

Original Research Article

<https://doi.org/10.20546/ijcmas.2019.805.080>**Cryogenic Biogas Enrichment Method for Use as a Vehicle Fuel**

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Energy content of biogas is directly proportional to the methane concentration thus, removing impurities increases the energy content of the gas so that it can be used as vehicle fuel or pooled into gas grid. Removing impurities is regarded as biogas upgrading, in doing so, cryogenic method is among different enrichment methods. Cryogenic method involves the subsequent compression and expansion of biogas until the suitable pressure and temperature is attained (in other words until the required purity is attained). With this method the suitable temperature and pressure to remove CO₂ and H₂S are calculated to be 215K and 1MPa respectively. At this point while CO₂ and H₂S are in their liquid state, CH₄ exists in its gaseous state. Under these conditions, CO₂ and H₂S are removed from the system under the action of gravity. The minimum work done to compress the gas is 0.5MJ/kg.

Introduction

The process of gas production from anaerobic degradation of organic substrates, namely manure, sewage sludge, organic household leftovers and industrial wastes is regarded as biogas production (Deublein and Steinhauser, 2008). The production of biogas and its efficient utilization would meet the fuel and energy demand (Kruczynski, *et al.*, 2012). Biogas released from reactors consists of varieties of impurities that reduce the efficiency of the gas and also causes adverse effects on the network during its use, ranging from the reactor to the point of its use. Biogas has wider industrial applications, for this reason upgrading is necessary. Biogas enrichment has been timely and suitable

option due to rapid growth in the price of fossil fuels (Kadam, and Panwar, 2017) and (Ogur, and Irungu, 2013). On this basis, impurities must be removed or reduced to a minimum level based on the purpose of use of biogas.

Apart from avoiding the adverse effects of impurities from the biogas, upgrading of biogas increases the concentration of methane, in other words enhances the calorific value or energy level of the biogas (Papacz, 2011). This is because; energy content of biogas is directly proportional to the methane concentration. Hence, removing carbon dioxide increases the energy content of the gas (Pettersson and Wellinger, 2009).

Materials and Methods

Biogas comprises of a number of gaseous impurities. Of all the impurities, method of removing of carbon dioxide (being the largest in proportion by volume) and hydrogen sulfide (being corrosive to metallic components), is aimed in this article.

Pooling biogas to a gas grid or using it as vehicle fuel demands the enrichment of methane to 95%, requiring the removal of CO₂ (Papacz, 2011). In other words, the volume of biogas is reduced by 40%. Various methods for removing CO₂ and H₂S from the mixture can be made based on different requirements. However a focus is only put on cryogenic biogas cleaning.

Cryogenic biogas cleaning

The science of low temperatures is one of the processes used to separate the gaseous components of biogas from each other. It uses the temperature difference properties of the type of gases. This process of biogas enrichment is used to create a gas or liquid containing mainly methane and light hydrocarbons. A simple (single stage) schematic diagram of upgrading process is shown below in figure 1.

The process begins with the compression of biogas up to 10Mpa. Several heat exchange steps are used progressively figure 2 to cool the biogas to a lower temperature, allowing CO₂ and H₂S to be liquefied and separated.

Cryogenic upgrading allows the use of various boiling points or sublimations of various gases, especially for the separation of carbon dioxide and methane. Raw biogas is cooled down to temperatures where carbon dioxide in the gas condenses or sublimates and gets separated as a liquid or solid while the methane is accumulated in the gas section.

Water and siloxanes are removed when the gas is cooled. Further availability of water is checked in the gas driers figure 1. The sublimation point of pure carbon dioxide is 194.65 K (Petersson and Wellinger, 2009). However, the methane content in biogas influences the characteristics of the gas, i.e. higher pressures and/or lower temperatures are necessary to condense or sublimate carbon dioxide when it is mixed with methane. Cooling usually takes place in several steps in order to extract the various gases in the biogas individually and optimize energy recovery. At the beginning of the process biogas is compressed, to a targeted pressure P₂ as a function of which the temperature T₂ is calculated by the expression below (Kirillin *et al.*, 1983).

$$T_2 = T_1 * \left(\frac{P_2}{P_1} \right)^{\frac{n-1}{n}} \quad (1)$$

The minimum work done to compress the gas to the required pressure is determined by equation (2) (Kirillin *et al.*, 1983).

$$L = - \frac{n}{n-1} * R * T_1 * \left[\left(\frac{P_2}{P_1} \right)^{\frac{n}{n-1}} - 1 \right] \quad (2)$$

$$\mu R = 8.314 MJ / (mol.K) \quad (3)$$

where R is the universal gas constant, and μ is the molecular weight of a mixture of CH₄ and CO₂

Results and Discussion

From the reactor, biogas is compressed and gets expanded through a nozzle into a separator figure 3. As a result, its temperature decreases from a temperature range of (300 - 450K) to (215 -270K), and pressure decreases from 10 to 1 MPa (Fig. 5). Under these conditions, CO₂ and H₂S are condensed and

liquefied and removed from the system under the action of gravity (Xu *et al.*, 2014). The minimum work done to compress the gas is: 0.5-0.6MJ/kg.

Enriched biogas under pressure of 1MPa and a temperature of 215 -270 K is compressed again to a pressure of 20 - 25 MPa and filled into balloons or fed into gas grid.

The figure 4 shows biogas compression and expansion processes at different polytropic indices. The solid lines describe compression, while the broken lines show the results of subsequent gas expansion. This graph is then superimposed on the CO₂ and H₂S phase diagrams (Lange *et al.*, 2016) and (Goos *et*

al., 2011) (Fig. 5), to find the best point that fits well in the shaded area.

CO₂ and H₂S are gases under normal conditions. The triple points where CO₂ and H₂S exist in all three states (solid, liquid and gas) at equilibrium are 216.4K and pressure of 0.52MPa and 187K and 0.02 MPa respectively. Figure 4 shows the overlap of the thermodynamic properties (phase diagram) of both CO₂ and H₂S and compression-expansion process of biogas. As it can be noted, the liquid state region of CO₂ lies well within that of H₂S, indicating common liquid state range of pressure and temperature.

Fig.1 Schematic diagram of cryogenic enrichment

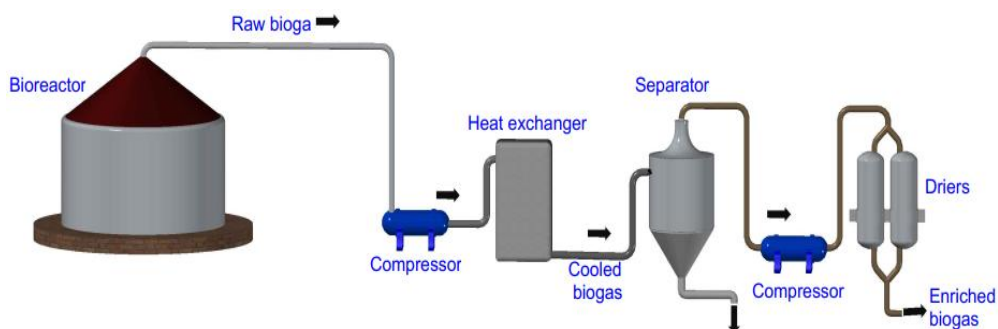


Fig.2 Sectional view of heat exchanger

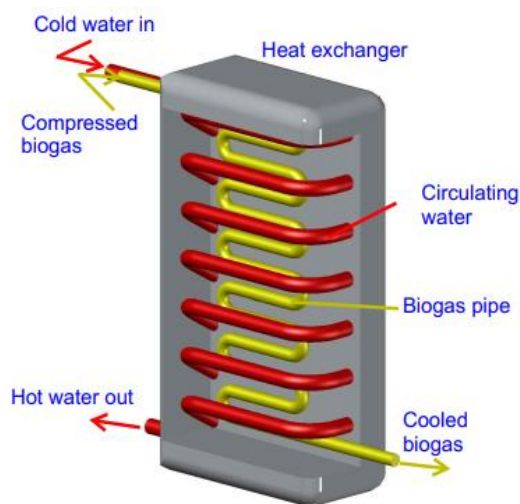


Fig.3 Sectional view of a separator

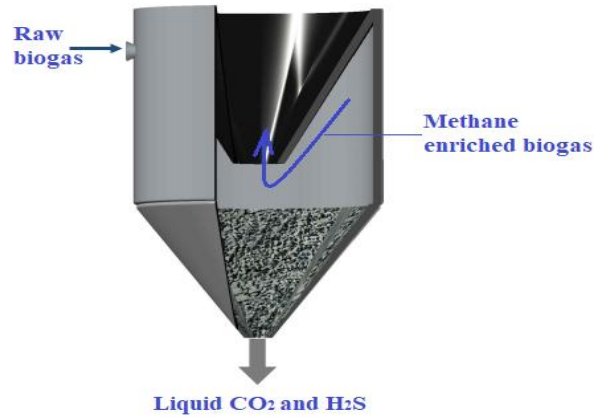


Fig.4 Compression and expansion of biogas

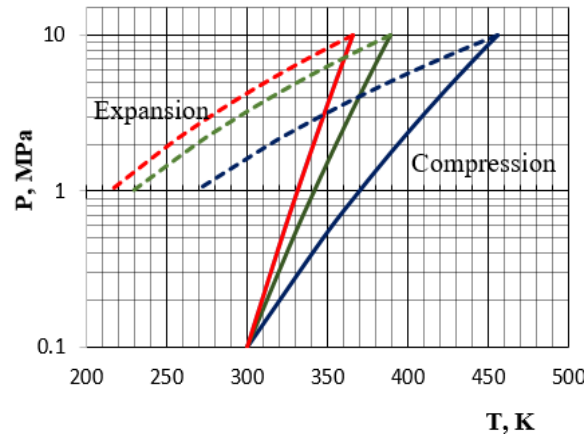
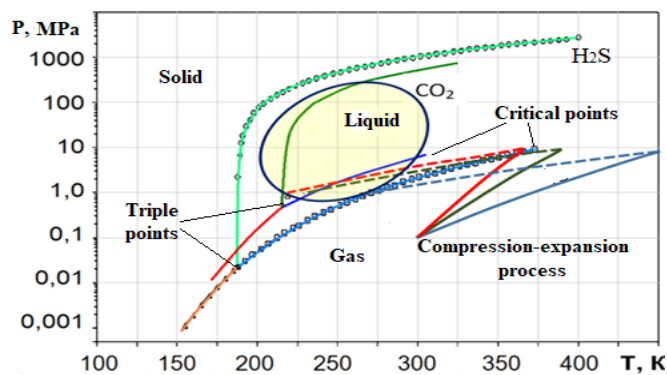


Fig.5 Phase diagrams of CO₂ and H₂S, and compression-expansion of biogas overlaps



The aim of this article is to address the separation of CO₂ and H₂S from the biogas by cryogenic method. Since this method requires compression of gas mixture, it demands energy

source to compress the gas and bring the targeted impurities to their liquid phase so that they can easily flow and get separated from the system. Taking these two key ideas into

account the optimal point where CO₂ and H₂S are liquefied with a minimum energy of 0.5MJ/kg is at a pressure and temperature of 1MPa and 215 K respectively (Fig. 5).

After removing CO₂ and H₂S, methane is re-compressed to fill it into cylinders. While compressing a biogas, naturally, a considerable amount of heat, which can be used to supplement heat to the bioreactor (when required) or other purposes, is generated.

In conclusion, depending on the temperature of the process, various degrees of purity can be achieved. A lower temperature results in higher carbon removal efficiency.

The advantage of the cryogenic method is that its operation does not require water or an absorbent, although it requires external cooling equipment, such as a refrigeration cycle or the addition of liquid nitrogen as a coolant.

In this study, however, circulating water is used as a cooling means. Whenever bioreactors are in cold climatic regions, hot water circulation around the substrates of the bioreactor is used to maintain the temperature of the reactor. Therefore, heat removed from the compressed biogas can be carried from the heat exchanger through the circulating water to the bioreactor as a supplementary heating system.

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