

2016

# Energy And Exergy Analysis For Low Temperature Refrigeration System For Biogas Upgrading

Joseph Bassila

*Cryopur, France / Laboratoire de Chimie Moléculaire, Génie des Procédés Chimiques et Energétique (CNAM), France,*  
joseph.bassila@cryopur.com

Joseph Toubassy

*Cryopur, France,* joseph.toubassy@cryopur.com

Denis Clodic

*Cryopur, France,* denis.clodic@cryopur.com

Amelie Danlos

*Laboratoire de Chimie Moléculaire, Génie des Procédés Chimiques et Energétique (CNAM), France,* amelie.danlos@cnam.fr

Follow this and additional works at: <http://docs.lib.purdue.edu/iracc>

---

Bassila, Joseph; Toubassy, Joseph; Clodic, Denis; and Danlos, Amelie, "Energy And Exergy Analysis For Low Temperature Refrigeration System For Biogas Upgrading" (2016). *International Refrigeration and Air Conditioning Conference*. Paper 1572.  
<http://docs.lib.purdue.edu/iracc/1572>

This document has been made available through Purdue e-Pubs, a service of the Purdue University Libraries. Please contact [epubs@purdue.edu](mailto:epubs@purdue.edu) for additional information.

Complete proceedings may be acquired in print and on CD-ROM directly from the Ray W. Herrick Laboratories at <https://engineering.purdue.edu/Herrick/Events/orderlit.html>

# Energy And Exergy Analysis For Low-Temperature Refrigeration System For Biogas Upgrading

Joseph BASSILA<sup>1\*</sup>, Joseph TOUBASSY<sup>1</sup>, Denis CLODIC<sup>1</sup>, Amélie DANLOS<sup>2</sup>

<sup>1</sup>Cryo Pur,  
3 rue de la Croix Martre 91120, Palaiseau, France  
+33 1 80 38 41 54, joseph.bassila@cryopur.com

<sup>2</sup>Conservatoire National des Arts et Métiers, Laboratoire de Chimie Moléculaire, Génie des Procédés Chimiques et Energétique (CNAM – CMGPCE EA 7341),  
292 rue Saint Martin 75003, Paris, France  
+33 1 40 27 21 77, chaires.turbomachines-moteurs@cnam.fr.

## ABSTRACT

Low-temperature refrigeration system for biogas upgrading has been developed by the Cryo Pur company based on cooling biogas in three steps: removing most of the water content at -40°C, removing siloxanes and SVOCs at -85°C and frosting CO<sub>2</sub> at temperatures varying from -90°C to -120°C. This process transforms biogas containing typically 60% methane, 35% CO<sub>2</sub>, 5% water vapor in methane containing 2.5% of CO<sub>2</sub>. This paper studies how a single low-temperature refrigeration system is able to cool biogas with an indirect system using low-temperature heat-transfer fluids. The exergy study defines the exergy losses and served as guidance for the energy/pinch analysis that is used for the design of the heat-exchanger series and the appropriate heat recovery. An optimal system could save up to 40% of the electric consumption of the refrigeration system.

## Keywords

*Biogas, Upgrading, Exergy Analysis, Pinch Analysis, Refrigeration Cascade*

## 1. INTRODUCTION

Biogas is produced from organic waste by means of bacteria in an anaerobic environment. Bigadan (2001) expedites this process at an operating temperature of 38°C (mesophilic) or 52°C (thermophilic) in the plant's digester. The biogas plant receives all kinds of organic waste - typically livestock manure and organic industrial waste. The manure and waste are mixed in the plant's receiving tank before being heated to 38-52°C and pumped into the digester in which the biogas is produced.

**Table 1:** Composition of biogas, Gerlach et al. (2013)

Component	Formula	Concentration
Methane	CH <sub>4</sub>	50-75%
Carbon dioxide	CO <sub>2</sub>	25-45%
Water vapor	H <sub>2</sub> O	2-7%
Hydrogen Sulphide	H <sub>2</sub> S	0.002-2%
Nitrogen	N <sub>2</sub>	<2%
Ammonia	NH <sub>3</sub>	<1%
Hydrogen	H <sub>2</sub>	<1%
Trace gases		<2%

Ryckebosch et al. (2011) mentioned many techniques that have been developed to remove H<sub>2</sub>S, H<sub>2</sub>O, trace components and CO<sub>2</sub> from biogas. IEA GHG (1993) introduce the cryogenic separation to capture the carbon dioxide from flue-gas stream by condensation.

Cryo Pur (2015) developed a low-temperature refrigeration system for biogas upgrading. This system is based on 3 main steps. The biogas passes through a first subsystem at -40°C to remove mainly water vapor, then a second subsystem at -85°C separates all components in order to obtain at exit 40% CO<sub>2</sub> and 60% CH<sub>4</sub>. In the third subsystem, CO<sub>2</sub> is captured by frosting. After this last step, upgraded methane (98% purity) is liquefied then used as fuel, in the industry and in a natural gas grid injection. In this paper a single low-temperature refrigeration system using low-temperature heat-transfer fluid is studied and compared to four refrigeration sub-systems.

The pinch analysis is used in order to identify possible transfers between heats sources and heat sinks.

## 2. ENERGY STUDY

### 2.1 Separation of H<sub>2</sub>O and CO<sub>2</sub>

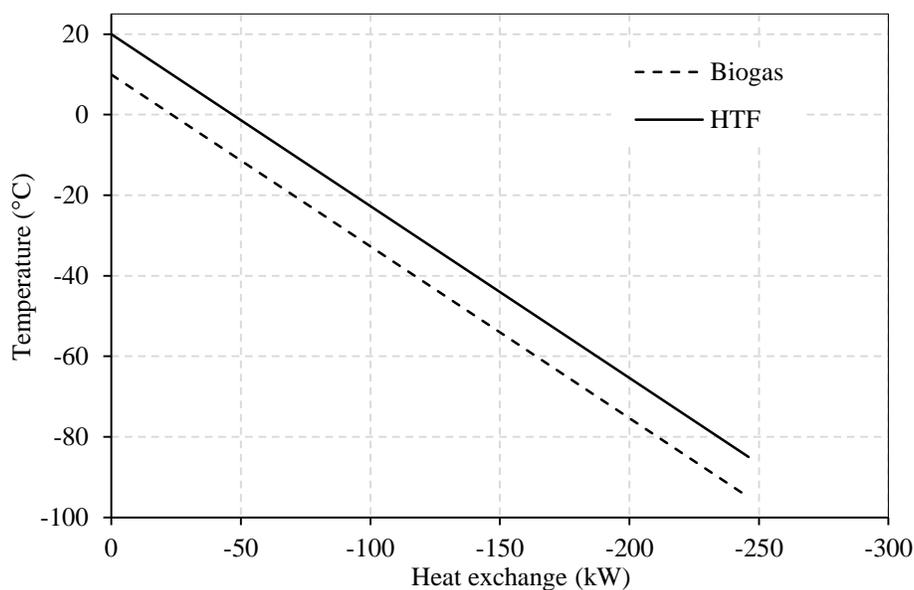
The required cooling capacities to remove water and carbon dioxide from methane are first calculated. In order to be generic, the dry biogas mass flow rate is fixed at 1 kg/s. Water is removed at two temperature levels -40°C and -85°C in the current system, but for the new system, the biogas will be cooled from 20°C to -85°C in one stage as shown in Figure 1. The biogas enters at volumetric composition of 56.8% methane, 37.7% carbon dioxide and 5.5% water, and exits at 60% methane and 40% carbon dioxide. For CO<sub>2</sub> removal, the heat transfer fluid cools the biogas at -116°C in order to reduce the CO<sub>2</sub> content at 2%. As shown in Figure 2, the volumetric composition is 98% methane and 2% carbon dioxide, then methane is liquefied at 1,5 MPa as shown in Figure 3.

The enthalpy difference of each fluid is calculated using Equation (1) while heat exchanged is calculated using Equation (2).

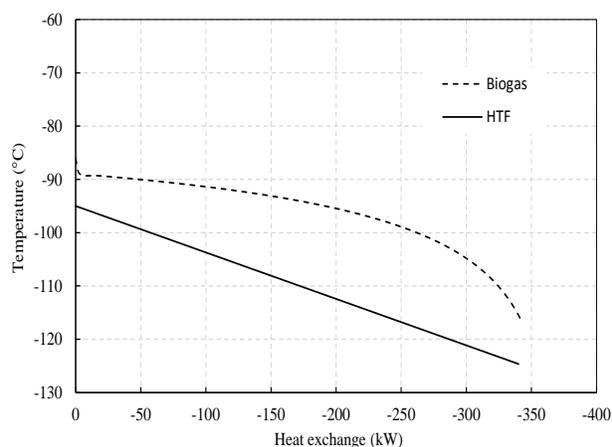
$$Q = \Delta H_{sen} + H_{lat} \quad (1)$$

$$P = \dot{m}_{exit} \cdot \Delta H_{sen} + \dot{m}_{sol} \cdot H_{lat} \quad (2)$$

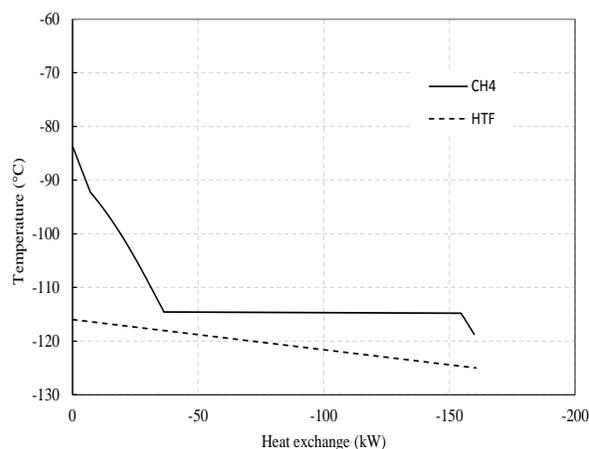
The heat exchanged ( $P$ ) is function of the flow rate at the exit of heat exchanger ( $\dot{m}_{exit}$ ), the sensible enthalpy difference ( $\Delta H_{sen}$ ), the mass of frost captured ( $\dot{m}_{sol}$ ) and the latent heat of sublimation ( $H_{lat}$ ).



**Figure 1:** T-P Diagram for removing water.



**Figure 2:** T-P Diagram for removing Carbon dioxide.



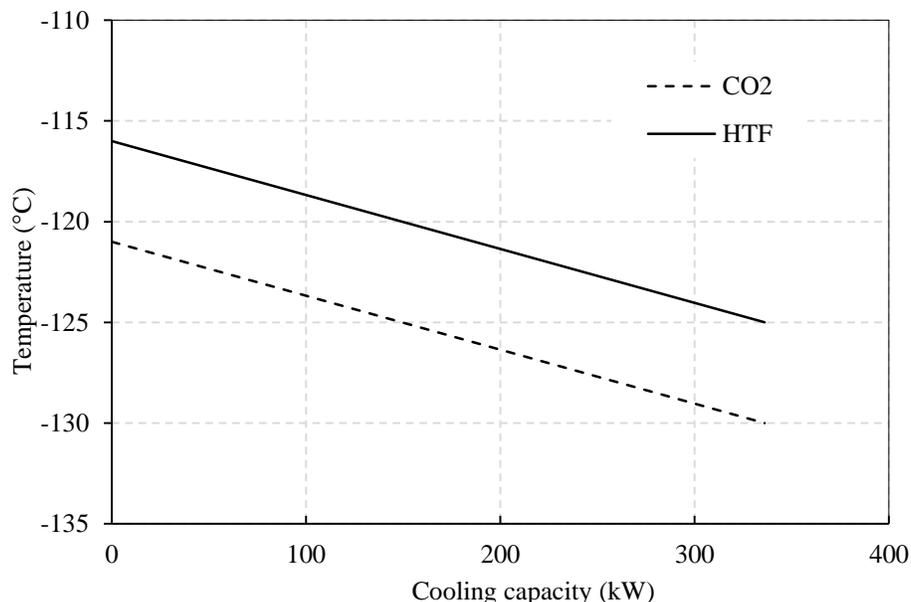
**Figure 3:** T-P Diagram for liquefying biomethane.

## 2.2 The strategy for energy recovery (Theoretical gain)

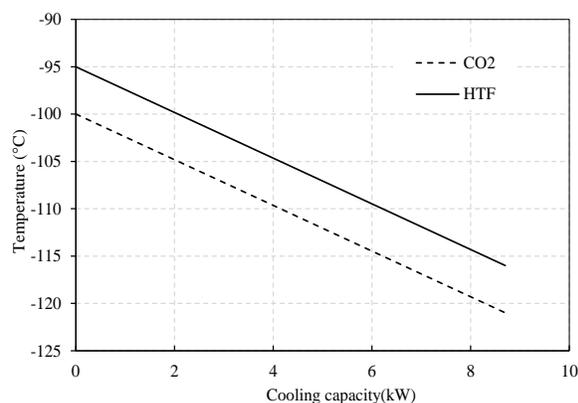
The system works using Frosting/Defrosting heat exchangers for water and CO<sub>2</sub>. The current Cryo Pur system defrosts carbon dioxide above the triple point (-56.6°C, and 0.52 MPa). The new strategy recovers the energy by sublimation of carbon dioxide in order to cool the heat-transfer fluid and by so reducing the cooling capacity of the refrigeration system.

The available energy recovery by sublimation of carbon dioxide is shown in Figure 4. CO<sub>2</sub> in gaseous state passes through two heat exchangers to cool the heat-transfer fluid as shown in Figure 5 and Figure 6.

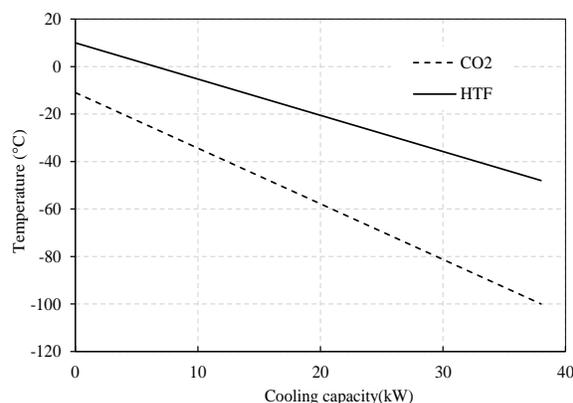
So the theoretical gain is about 336 kW for 1 kg/s of dry biogas.



**Figure 4:** T-P Diagram for recovering power by sublimation of carbon dioxide.



**Figure 5:** T-P Diagram for heat recovery.



**Figure 6:** T-P Diagram for heat recovery.

### 3. EXERGY LOSSES WITH LOW TEMPERATURE HEAT TRANSFER FLUID

The exergy Equation 3 is the maximum theoretical work than can be obtained from an amount of energy.

$$Ex = m(\Delta h - T_a \cdot \Delta s) + \sum_i W_i + \sum_i Q_i \cdot \left(1 - \frac{T_a}{T_i}\right) \quad (3)$$

It is calculated as a function of the mass flow rate ( $m$ ), the enthalpy difference ( $\Delta h$ ), the entropy difference ( $\Delta s$ ), the mechanical work ( $W_i$ ), the heat transferred ( $Q_i$ ), the ambient temperature ( $T_a$ ) and the fluid temperature ( $T_i$ ).

To improve the heat exchange in the system, exergy losses should be minimized.

Comparing many low-temperature heat-transfer fluids (HTF), they have the same exergy losses. We conclude that the only difference between them is their flowrates in each heat exchanger. So they are compared by their heat capacity; once the heat capacity is higher, the flow rate decreases, the power and the consumption of the cryogenic pump decrease.

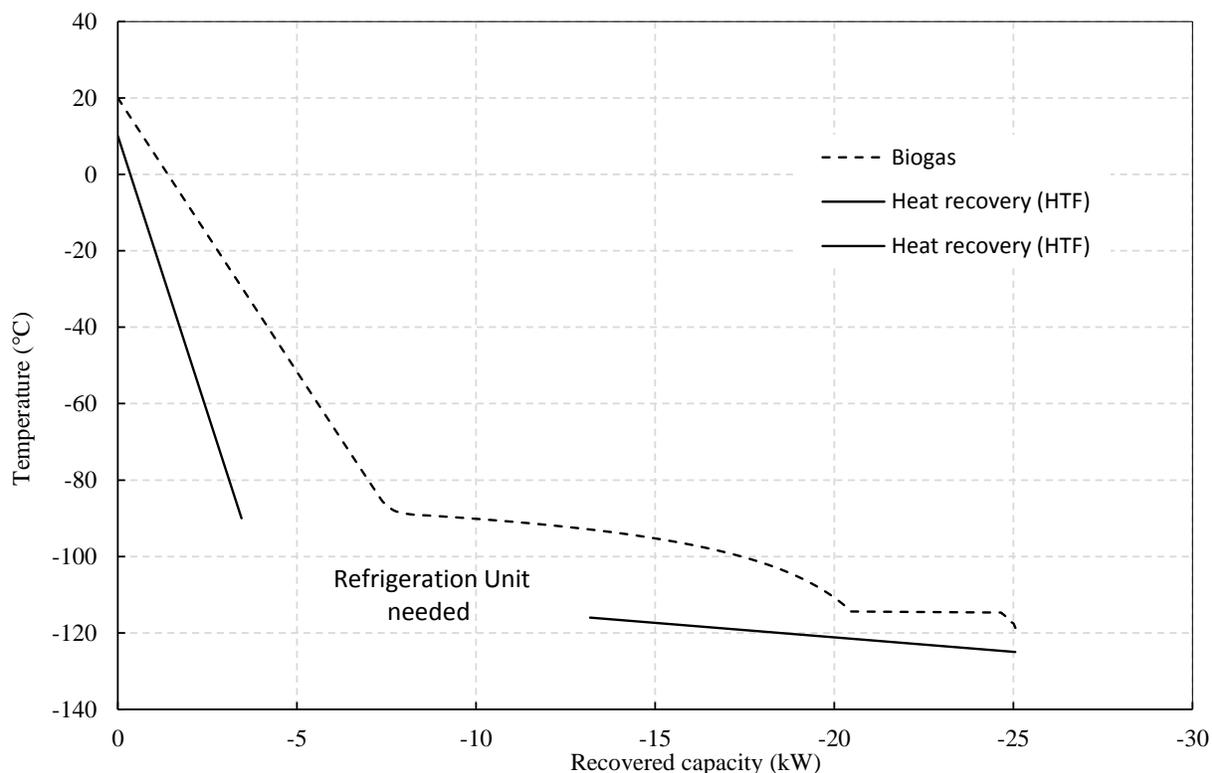
#### 4. ENERGY ANALYSIS

In order to obtain the minimum power consumption of the system, the required energy to upgrade the biogas and liquefy the methane is calculated, then the recovered energy from sublimation of carbon dioxide is calculated from -130°C to -11°C.

The HTF flows returning from the different subsystems at different levels of temperature are mixed. Using Equation (4), the total enthalpy ( $h$ ) is calculated as function of the mass flow rates ( $m_i$ ) and the enthalpy at each level temperature ( $h_i$ ), so the average temperature of the HTF flows is determined and then the required cooling capacity of the refrigeration system is calculated.

$$h = \frac{\sum_i (m_i \cdot h_i)}{\sum_i m_i} \quad (4)$$

Figure 7 represents the (T-P) diagram, the possible heat recovery and the necessary cooling capacity to remove water and carbon dioxide, to liquefy methane, and the theoretical available energy to be recovered by CO<sub>2</sub> sublimation from -130°C to -11°C. Referring to Figure 7, for the reference flow of 1 kg/s of biogas, the refrigeration system should generate 276 kW for biogas upgrading and methane liquefaction.



**Figure 7:** Energy recovery in the T-P diagram.

In order to optimize the heat-exchanger network for heat recovery, the Pinch analysis is carried out. The Pinch analysis aims at identifying the heat recovery opportunities by heat exchange in complex thermal processes. Linhoff (1970) developed a graphical method to calculate the minimum energy requirement of a process and design the heat

recovery exchanger network. The possible heat exchange is limited by the approach temperature between the hot and the cold stream in the heat exchanger. Marechal (2007) shows that when the approach temperature is small, the energy savings are high but the investment required is also high; when the approach temperature is higher, the investment decreases while the operating costs increase. The minimum temperature approach is the smallest temperature difference between the hot and the cold streams in the heat exchanger. The minimum temperature difference can be used as a parameter to determine the optimal size of the heat exchanger.

## 5. RESULTS AND CONCLUSION

In the current Cryo Pur system, 0.63 kW/Nm<sup>3</sup> of biogas at 55% CH<sub>4</sub> and 45% CO<sub>2</sub> is needed to obtain methane with 98% purity, with the new recovery option, 0.36 kW/Nm<sup>3</sup> could be reached. So the new strategy for Cryo Pur could save up to 40% of the electrical consumption.

This new architecture requires many developments in order to limit pressure losses and to define the optimal sublimation pressures.

## NOMENCLATURE

SVOC	Sulfuric Volatile Organic Compound
HTF	Heat Transfer Fluid
P	Power (kW)
$\dot{m}$	Flowrate (kg/s)
T	Temperature (K)
s	Entropy (kJ/kg. K)
Ex	Exergy (kJ)
w	Work (kJ)
H	Enthalpy (kJ/kg)
Q	Heat (kJ/kg)
$\Delta$	Difference
out	Outlet
in	Inlet
sol	solid
lat	latent
sen	sensible
a	Ambient
i	Index

## REFERENCES

- Ryckebosch et al. (2011). Techniques for transformation of biogas to biomethane. *Biomass and Bioenergy*, 35(5), 1633-1645
- IEA GHG (1993). The capture of carbon dioxide from fossil Fuel Fired Power Stations, Cheltenham, UK
- Bigadan (2001). Biogas technology. From <http://bigadan.com/p/biogas-technology/how-to-make-biogas>.
- Cryo Pur, (2015). Valorize biogas with bio-LNG and bioCO<sub>2</sub>. From <http://www.cryopur.com/>.
- Gerlach et al. (2013). *Sustainable Biogas Production*, Frankfurt, Germany: FiBL Projekte GmbH Publisher
- Linhoff (1970). Introduction to Pinch Analysis. Robert F. Boshm (Eds), *Developments in Design of Thermal System* (122-138). Cambridge, UK: Cambridge University Press
- Marechal (2007). *Pinch Analysis*. Ecole Polytechnique Fédérale de Lausanne, Switzerland

## ACKNOWLEDGEMENT

This work was supported by Cryo Pur and Cnam (Conservatoire National des Arts et Métiers). We thank Georges DESCOMBES for his comments that greatly improved the manuscript.

