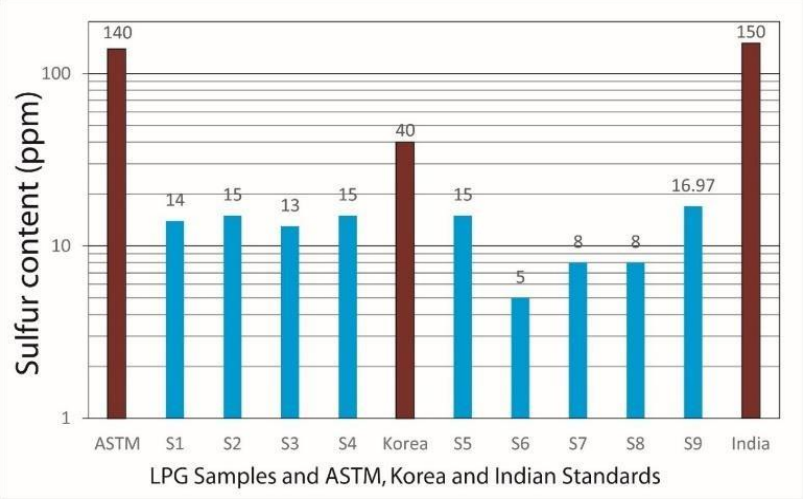


Fig. 2 The sulfur contents of LPG mixtures available in Bangladesh compared to ASTM, Korea and Indian standards

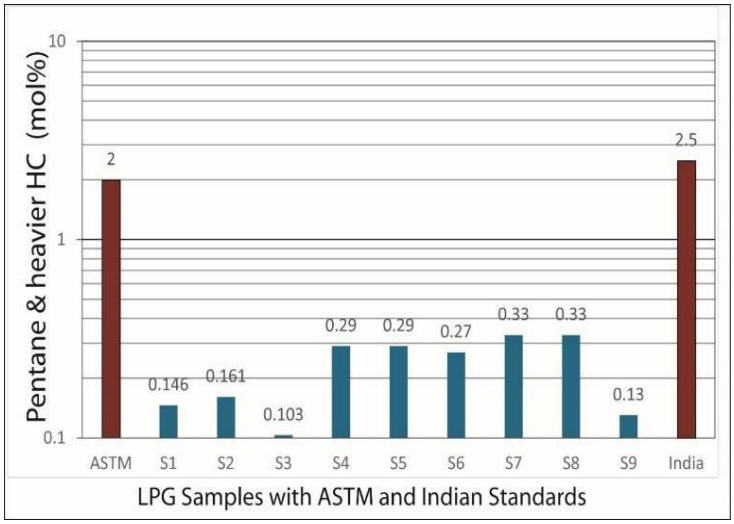


Fig. 3 The pentane contents of LPG mixtures available in Bangladesh compared to ASTM and Indian standards.

The butadiene is a minor component of the LPG product. Butadiene is a dangerous chemical that can cause cancer and genetic defects when inhaled [24] - [26]. The LPG plant operator are facing problem that imports LPG compositions from external sources containing quantities of butadiene that exceed the legal limit of 0.5%. LPG compositions containing a butadiene content of more than 0.5% are considered out of specification, while butadiene contents of less than 0.5% are considered to be on specification [27]. Another reason for limiting the butadiene content in LPG is that butadiene has a higher explosion limit than propane and butane. The higher the butadiene concentration in the liquid gas, the higher the explosion limit [28]. Table-3 shows that the concentrations of butadiene in all samples are less than 0.01% (mole) which are within the safe limit.

According to ASTM and Indian Standards, the LPG mixtures should have no free water content. The examined samples have no free water content as shown in table-3.

The composition of the LPG residue varies with the variation of the LPG composition, especially its impurities and their content[29]. According to ASTM standards, the maximum limit the residue on evaporation of 100 mL LPG at 38 degree Celsius is 0.05 mL. All the samples show the residue on mentioned condition remains below 0.05 mL (table-3).

LPG has a distinct smell that warns of leaks. Generally, LPG is odourized by adding mercaptans. Its odour is detectable in the air at concentrations up to one fifth of the lower flammability limit. In other words, it can be smelled enough before it becomes dangerous enough to catch fire [30]**.** All the samples had the detectable odour.

The copper corrosion limits are designed to ensure that there are no problems with deterioration of copper and copper alloy fittings and connections commonly used in many types of utility, storage and transportation equipment. The copper corrosion test detects the presence of hydrogen sulfide, which is highly corrosive. However, the result of copper corrosion has no connection with the total sulfur content. The copper corrosion limits also provide assurance that the LPG does not contain H_2S in quantities such that it poses a health and safety risk when the product is known not to contain corrosion inhibitors or other chemicals that reduce the reaction with the copper srtip [23]. According to ASTM and Indian standards, the Copper strip corrosion at 38°C for 1

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hour should not worse than rating No. 1. All the samples show rating No. 1A for the copper corrosion test at 38°C for 1 hour. The samples don't have H_2S in quantities such that it poses a health and safety risk.

Although there are no specific requirements for density or relative density, it may be needed for other purposes and should be reported. In addition, the specific gravity of the propane and butane mixture is needed to determine the allowable maximum vapor pressure [23]. The relative density @37.8 Deg C (kg/L) have been reported for all the samples with a range from 0.549 to 0.555 kg/L (table 4).

Vapor pressure is an indirect measure of the most extreme low temperature conditions under which initial vaporization is expected. It can be viewed as a semi-quantitative measure of the amount of the most volatile material present in the product. It can also be used as a means of predicting the maximum pressures that may occur at LPG cylinder temperatures. The maximum standards of vapor pressure at 37.8 °C (100 °F) in LPG mixture is about 1435 kPa [23]. The analyzed samples had the vapor pressure at 37.8 °C (100 °F) in the range between 544.69 and 667.8 kPa. A diagram of vapor pressure vs relative density of the analyzed samples is illustrated in figure 4 which shows very low linear relationship between them.

The calorific value (sometimes called as heating value) is the quantity of heat produced by its combustion at constant pressure and under normal temperature and pressure. Figure 5 shows that the calorific values against the butane content of the examined samples have the Pearson's ratio 0.78 which indicates there is high positive linear correlation among them. While the propane content has high negative linear relationship with calorific values (figure 6). The calorific values against the relative density of the tested samples show moderate positive linear relationship (figure 7).

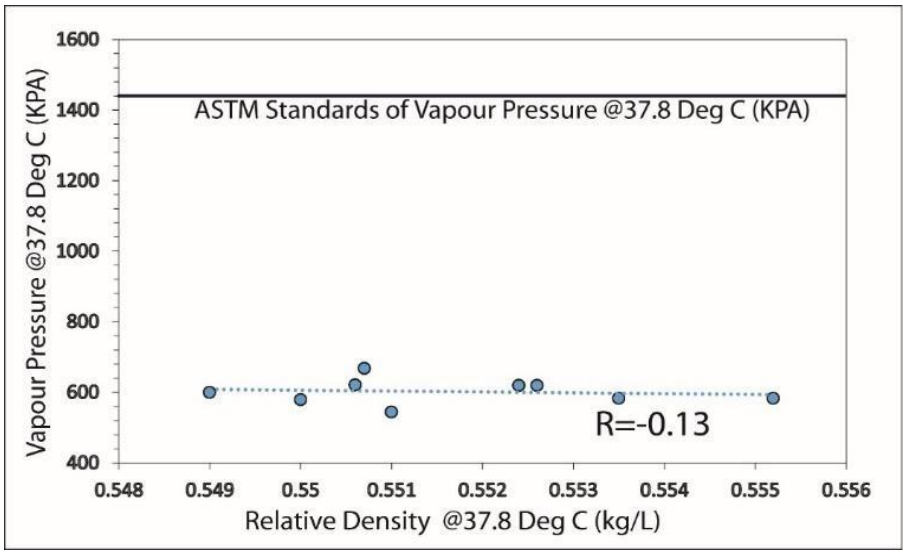


Fig. 4 The vapor pressure against relative density of the examined LPG mixture.

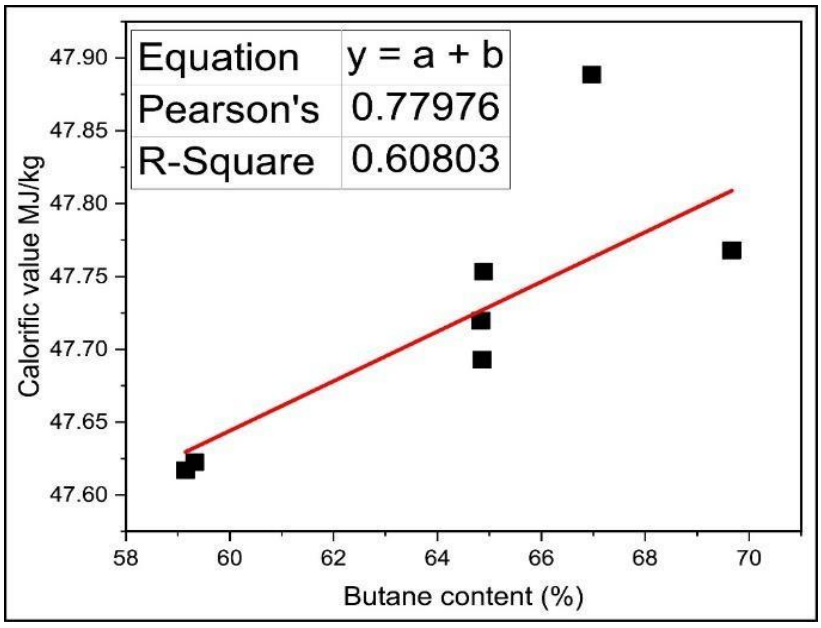


Fig. 5 The calorific value of the examined LPG against butane content.

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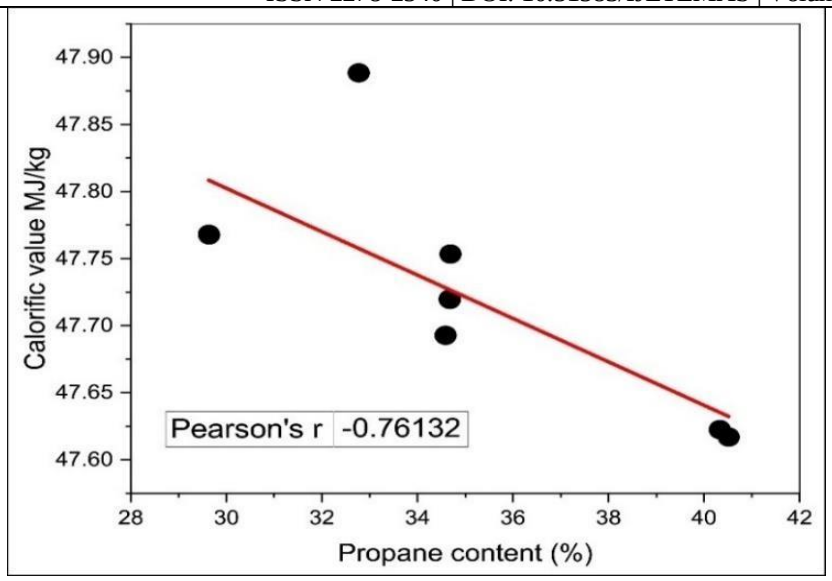


Fig. 6 The calorific value of the examined LPG against propane content.

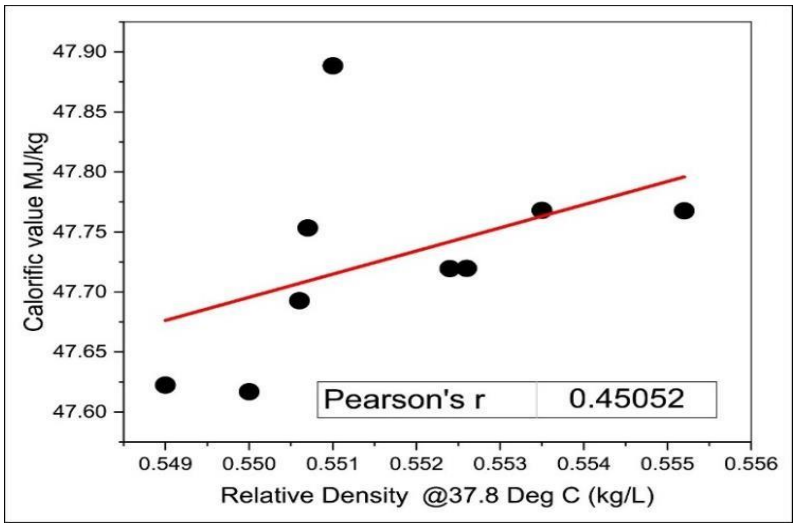


Fig. 7 The calorific value of the examined LPG against relative density.

V. Conclusion

Considering the weather of Bangladesh, the propane-butane ratio of LPG is kept within the range of 40:60-30:70 which is nearly similar to Indian tradition of using propane-butane ratio. There is no free water content. There is very low sulfur content in the LPG mixtures in comparison to the ASTM, Korea and Indian standards. The pentane and heavier hydrocarbon content are much lower than the Indian and ASTM standards. The butadiene has been detected very less than its tolerable limit. The distinct smell of the LPG is easily detectable. The residue on evaporation is within the permissible limit. The LPG does not contain highly corrosive H_2S in quantities such that it poses a health and safety risk. The vapor pressure exerts within the permissible limit and has very low linear relationship with the relative density. But the relative density has moderate positive linear relationship with the calorific value of the tested LPG mixture. The calorific value is also highly correlated positively with butane content and negatively with propane content. Comparison of standard value and existing value of composition and other physicochemical properties of LPG available in cylinders in Bangladesh shows that the LPG mixtures are within safe limit.

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