Adaptation and Evaluation of two Petrochemical Residual Streams to be Useful as Fuels in a Gas Boiler

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ABSTRACT

A practical study was performed to evaluate potential use as fuels and disposal alternative of Pyrolysis Liquids and Butane 1,3 Butadiene. Potential evaluation include: Chemical characterization of residual streams, combustion aspects, practical adaptations for these streams and standard burners for liquid fuels into the existing gas boiler, considering flame length, caloric content, combustion chamber design, control during burning operation, analysis of stack gases and economical aspects.

Chromatographic analysis results shown that this alternative was considered for managing these streams.

INTRODUCTION

In order to obtain Ethylene, fresh and recicled Ethane and steam are feed into furnaces heated to 1562 °F., the reaction yields besides Ethylene, Hydrogen, Methane, Propylene, Pyrolisis Liquids.and 1,3 Butadiene [1]. These last two streams are byproducts harmful to health Table 1 [2]. Pyrolisis Liquids contain 41.15 mol % of Benzene, among other hydrocarbons Table 2 and the stream 1,3 Butadiene contains 45.11 mol % of 1,3 butadiene among other components Table 3. Due that the quantities of these byproducts , (700 BBL/day of each one), are not enough to justify a processing facilty they must be stored.

One option for eliminating these byproducts utilizing also its caloric content is burning them suppling these streams to combustion systems equipped with gun burners for Pyrolisis Liquids, gas burners for 1,3 Butadiene or for both streams dual burners oil and gas .

In this study the combustion test for Pyrolisis Liquids and 1,3 Butadiene were carried out in separate boilers each one producing 200 ton/hr of steam at 853 psia., pertaining to a Petrochemical facilities located at the South of Mexico.

Previously to combustion, the Pyrolisis Liquids and 1,3 Butadiene streams were analysed on its composition for knowing the harmful compounds to be eliminated. Its physical and chemical properties were useful for estimating: the fuels managment, caloric content, air combustion requirements, emissions expected as CO_2, H_20, O_2, SO_2 , NO_X , burners operation and flame lenghts [3].

The drawings of combustion chambers for both boilers were reviewed on its dimensions in order to know the longest flame permissible without damaging the front wall pipes [4].

Taking into account all the above, the residual streams were supplied to the available burners to carry on the tests, all combustion parameters were controlled and the stack gas was sampled for analysing CO_2, H_20 and O_2 , until the combustion system reached the desired operation conditions and when they remained constant, the flue gas were sampled according to the method 18 of the Code of Federal Regulation Part 60 [5], the samples were sent to chromatographic analysis and the results were useful to determinate the efficiency of elimination for all compounds present in the streams.

The combustion test results of Pyrolisis Liquids and 1,3 Butadiene shown that these streams can be supplied as fuels to boilers and at the same time being eliminated. Economical comparison based on caloric content was made.

EXPERIMENTAL

The combustion experiments were carried out in two steam generators from Foster Wheeler Limited on duty in a Petrochemical facility located in the South of Mexico. The boilers specifications indicated in Table 4, includes among others the requirements of natural gas to reach the peak capacity and the distance between the burner´s refractory and the front wall of pipes. The boilers are equipped with dual burners oil and gas, from Peabody Engineering Type H-30, with twin fluid atomisers, the cone angle physically set by the method of construction and firing rates from 18 million to 160 million BTU/hour according with specifications given in Table 5.

In order to know the composition and their physical and chemical properties of Pyrolisis Liquids and 1,3 Butadiene, representative samples of both streams were analysed giving the following results: Pyrolisis Liquids Table 6, and 1,3 Butadiene, Table 7.Hereinafter these streams will be refered to as PL and 1,3 B respectively.

The Physical and Chemical results were useful for estimating the fuels management, air combustion requirements, emissions expected as CO_2, H_20, O_2, SO_2, NO_X, burners operation and flame lenghts. The composition was useful for knowing the harmful compounds to be eliminated.

Pyrolisis Liquids combustion

Ethylene plant produces 700 BBL/day of PL and due that the boiler was on duty three lower burners were available to handle the PL volume and the upper three remained operating with natural gas. In order to comply with the environmental standard, the combustion air and stack gas composition were estimated to obtain 4 vol. % of oxygen in stack dry gas Table 8, the practical combustion test was carried under these conditions.

From the above basis, the gun burners fired PL , from 57.06 gal/hr to 220.07 gal/hr (5.2 BBL/hr), in each burner, this last figure gives a firing rate of 25 million BTU/hr for each burner locating them within specification. Flow rate measurement was made using a portable ultrasonic flowmetering system, from Panametrics with accuracy of ± 1 % in velocity (m/sec) and volumetric (lt/hr).

During the combustion test to measure the stack gases: oxygen (O_2), carbon monoxide (CO), and carbon dioxide (CO_2), a portable analyser with standard electrochemical sensors from Enerac 2000, calibrated with gases of known concentration was utilized. The steam output decreased 6 Ton/hr, (from 174 to 168 tons/hr), in spite of this fact the natural gas supply was not increased and when reached desired operation conditions and they remained constant, the flue gas were sampled according to the method 18 “Measurement of gaseous organic compound emissions by gas chromatography” of the Code of Federal Regulation Part 60, utilizing the integrated bag sampling train consisting of Probe (stainless steel), filter (glass wool), teflon sample line, ball check, vaccum line, vaccum pump, charcoal tube, needle valve, tedlar bag (12”x 19”) and flowmeter Figure 1

1,3 Butadiene combustion

Ethylene plant produces 700 BBL/day of 1,3 Butadiene. For 1,3 B same as for PL, the combustion air and stack gas composition were estimated to obtain 4 vol. % of oxygen in stack dry gas, Table 9 , The volume of 1,3 B, was distributed in two gun burners with the same design specifications as those used for burning the PL. and due its low viscosity they do not need atomization fluid. The gun burners fired 1,3 B, at a rate of 116.25 gal./hr, (2.77 BBL/hr) in each one, giving a firing rate of 11.44 million BTU/hr, locating them out of burners specifications. During this test due that natural gas supply was not reduced in a significant quantity the steam output keeped constant .The test procedure for firing 1,3 B and measurement of flue gas were the same as for PL.

RESULTS AND DISCUSSIONS

Initially the chromatographic analysis of PL detected 105 compounds and during combustion the stack gas chromatography detected small quantities of 12 original compounds, from those, 6 were part of original PL stream, none of them were Benzene, the rest pertained to natural gas Table 10. These 12 remaining components constituted 3.63 wt. % of total stack gas where methane from natural gas contributed with 1.40 wt %.

During PL combustion test, the flue gas monitored gave the results showed in Table 11. The high value of CO_2 expected in flue dry gas was 10.6 vol. % Table12, the value obtained from stack gas was 12.3 vol. % , this figure shows that the combustion efficiency was high.

The gun burners fired 1,3 B., at a rate of 116.25 gal./hr, (2.77 BBL/hr) in each one, this figure gives a firing rate of 11.44 million BTU/hr, locating them out of burner´s specifications, however, in spite of this fact the stack gas results shown that the combustion was complete and the main purpose of eliminating this compounds was achieved, Table 13. The test carried with 1,3 B stream show that it can be supplied and burned through gun burners with good results and in basis of these combustion test results the Petrochemical facility will made the necessary arrangements for vaporizing and mixing 1,3 B with air previously to supply it to the gas burners, with this arrangements it is expected to keep the performance of boiler´s gas burners avoiding adjustments or changes in the heating cycle.

Initially the chromatographic analysis of 1,3 B, detected 18 compounds and during combustion the stack gas chromatography detected small quantities of 11 original compounds, from those, 10 were part of the original 1,3 B stream, none of them were 1,3 Butadiene and 1 pertained to natural gas Table 13. These 11 remaining compounds constituted 6.63 wt. % of the total stack gas, where the olefines C_5 and C_6 plus heavier components contributed with 2.58 wt %.

During 1,3 B combustion test, the flue gas sample gave the results showed in Table 14. The value of CO_2 expected in flue dry gas was 10.15 vol.% Table 15 and the value obtained during the test was 13.8 vol. %, showing that the combustion efficiency was high.

In order to set the sales value for PL and 1,3 B, this streams, were compared with Natural Gas price, based on their caloric contents Table 16.

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Table 1. How can 1,3-butadiene affect the human health.

The Environmental Protection Agency (EPA) requires that discharges or spills into the environment of 1 pound or more of 1,3-butadiene be reported. The Occupational Safety and Health Administration (OSHA) has set an occupational exposure limit of 1,000 parts of 1,3-butadiene per million parts of air (1,000 ppm) codified as 29 CFR 1910.1000, Table Z-1 in 1971 under the authority of Section 6(a). The National Institute for Occupational Safety and Health (NIOSH) recommends that 1,3-butadiene be kept to the lowest feasible concentration because of its potential to cause cancer.
Breathing very high levels of 1,3-butadiene for a short time can cause central nervous system damage, blurred vision, nausea, fatigue, headache, decreased blood pressure and pulse rate, and unconsciousness. There are no recorded cases of accidental exposures at high levels that caused death in humans, but this could occur.
Breathing lower levels may cause irritation of the eyes, nose, and throat. Studies on workers who had longer exposures with lower levels have shown an increase in heart and lung damage, but these workers were also exposed to other chemicals. We don't know for sure which chemical (or chemicals) caused the effects. We also do not know what levels in the air will cause these effects in people when breathed over many years.
Animal studies show that breathing 1,3-butadiene during pregnancy can increase the number of birth defects. Other effects seen in animals that breathed low levels of 1,3-butadiene for one year include kidney and liver disease, and damaged lungs. Some of the animals died
There is no information on the effects of eating or drinking 1,3-butadiene. Skin contact with liquid 1,3-butadiene can cause irritation and frostbite.
The Department of Health and Human Services has determined that 1,3-butadiene may reasonably be anticipated to be a carcinogen. This is based on animal studies that found increases in a variety of tumor types from exposure to 1,3-butadiene.
Studies on workers are inconclusive because the workers were exposed to other chemicals in addition to 1,3-butadiene.

Table 2. Composition of Pyrolisis Liquids

Compound	wt. %	vol. %
butene-1 + isobutilene	0.04	0.05
n-butene	0.22	0.30
transbutene-2	1.62	2.10
2,2-dimethylpropane	0.02	0.02
cisbutene-2	1.69	2.13
1,2-butadiene	0.54	0.66
3- methylbutene-1	0.13	0.16
Isopentane	0.08	0.10
pentene-1	2.54	3.14
2- methylbutene-1	0.15	0.19
n-pentane	0.07	0.08
Isoprene	1.04	1.20
Transpentene	1.51	1.83
Cispentene	0.66	0.79
2-methylbutene-2	0.45	0.53
t-1,3-pentadiene	1.57	1.82
c-1,3-pentadiene	5.44	6.18
2,2- dimethylbutane-2	0.98	1.18
Cyclopentene	6.16	6.25
Cyclopentane	0.28	0.29
3-methylpentane	0.01	0.01
olefines c6	2.90	3.36
n-hexane	0.11	0.13
2,2- dimethylpentane	0.10	0.11
Methylcyclopentane	0.05	0.05
2,2,3-trimethylbutane	0.00	0.00
Benzene	46.09	41.15
Cyclohexane	0.01	0.01
2- methylhexane	0.01	0.01
1,1-dimethylcyclopentane	0.01	0.01
3- methylhexane	0.01	0.01
3- ethylpentane	0.01	0.01
2,2,4-trimethylpentane	0.10	0.11
olefines C_7	3.07	3.43
n-heptane	0.02	0.02
Methylcyclohexane	0.05	0.05
Ethylcyclopentane	0.01	0.01
2,5- dimethylhexane	0.01	0.01
2,4- dimethylhexane	0.21	0.23
t-c-1,2,4-trimethylcyclopentane	0.16	0.17
2,3,4-trimethylpentane	0.02	0.02
Toluene	4.92	4.46
c-t-1,2,4-trimethylcyclopentane	0.02	0.02
2-methylheptane	0.02	0.02
4-methylheptane	0.03	0.03
3,4-dimethylhexane	0.05	0.06

Table 2. Composition of Pyrolisis Liquids (Cont.)

3-methylheptane	0.02	0.03
c-1,3-dimethylcyclohexane	0.01	0.01
2,2,4,4-tetramethylpentane	0.03	0.03
c-1-ethyl-3-methylcyclopentane	0.05	0.06
t-1-ethyl-2-methylcyclopentane	0.01	0.01
t-1,2-dimethylcyclohexane	0.01	0.01
olefines C_8	0.12	0.13
n-octane	0.04	0.05
2,4,4-trimethylhexane	0.02	0.02
Isopropylcyclopentane	0.01	0.01
c-1-ethyl-2-methylcyclopentane	0.01	0.01
c-12-dimethylcyclohexane	0.05	0.04
n- propylcyclopentane	0.01	0.01
113-trimethylcyclohexane	0.01	0.01
Ethylbenzene	0.63	0.57
meta-xilene	0.09	0.08
para-xilene	0.07	0.06
2,3-dimethyl-3-ethylpentane	0.01	0.01
4-methyloctane	0.01	0.01
2-methyloctane	0.01	0.01
orto-xilene	0.06	0.05
1-methyl-2-propylcyclopentane	0.01	0.01
c-1-ethyl-3-methylcyclohexane	0.01	0.01
t-1-ethyl-4-methylcyclohexane	0.02	0.02
Naphthene C_9	0.02	0.02
olefines C_9	0.02	0.02
n-nonane	0.02	0.03
olefine C_{10}	0.01	0.01
t-1-ethyl-3-methylcyclohexane	0.01	0.01
isopropylbenzene	0.02	0.01
3,5-dimethyloctane d/l	0.01	0.01
naphthene C_9	0.01	0.01
n-butylcyclopentane	0.01	0.01
n-propylbenzene	0.07	0.06
3,3-dimethyloctane	0.04	0.06
meta-ethyltoluene	0.02	0.01
para-ethyltoluene	0.01	0.01
1,3,5-trimethylbenzene	0.01	0.01
5-methylnonane	0.07	0.07
naphthene C_{10}	0.03	0.03
isobutylbenzene	0.01	0.01
n-decane	0.04	0.05
1,2,3-trimethylbenzene	0.09	0.09
parafine C_{11}	8.06	8.55
secbutylcyclohexane	0.01	0.01
1-methyl-2-isopropylbenzene	0.03	0.03
1,3-diethylbenzene	0.07	0.07

Table 2. Composition of Pyrolisis Liquids (Cont.)

1-methyl-2-propylbenzene	0.19	0.18
n-butylbenzene	0.01	0.01
1,3-dimethyl-5-ethylbenzene	0.02	0.02
1-methyl-2-propylbenzene	0.01	0.01
n-undecane	0.61	0.65
parafine C_{12}	0.00	0.00
aromatic C_{11}	0.04	0.04
aromatic C_{11}	0.01	0.01
parafine C_{12}	0.09	0.09
aromatic C_{11}	0.23	0.21
aromatic C_{11}	0.16	0.14
n-dodecane	0.18	0.19
weight fractions	0.47	0.46
not identified	4.84	5.09
total	100.00	100.00

Table 3. Composition of 1, 3 Butadiene stream

Compounds	mol %
Propane	1.40
iso-butane	0.04
n-butane	12.82
iso-pentane	< 0.01
n-pentane	0.05
cyclo-pentane	< 0.01
Propadiene	< 0.01
Acetylene	0.04
Propylene	2.37
1- Butene	12.54
cis-2-Butene	6.22
trans-2 -Butene	15.62
iso-Butene	0.67
1-2- Butadiene	0.61
1-3-Butadiene	45.11
Ethyl-Acetylene	0.22
Vinyl-Acetylene	1.18
Olefines C_5 and C_6 + Heavier Fractions	1.10
Total	100 .00

Table 4. Main features in the gas boiler used for the combustion test

Feature	Description
Peak Capacity	544,000 lb/hr
Normal Capacity	495,000 lb/hr
Boiler H.P.	3,395 bhp
Operation Pressure	650 psia
Design Pressure	850 psia
Number of burners	6
Distance from burner's tile to frontal wall of pipes	22.96 ft.
Distace between lateral burner and the wall of pipes	6.56 ft.
Volume of Combustion Chamber	26,553.12 ft^3
Volume of Convection area	14,830.2 ft^3
Requeriments of natural gas to reach the peak capacity	732,612 ft^3

Table 5. Burner´s specifications.

Feature	Description
Make	Peabody Engineering
Model	Type H Register Burners
Burner Type	Dual oil and gas
Firing rate	18 million to 200 million BTU/hour
Throat diameter	30 in.
Type of atomizer	Twin fluid atomizer: Air or steam with internal mix
Atomizer nozzle	6 holes ϕ 9 mm. 30 ° angle
Liquid fuel outlets	6 holes ϕ 10 mm
Atomizing steam or air outlets	19 holes ϕ 9 mm
Gun length	6.56 ft.
Fuel liquid Inlet pressure	7.11 to 21.33 psia.

Table 6. Properties of Pyrolisis Liquids

Properties	Value
Viscosity @ 37.8 °C, SSU	27.6
Specific Weight 20/4 °C	0.8245
API Gravity, °API	39.50
Net Heat Capacity, Btu/lb	16726
Raw Heat Capacity, Btu/lb	17745
Carbon, wt.%	90.00
Hydrogen, wt. %	9.3
Nitrogen, wt. %	0.005
Oxygen, wt. %	0.33
Sulfur, wt. %	0.071
Vapor Pressure. Lb/in^2	9.0
Conradson Carbon, wt. %	0.18
Ramsboton Carbon, wt. %	0.22

Table 7. Properties of 1,3 Butadiene

Properties	Value
Specific Weight 20/4 °C	1.916
Carbon, wt.%	85.55
Hydrogen, wt. %	14.40
Nitrogen, ppm wt.	32.4
Sulfur, wt. %	0.002646
Net Heat Capacity, Btu/ft^3	7.36 X 10^5
Vapor Pressure. lb/in^2	55

Table 8. Operation conditions required for Pyrolisis Liquids combustion test

Parameter	Burners 1,2 and 3 (lower location)	Burners 4,5 and 6 (upper location)
Fuel Type	Pyrolisis Liquids	Natural gas
Flow	88.275 ft^3/hr	438,020.5 m^3/hr
Air feed temperature	356 °F	356 °F
Stoichiometric air requirement	836,599.8 ft^3 at 77 °F 1,271,760.27 ft^3 at 356 °F	5,412,458 ft^3 at 77 °F 8,227,653.7 ft^3 at 356 °F
Excess % Air requirement to obtain 4 % O_2 in flue dry gas	21.37 %	21.37 %
Air requirement to obtain 4 % O_2 in flue dry gas	178,774.5 ft^3 at 77 °F 271,781 ft^3 at 356 °F	1,156,614 ft^3 at 77 °F 1,758,226 ft^3 at 356 °F
Relation air/fuel	16.39 lb air/lb fuel	19.28 lb air/lb fuel
Fuel pressure in the head	14.22 psia.	-------
Fuel pressure at the burner inlet	8.53 psia.	7.11 psia.
Total air requirement	1,015,374 ft^3 at 77 °F 1,543,541 ft^3 at 356 °F	6,569,072 ft^3 at 77 °F 9,985,879 ft^3 at 356 °F
Atmospheric pressure	760 mmHg	760 mmHg
Ambient temperature	77 °F	77 °F
% O_2 atmospheric	21 %	21 %

Table 9. Operation conditions required for 1,3 Butadiene combustion test

Parameter	Burners 1 and 3 (lower location)	Burners 4,5 and 6 (upper location)
Fuel Type	Butadiene	Natural gas
Flow	31.07 ft^3/hr	501,649 ft^3/hr
Air feed temperature	356 °F	356 °F
Stoichiometric air requirement	243,003 ft^3 at 77 °F 369,378 ft^3at 356 °F	6,199,094 ft^3 at 77 °F 9,423,427 ft^3 at 356 °F
Excess % Air requirement to obtain 4 % O_2 in flue dry gas	21.09 %	21.09 %
Air requirement to obtain 4 % O_2 in flue dry gas	51,234 ft^3 at 77 °F 77,894 ft^3 at 356 °F	1,307,388 ft^3 at 77 °F 1,987,388 ft^3 at 356 °F
Relation air/fuel	17.88 lb air/lb fuel	19.23 lb air/lb fuel
Fuel pressure at the burner inlet	18.49 Kg/cm^2	7.11 kg/cm^2
Total air requirement	294,238 ft^3 at 77 °F 447,272 ft^3 at 356 °F	7,506,482 ft^3 at 77 °F 11,410,815 ft^3 at 356 °F
Atmospheric pressure	760 mmHg	760 mmHg
Ambient temperature	77 °F	77 °F
% O_2 atmospheric	21 %	21 %

Table 10. Pyrolisis Liquids combustion products

Compound	mol %	wt. %
Metane	2.57	1.40
Etane	0.05	0.05
Propane	0.08	0.13
Iso-butane	0.12	0.24
n-butane	0.36	0.71
Iso-pentane	0.14	0.35
n-pentane	0.04	0.10
etylene	0.08	0.08
1-Butene	0.13	0.25
Cis-2-butene	0.04	0.09
Trans-2-butene	0.06	0.11
Iso-butene	0.06	0.12
Carbon Dioxide	13.90	15.18
Oxygen	4.57	6.86
Nitrogen	77.79	74.34
Total	100.00	100.00

Table 11. Combustion emissions of flue gas, obtained during the combustion test of Pyrolisis Liquids

Compound	Dry gas
CO_2, vol.%	12.3
CO, vol.%	0.00
O_2, vol.%	4.4
Stack temperature, °F	406.4
Combustion efficiency %	92
Flame length ft.	10

Table 12. Theoretical emissions in flue gas. Pyrolisis Liquids combustion test

Compound	Pyrolisis Liquids		Natural gas		Pyrolisis Liquids + Natural gas	
	Wet gas	Dry gas	Wet gas	Dry gas	Wet gas	Dry gas
CO_2, mol %	12.6063	13.7060	8.59	10.09	9.1095	10.5894
SO_2, mol %	0.0037	0.0040	0.00	0.00	0.0005	0.0006
H_2O, mol %	7.9900	0.0000	14.88	0.00	13.9900	0.0000
O_2, mol %	3.5700	3.8800	3.42	4.02	3.4400	4.0000
N_2, mol %	75.8200	82.4100	73.11	85.89	73.4600	85.4100
Total	100.0000	100.0000	100.00	100.00	100.0000	100.0000

Table 13. 1,3 Butadiene combustion products

Compound	mol %	wt. %
Metane	1.11	0.58
Propane	0.04	0.06
Iso-butane	0.12	0.23
n-butane	0.77	1.47
Iso-pentane	0.03	0.08
n-pentane	0.04	0.10
1-Butene	0.33	0.60
Cis-2-butene	0.14	0.27
Trans-2-butene	0.25	0.47
Iso-butene	0.10	0.19
Carbon Dioxide	7.76	11.22
Oxygen	7.41	7.79
Nitrogen	80.84	74.36
Olefines C_5 and C_6 + Heavier Fractions	1.03	2.58
Total	100.00	100.00

Table 14. Combustion emissions of flue gas, obtained during the combustion test of 1,3 Butadiene

Compound	Dry gas
CO_2, vol.%	13.8
CO, vol.%	0.00
O_2, vol.%	2.2
Stack temperature, °F	415.4
Combustion efficiency %	92
Flame length ft.	8

Table 15. Theoretical emissions in flue gas. 1,3 Butadiene combustion test

Compound	Pyrolisis Liquids		Natural gas		Pyrolisis Liquids + Natural gas	
	Wet gas	Dry gas	Wet gas	Dry gas	Wet gas	Dry gas
CO_2, mol %	10.8940	12.2406	8.61	10.11	8.69	10.18
SO_2, mol %	0.0001	0.0001	0.00	0.00	<1 ppm	<1 ppm
NO, mol %	0.0004	0.0004	0.00	0.00	<1 ppm	<1 ppm
H_2O, mol %	11.0019	0.0000	14.91	0.00	14.77	0.00
O_2, mol %	3.4575	3.8850	3.38	3.98	3.39	4.00
N_2, mol %	74.6461	83.8739	73.10	85.91	73.15	85.82
Total	100.0000	100.0000	100.00	100.00	100.00	100.00

Table 16. Theoretical emissions in flue gas. 1,3 Butadiene combustion test

Fuel	Caloric content BTU/ft^3	BTU/USDlls.	Price USDlls./ft^3
Natural Gas	1,000	478,029.04	2.61
Pyrolisis Liquids	849,617.67	478,029.04	2,216.05
1,3 Butadiene	736,335.31	478,029.04	1,920.51

Figura 1. Integrated Bag Sampling Train