Ethanol dehydration via azeotropic distillation with gasoline fraction mixtures as entrainers: A pilot-scale study with industrially produced bioethanol and naphta.

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Abstract

Various hydrocarbons (n-hexane, cyclohexane, toluene, isooctane) and mixtures of them (binary, ternary or quaternary), as well as two different types of industrially produced naphtha (one obtained by direct distillation and the other from a catalytic cracking process), have been tested as candidate entrainers to dehydrate ethanol. The tests were carried out in an azeotropic distillation column on a semi pilot plant. The results show that it is possible to dehydrate bioethanol using naphtha as entrainer, obtaining as a result a fuel blend with negligible water content and ready for immediate use in motor vehicles.

1. Introduction

In order to reduce the greenhouse gas emissions produced by the transportation sector, the use of biomass as a fuel or in a mixture with fossil fuels has been promoted by government legislative and financial initiatives. Among the various biofuel technologies, bioethanol has advanced the most. However, bioethanol obtained from the fermentation of biomass, which can come from different sources, needs to undergo several steps of purification. This bioethanol is produced in an aqueous media and must thus be dehydrated before it can be used as a fuel. European legislation permits a maximum of 0.3% water by weight fraction in the bioethanol to be blended with gasoline [1].

Even though the presence of a binary azeotrope in the water + ethanol mixture makes it impossible to separate ethanol and water in a single distillation step, there exists many other techniques [2] that can be used, alone or in combination, to dehydrate ethanol. Some of them include, but are not limited to: adsorption on molecular sieves [3], azeotropic distillation [4], pressure swing distillation [5], pervaporation [6], extractive distillation with ionic liquids [7], etc.

Concretely, in recent years several papers have treated bioethanol dehydration through only simulation testing different techniques such as pressure swing absorption [8], hybrid processes (distillation/adsorption/vapor permeation [9], based on liquid-liquid extraction [10], heteroazeotropic distillation using a gasoline additive as entrainer [11], extractive batch distillation [12] or a heat-pump-assisted extractive distillation in a single step [13] and comparing them to the conventional process. Though all the techniques presented in these papers seem viable alternatives and arouse improvements in bioethanol dehydration process, the results presented are almost all based on

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simulation and lack in the majority of cases experimental validation. Thus it is necessary experimental data regarding non-conventional processes for ethanol dehydration.

These techniques are more or less energy demanding depending on the technique and the separation requirements. Traditionally, the ethanol is purified completely before being used in a fuel blend. However, in the novel process proposed in a previous paper [14], based on heterogeneous azeotropic distillation, the immediate product already is a fuel blend containing little water, meaning that it can be used directly or added to another fuel on the contrary of the other processes proposed in papers where dehydrated ethanol has to be mixed afterwards and thus intensifying the whole process.

In a previous study, several pure hydrocarbons (hexane, cyclohexane, toluene and isooctane) [14] were tested as entrainers in the ethanol dehydration process. The results showed that it was possible to obtain a hydrocarbon + ethanol mixture with a negligible water content. The continuation of that research is the subject of the present article: a study of the possibility of using hydrocarbon mixtures as entrainers in the industrial bioethanol dehydration process, with a view to assessing the viability of a process where a gasoline and ethanol mixture containing a negligible amount of water can be obtained directly.

The research has been carried out in two stages:
1. Several binary and ternary hydrocarbon mixtures, combinations of those investigated previously, [14] were tested as entrainers to verify their suitability for the ethanol dehydration process.
2. Two different types of naphtha, obtained from a refinery, were tested in combination with industrial bioethanol in order to investigate the possibility of using a more complex mixture as entrainer and so, by the above process, directly manufacture a complex fuel blend.

2. Experimental procedure

2.1. Chemicals

For the first stage of the study, analytical grade hydrocarbons supplied by PANREAC AppliChem (ITW reagents) provider were used. Their purities were as follows: hexane (min 98.5%), cyclohexane (min 99.5%), isooctane (min 99%) and toluene (min 99%) as stated by provider. The analytical grade ethanol and 2-propanol used to prepare the standards were supplied by MERCK provider, (with purities min 99.9 and 99.8%, respectively as stated by provider). The deionized water that was used had a measured conductivity of 3 µS/cm.

For the second stage, bioethanol produced in a single distillation step on an actual fermentation plant, was used directly (Abengoa Plant, Valle de Escombreras-Cartagena (Spain)). Bioethanol water content was checked against the Karl-Fischer (Mettler-Toledo model DL-31) technique and was, on average, 8.3% by weight fraction. The degree of purity of the bioethanol was measured in a gas chromatograph (Agilent 7890B) coupled to a high resolution mass spectrometer (Agilent 7200(Q-TOF) . It was found to contain 1-propanol, isobutanol and ethyl acetate in quantities less than 1% by mass each. Other compounds were also detected during the analysis, but the sum total

99   of their concentrations amounted to less than 1%.

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101 The bioethanol was analyzed in an Ionized Coupled Plasma detector coupled to a mass
102 chromatograph (Perkin Elmer, Model: 7300 DV (with dual vision)) to determine its
103 mineral content in S, P and Cu. Furthermore, ionic chromatography (Metrohm 850
104 ProfIC AnCat- MCS) was used to analyze for SO_4 and Cl and determine their
105 concentrations. The results of the analyses are presented in Table 1.

106

107 As can be seen, the inorganic content of the bioethanol is far below legal limits. The
108 limits quoted in Table 1, as per the European regulation EN 15376:2011 [1], refer to
109 ethanol that is directly mixed with fuel. In the case of the present study, the ethanol is
110 dehydrated during the process itself. As a result, the limit for inorganics should
111 correspond to that of the final fuel. That is, if there can be at most 10% ethanol in the
112 final fuel then the concentration of the inorganics in the bioethanol + fuel mixture
113 should not exceed a limit of 10% in the dehydrated ethanol.

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115 The two types of naphtha that were used were provided by Repsol (Cartagena plant -
116 Spain). Naphtha 1 was obtained by direct distillation of petrol whereas naphtha 2 was
117 obtained by the catalytic cracking of higher boiling point petrol fractions. The
118 characteristics of both are recorded in Table 2 and Table 3. These characteristics are
119 quoted by the provider and were checked in this work by means of a 1:100 split-
120 injection mass chromatography analysis on both naphtha types, during which a
121 temperature ramp of 5ºC/min was applied, following the procedure outlined in
122 regulation ISO 22854:2014 [15]. A high resolution mass spectrometer (Agilent 7200(Q-
123 TOF)) coupled to a gas chromatograph (Agilent 7890B) was employed to this end. As
124 can be seen in Table 3, naphtha 2 has a higher than usual content in olefins and
125 aromatics as would be expected from the cracking process.

126

127 The distillate curves of the two naphtha types, presented in Figure 1a, have been
128 obtained in keeping with regulation ISO 3405:2011 [16]. Figure 1b shows the
129 chromatographs of both naphtha types. Their volatilities are fairly similar even though
130 naphtha 1 has a slightly wider temperature range between the initial and final
131 evaporation points than naphtha 2, as can be appreciated from Figure 1a.

132

133 **2.2. Procedure**

134

135 The first part of the study followed the procedure described in a previous paper ([14]).
136 A 50 mm diameter Armfield UOP3CC column built to the scale of a semi pilot plant,
137 and fitted with eight plates and covered with Armaflex AF (Armacell Advanced
138 Insulation) insulating material, was used to this end. Two preheated feeds were let onto
139 the first plate, while the heating power in the boiling chamber was varied. A decanter
140 with two outlets after the condenser permitted one of the condensed vapor phases to
141 return to the column.

142

143 The flow diagram of the process is presented in Figure 2. The two feeds are pumped
144 inside the column thanks to two peristaltic pumps. The hydrocarbon mixture goes
145 through a heat exchanger (HE-6) where it is preheated with the bottom product of the
146 column. Later on this feed and the ethanol one go through two heat exchangers where
147 they are preheated thanks to an oil heating flux (HE-4 and HE-5). Those two preheated
148 feeds enter the column at the top stage. The vapor exiting the top stage of the column

149 condensates inside the condenser (HE-2) where cold water acts as a refrigerant. The
150 condensate arrives to a decanter where it splits in two phases. The organic phase returns
151 to the column after a heating step (HE-3) in the first stage. The aqueous phase
152 containing the majority of water is discharged. The bottom product is the desired
153 product the ethanol + hydrocarbon mixture with little water content and exits from the
154 boiling chamber (HE-1) at half of the boiling chamber height The TM symbols in the
155 figure correspond to thermopar sensors connected to the main control equipment for
156 monitoring and saving the temperature data all along the process.
157
158 In Table 4 are presented the experimental conditions (flow rates, compositions,
159 temperatures…) for each of the experiments on the different mixtures.
160
161 The feed flow rate was maintained constant between experiments. The organic phase
162 from the decanter was returned to the column while the aqueous phase was treated
163 separately. The flow rates of ethanol and entrainer presented in Table 4 permits the
164 obtaining of a fuel blend with an ethanol content of up to 10% by mass (objective for
165 biofuel in transport fuel in the EU for the year 2020)[17].
166
167 The samples collected from the distillate and bottom product were analyzed by gas
168 chromatography in a Shimadzu GC-14B gas chromatograph with a PORAPACK Q
169 column coupled to a Thermal Conductivity Detector (TCD). The carrier gas was helium
170 at a flow rate of 50 mL/min and an oven temperature of 170 ºC. As the obtained
171 samples were very complex (many different hydrocarbons coming from the naphtha
172 feed), only the ethanol and water content were quantified. The water content was
173 checked against the Karl Fischer technique.
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176 **3. Experimental results**
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178 **3.1. Hydrocarbon mixtures**
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180 Figures 3 through 6 are plots of different variables against the power used in the boiling
181 chamber, and correspond to the experimental data that have been obtained. Figures 3a,
182 4a, 5a and 6a show the flow rates of the distillate and bottom product. Figures 3b, 4b, 5b
183 and 6b show the hydrocarbon and water content in the distillate. Figures 3c, 4c, 5c and
184 6c show the hydrocarbon composition of the bottom product. Finally, figures 3d, 4d, 5d
185 and 6d show the water content in the bottom product.
186
187 As can be deduced from the figures, there exists a heating power for which it is possible
188 to obtain a fuel blend containing less than 100 ppm water, accompanied by a variation
189 in ethanol content of between 5 and 10%.
190
191 When the power is increased, the ethanol content in the bottom product tends to fall as
192 the water content does, except for the experiments with mixtures containing hexane. In
193 these two cases (Figures 5c and 6c) it is observed that the ethanol content remains
194 approximately constant in the power range tested. An optimal power appears to result in
195 a maximum ethanol concentration in the bottom product, corresponding to a water
196 content that is below the legal limit. Obtaining a fuel blend where the water/ethanol
197 mass content ratio is 0.003 would be an optimum situation. To achieve this situation, for
198 instance, in the experiments with the hexane-cyclohexane-isooctane a power of around

199 90 W is required. Based on the flow rates used and experimental conditions (preheating
200 step, heat loses...) 1.7 kJ/g ethanol or 0.47 kWh/kg ethanol dehydrated is required.
201 While this energy cost is low compared to conventional process (e.g. 2.07 kWh/kg [13])
202 it takes only into account the experimental heat duty in the boiling chamber. This step
203 should be integrated in the heat exchanger network of a refinery to analyze its industrial
204 viability.
205
206 The hydrocarbon content in the bottom product increases as the power does (except for
207 experiment 3 and 4 where the hydrocarbons/ethanol proportion remains almost constant
208 as stated before). An increase in the hydrocarbon content of the distillate is also
209 observed in all of the experiments. This indicates somewhat of a loss of volatile
210 compounds in the distillate as the power increases. It also highlights the existence of an
211 optimal power for the process that avoids hydrocarbon losses in the distillate. For
212 instance, for the optimal power shown previously the ethanol recovery rate in the
213 bottom product is 78.4%, while for the hexane, cyclohexane and isooctane is 97.5, 98.4
214 and 99.4% respectively.
215
216 Moreover, while it is not shown in the figures, the existence of a maximum heating
217 power beyond which the desired separation no longer takes place, has been observed. In
218 fact, when the power is too high the vapor obtained in the distillate becomes
219 homogeneous and thus the decanter cannot separate an aqueous rich fraction from the
220 column.
221
222 **3.2. Naphtha feed**
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224 In figures 7 and 8, the same variables as in the experiments on the hydrocarbon
225 mixtures are plotted, again as a function of the power used in the boiling chamber.
226 However, due to the complexity of the mixtures, in this case only ethanol and water
227 content have been plotted.
228
229 As in the earlier case, it can be seen that an increase in the power employed increases
230 the distillate flow rate and reduces that of the bottom product. More power reduces the
231 water content in the bottom product to below the detection limit of 50 ppm.
232
233 On the other hand, the greater the power used the more ethanol and naphtha compounds
234 appear in the distillate. As a result, an optimum power exists for which the water
235 content in the bottom product is below the limit and the flow rates of the ethanol and
236 naphtha compounds in the distillate are minimized, avoiding in this case the loss of
237 valuable compounds. Nevertheless, subjecting the distillate to another separation step
238 might make it possible to recover of some of those compounds.
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241 **5. Conclusions**
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243 In this paper, on the one hand, it has been studied the performance of four hydrocarbons
244 (n-hexane, cyclohexane, toluene, isooctane) and their binary, ternary and quaternary
245 mixtures to dehydrate ethanol obtaining a fuel blend. On the other hand, the
246 performance of two different types of industrially produced naphtha (one obtained by
247 direct distillation and the other from a catalytic cracking process) has been checked.
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Experiments under similar conditions have been performed in an azeotropic distillation column on a semi pilot plant scale to compare the technical viability of the hydrocarbons to dehydrate ethanol. In these experiments, the flow rates and the composition has been analyzed with the power increase. It has been observed that in most of the experiments carried out, there is a maximum heating power beyond which the desired separation no longer takes place.

It has been proved that the use of a heterogeneous azeotropic process to dehydrate bioethanol and directly obtain a fuel blend is possible for many hydrocarbon mixtures, including complex ones such as naphtha.

The next step in the research would be to analyze the costs (material and energy) of the proposed process and then to integrate it into a refinery/biofuel production plant in order to verify to what extent the dehydration of ethanol is improved relative to conventional processes.

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314   20% by 2020 (2008) Retrieved 15 January, 2015, from www.erec.org
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325   Table 1. Inorganic Content of bioethanol
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Compound	Content (ppm)	Legislation limit (ppm)
S	<3	10
SO_4	Not detected	4.0
P	No detected	0.15
Cu	0.023	0.1
Cl	0.25	6.0
NO_3^-	0.28	No limit

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330   Table 2. Composition of the two naphtha types, in mass percent, as a function of their
331   carbon number in the molecules.
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	%w/w	
	naphtha 1	naphtha 2
C5	0	0.068
C6	8.712	3.805
C7	33.502	33.936
C8	37.603	52.415
C9	18.405	9.236
C10	1.653	0.503
C11	0.096	0
C12	0	0.022

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357 Table 3. PIONA (w/w%) analysis of the two naphtha types

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	w/w%	
Type of hydrocarbon	naphtha 1	naphtha 2
p6	3.916	0.670
p7	18.369	4.547
p8	18.385	11.459
p9	10.674	2.981
p10	1.288	0.503
p11	0.096	0.000
N5	0.000	0.000
n6	4.155	0.482
n7	10.745	5.157
n8	11.877	6.007
n9	6.772	1.834
n10	0.226	0.000
n11	0.000	0.000
a6	0.641	0.105
a7	4.388	9.277
a8	7.341	21.562
a9	0.959	2.395
a10	0.139	0.000
a11	0.000	0.000
O6	-	2.654
O7	-	14.955
O8	-	13.386
O9	-	2.025
O10	-	0.000

359

	P%	N%	A%	O%			
naphtha 1	52.728	33.775	13.468	-			
naphtha 2	20.16	13.48	33.34	33.02			
	% n-parafins	% isoparafins	% n-olefins	% iso-olefins	% naphteno-olefins	% di-olefins	% Other olefins
naphtha 1	17.83	34.90	-	-	-	-	-
naphtha 2	2.27	17.89	6.3	12.22	12.84	1.21	0.45

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371   Table 4: Experimental conditions in each of the experiments

Experiment	Feed 1	Feed 2	Flow rate Feed 1 g/min	Flow Rate Feed 2 g/min	Water %w/w in Feed2	T (ºC) Feed Stage
1.Binary entrainer	50% w/w Hexane; 50% w/w Isooctane	Ethanol with water	41.6	4.34	6	62
2.Binary entrainer	50% w/w Cyclohexane, 50% w/w Isooctane	Ethanol with water	41.63	4.38	10	65
3.Ternary entrainer	33 % w/w Hexane, 33% w/w Cyclohexane 33% w/w Isooctane	Ethanol with water	41.38	4.4	7	62
4.Quaternary entrainer	25% w/w Hexane, 25 % w/w Cyclohexane, 25% w/w Isooctane, 25% w/w Toluene	Ethanol with water	42.34	4.34	6	68
5. Complex mixture	Naphtha 1 from direct distillation	Bioethanol	43	4.3	8.3	63
6. Complex mixture	Naphtha 2 from FCC process	Bioethanol	44.5	4.3	8.3	63

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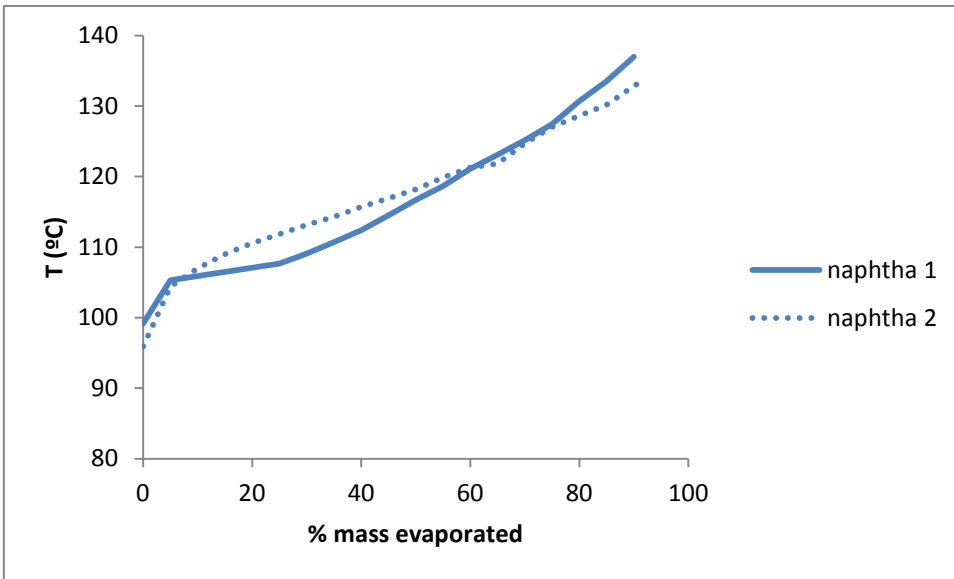
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387 Figure 1a. Distillation curves of the two types of naphtha studied.
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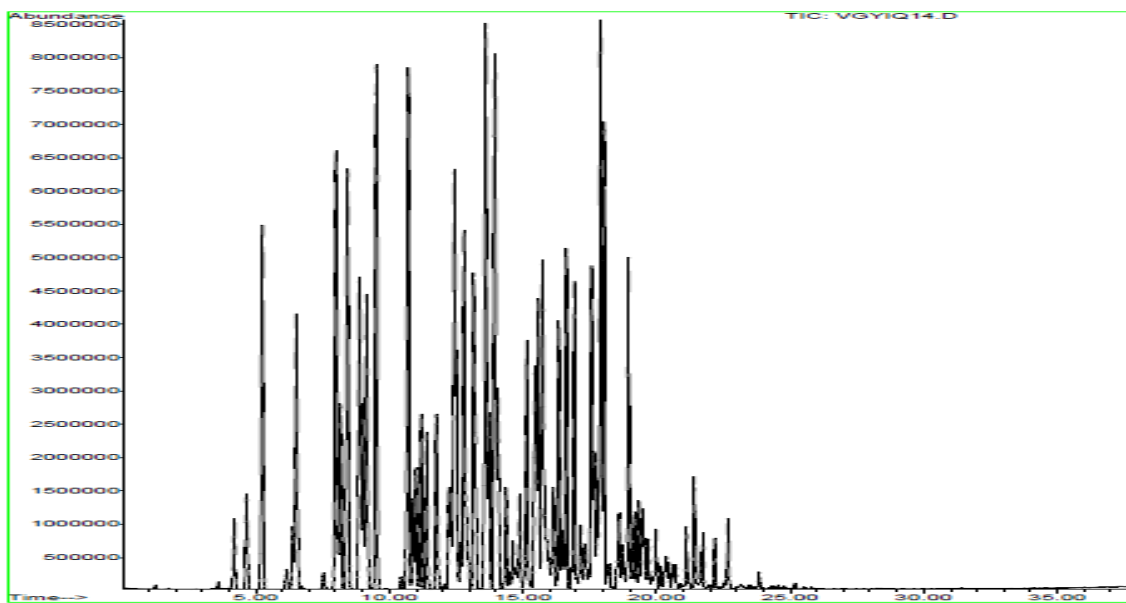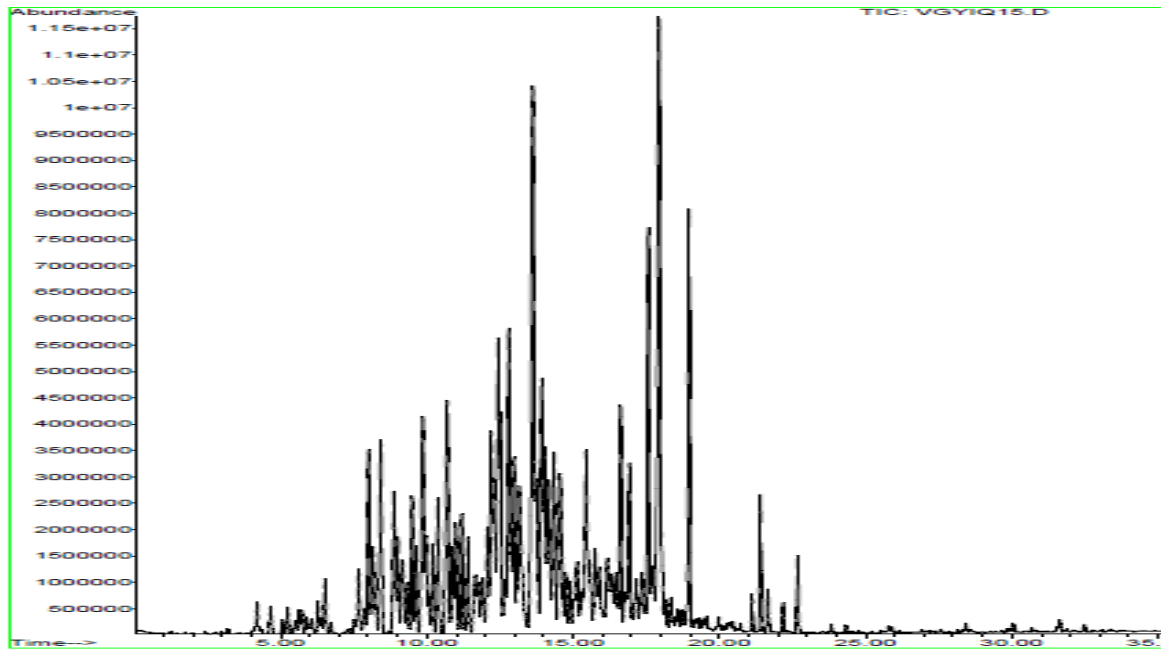
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406 Figure 1b. Chromatographs with mass analysis for naphtha 1 (above) and naphtha 2
407 (below).
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412   Figure 2. Experimental flow diagram of the tested process
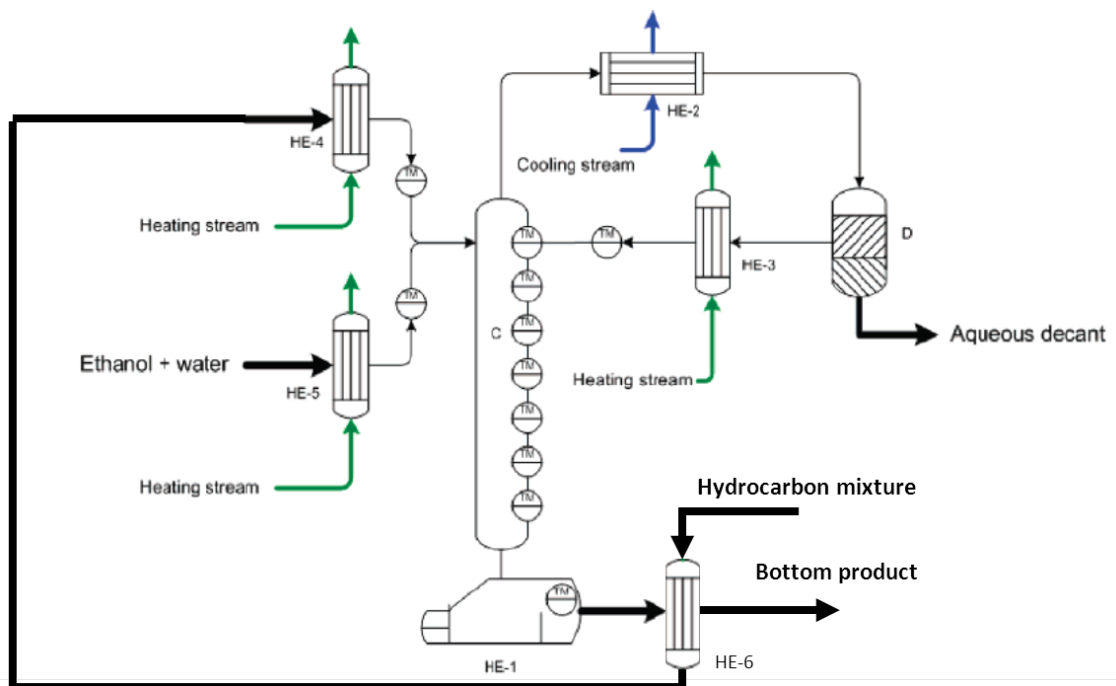
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420 Figure 3. Mixture containing hexane and isooctane. Plot of several variables against reboiler
421 heat duty: a) Experimental top (♦) and bottom (■) product flow rates. b) Experimental
422 composition (w/w) of water, ethanol hexane and isooctane in the column distillate (aqueous
423 phase). c) Experimental composition (w/w) of ethanol, hexane and isooctane in the column
424 bottom. d) Experimental composition (w/w) of water in the bottom product.
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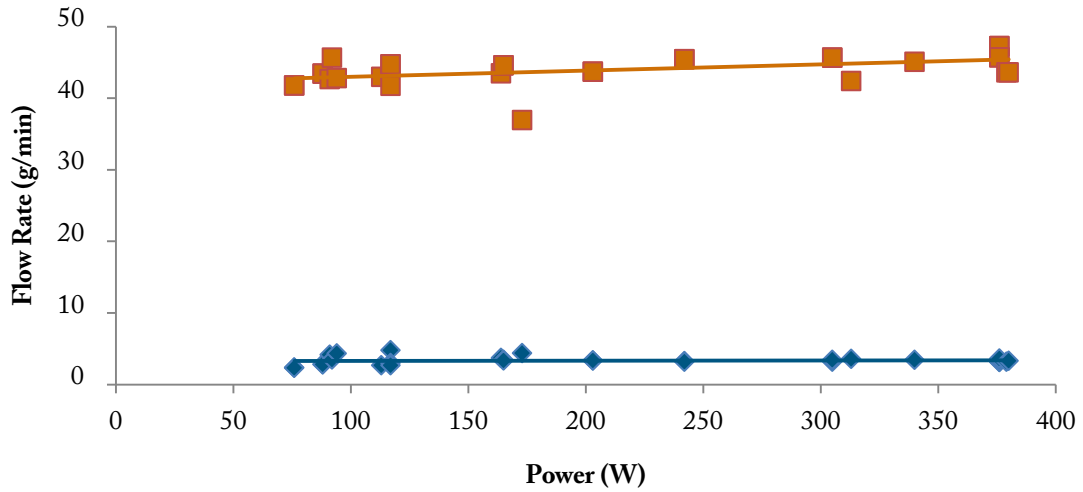
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427 a)
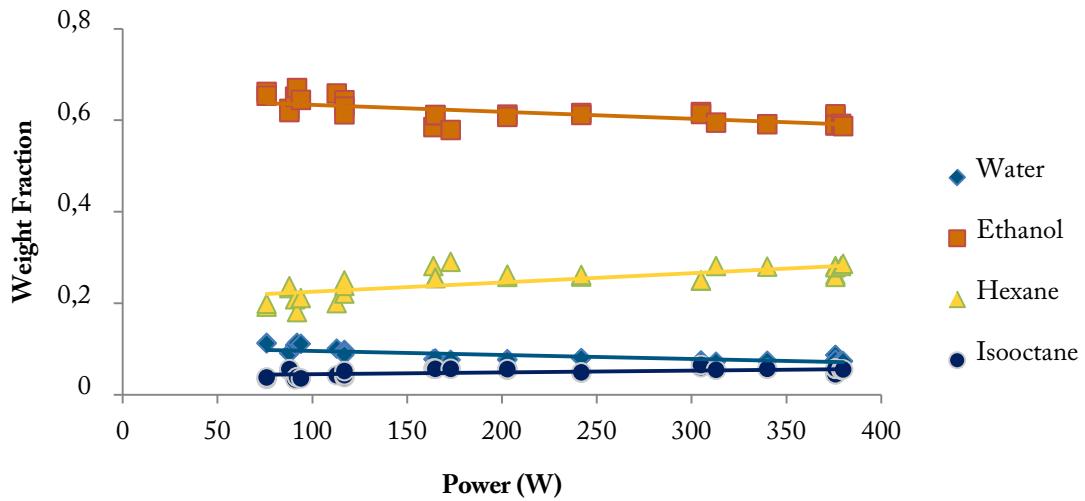
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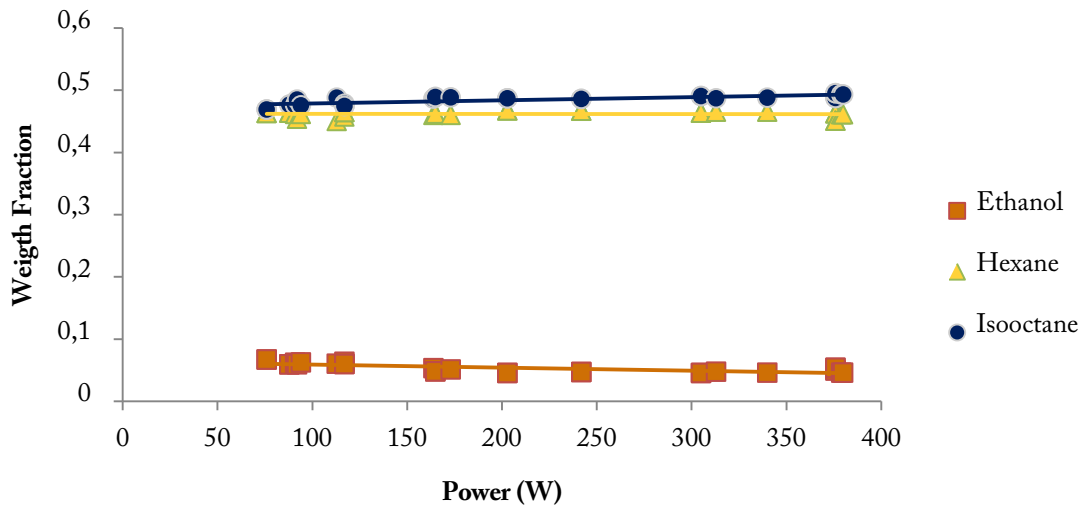
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431   c)
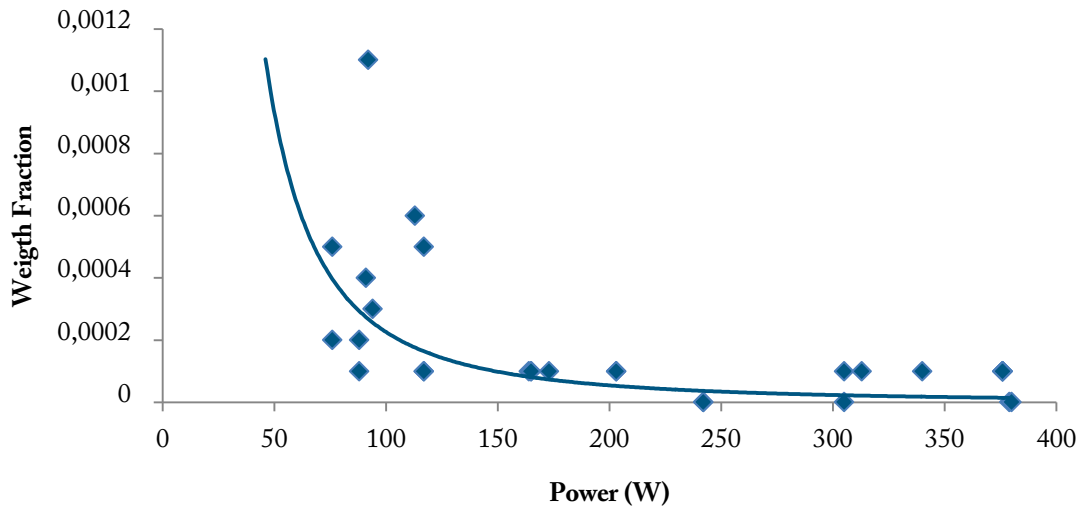
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433   d)
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436   Figure 4. Mixture containing cyclohexane and isooctane. Plot of several variables against

437   reboiler heat duty: a) Experimental top ($\blacklozenge$) and bottom ($\blacksquare$) product flow rates. b) Experimental
438   composition (w/w) of water, ethanol cyclohexane and isooctane in the column distillate
439   (aqueous phase). c) Experimental composition (w/w) of ethanol, cyclohexane and isooctane in
440   the column bottom. d) Experimental composition (w/w) of water in the bottom product.
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444   a)
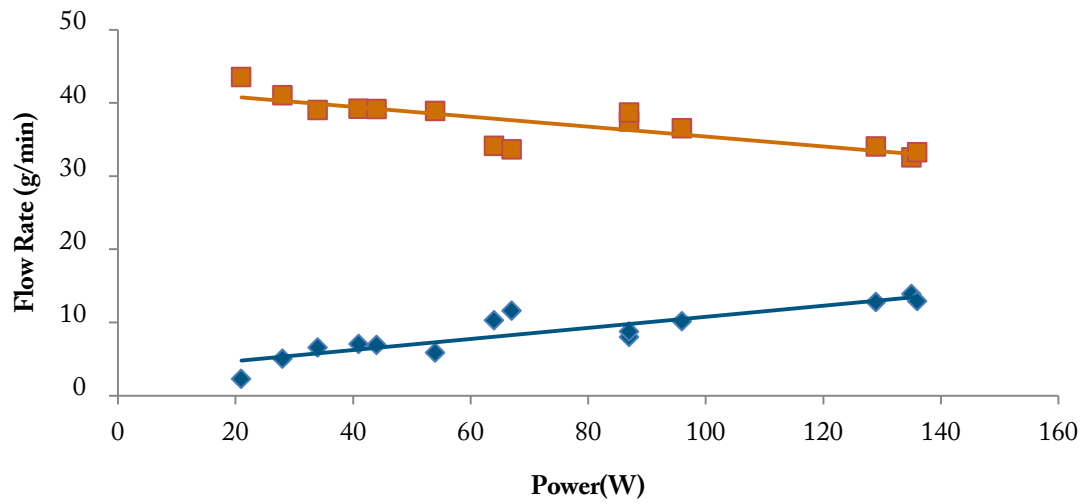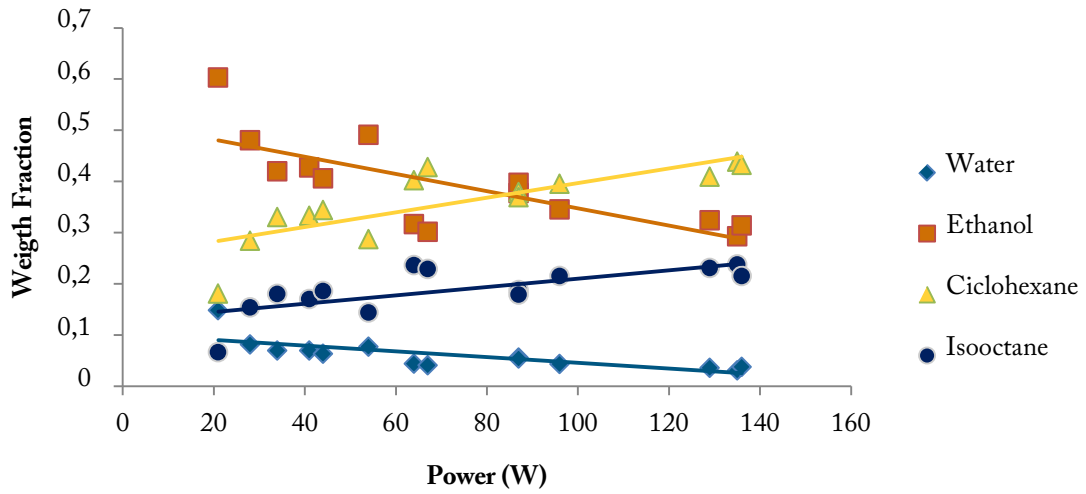
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446   b)
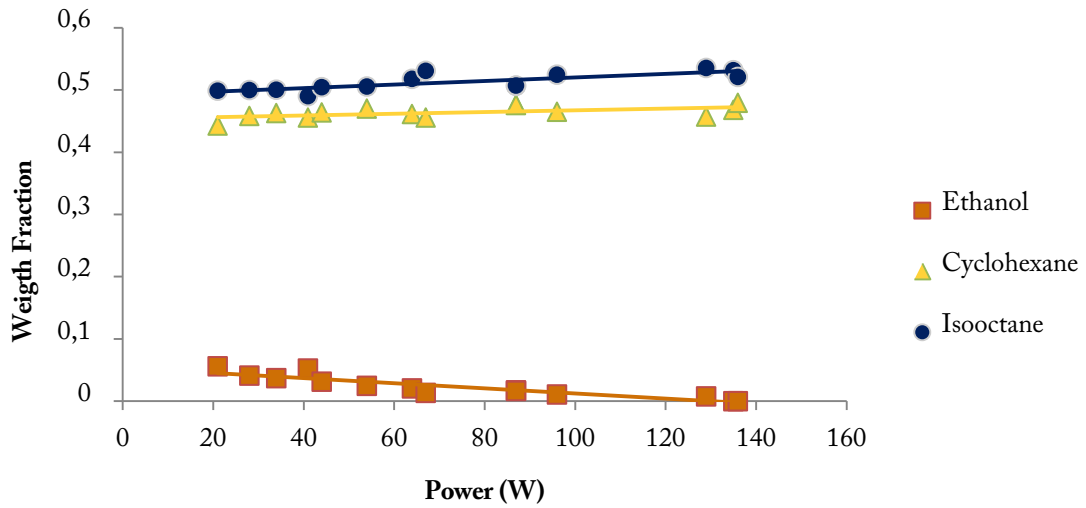
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448   c)
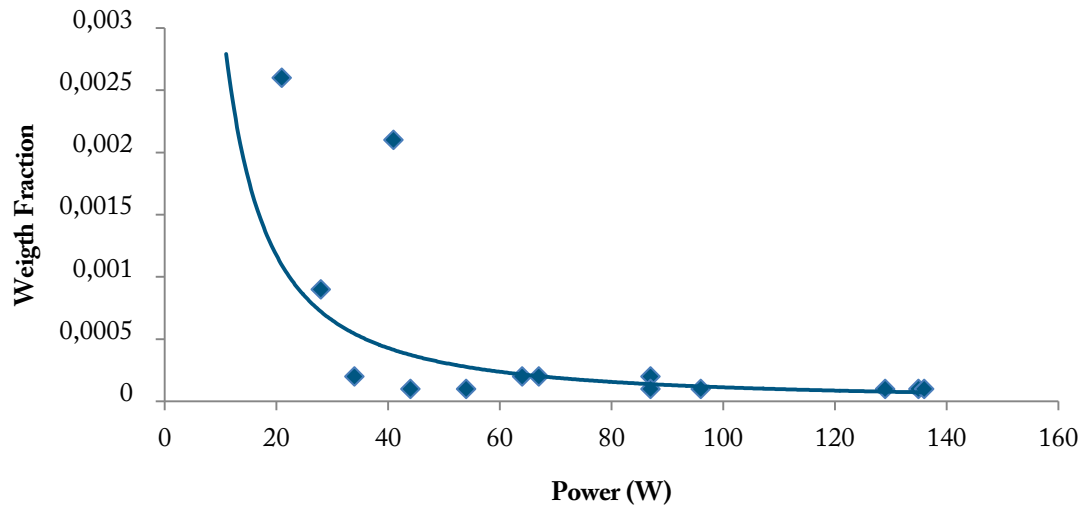
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454   Figure 5. Mixture containing hexane, cyclohexane and isooctane. Plot of several variables

455   against reboiler heat duty: a) Experimental top (♦) and bottom (■) product flow rates. b)
456   Experimental composition (w/w) of water, ethanol, hexane, cyclohexane and isooctane in the
457   column distillate (aqueous phase). c) Experimental composition (w/w) of ethanol, hexane,
458   cyclohexane and isooctane in the column bottom. d) Experimental composition (w/w) of water
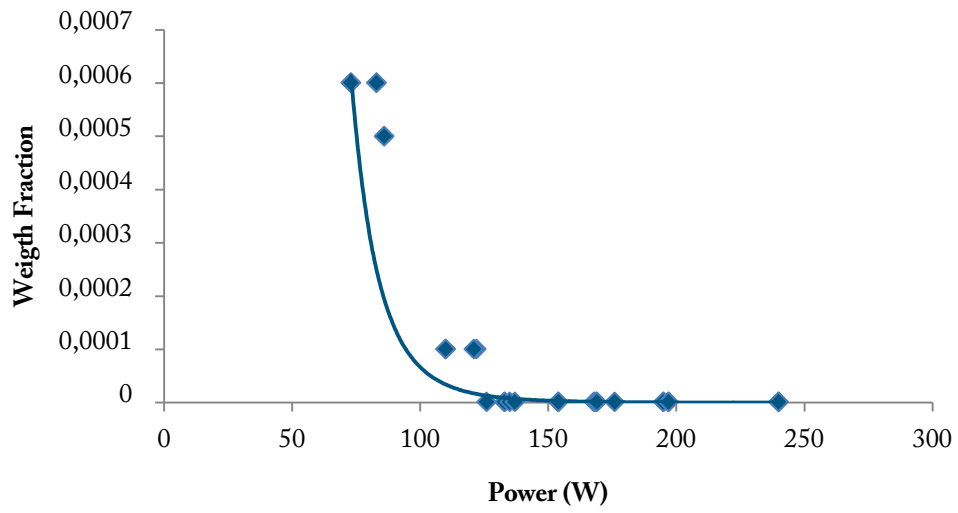459   in the bottom product.
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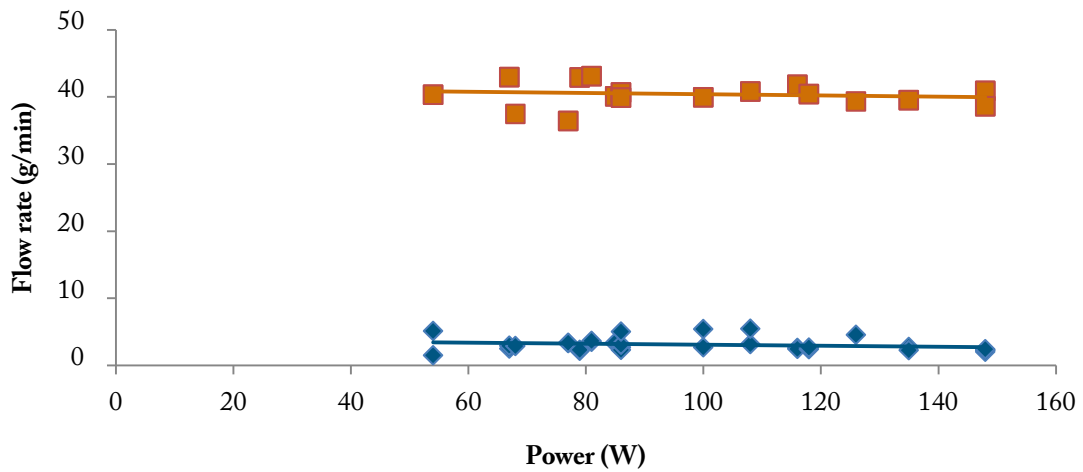
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464   a)
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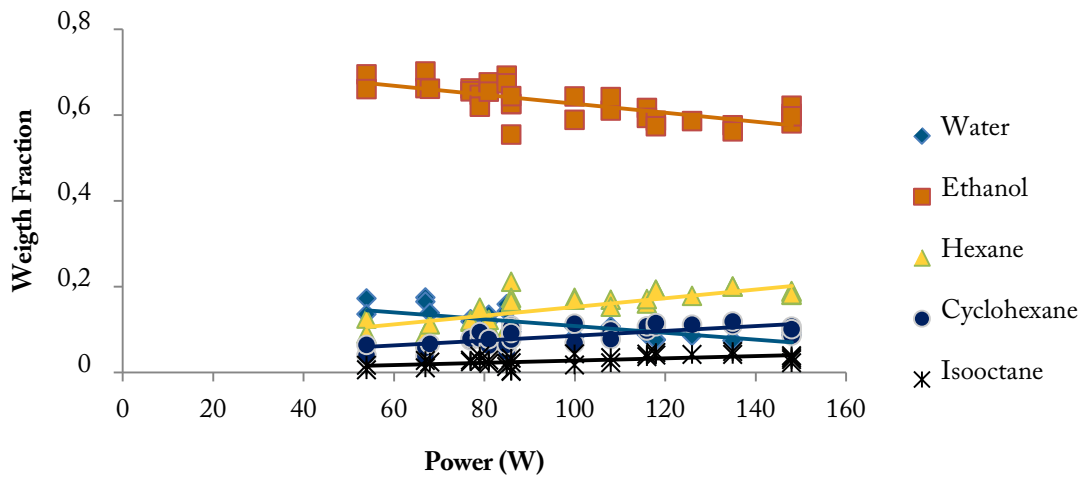
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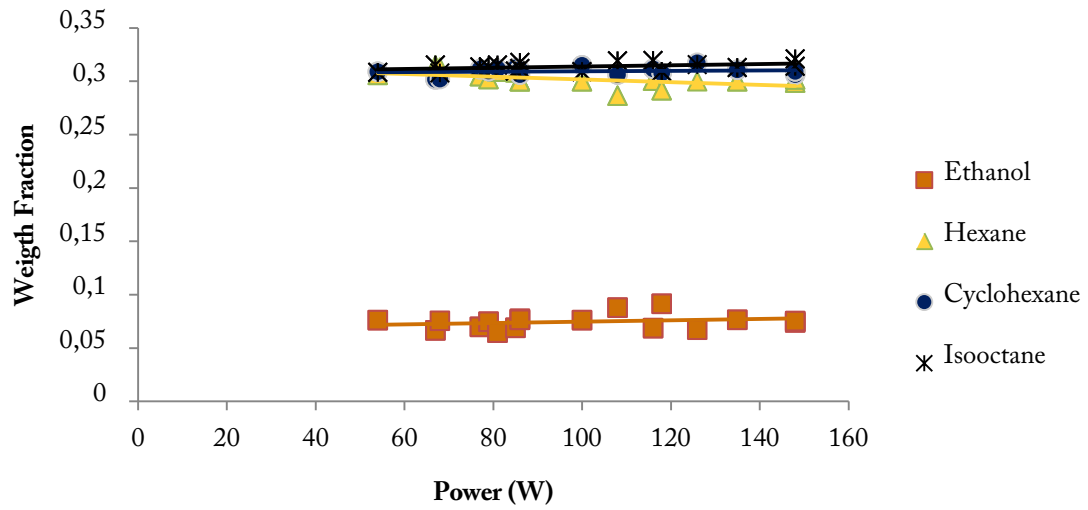
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469   c)
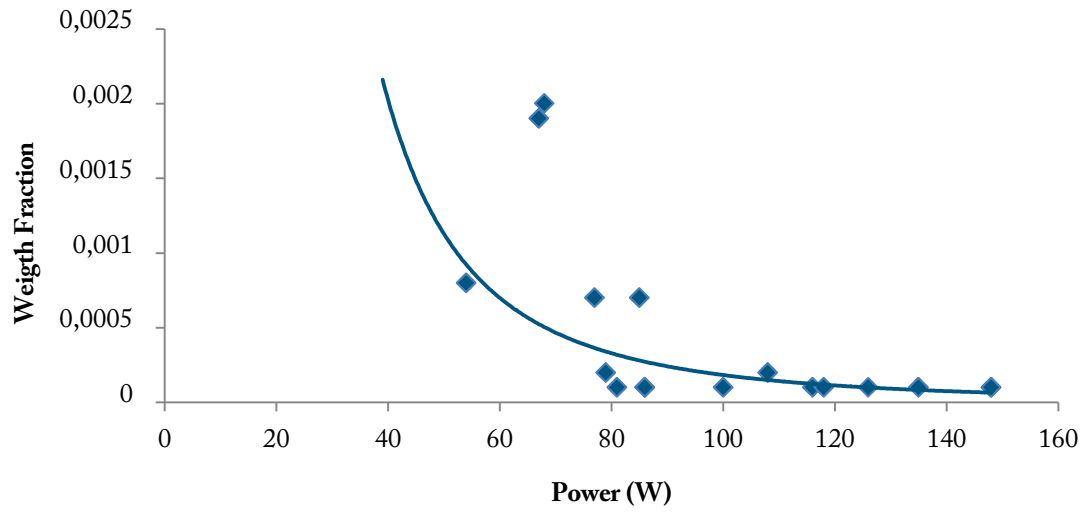
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471   d)

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479   Figure 6. Mixture containing hexane, cyclohexane, isooctane and toluene. Plot of several
480   variables against reboiler heat duty. a) Experimental top (◆) and bottom (■) product flow
481   rates. b) Experimental composition (w/w) of water, ethanol, hexane, cyclohexane, isooctane and
482   toluene in the column distillate (aqueous phase). c) Experimental composition (w/w) of ethanol,
483   hexane, cyclohexane, isooctane and toluene in the column bottom. d) Experimental composition
484   (w/w) of water in the bottom product.
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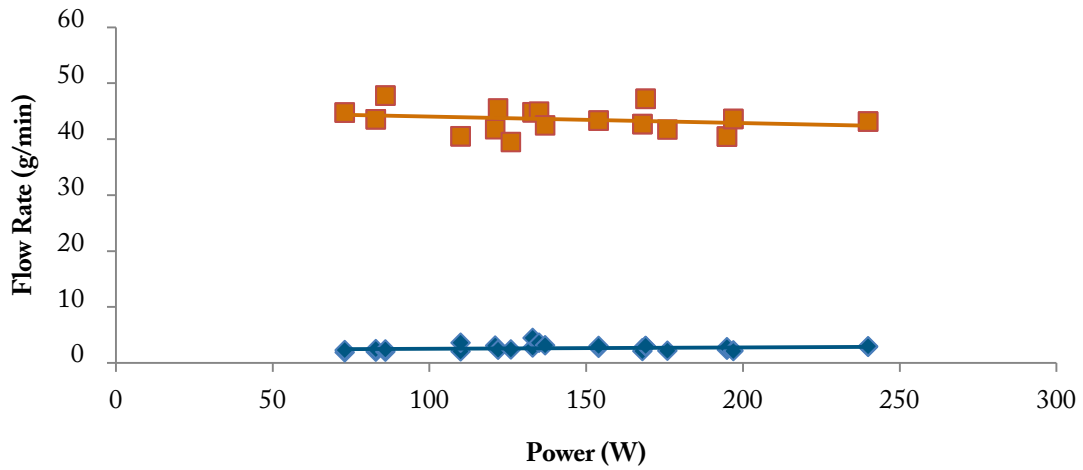
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488   a)
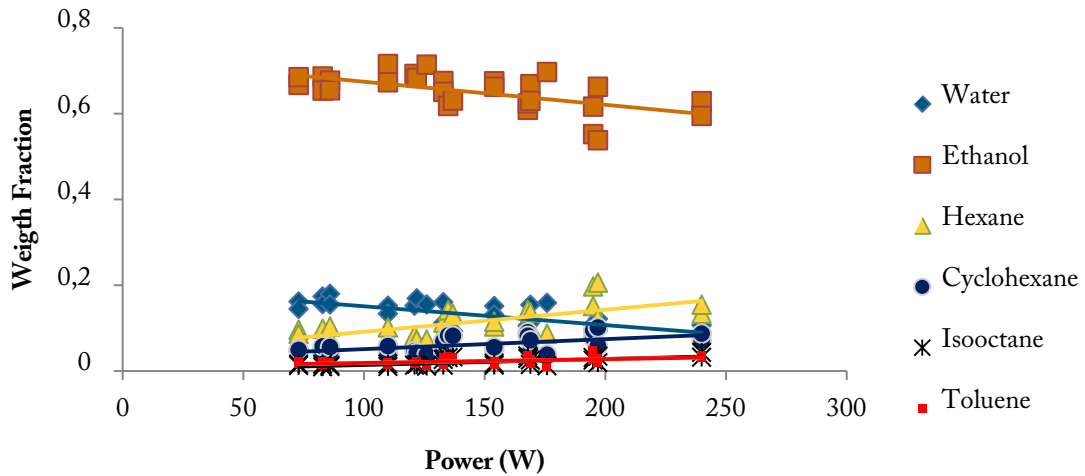
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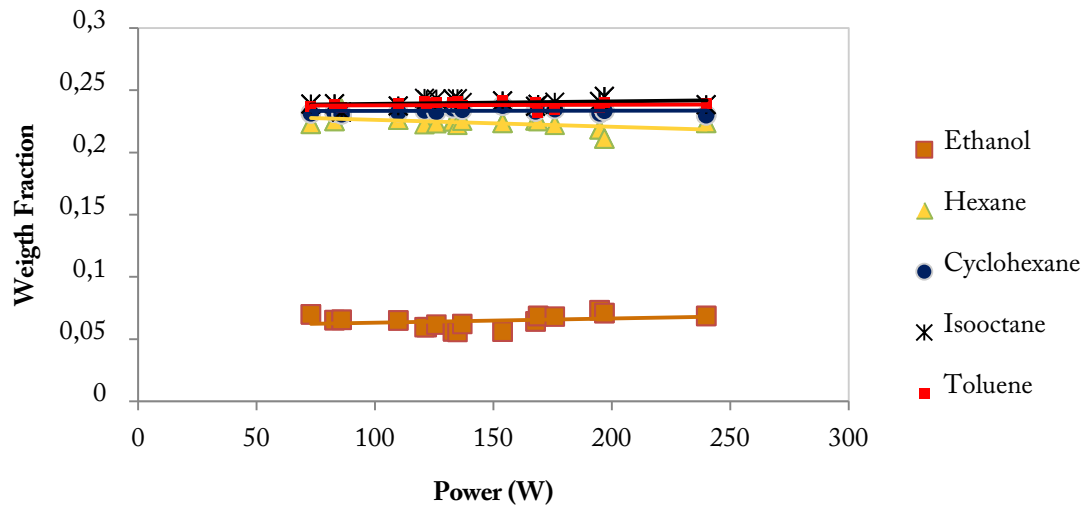
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492   c)

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494   d)

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498   Figure 7. Naphtha 1 Mixture. Plot of several variables against reboiler heat duty: a)
499   Experimental top (◆) and bottom (■) product flow rates. b) Experimental composition (w/w)
500   of water (◆) and ethanol (■), in the column distillate (aqueous phase). c) Experimental
501   composition (w/w) of ethanol, in the column bottom. d) Experimental composition (w/w) of
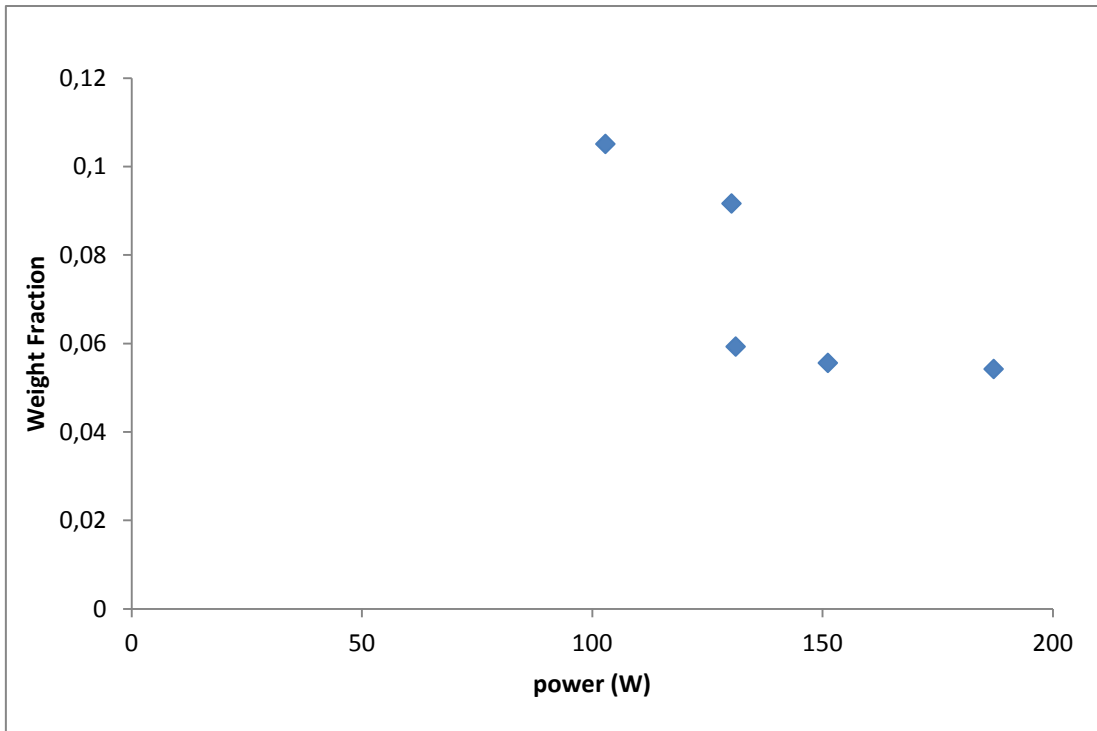502   water in the bottom product.
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506   a)
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511   b)
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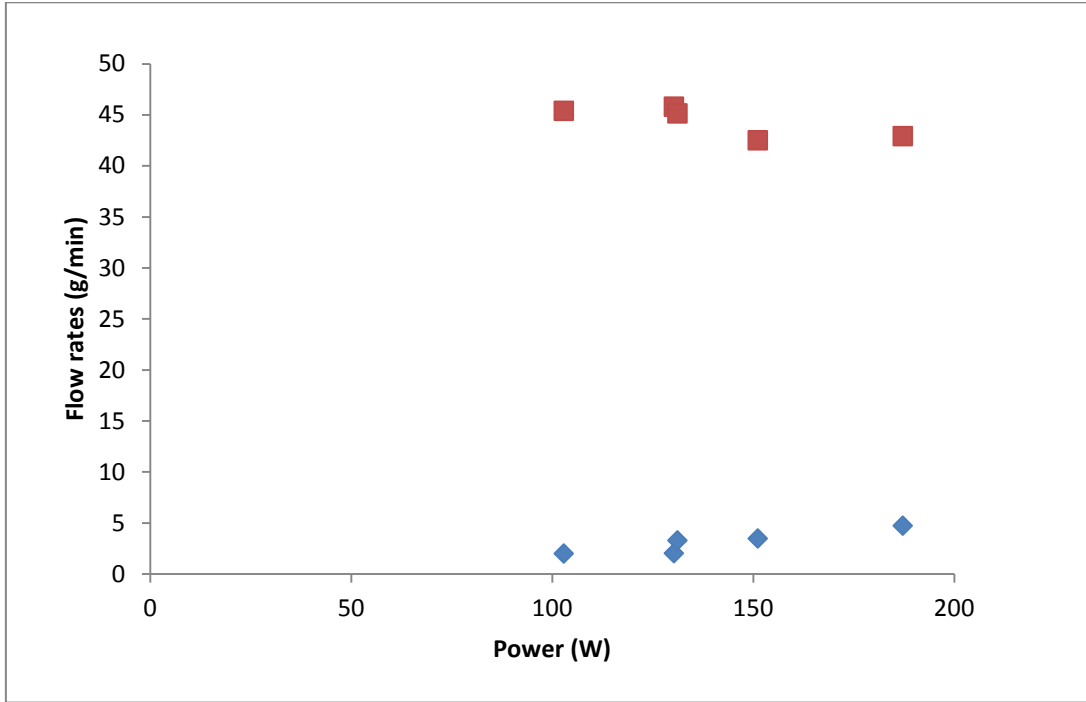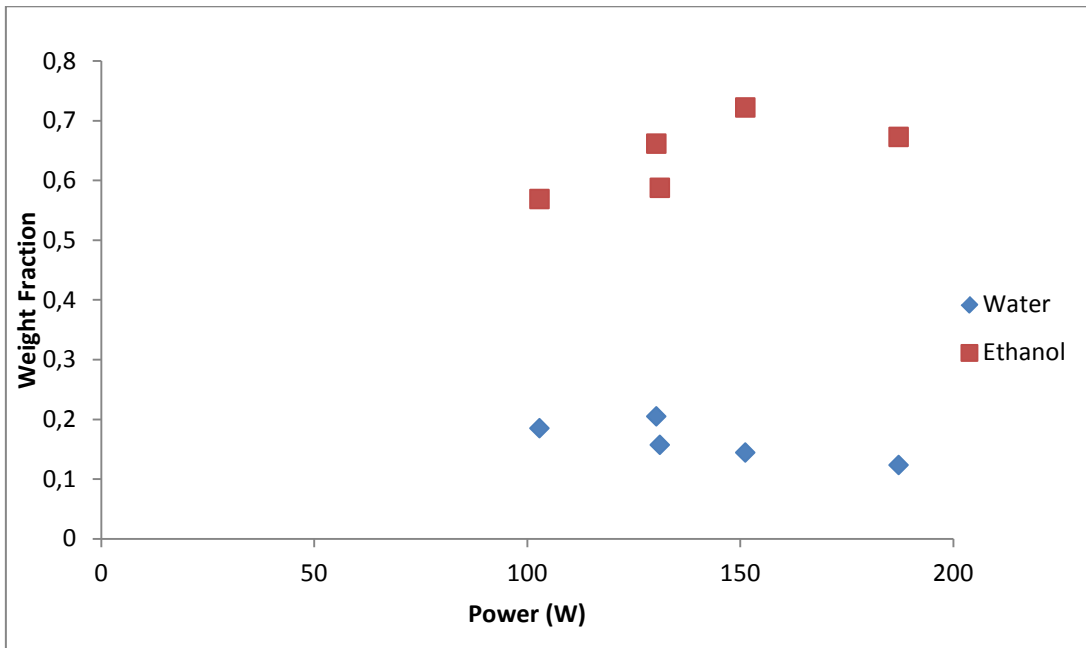
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515   c)

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518   d)

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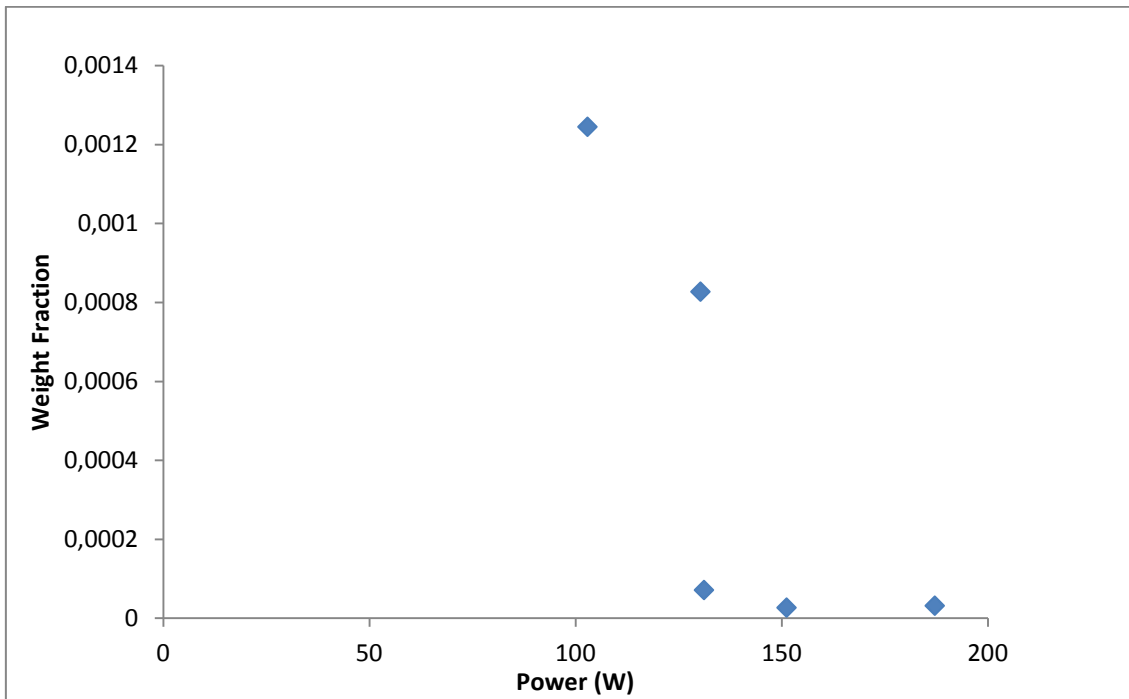
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524   Figure 8. Naphtha 2 Mixture. Plot of several variables against reboiler heat duty: a)

525   Experimental top (♦) and bottom (■) product flow rates. b) Experimental composition (w/w)

526   of water (♦) and ethanol (■), in the column distillate (aqueous phase).c) Experimental
527   composition (w/w) of ethanol, in the column bottom. d) Experimental composition (w/w) of
528   water in the bottom product.
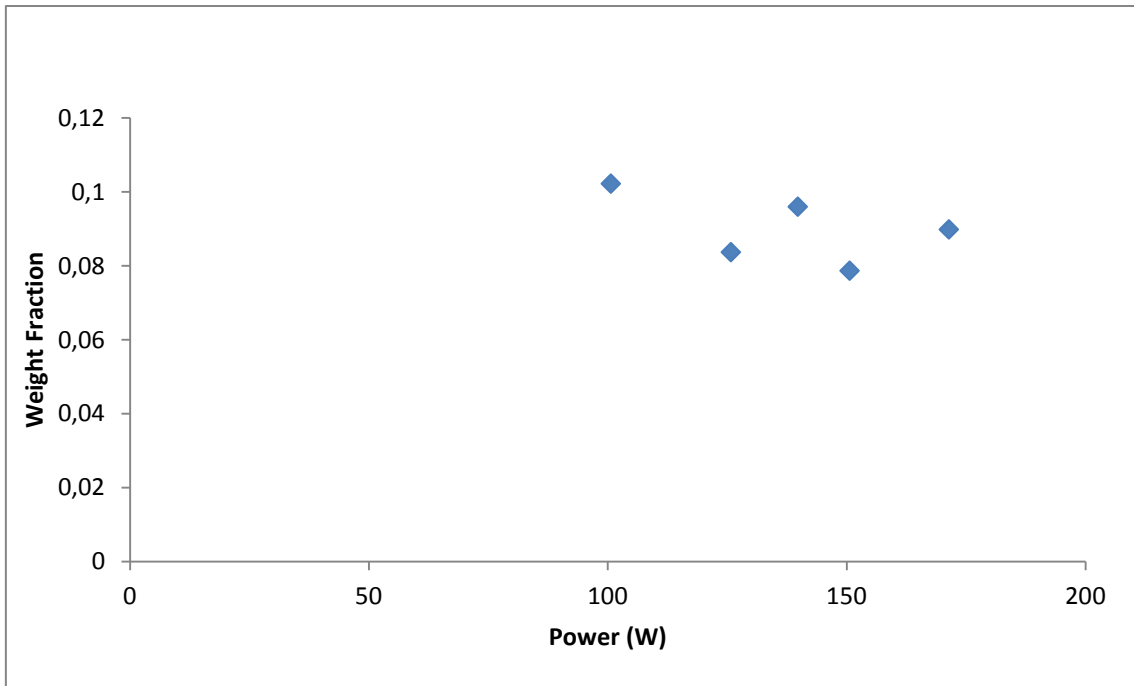529
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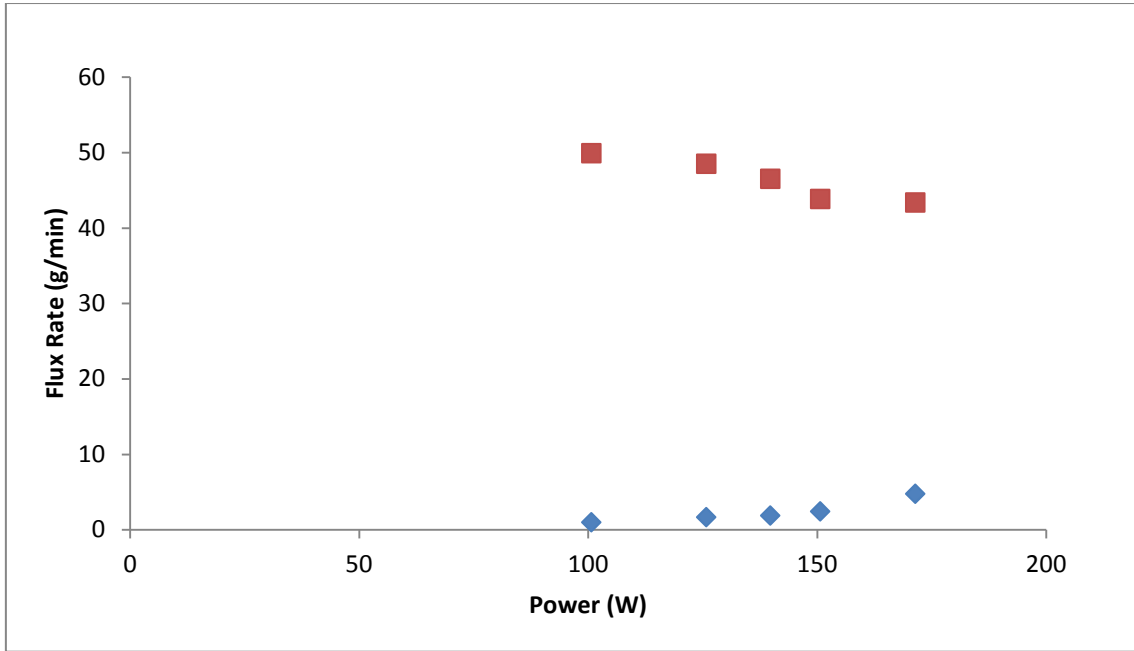
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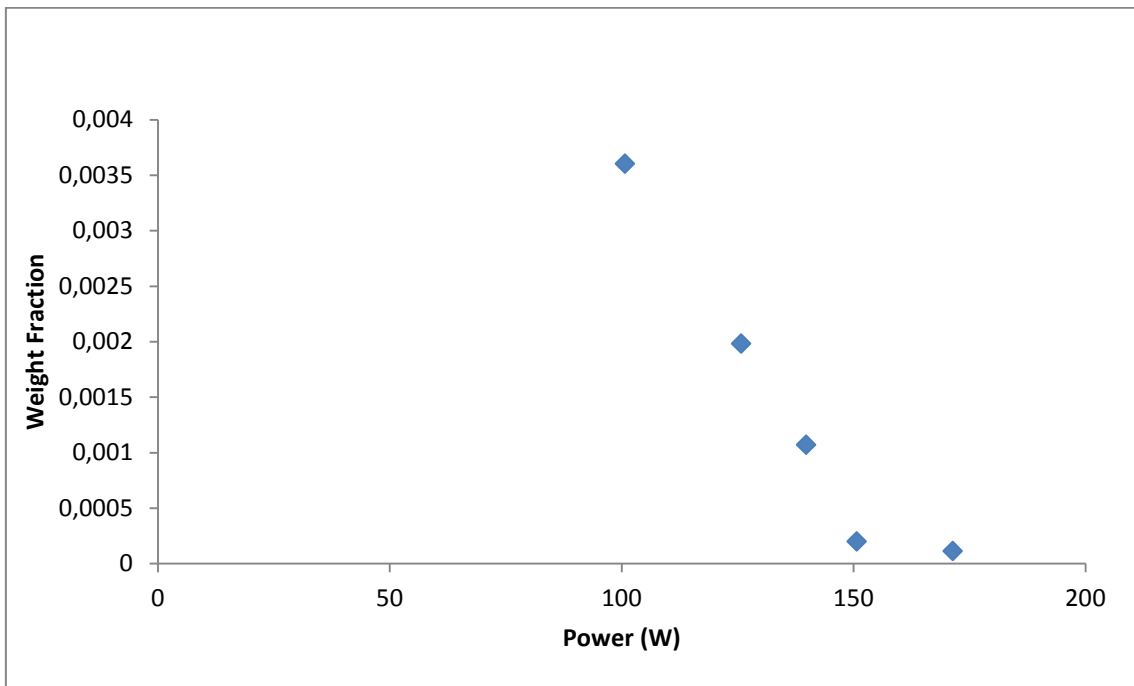532
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534   a)

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537   b)

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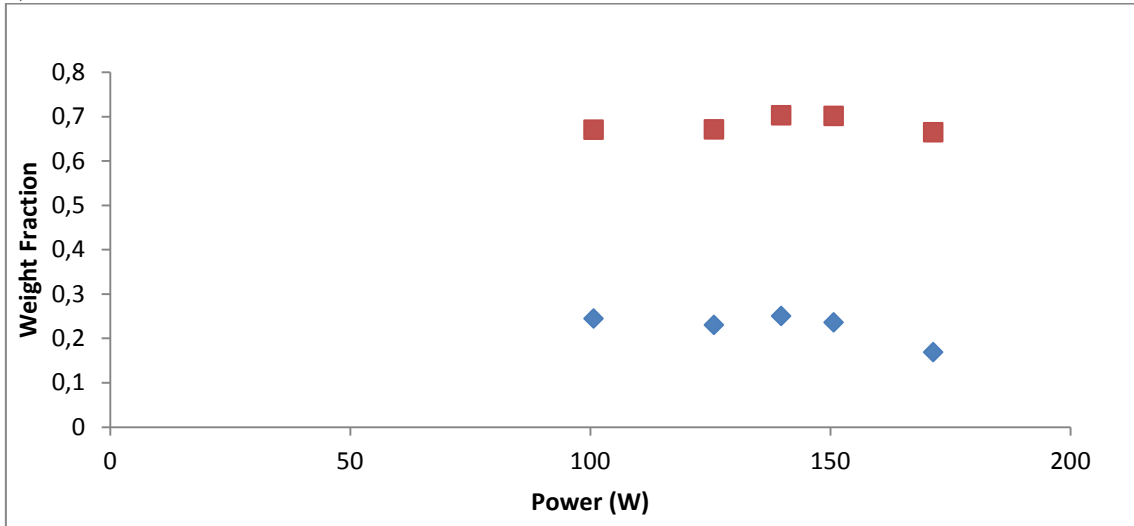
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543   c)

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548   d)

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