Biobased Chemistry and Technology

Thesis Biobased Chemistry and Technology

Analysing the robustness and flexibility of lipid biorefineries using response surface methodology

Joost Goedhart

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Supervisor(s)	:	Ton van Boxtel, Farnoosh Fashaei
Examiners	:	Ton van Boxtel, Ellen Slegers
Group	:	Biobased Chemistry and Technology
Address	:	Bornse Weilanden 9 6708 WG Wageningen the Netherlands

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Preface

I would like to thank Farnoosh Fasaei, Ton van Boxtel, and the department of BCT for providing me with feedback and support during my BSc Thesis. I would also like to thank Ellen Slegers .

Abstract

In the biobased economy algae are regarded as an important source of materials. Lipid-biorefineries are suited to yield biodiesel from algae. Biorefineries consist of several process steps, each with different options for operating units. The models for the operating units are presented by Slegers et al. (2014). In the research of Slegers et al. (2014) the optimal operating chain is derived. The algae broth input is constant and control factors are optimized.

Since algae do not constant grow constant throughout the year, variation in the feed flow occurs. A robust biorefinery is needed to keep the output constant with variation in the algae broth. Furthermore lipid biorefineries want to fulfil varying demands, a flexible biorefinery is needed. Control over the system through control factors is desired to fulfil these demands.

With the use of response surface methodology (RSM), the operating units and operating chains of lipid biorefineries are analysed. The RSM provides a quantitative approach in analysing the systems and insight whether a system is flexible or robust. Furthermore it indicates which characteristics of the algae broth or control factors influence the process.

With the classification of the operating units, robust or flexible operating chains can be created. The most robust operating chain is created by linking the most robust operating units. The most flexible operating chain is created by linking the most flexible operating units. In this study the most flexible, robust, and the most efficient operating chains presented by Slegers et al. (2014) are studied.

In this research a robust system is classified as a system where the output will be minimal influenced by the variation of the input. The RSM is created by linking the output with the input. Low parameters from the RSM indicate a robust system that is minimal influenced by the varying flow and concentrations of algae. The control factors are optimized. For the operating chains a different operating point is studied to investigate if the robustness is improved.

Flexible systems are indicated with high control over the output through control factors. A RSM will be created by linking the output with the control factors. High parameters from the RSM indicate much influence of the control factors, thus can be used to influence the output of the system. With the use of a non-square relative gain array (NRG) control over the flexible

operating chain can be reached. The NRG indicates which control factor favours which output.

The RSM is successfully applied to create a more flexible or robust processing chain. Also more insight can be gained over the process, indicating which control factors have influence on the output. From this information it is possible to pair control factors with output. The framework created for this thesis can be applied on any system.

1. Introduction

1.1 Problem statement

Depletion of oil reserves requires the investigation for alternatives choices. The intergovernmental panel on climate change wants all fossil fuels to be banned in 2100 to stop global warming(IPCC, 2014). Biobased economy is an economy independent of fossil fuels. This economy can be reached through effectively using biomass. Currently biomass is mostly used for feed and food, however in the biobased economy it will be used as a chemical, material, and fuel. For the biobased economy algae are regarded as an important source for these products(Wolkers et al., 2011).

Algae produce biomass from CO_2 and solar energy. Algae can efficiently use 3-8% of the solar energy while plants can only use 0.5%(Lardon et al., 2009). Also algae are more suitable at producing biofuel than plants(Chisti, 2008). Beside CO_2 and light, algae need nutrient rich water to grow(Chisti, 2007). Algae can be cultivated on waste water and marine conditions and are not competitive for fresh water resource (de la Noüe et al., 1992; Pittman et al., 2011; Wang et al., 2010), making it suitable for cultivation on regions not suited for crops.

Algae production is dependent on different variables: temperate, light, wind velocity, relative humidity, reactor geometry, algae characteristics, biomass characteristics, and wall material(Slegers et al., 2011a; Slegers et al., 2013; Ugwu et al., 2008). Therefore the production of algae won't be constant year-round.

Products from algae are a hot-topic currently, it is possible to extract oils, proteins, starch and pigments from algae with the use of biorefineries(Wolkers et al., 2011). IEA Bioenergy task 42(Cherubini et al., 2007) defines biorefineries as the sustainable processing of biomass into a spectrum of marketable products and energy. However current algae biorefineries are not yet economic feasible (Williams and Laurens, 2010). Production costs must be reduced ten times and efficiency increased three times (Wolkers et al., 2011). Another way to increase the economic feasibility of algae is to coproduce products along the biofuel(Langeveld et al., 2010; Li et al., 2008) or to lower external energy use by recovering and utilizing the non-lipid portion of the algal(Chowdhury et al., 2012).

In the project of TKI-biorefinery several scenarios for biorefineries are considered. These biorefineries produce products from algae. Biorefineries consist of several process steps. These process steps can be categorized into harvesting, dewatering, disruption, extraction and conversion. Within the process steps several unit operations are possible, each with their own configurations. The configurations of the unit operations are optimized with respect to efficiency. In the model an optimum operation chain has been designed with an inflow of 5000 L h^{-1} and an concentration of 2 g L^{-1} algae. Optimisation has been reached through maximizing the net energy ratio(NER) or yield of biodiesel.

The quantity of algae broth production varies throughout the year, causing the output to shift(Slegers et al., 2011b). The variation of algae can be feed flow rate, concentration, and substance. These all influence the product yield and NER(Scott et al., 2010). As a consequence, a system optimized for a given condition may perform below expectations under different circumstances. Ideally a lipid refinery can compensate for these variations in algae. Compensation can be done through changing control factors (for example concentration factor, chitosan concentration, bead filling, extraction temperature, and methanol flow) in the process.

Furthermore the demand for product can change over time. Making flexible biorefineries will be able to fulfil these new demands. This flexibility can be done through adjustment of the control factors of the operating chain.

Phadke (1995) defines robustness as 'system parameters (variables) that are permanently set in such a way as to minimize the effect of unforeseeable changes in the operating environment on the performance of the system without eliminating the cause of the changes themselves.' Whereas in this thesis robustness does not fix the system parameters but let them change to keep the optimal performance. A robust system is defined as a system that has the capability to compensate the influence of disturbance through control factors.

Olewnik et al. (2004) defines a flexible system as 'a system designed to maintain a high level of performance through real time changes in configuration when operating conditions or requirements change in a predicable or unpredictable way.' While a flexible system in this research is defined as a system that can change its process, within the allowable constraints, to fulfil alterations in the outflow demand. A flexible system has a high controllability.

Currently no research has been done around the robustness or flexibility of algae biorefineries. Sensitivity analysis, as seen in Yao (2015), have been performed on the models of algae biorefineries by Slegers et al. (2014).

1.2 Objectives

The objective of this thesis is to develop an effective framework to keep the desired production in lipid biorefinery. This can be either a robust system, where the influence of the input on the output is minimal, or flexible, where it is easier to adapt the systems output. An operating chain consist of all processing unit used from begin- to end-product. In this research four different operating chains will be analysed. These four operating chains consist of the most robust, flexible and efficient with regard to biodiesel yield and NER.

Each processing unit is analysed on its robustness or flexibility, from this study the most robust or flexible operating units can be found. The robust operating chain will be created by linking the most robust operating units, for a flexible operating chain the most flexible operating units will be linked. In the paper of Slegers et al. (2014) the most effiecient operating chain with respect to biodiesel yield and NER are given.

Investigating how the output changes with a varying input will provide information on the robustness of the system. Analysing the influence of the control factor on the output will gather more information about the flexibility of the system. Control factors are the operating conditions of the processing units. These influence how the system operates. In this research there are two production objective: flow of biodiesel and net energy ratio (NER). The NER is the energy gained from the biodiesel divided by the total energy usage from the production of the biodiesel.

1.3 Approach

For the design of a robust or flexible lipid biorefineries a step-wise approach is chosen. Since each step in the process of the lipid biorefinery has multiple options for operating units, first all the different processing units will be analysed on their robustness or flexibility. Robustness is analysed by linking the input of the algae broth with the output. It indicates how variation in the algae broth influences the output. Flexibility is analysed through linking the control factors with the output. It indicates how much influence on the output is reached with the control factors. The output in this thesis consist of flow of biodiesel and NER. The processing units are provided from the paper of Slegers et al. (2014).

The next step after analysing the processing is classifying them on whether they are robust or flexible.

After classifying the operating units an operating chain will be created with them. Linking the most robust operating units creates the most robust operating chain. Linking the most flexible operating units creates the most flexible operating chain.

Linking the operating units black box models will yield a new black box model as can be seen in Figure 1, where the different black box models linked together form one black box model of the operating chain. Output will only consist in flow of biodiesel and NER for the operating chain. Input will be the algae broth. The control factors depends on the operating chain.

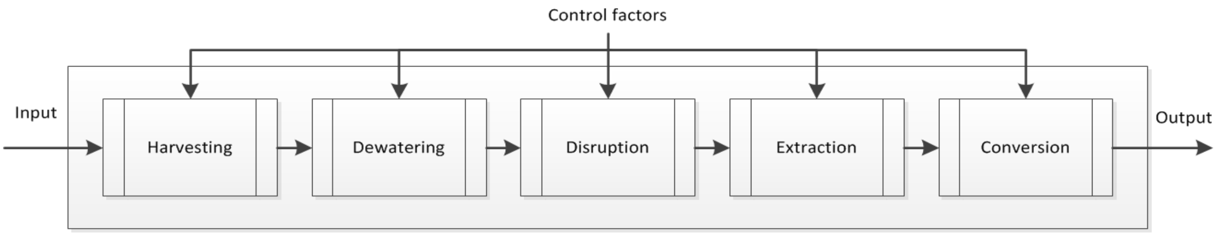

Figure 1 A black box model of an operating chain of an algae biorefinery where the operating units also consist of black box models.

Since only the relation between output and input or control factors are relevant a black box model is created for each processing unit. Every process unit has a different input and output. These black box models will be created through response surface methodology (RSM). The RSM provides a full quantitative results of the system, while most sensitivity test only give semi quantitative results. The quantitative results are used to classify processing units whether they are robust or flexible. Furthermore the quantitative results can be used to control the system. Another advantage of RSM is that no information about the initial condition of the model is needed. Also the polynomial model formed from the RSM gives information about the system without solving the model (Ivakhnenko, 1971).

Robustness is analysed by linking the input with the output while keeping the control factors optimal. Flexibility through linking the control factors with the output and keeping the algae broth fixed. Processes without control factors or which have no influence on the output will not be analysed on their flexibility. Processes were the output is constant will not be analysed on their robustness. These are already robust and an RSM will not yield any new information.

For robust operating chains the control factors are optimized. However performing at different operating points can influence the robustness. In this study performing with non-optimal configurations is also investigated.

For flexible systems control over the outputs is preferred. Since the flexible systems have interaction, changing one input can influence multiple outputs, knowing which input influence which output the best, a more controllable system can be created. The relative gain array (RGA) gives insight in the amount of interaction. Not only will it tell the amount of interaction but also which control factor can be best used to control which output. Thereby optimal control over the system can be reached.

In this thesis four different operating chains will be analysed. The most robust operating chain will be analysed on its robustness. The most flexible operating chain will be analysed on its flexibility. The other two operating chains that will be analysed are those with the highest NER and flow of biodiesel according to the paper of Slegers et al. (2014). Reason for this is that a high flow of biodiesel or NER can be preferred more than a robust or flexible operating system. Slegers et al. (2014) never analysed how robust and flexible these systems are. However just analysing the operating chain with the higher NER and flow of biodiesel will not yield much information. By comparing them with the most robust and flexible operating chain more information can be gained.

2. Materials and methods

2.1 Processing units

Throughout the process chain the algae broth is processed to biodiesel. For this process several steps are needed as seen in Table 1 and in Figure 21. Every process has multiple inputs and outputs, however not all are relevant for this research. The focus of this research lies on NER and biodiesel yield. Some properties of algal broth that has influence on the products are considered, flow, concentration, and lipid fraction. Broth flow gives an indication for the product yield per hour and the energy demand to run the process chain. Concentration value shows the amount of lipid in the system and lipid content illustrate the potential amount of biodiesel. A concluded list of the relevant inputs and outputs for each process step can be found in Table 1.

For every output a RSM is created to analyse the processing units. From this RSM classification can be done to indicate robustness or flexibility. For the calculation of the RSM the input values have to be given. Each process step has different inputs with different values. All the possible values of these inputs must be determined from the model to create an accurate RSM. This will be done by finding the minimum and maximum values of the output of the previous step in the process, these values will be the input of the next processing unit. These minimum and maximum values can be found in appendix A. The feed flow rate for the flexible systems are fixed values and not considered as a range. Therefore the average of minimum and maximum values of the outputs are taken.

The output concentration of dewatering which is the input of disruption step is fixed at 100 g L^{-1}. It is assumed that in harvesting and dewatering only the flow has an influence on the end product. In bead-milling unit there is no change in algae flow and concentration, consequently only a RSM of energy usage will be created. After harvesting and dewatering lipids of the algae will be extracted to be converted to biodiesel. Flow of algae broth still remains relevant for input since this has an influence on the energy demand of the processing unit. The only relevant outflow in the final-step in the processing chain is biodiesel-flow and energy usage, the process chain is focused on yielding biodiesel from algae. All other outputs are not relevant in the final-step, these have no influence on the production objective.

NER evaluation is not a step wise approach and can only be calculated from the whole operating chain. The NER is calculated from the energy usage of processing units and the biodiesel yield. In order to test the robustness or flexibility of the processing units with regards to the NER, energy usage is used.

Table 1 All relevant input and outputs of the process steps

Process step	Input		Output	
Harvesting	Flow of algae broth	(m^3 h^{-1})	Flow of algae broth	(m^3 h^{-1})
	Alga concentration	(kg m^{-3})	Energy usage	(J)
Dewatering	Flow of algae broth	(m^3 h^{-1})	Flow of algae broth	(m^3 h^{-1})
	Alga concentration	(kg m^{-3})	Energy usage	(J)
Disruption	Flow of algae broth	(m^3 h^{-1})	Flow of algae broth	(m^3 h^{-1})
			Concentration lipids	(kg m^{-3})
			Energy usage	(J)
Extraction	Flow of algae broth	(m^3 h^{-1})	Mass lipids	(kg h^{-1})
	Concentration lipids	(kg m^{-3})	Energy usage	(J)
Conversion	Flow of algae broth	(m^3 h^{-1})	Flow of biodiesel	(L h^{-1})
	Mass lipids	(kg h^{-1})	Energy usage	(J)

2.1.1 Control factors

Table 2 shows the selected control factors. The values for the lower and upper bound are given in Slegers et al. (2014) and provided in Table 2. According to Slegers et al. (2014) the control factor for the dewatering step is concentration factor. The concentration factor influence the amount of reduction in the volume and increase in the concentration of the algae broth. Since concentration of the algae broth is fixed at 100 g L^{-1} after dewatering, the concentration factor is dependent on the concentration of algae going into the process. Thus the concentration factor in dewatering is not considered as an control factor in this research. Since the concentration factor of dewatering has a maximum of 40 times, the concentration of algae inflow has to have a minimum value of 2.5 g L^{-1} to ensure that the 100 g L^{-1} is reached after dewatering.

Table 2 Control factors given with their lower and upper bound.

Control factor	Unit	Lower bound	Upper bound
Concentration factor harvesting	(-)	2	40
Concentration factor drying	(-)	4	160
Concentration chitosan flocculation	(g m^{-3})	150	250
Concentration poly-glutamate flocculation	(g m^{-3})	10	60
Bead filling	(%)	70	90
Temperature $SCCO_2$	(K)	313	333
Pressure $SCCO_2$	(bar)	150	300
Flow of methanol	(m^3 h^{-1})	0.3	0.9
Temperature supercritical methanol wet conversion	(K)	513	533

2.2 Operating chains

Connecting the processing units together creates an operating chain. The output of a processing unit is the input of the next processing unit. Operating chains will be analysed through RSM creating a black box mode as seen in Figure 1. A second order model for each unit will be fitted to link the input with the output. A second order model is chosen as stated in 2.3 Response surface methodology. The operating chain will be analysed the same way as the operating units.

For energy usage the output of an operating unit is not the input of the next processing unit. Energy usage is added up after each process step to calculate the total energy usage.

2.3 Response surface methodology

With the use of the response surface methodology (RSM) it is possible to study the processing units and operating chains. RSM shows how the input or control factor influences the output. RSM is calculated by changing the conditions one at a time and computing the different outcomes. From this data a RSM can be created. Since not all process are linear, a second order model is used for RSM to ensure a good fit for the RSM. A third order model or higher has complex computation so second order model is assumed to be sufficient for the aim of this study. A second order model of RSM is :

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i U_i + \sum_{i=1}^{k} \beta_{ii} U_i^2 + \sum_{i<j} \sum_{j=2}^{k} \beta_{ij} U_i U_j + \epsilon \qquad (1)$$

Where y is the chosen outcome, U the changing conditions i that is applied on the processing units or operating chains, β the parameter, k the amount of conditions that can be changed and ϵ the error.

The parameters give an indication about the influence of U on the outcome. Parameters close to zero indicate that U has a low influence on the outcome, parameters bigger than zero indicate that U has a higher influence on the outcome. The parameters can be used to see if a system is robust or flexible. β_0 has no influence on the robustness or flexibility as it is not related to a changing condition.

The RSM is created with a computer simulation and not with experimentation where variation may occur between results. Doing the calculation with the same data will always yield the same results thus the error is obsolete.

The changing conditions have different scale of size, comparison of the parameters can only be done if rescaling is applied. Moreover it is also common to rescale the conditions before applying the RSM, this ensures the RSM is more accurate(Myers, 1976). Rescaling is done through recoding with the following formula:

$$U_{ij} = \frac{\xi_{ij} - [\max(\xi_{ij}) + \min(\xi_{ij})]/2}{[\max(\xi_{ij}) - \min(\xi_{ij})]/2} \qquad (2)$$

$$-1 \leq U_{ij} \leq 1$$

In which U_{ij} is coded value of the *i*th observation of variable *j*, ξ_{ij} is true value of the *i*th observation of variable *j*. $\text{Max}(\xi_{ij})$ is the maximum of all observations *i* of variable *j*. $\text{Min}(\xi_{ij})$ is the minimum of all observations *i* of variable *j*.

From this point all changing conditions will be given in coded values, since these are used for the RSM. This will be indicated with labelling them as coded values.

2.3.1 Statistical test

To ensure the RSM is a proper fit to the processing unit and operating chain a statistical test is performed. This statistical test indicates the fit of the RSM to the processing unit. The statistical test is to ensure the parameters represent the processing unit and can be used to classify the processing unit.

For each RSM the fit will be calculated with the correlation coefficient(R^2) and the adjusted correlation coefficient. The correlation coefficient is an indication on the fit of the RSM towards the modelled data. R^2 increases when a new variable is added, even if this variable does not hold new information. The adjusted R^2 does not hold this problem, only variables with predictive capability increase the adjusted R^2. The adjusted R^2 is more suited to see if the fit of the RSM is correct. The expression for adjusted R^2 is:

$$R^2_{adjusted} = (1 - R^2)\frac{n-1}{n-m-1}$$

Where R^2 is the normal correlation coefficient, n the number of data points and m the amount of independent variables.

Processing units are classified on their parameters. The RSM always gives a value for a parameter even if the changing condition has no influence on the result. To ensure parameters are correct a student's distribution test is performed on each parameter. $p < 0.05$ indicates that the coefficient is statistical significant. Parameters where $p > 0.05$ will not be used to investigate the robustness or flexibility of the system. The student's distribution test can reduce the amount of parameters that have influence on the output, this will not influence the robustness or flexibility of the processing unit. The robustness or flexibility is dependent on the value of the parameters, not the amount of parameters.

More information about the calculation of the student's distribution test can be found in appendix E.

2.4 Robustness

For finding a robust system all the processing units will be studied with varying inputs. The feed flow for harvesting will range from 4500 to 5500 L h^{-1} and the concentration from 0.5 to 1.5 g L^{-1}. The range of concentration is chosen because of the processing unit poly-glutamate flocculation, its input cannot exceed 1.5 g L^{-1}. Further explanation about this limitation can be found in section 4. Discussion For the other process steps the range of the feed flows can be found in appendix A.

In this research a robust system is classified as a system where the output will be minimal influenced by the variation of the input. Low parameters indicate a robust system that is minimal influenced by the varying flow and concentrations.

The control factors for the processing units are optimized with regards to their configuration when analysing them individually. Operating units are not individually analysed on their performance with non-optimal configurations. The performance of each processing unit is dependent on its configuration, some configurations improve the robustness of the processing unit. For example a concentration factor of 5x for the processing unit centrifugation in harvesting yields an outflow concentration from 2.5 to 7.5 g L^{-1}, while a 40x concentration factor increases the range of the outflow concentration from 20 to 60 g L^{-1}. This indicates that a low concentration factor makes the processing unit robust, whereas a high concentration factor indicates a low robust system since it is more influenced by the input. Analysing all possible configuration of the processing unit could reduce the influence of the configuration. Doing this increases the complexity of the system, and increases complex computational. Reduction of computational cost is not part of this study however using optimal configuration is sufficient in this study. Analysing the operating units with all possible configuration is done for testing them on flexibility. The inputs will only be control factors and the variation of the algae broth will be constant, thus complex computational will be low.

The performance of operating chains has been analysed with non-optimal configuration as stated in the introduction, to indicate if the condition below optimum increases the robustness. The non-optimal configuration will be set to the average of the minimum and maximum configuration of the processing unit. This will only be applied to robust operating chains and not individually operating units as stated above.

Linking robust processing units together will create a robust operating chain. Proof of this theorem can be found in 2.6 Linking processing units.

2.5 Flexibility

Flexible system are indicated with high control over the output through control factors. A RSM will be created with all the possible configurations of the control factors. The inflow of algae broth is fixed at 5000 L h^{-1} with a concentration of 1 g L^{-1} for the process step harvesting. For the other process steps the average of the flows in appendix A are taken.

Parameters with higher values from the RSM indicate higher influence of the control factors which can be used to influence the output of the system. Low parameters indicate that the control factor has no influence on the output of the system. The influence of a control factor on the output will be visualized with the use of a perturbation plot. In a perturbation plot only one control factor is changed, while the others are set to zero, and plotted how this alters the output. Placing all the control factors in one plot indicates how a control factor influence the output compared to the other control factors. From the RSM it is only seen which control factor have high influence, from the perturbation plot it is more clear how this control factor influence the output. This will only be done for operating chain and not operating units. Most operating units only have one control factor, the perturbation plot is used to compare control factors within the system.

Control over the systems cannot be reached with the RSM and perturbation plots only. The systems have multiple outputs with interaction, the RSM and perturbation plot do not hold information about interaction. In 2.7 Pairing this will be taken into account to indicate which control factor should control which output.

Linking flexible processing units together will create a flexible operating chain. Proof of this theorem can be found in section 2.6.

2.6 Linking processing units

It is stated previously that linking robust processing units together creates a robust operating chain and the same for flexible operating chains. To prove this statement, information is needed from the RSM. Parameters from the RSM influence the output, low parameters give an indication that the output of the operating unit has a small range, which can be seen in Figure 2, while an operating unit with high parameters has a large range that can be seen in Figure 4.

These output ranges can be indicated using the propagation of error (POE). The POE calculates the variance of the input to the output. This variation can be used to find the optimal points and also to find the points with large variation. A low POE indicates low variation, an high POE indicates high variation. The expression for the POE is as follow:

$$POE = \sqrt{\sum_i \left(\frac{\delta f}{\delta U_i}\right)^2 \ \delta_{U_i}^2 + \delta_{residual}^2} \qquad (3)$$

Where f is the function, U_i the individual factor i, $\delta_{U_i}^2$ the variance of the input factor U_i, and $\delta_{residual}^2$ the residual variance. Since the models always produce the same outcome the residual variance can be neglected.

Since the first derivative is taken from the RSM, β_0 has no influence on the POE. The variation of the outcome is determined by the variance of the input factor U_i and the coefficients β_i, β_{ii}, and β_{ij}. High coefficients cause a high POE, and low coefficients cause a low POE.

The variance of the input factor U_i is determined by the processing unit variance before the current one. If the previous processing unit had a large POE, its variation of the input factor U_i will be high. This will increase the POE of the current processing unit.

Flexible operating units have high parameters and high variance of the input factor, thus linking flexible operating units will create a system with an high POE. Combining flexible operating units creates a flexible operating chain. For robust operating units the variance of the input factor and the coefficients are low, thus giving a low POE. Combining these operating units will create a robust operating chain.

This all can also be visualized with graphs. The first two process steps of the lipid biorefinery are taken as an example, harvesting and dewatering. These two first steps of two different operating chains are visualized in figure 3-6. For the harvesting step the operating units poly-glutamate flocculation and chitosan flocculation are taken, poly-glutamate flocculation has low parameters and chitosan flocculation has high parameter. For the dewatering step the operating unit centrifugation has been chosen.

Figure 2 and Figure 4 show the response of the processing unit poly-glutamate flocculation and chitosan flocculation on a varying concentration algae broth to the outflow of algae. Δy_1 depicts the range of outflow of algae from the processing unit poly-glutamate flocculation, Δy_2 is the range of outflow of algae from the processing unit chitosan flocculation. Figure 3 and Figure 5 show the response of the processing unit centrifugation to a varying inflow of algae to the outflow of algae.

For graphical presentation the flow of algae in the first process step is fixed at 5000 L h^{-1}. On the y-axis the coded values are given for the range, not the true values. The true values of the varying concentration algae in Figure 2 and Figure 4 is 0.5 – 1.5 g L^{-1}, and for the inflow of algae in Figure 3 and Figure 5 is 0.002 – 2.6 L h^{-1}.

The outflow of Figure 2 is the inflow of Figure 3 and the outflow of Figure 4 is the inflow of Figure 5. However Figure 3 and Figure 5 depict all the possible inflow for that processing unit. The processing unit in harvesting limits the possible range of the inflow for centrifugation. In Figure 3 Δy_3 depicts the range of outflow of the processing unit centrifugation if poly-glutamate flocculation is the previous processing unit. It is seen that the range is low. In Figure 5 Δy_4 is the range of outflow of the processing unit centrifugation if chitosan flocculation is the previous processing unit. It is seen that the range is high.

Figure 3-6 are indicating that choosing a robust processing units greatly influences the next operating unit. The same process can be done to see that flexible processing unit have influence on the next processing unit.

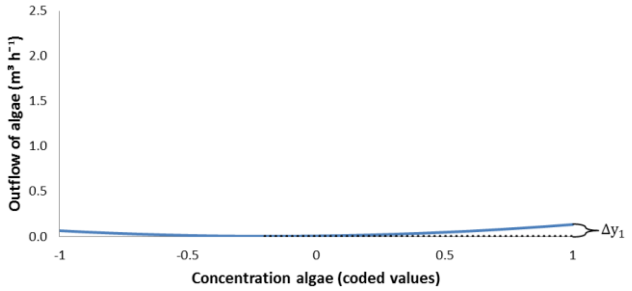

Figure 2 Response of poly-glutamate flocculation on varying concentration algae broth, where Δy_1 indicate the range of the outflow.

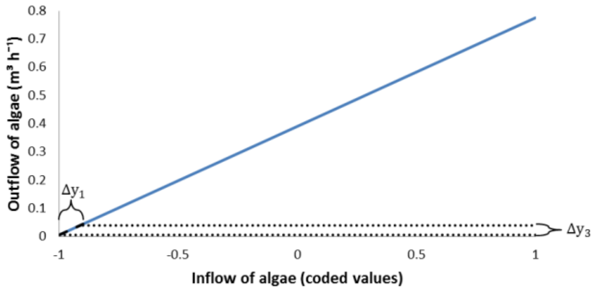

Figure 3 Response of centrifugation on varying inflow of algae broth. Δy_1 indicate the range of outflow of the processing unit poly-glutamate flocculation and Δy_3 the range of centrifugation with poly-glutamate flocculation as the previous processing unit.

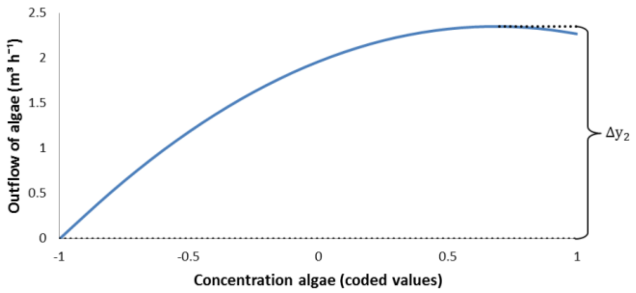

Figure 4 Response of chitosan flocculation on varying concentration algae broth, where Δy_2 indicate the range of the outflow.

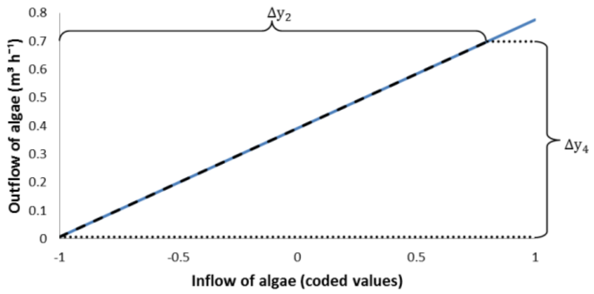

Figure 5 Response of centrifugation on varying inflow of algae broth. Δy_2 indicate the range of outflow of processing unit chitosan flocculation and Δy_4 the range of centrifugation with chitosan flocculation as the previous processing unit.

For energy usage the output of the processing unit is not the input for the next processing unit. Energy usage is added up and there are no interactions between the operating units. Therefore only the range of the energy usage of a processing unit is being investigated. Each processing unit has a range for its energy usage. These ranges of energy usage are added up, where a large range of the total energy usage of the operating chain is created if the processing unit have large ranges. If the range of the operating chain is large, the system would be classified as flexible. The large ranges of operating units are caused by high parameters, which are classified as flexible operating units. Thus linking operating units with flexible energy usage creates a flexible operating chain in energy usage. The same trend as for flexible system is for robust operating chains. Adding operating units that have a small range in energy usage will yield an operating chain that will have a small range in energy usage. Small ranges are indicated with low parameters, which are classified as robust operating units.

2.7 Pairing

For flexible systems control over the output by configuring the control factors is desired. The system has multiple outputs, interaction between the outputs occur. Interaction can be reduced by knowing which control factor to use for controlling one output while not altering the other outputs.

The RSM shows how much influence a changing condition has on the outcome. This information can be used to reach the desired output with the right configuration of the RSM. However the RSM does not hold information about how it influences the other outputs. The relative gain array (RGA) holds information about which control factor influence which output while not altering the other outputs. Consequently the RGA indicates which control factor is preferred to influence an output with regards to the other control factors. From this RGA a controllable system can be created.

The RGA calculations will be done with the RSM. Through rescaling it is more apparent which control factor has a higher influence on the system, since the control factors have different scale of size. Each element in the matrix indicates how the control factor can influence the output.

The RGA only works for linear time-invariant nxn MIMO systems. The flexible systems described in this study consist of multivariable, non-linear and non-square (nxm) systems. Chang and Yu (1990) describes the calculations of the multivariable non-square relative gain array (NRG). More information can be found in appendix F. For the calculation the system is linearized.

3. Results

3.1 Processing units

From the coefficients of the RSM a classification can be made about the flexibility or robustness of a processing unit. The coefficients from the response surface methodology can be found in appendix B and C. Classification of the processing units can be found in Table 3. An example for this classification is being followed in the next paragraph.

Example 1: The RSM with respect to robustness of centrifugation and pressure filtration in harvesting are given below:

Centrifugation:

$$\mathrm{Flow_{algae}} = 2.38 + 0.238 \cdot \mathrm{U_1} - 1.20 \cdot 10^{-15} \cdot \mathrm{U_1^2} + 1.37 \cdot 10^{-16} \cdot \mathrm{U_2} - 7.03 \cdot 10^{-16} \cdot \mathrm{U_2^2} - 5.27 \cdot 10^{-17} \cdot \mathrm{U_1U_2}$$

$$\mathrm{Energy\ usage} = 5.10 \cdot 10^{3} + 499 \cdot \mathrm{U_1} + -0.0012 \cdot \mathrm{U_1^2} + 880 \cdot \mathrm{U_2} - 237 \cdot \mathrm{U_2^2} + 88.0 \cdot \mathrm{U_1U_2}$$

Pressure filtration:

$$\mathrm{Flow_{algae}} = 2.38 + 0.238 \cdot \mathrm{U_1} - 1.20 \cdot 10^{-15} \cdot \mathrm{U_1^2} + 1.37 \cdot 10^{-16} \cdot \mathrm{U_2} - 7.03 \cdot 10^{-16} \cdot \mathrm{U_2^2} - 5.27 \cdot 10^{-17} \cdot \mathrm{U_1U_2}$$

$$\mathrm{Energy\ usage} = 2.78 \cdot 10^{3} + 278 \cdot \mathrm{U_1} + 1.19 \cdot 10^{-4} \cdot \mathrm{U_1^2} + 1.94 \cdot 10^{-5} \cdot \mathrm{U_2} + 9.07 \cdot 10^{-7} \cdot \mathrm{U_2^2} + 3.86 \cdot 10^{-6} \cdot \mathrm{U_1U_2}$$

With U_1 = Flow in (m^3 h^{-1}), and U_2 = Concentration algae (kg m^{-3}) both encoded.

Pressure filtration and centrifugation have the same values from the RSM for the outflow but the higher coefficients for the energy usage in the centrifugation unit make the pressure filtration more robust than centrifugation.

Example 2: The RSM with respect to flexibility of chitosan flocculation and poly-glutamate flocculation in harvesting are given below:

Chitosan flocculation:

$$\mathrm{Flow_{algae}} = 0.281 - 0.405 \cdot \mathrm{U_1} + 0.343 \cdot 10^{-15} \cdot \mathrm{U_1^2} - 0.438 \cdot \mathrm{U_2} - 0.0268 \cdot \mathrm{U_2^2} + 0.403 \cdot \mathrm{U_1U_2}$$

$$\mathrm{Energy\ usage} = 2.50 \cdot 10^{3} + 0.0103 \cdot \mathrm{U_1} - 0.0085 \cdot \mathrm{U_1^2} + 0.0120 \cdot \mathrm{U_2} + 0.0010 \cdot \mathrm{U_2^2} - 0.0101 \cdot \mathrm{U_1U_2}$$

With U_1 = concentration factor (-), and U_2 = chitosan concentration (g m^{-3}) both encoded.

Poly-glutamate flocculation:

$$\mathrm{Flow_{algae}} = 0.093 - 4.92 \cdot 10^{-8} \cdot \mathrm{U_1} - 6.91 \cdot 10^{-14} \cdot \mathrm{U_1^2}$$

$$\mathrm{Energy\ usage} = 600 + 2.37 \cdot 10^{-10} \cdot \mathrm{U_1} + 2.85 \cdot 10^{-26} \cdot \mathrm{U_1^2}$$

With U_1 = poly-glutamate concentration (g m^{-3}) encoded.

The coefficient of ploy-glutamate flocculation are lower, and thus chitosan flocculation is more flexible.

Table 3 Classification of processing units on its robustness or flexibility

Process step	Processing unit	Robust	Flexible
Harvesting	Centrifugation	+/-	+/-
	Pressure filtration	+	+/-
	Vacuum filtration	+	+/-
	Ultrasound	+	
	Chitosan flocculation	-	+
	Poly-glutamate flocculation	-	-
Dewatering	Centrifugation	-	
	Pressure filtration	+/-	
	Vacuum filtration	-	
Drying	Drying	+/-	+
Disruption	Bead milling	+/-	+/-
Extraction	Hexane	+/-	-
	Supercritical CO_2	+/-	+
Conversion	Acidic	+	-
	Alkaline	+/-	-
	Enzymatic direct conversion	+/-	-
	Microwave assisted dry conversion	-	+/-
	Supercritical methanol wet conversion	-	+/-

The RSM shows which control factors have influence on the process. Ultrasound shows no flexibility since the model for the processing unit does not have any control factors. For dewatering there is no flexibility, the value of the control factor is determined by the concentration of the flow in. Bead filling percentage has only influence on the mass of lipids, not on energy consumption or flow. The flow of methanol only has influence on the flow of biodiesel for enzymatic direct conversion, microwave assisted dry conversion, and supercritical methanol wet conversion. It does have influence on the energy consumption for all the processing units in conversion.

3.2 Flexible and robust operating chain

For the creating of robust or flexible operating chains Table 3 is used. Linking robust processing units together creates a robust processing chain, the same goes for flexibility as stated in 2.6 Linking processing units.

Poly-glutamate flocculation will not be included in any of the following operating chains, consequently the concentration of the algae in the operating chains will be altered slightly. Since the processing unit poly-glutamate flocculation could not handle a concentration exceeding 1.5 g L^{-1}, the new concentration of algae for robustness will have a range of 1.8 – 2.2 g L^{-1} and for flexibility it will be 2.0 g L^{-1}. This is caused by Slegers et al. (2014) who reported the concentration of 2.0 g L^{-1} as feed concentration.

3.2.1 Robust

Figure 6 depicts the most robust operating chain which is created by linking the most robust operating units. The operating chain consist of five operating units and has two control factors, bead filling percentage (%) in the processing unit bead milling and flow of methanol (L methanol kg^{-1} lipids) in the processing unit acidic conversion. The range of the in- and outflow is given.

Figure 6 Most robust operating system with the corresponding in- and outflows and the possible configurations for the control factors.

Table 4 depicts the result of the RSM for flow of biodiesel. U_1 is the flow of algae (m^3 h^{-1}) and U_2 the concentration of algae (g L^{-1}).

Table 4 RSM results for flow of biodiesel from the operating chain in Figure 6.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Flow of biodiesel (L h^{-1})	1.23	0.135	-0.0010	0.135	-0.0010	0.0175	0.994	0.994
***p*-value**	*0*	*0*	*9.95e-14*	*0*	*7.29e-14*	*0*		

Table 4 indicates that all coefficients have statistical significant, all p-values are below 0.05. Both U_1 and U_2 have a significant influence on the output. R^2 and adjusted R^2 confirm that the RSM is a good fit.

Table 5 RSM results for NER from the operating chain in Figure 7.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
NER (-)	0.344	0.0135	-8.33e-04	0.0219	-8.50e-04	0.0020	0.975	0.975
***p*-value**	*0*	*0*	*3.70e-99*	*0*	*3.65e-103*	*0*		

Table 5 depicts the results of the RSM for NER. It shows that all coefficient have statistical significant, all p-values are below 0.05. Both U_1 and U_2 have a significant influence on the output. R^2 and adjusted R^2 confirm that the RSM is a good fit.

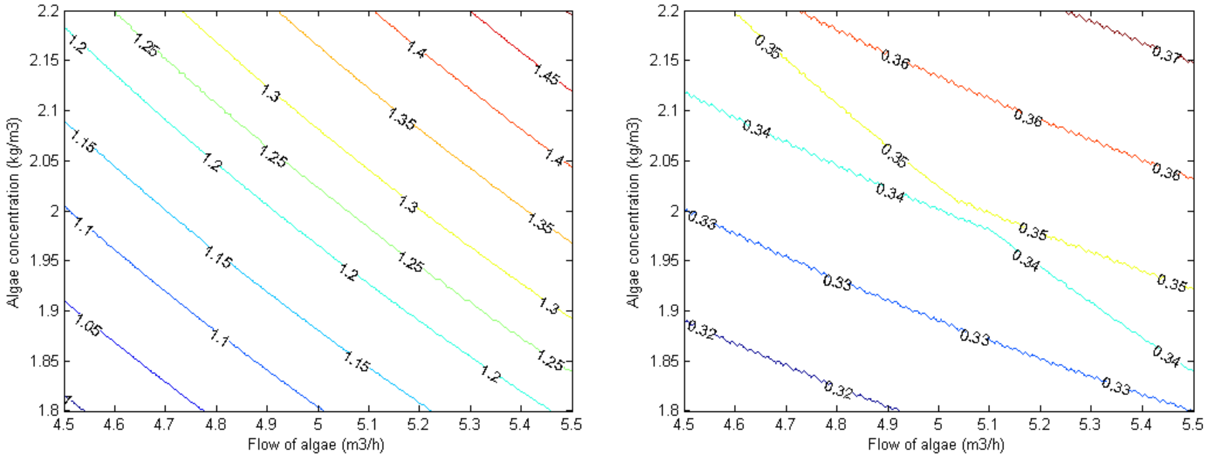

Figure 7 Contour plot of algae concentration (kg m^{-3}) and flow of algae (m^3 h^{-1}) against flow of biodiesel(L h^{-1}) and NER(-) respectively.

Figure 7 depicts a plot figure of the algae concentration (kg m^{-3}) and flow of algae (m^3 h^{-1}) against flow of biodiesel (L h^{-1}) and NER (-). The right plot figure shows a small change in the middle of the plot. The flow of biodiesel flow increases faster than normal and for the NER a small offset is shown. The cause lies in the processing unit bead-milling. For the excess flow of lipids an extra bead milling unit is needed. This extra unit uses extra energy and does increase the flow of biodiesel since the efficiency of the bead milling is increased if the flow is lowered.

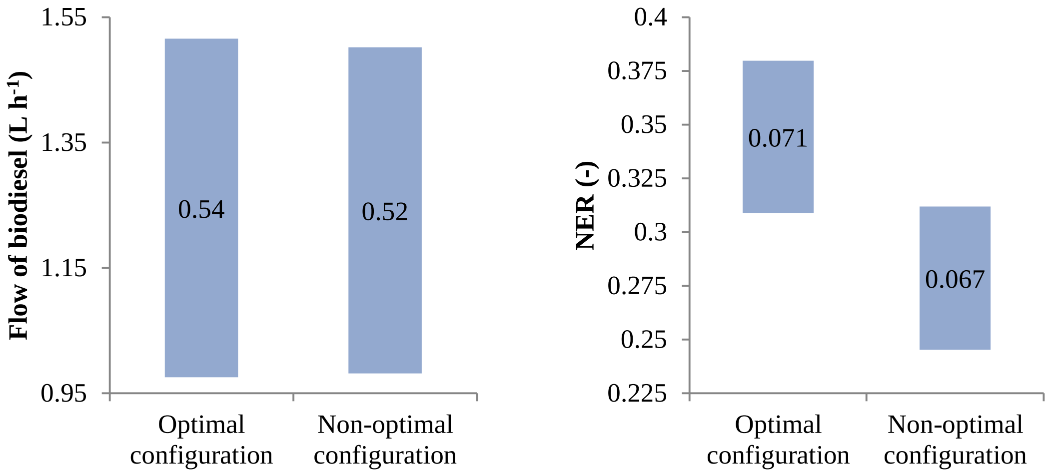

Figure 8 Range of the output with optimal and non-optimal configurations.

In section 1 it was discussed that the robust operating chains will be analysed on two different operating points. Figure 8 depicts the influence of these two operating points, optimal configuration against non-optimal configuration. The range of the outputs are given in the figure. The range for the output is decreased using non-optimal configuration increasing the robustness of the system. Using non-optimal configurations lowers the value of the NER.

3.2.2 Flexible

Figure 9 depicts the most flexible operating system which is created by linking the most flexible operating units. It consists of three operating units and has four control factors. From this operating chain a RSM is created in which U are the control factors. U_1 is the concentration factor of chitosan flocculation(-), U_2 is the chitosan concentration (kg m^{-3}), U_3 the temperature of supercritical methanol wet conversion(K), and U_4 the flow of methanol(L methanol kg^{-1} lipids). The range of the outflow is given.

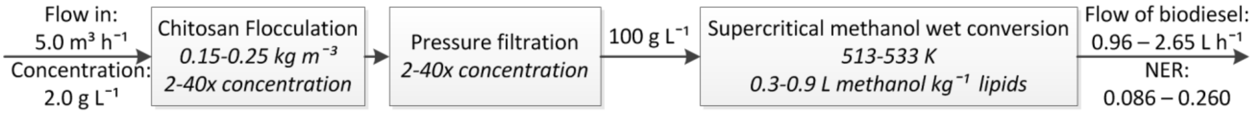

Figure 9 Most flexible operating system with the corresponding in- and outflows and the possible configurations for the control factors.

Table 6 depicts the result of the RSM for flow of biodiesel.

Table 6 RSM results for flow of biodiesel from the operating chain in Figure 10.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_3	β_{33}	β_4	β_{44}
Flow of biodiesel (L h^{-1})	2.11	-1.33e-15	3.20e-16	0.0438	-0.0700	-0.365	0.138	-0.0156	-0.596
***p*-value**	0	1	1	1.10e-197	3.89e-186	0	0	3.33e-49	0
	β_{12}	β_{13}	β_{14}	β_{23}	β_{24}	β_{34}	R^2		Adjusted R^2
Flow of biodiesel (L h^{-1})	6.46e-16	1.87e-15	1.30e-15	-8.7e-03	-3.71e-04	-7.38e-03	0.998		0.998
***p*-value**	1	1	1	4.47e-10	0.786	1.11e-234			

Table 6 concludes that U_1 is not statistical significant to the *p*-value. Any interaction with U_1 is non-significant, all interaction terms (β_{1X}) show a *p*-value above 0.05. All other control factors are statistical significant on the outflow of biodiesel except for the interaction coefficient between U_2 and U_4, this coefficient has no statistical significant. R^2 and adjusted R^2 confirm that the RSM is a good fit.

Table 7 RSM results for the NER from the operating chain in Figure 9.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_3	β_{33}	β_4	β_{44}
NER(-)	0.157	6.21e-04	-7.36e-04	0.0019	-0.0030	-0.0725	0.0317	-0.0044	-0.0460
***p*-value**	0	0.0342	0.137	1.45e-10	1.50e-09	0	2.66e-273	3.00e-44	0
	β_{12}	β_{13}	β_{14}	β_{23}	β_{24}	β_{34}	R^2		Adjusted R^2
NER(-)	1.53e-05	-4.74e-04	-3.17e-05	-4.95e-04	-5.07e-05	-0.0042	0.993		0.993
***p*-value**	0.970	0.252	0.939	0.232	0.902	2.75e-22			

Table 7 depicts the result of the RSM for NER. U_1 only shows a linear relation with the output, the second order term β_{11} shows no influence on the output. All other coefficients are statistical significant except for the interaction coefficients. The interaction coefficients β_{34} is statistical significant. The control factors related to β_{34} are from the same processing unit, as a result they influence each other. U_1 and U_2 are also from the same processing unit however U_1 has a small influence on the output, consequently β_{12} even less. This can be further seen in Figure 10. R^2 and adjusted R^2 confirm that the RSM is a good fit.

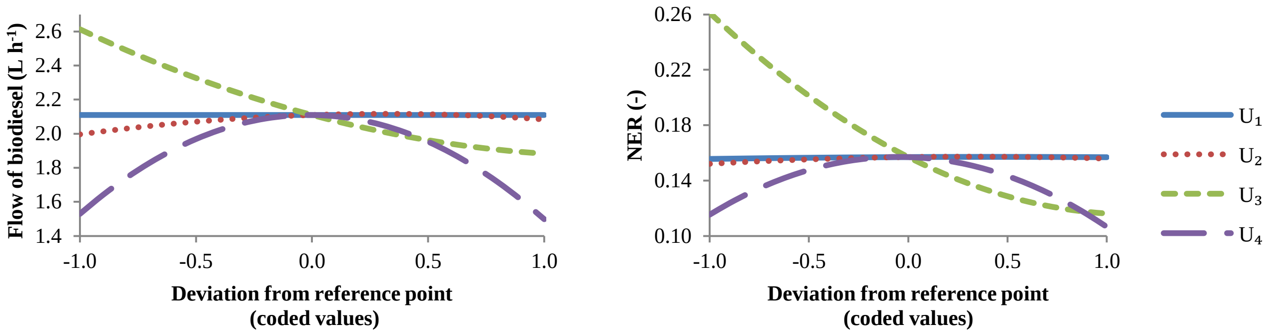

Figure 10 Perturbation plot of flow of biodiesel (L h^{-1}) and NER(-) for the operating chain given in Figure 9.

The perturbation plot shows the change of one control factor on the output while keeping the other control factors at zero. The perturbation plot shows which control factor has an influence on the output. The type of response of the control factor on the output is also visualized. Figure 10 depicts that U_1 and U_2 show little influence on the output with regards to U_3 and U_4.

3.3 Operating chains from Slegers et al. (2014)

In Slegers et al. (2014) a list of operating chains are given. Included are those with the highest NER and highest flow of biodiesel. In this study these two are analysed to get more insight on their flexibility and robustness.

3.3.1 Highest NER

3.3.1.1 Robust

The operating chain with the highest NER is identical to the one discussed in section 3.2.1. There it was analysed on its robustness but not on its flexibility. The following section will analyse the operating chain on its flexibility.

3.3.1.2 Flexible

Figure 11 depicts the operating chain with the highest NER according to Slegers et al. (2014). It consists of five operating units and has two control factors. From this operating chain a RSM is created in which U are the control factors. U_1 is the bead filling percentage(%) from the processing unit bead milling and U_2 the flow of methanol (L methanol kg^{-1} lipids) of the process unit acidic conversion. The range of the outflow for flexibility is given.

Figure 11 Operating chain with the highest NER according to Slegers et al.(2014). Given are the corresponding in- and outflows for flexibility and the possible configurations for the control factors.

Table 8 depicts the result of the RSM for flow of biodiesel.

Table 8 RSM results for flow of biodiesel from the operating chain in Figure 11.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Flow of biodiesel (L h^{-1})	1.21	0.0117	-6.75e-04	2.23e-17	-5.07e-16	-6.02e-17	1.00	1.00
***p*-value**	2.68e-91	1.05e-55	7.08e-28	1	1	1		

Table 8 concludes that only U_1 is statistical significant. Reason is that the flow of methanol in acidic conversion has no influence on the flow of biodiesel as stated in 3.1 Processing units. R^2 and adjusted R^2 confirms that the RSM is a good fit.

Table 9 depicts the results of the RSM for NER.

Table 9 RSM results for NER from the operating chain in Figure 12.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
NER (-)	0.344	0.0135	-8.33e-04	0.0219	-8.50e-04	0.0020	0.975	0.975
***p*-value**	*0*	*0*	*3.70e-99*	*0*	*3.65e-103*	*0*		

Table 9 concludes that both U_1 and U_2 are statistical significant. R^2 and adjusted R^2 confirms that the RSM is a good fit.

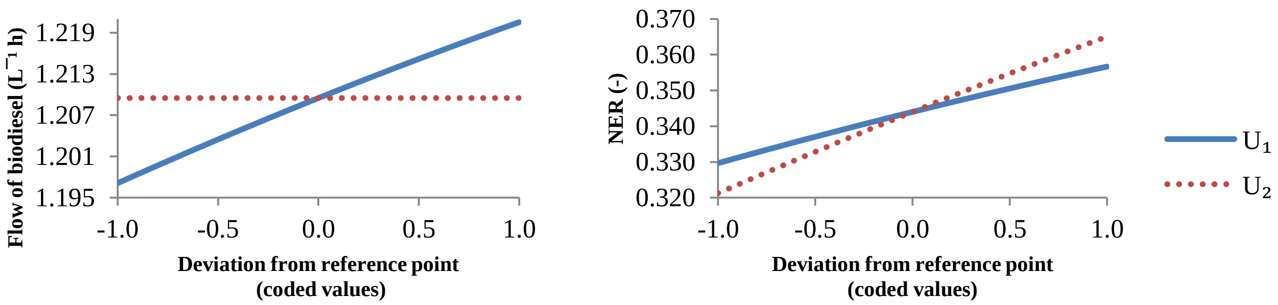

Figure 12 Perturbation plot of flow of biodiesel (L h^{-1}) and NER(-) for the operating chain given in Figure 11.

The perturbation plots in Figure 12 show a linear relation between U_1 and U_2 with the output. The RSM from Table 8 and Table 9 indicates a non-linear relation, the *p*-values indicate that second order parameters, β_{11} and β_{22}, have significant influence on the output. But value β_{11} and β_{22} are too low to depict a non-linear relationship in the perturbation plot. The perturbation plot of flow of biodiesel indicates that U_1 has no significant influence on the output, previous stated by the RSM.

3.3.2 Flow of biodiesel

3.3.2.1 Robust

Figure 16 shows the optimal operating chain for the maximum flow of biodiesel with the in- and outflows for robustness.

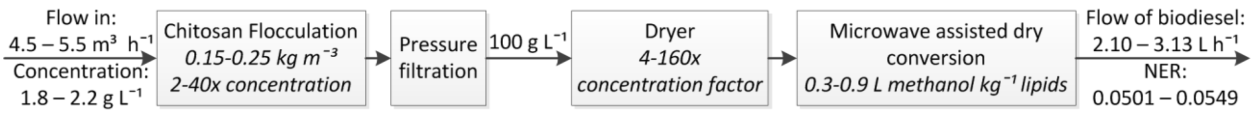

Figure 13 Operating chain with the highest flow of biodiesel according to Slegers et al.(2014). Given are the in- and outflow for robustness.

Table 10 depicts the results of the RSM for flow of biodiesel.

Table 10 RSM results for flow of biodiesel from the operating chain in Figure 13.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Flow of biodiesel (L h^{-1})	2.59	0.259	1.68e-05	0.259	1.72e-05	0.0259	1.00	1.00
***p*-value**	8.27e-115	1.59e-98	8.47e-15	1.59e-98	5.93e-15	1.13e-76		

Table 10 indicates that all coefficients are statistical significant. Both U_1 and U_2 have a significant influence on the flow of biodiesel. R^2 and adjusted R^2 confirm that the RSM is a good fit.

Table 11 shows the results of the RSM for NER.

Table 11 RSM results for NER from the operating chain in Figure 16.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
NER (-)	0.0527	0.0012	-9.14e-05	0.0012	-9.21e-05	-6.41e-05	1.00	1.00
***p*-value**	3.89e-78	1.74e-49	4.48e-24	1.42e-49	3.93e-24	1.26e-22		

Table 11 indicates that all coefficients are statistical significant. U_1 and U_2 have a significant influence on the flow of biodiesel. R^2 and adjusted R^2 ensure that the RSM is a good fit.

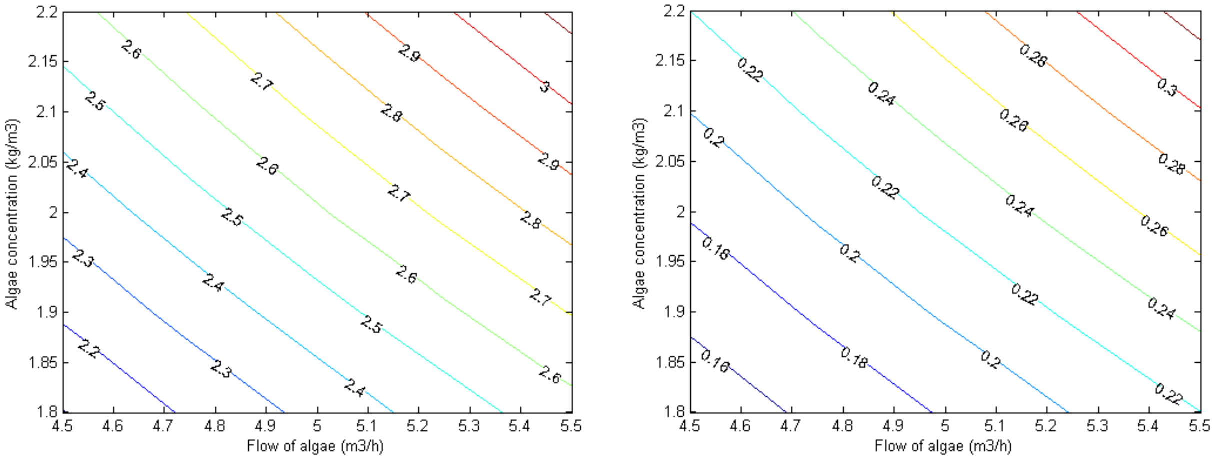

Figure 14 Contour plot of algae concentration (kg m^{-3}) and flow of algae (m^{3} h^{-1}) against flow of biodiesel(L h^{-1}) and NER(-) respectively.

Figure 14 show that both flow of biodiesel and NER show a slight non-linear relation with algae concentration and flow. Increasing both the concentration and algae flow increases both the flow of biodiesel and the NER.

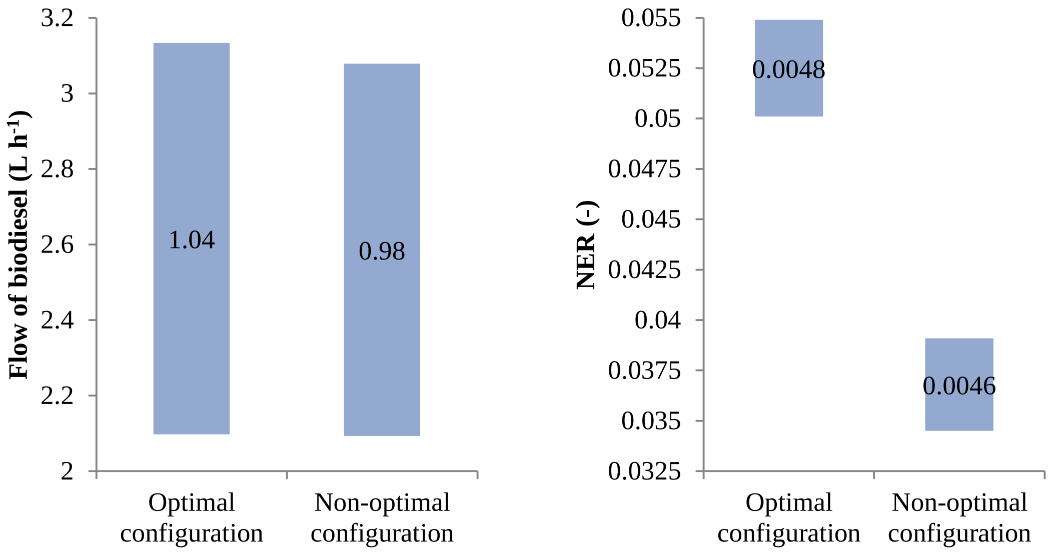

Figure 15 Minimum and maximum values of the output with optimal and non-optimal configurations and their given range.

Figure 15 depicts the influence of using optimal configuration against non-optimal configuration. The range of the outputs are given in the figure. The range for the output is decreased using non-optimal configuration increasing the robustness of the system. Using non-optimal configurations lowers the value of the NER.

3.3.2.2 Flexible

Figure 16 depicts the optimal operating chain for the maximum flow of biodiesel with the corresponding input and output if the system is analysed on its flexibility. It consists of four operating units and has two control factors. From this operating chain a RSM is created in which U are the control factors. U_1 is the concentration factor(-), U_2 the chitosan concentration(kg m^{-3}), U_3 the concentration factor of drying(-), and U_4 the flow of methanol(L methanol kg^{-1} lipids). The range of outflow for flexibility is given.

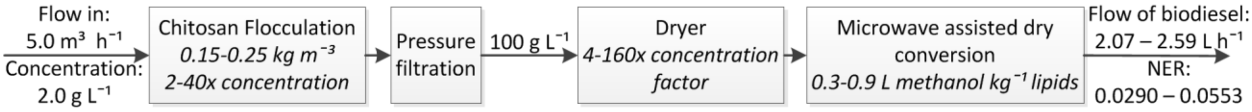

Figure 16 Operating chain with the highest flow of biodiesel according to Slegers et al.(2014). Given are the in- and outflow for flexibility.

Table 12 depicts the result of the RSM for flow of biodiesel.

Table 12 RSM results for flow of biodiesel from the operating chain in Figure 16.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_3	β_{33}	β_4	β_{44}
Flow of biodiesel (L h^{-1})	2.58	-2.54e-15	4.90e-16	0.0601	-0.0960	-9.15e-16	1.66e-15	-5.13e-04	2.74e-04
***p*-value**	0	1	1	0	0	1	1	0	0
	β_{12}	β_{13}	β_{14}	β_{23}	β_{24}	β_{34}	R^2	Adjusted R^2	
Flow of biodiesel (L h^{-1})	7.53e-16	1.97e-15	1.60e-15	1.71e-15	-2.40e-05	1.78e-15	1.00	1.00	
***p*-value**	1	1	1	1	5.18e-18	1			

U_2 and U_4 and their interaction parameters are statistical significant. U_1 and U_3 are not statistical significant, these control factors have no influence on the amount of lipids or the conversion of lipids into biodiesel. R^2 and adjusted R^2 ensure that the RSM is a good fit.

Table 13 shows the result of the RSM for NER.

Table 13 RSM results from the NER from the operating chain in Figure 16.

Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_3	β_{33}	β_4	β_{44}
NER(-)	0.0365	3.03e-05	-3.60e-05	2.88e-04	-4.58e-04	-0.0027	0.0035	-0.0063	0.0010
***p*-value**	0	0.5885	0.7039	3.50e-07	1.64e-06	6.57e-209	1.47e-159	0	2.68e-25
	β_{12}	β_{13}	β_{14}	β_{23}	β_{24}	β_{34}	R^2	Adjusted R^2	
NER(-)	4.39e-07	-4.44e-06	-9.89e-06	-4.01e-05	5.59e-05	9.07e-04	0.944	0.944	
***p*-value**	0.996	0.955	0.901	0.612	0.480	1.06e-27			

All the control factors are statistical significant on the NER except for U_1. Only one interaction coefficient shows significant influence on the outcome, the interaction coefficient between U_3 and U_4. β_{34} shows significant influence because the flow of methanol is related to the flow of the algae broth for the processing unit, with the processing unit dryer this flow can be reduced by increasing the concentration factor.

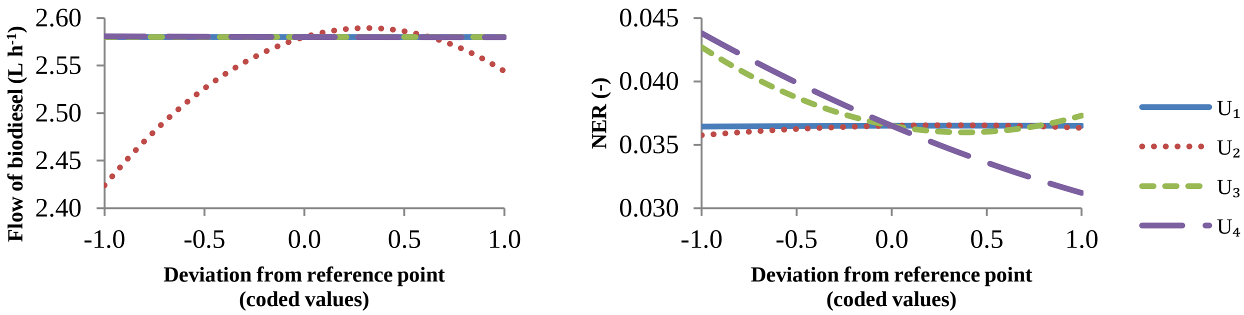

Figure 17 Perturbation plot of flow of biodiesel (L h^{-1}) and NER(-) for the operating chain given in Figure 16.

The perturbation plot in Figure 17 shows that for flow of biodiesel only U_2 has any influence on the output in contradiction to the RSM which noted that both U_2 and U_4 have significant influence on the output. From the perturbation plot of the NER it is seen that U_1 shows no influence on the output and U_2 a minimal influence.

3.4 Pairing

The relative gain array (RGA) can be used for pairing control factors with outputs. Only for the flexible systems a relative gain can be created since in these systems control factors have been used to influence the output. In robust systems the control factors are always set to optimal or fixed configurations. RGA is a method to investigate the possibility for input output pairing. The concept is designed for systems with an equal number of inputs as outputs variables. Since there are more control factors than outputs a non-square relative gain is needed.

In the relative gain the first column is for the first output, flow of biodiesel, and the second column for the second output, the NER. The rows indicate the different control factors for the operating chain. Each element in the matrix indicates how the control factor can influence the output of its column. For example in Matrix 1 in the first column and second row the value is 0.634, which is the highest number of this column. To influence the flow of biodiesel the second control factor should be used. The last row of the matrix are both low values, indicating that the control factor has low influence on both of the outputs. It is preferred to have only one large value per row so that a control factor influences one output and interaction is minimal.

The relative gain indicates which control factor is relatively better to control the output. The relative gain of a control factor is a percentage of the total control that can be reached on the output. All the individuals values of the relative gain for one output should be equal to one.

Matrix 1 depicts the non-square relative gain for the operating chain of Figure 9. This operating chain is the most flexible operating chain.

$$\begin{bmatrix} -1.8 \cdot 10^{-13} & 0.909 \\ 0.634 & 0.0156 \\ 0.366 & 0.0749 \\ 3.51 \cdot 10^{-4} & 2.11 \cdot 10^{-4} \end{bmatrix}$$

Matrix 1 Non-square relative gain for the operating chain of Figure 9.

U_1 , the concentration factor for harvesting, shows large influence on the NER. U_2 , chitosan concentration, and U_3, temperature supercritical methanol wet conversion, influence the flow of biodiesel. U_4, the flow of methanol, shows minimal influence on both outputs.

Matrix 2 depicts the square relative gain for the operating chain given for Figure 11. This operating chain has the highest NER according to Slegers et al. (2014).

$$\begin{bmatrix} 1.0 & 0 \\ 0 & 1.0 \end{bmatrix}$$

Matrix 2 Square relative gain for the operating chain of Figure 11

U_1, the bead milling percentage, only influences the flow of biodiesel. U_2, the flow of methanol, only influences the NER.

Matrix 3 depicts the non-square relative gain for the operating chain for Figure 16. This operating chain has the highest flow of biodiesel according to Slegers et al. (2014).

$$\begin{bmatrix} 5.41 \cdot 10^{-11} & 0.879 \\ 0.998 & 8.89 \cdot 10^{-4} \\ 5.93 \cdot 10^{-14} & 0.105 \\ 0.00239 & 0.0159 \end{bmatrix}$$

Matrix 3 Non-square relative gain for the operating chain of Figure 16

U_2, the chitosan concentration, shows a high control on the flow of biodiesel. For the NER U_1, the concentration factor for harvesting, indicates a high control. U_3, the concentration factor of drying, could be used to control the NER. U_4, the flow of methanol, shows minimal influence on both the outputs.

4. Discussion

In this study the different models provided by Slegers et al. (2014) were analysed. Using these models some limitation were experienced. These are discussed in section 4.1. Using the RSM for testing system on their robustness or flexibility has not yet been performed. Perspectives for future use of this method are also discussed in 4.2.

4.1 Models

The level of detail provided from the models differ. Some models were white box models while others were grey. To make better conclusions about the operating chains, the models have to be reconsidered.

The algae concentration in the feed flow was in the range of 0.5 to 1.5 g L^{-1}. The choice for the range algal concentration is due to the model of poly-glutamate flocculation. Exceeding this range makes the model unreliable which can also be seen in Figure 18, where the model becomes unstable around 2 g L^{-1}. However as this processing unit is not used in any of the operating chain as stated in section 3.2, it was chosen to increase the concentration of algae to the values as given by Slegers et al. (2014).

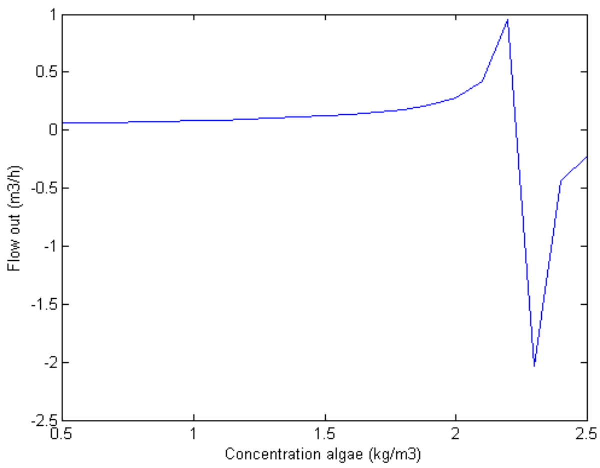

Figure 18 Flow out(m^3 h^{-1}) against concentration algae (kg m^{-3}) for poly-glutamate flocculation

One of the criticism with this frame work is using the same unit operation like centrifugation in both harvesting and dewatering step for one operating chain. In reality the desired concentration can be reached after one step of centrifugation which eliminate the need application of second centrifugation unit for dewatering step.

The correlation coefficient in bead milling models gives another reason to discuss the models. The correlation coefficient in the case of bead milling unit model is low which means not a good fit. There are some reasons to be discussed for this low correlation coefficient. First of all the main model reported by Doucha and Lívanský (2008) that bead milling model in this study has been originated from shows a low correlation coefficient of 0.862. Moreover as it can be seen in Figure 19, there are little spikes on a fixed interval. These spikes are the consequence of exceeding the maximum fixed value for flow of lipids per hour in bead milling unit. In this case, exceeding the maximum value each time creates an extra bead mill unit, therefore splitting the flow of algae over the extra unit. Therefore the efficiency of the bead mill units rises, causing spikes in the graph. Due to these spikes a good fit is hard to compute, since the RSM is always a smooth line.

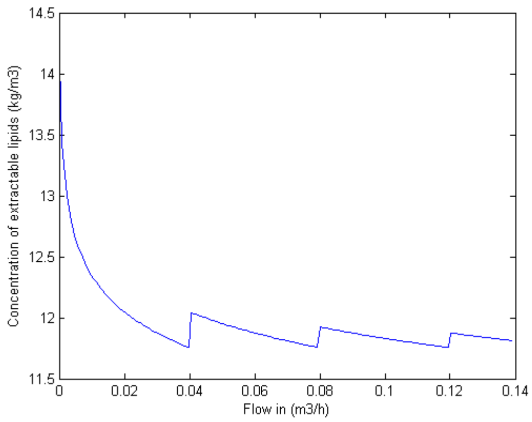

Figure 19 Concentration of extractible lipids (kg m $^{-3}$) against flow in (m^3 h^{-1}) for bead milling

Another issue regarding to the models is that processing units with as control factor the concentration factor show low correlation coefficient. Figure 20 depicts the flow out against the concentration factor, it illustrates a negative exponent function. The RSM consist of a second order model. A good fit with a second order model for a negative exponent function is hard to compute, resulting in a low correlation coefficient.

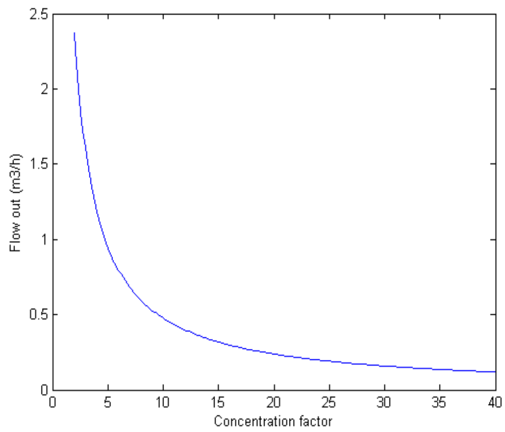

Figure 20 Flow out (kg m-3) against concentration factor for the processing unit dryer

It should be mentioned that assumptions were made as no real data was available about the variation in the algae broth. For the analysis of the processing units and operating chain on their robustness the minimum and maximum flow was set at 4500 L h^{-1} and 5500 L h^{-1} and for concentration to 1.8 and 2.2 g L^{-1}.

After the process step dewatering and harvesting the concentration of algae is fixed at 100 g L^{-1}. This limits the flexibility of the processing units in dewatering and harvesting, since they are configured to reach this concentration. It is seen from the RSM that the control factors in dewatering and harvesting show little influence on the end-product. Removing this limitation might increase the flexibility of the process.

4.2 Perspectives

The approach of this work was to demonstrate the usage of RSM to analyse systems on their robustness and flexibility. The benefit the RSM offers is the easy and quick insight into the system. This insight can be used to compute certain aspects of the system, like controllers and configurations.

The current operating chain only produces one product, however to increase the economic feasibility of algae, multiple products need to be extracted from the algae broth. Each product has different prices and analysing trade-offs between products can be performed.

Adding more products might alter the use of flexibility in this study, since here it is depicted as the range of the outflow. With more end-products more market demands can be fulfilled. To fulfil the different trends within these market demands, quick adaptions to the production chain needs to performed. In that case, flexibility would be about the adaption of the operating chain to fulfil new market demands.

Assumptions regarding the control factors are made. It is believed that control factors can be easily set to new configurations. However, some control factors might be difficult to configure. For example, the processing unit bead milling has as control factor bead filling, a percentage of beads in the unit. This percentage can be changed by adding or removing beads from the unit. The processing unit has to be shut down when changing the amount of beads. This may cause shutting down the whole operating chain. Therefore it might be more beneficial to use a processing unit that is easier to adapt.

More robustness or flexibility can be reached by including more control factors. A higher control can result in better performance.

In this study it was stated that performing below the optimum configurations could increase the robustness of the operating chain. This is examined by adjustment of the control factors and comparing the result with an operating chain with optimal configuration. In the current work focus was on creating robust operating chain, but the optimal configuration for the robust operating chain has not been examined.

For the robust operating chain compared to the operating chain with optimal biodiesel yield the output range of NER is larger. The main reason are trade-offs performance in the classifications of the processing units. Since most processing units have RSM's for multiple outputs, classification of a processing unit on its robustness or flexibility is not straightforward. In some cases RSM’s show contradicting information, where one RSM holds information that a system was flexible while the other RSM indicates contradictory results. The RSM’s with information about the algae broth and the process to convert the algal broth to biodiesel are preferred for the classification as opposed to the RSM about the energy usage of the processing unit.

5. Conclusion

In this work, a response surface methodology (RSM) based approach for the analysis of robustness or flexibility in lipid biorefineries is introduced. With the use of the RSM it is possible to analyse processing units and operating chains. The RSM provides information about which inputs or control factors are relevant for the process and the scope of influence.

The robust operating chain compared to the operating chain with optimal biodiesel yield has a smaller output range of biodiesel yield. Choosing robust operating units increase the robustness of the operating chain because of decreasing the influence of input on output. However for a robust operating chain the flow of biodiesel is substantially lower.

Choosing a non-optimal configuration decreases the range of output slightly as a result making the system more robust. However it lowers the value of the biodiesel flow and NER. A trade-off analysis between optimal output and optimum robustness was not done in this study.

It is observed that a flexible operating chain has a higher range, therefore a higher flexibility, compared to non-flexible operating chain.

Pairing of the outputs with the control factors is not straightforward. Net energy ration (NER) and flow of biodiesel are two output that show interaction, since the NER is partially derived from the flow of biodiesel. However the non-square relative gain (NRG) shows that it is possible to make choice which control factor has the best influence on the output. Using this information, more control over the system can be reached. Adding more final products will make the NRG more beneficial for control over the system since it is more complicated to make trade-offs from the information presented by the RSM without the NRG.

The RSM is successfully applied to create a more flexible or robust processing chain. Also more insight can be gained over the process, indicating which control factors have influence on the output. It can be concluded that with the RSM an effective framework for the analysis of a system on its robustness or flexibility can be created.

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Appendices

Appendix A. Processing units

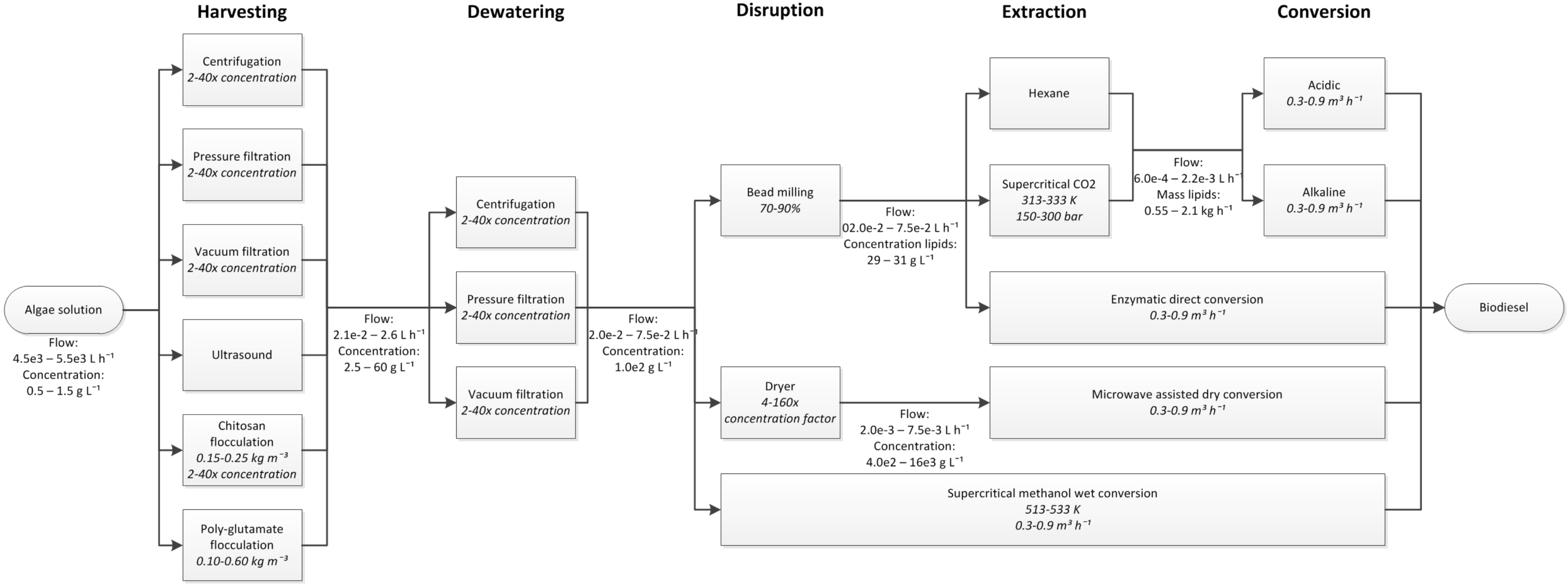

Figure 21 All the processing units and the possible configurations for an operating chain. Before each process step the minimum and maximum values are given.

Appendix B. Result processing units robustness

Table 14 Robustness of the processing units in harvesting with U_1 = Flow in ($m^3\,h^{-1}$), and U_2 = Concentration algae ($kg\,m^{-3}$) encoded

Harvesting	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Centrifugation	Flow of algae broth ($m^3\,h^{-1}$)	2.3750	0.2375	-1.1981e-15	1.3738e-16	-7.0276e-16	-5.2736e-17	1	1
	p-value	4.6871e-291	9.1446e-275	0.0053	0.5488	0.0803	0.8702		
	Energy usage (J)	5.1041e+03	498.5537	-0.0012	880.1164	-237.1690	88.0117	0.9993	0.9991
	p-value	5.9680e-42	1.7725e-25	0.9999	3.6880e-30	3.7755e-15	5.5295e-09		
Pressure filtration	Flow of algae broth ($m^3\,h^{-1}$)	2.3750	0.2375	-1.1981e-15	1.3738e-16	-7.0276e-16	-5.2736e-17	1	1
	p-value	4.6871e-291	9.1446e-275	0.0053	0.5488	0.0803	0.8702		
	Energy usage (J)	2.7778e+03	277.7801	1.1858e-04	1.9405e-05	9.0688e-07	3.8616e-06	1	1
	p-value	3.0502e-196	5.9505e-180	1.3486e-54	5.4273e-44	1.4950e-14	8.1182e-28		
Vacuum filtration	Flow of algae broth ($m^3\,h^{-1}$)	2.3750	0.2375	-1.1981e-15	1.3738e-16	-7.0276e-16	-5.2736e-17	1	1
	p-value	4.6871e-291	9.1446e-275	0.0053	0.5488	0.0803	0.8702		
	Energy usage (J)	6.1111e+03	611.1135	1.1858e-04	1.9405e-05	9.0688e-07	3.8616e-06	1	1
	p-value	9.5132e-203	1.8560e-186	1.3486e-54	5.4274e-44	1.4950e-14	8.1184e-28		
Ultrasound	Flow of algae broth ($m^3\,h^{-1}$)	0.1000	0.0100	-3.7004e-17	3.1402e-18	-2.2275e-17	-1.1935e-17	1	1
	p-value	1.4059e-295	2.7429e-279	7.7209e-04	0.5732	0.0265	0.1399		
	Energy usage (J)	8.3400e+03	840.0027	1.3272e-04	2.8999e-05	2.6156e-07	5.7805e-06	1	1
	p-value	1.5754e-204	2.6822e-188	9.6674e-55	1.6011e-46	2.8297e-05	2.3370e-30		
Chitosan flocculation	Flow of algae broth ($m^3\,h^{-1}$)	1.9603	0.1545	-6.1308e-16	1.1378	-0.8303	0.1138	0.9959	0.9948
	p-value	4.1587e-25	8.7062e-08	1	2.4599e-23	1.7270e-16	3.4341e-04		
	Energy usage (J)	2.5003e+03	50.0099	0.0014	-0.0024	0.0152	3.4855e-04	1.0000	1.0000
	p-value	7.5329e-102	2.7995e-72	0.6950	0.2726	5.1872e-04	0.9100		
Poly-glutamate flocculation	Flow of algae broth ($m^3\,h^{-1}$)	0.0924	0.0097	-3.8318e-17	0.0345	0.0084	0.0034	0.9985	0.9981
	p-value	3.3980e-33	2.3652e-17	1	8.8629e-28	4.6841e-12	4.5850e-07		
	Energy usage (J)	600.0194	50.0032	1.3417e-04	-1.2270e-04	-5.2248e-05	-2.2269e-05	1.0000	1.0000
	p-value	8.4161e-148	5.2426e-130	7.2558e-20	1.9655e-23	2.4912e-12	1.6988e-07		

Table 15 Robustness of the processing units in dewatering with U_1 = Flow in ($m^3 h^{-1}$), and U_2 = Concentration algae (kg m^{-3}) encoded

Dewatering	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Centrifugation	Flow of algae broth ($m^3 h^{-1}$)	0.3910	0.3846	-4.6740e-19	0.3597	-1.3646e-16	0.3538	1.0000	1.0000
	p-value	5.4591e-289	1.4540e-291	0.9954	5.1929e-291	0.1061	5.1332e-288		
	Energy usage (J)	2.8503e+03	2.5930e+03	0.0017	629.2376	-429.1152	619.0133	0.9936	0.9919
	p-value	5.4968e-20	6.7271e-22	1.0000	1.2352e-10	6.9935e-05	4.5546e-08		
Pressure filtration	Flow of algae broth ($m^3 h^{-1}$)	0.3910	0.3846	-4.6740e-19	0.3597	-1.3646e-16	0.3538	1.0000	1.0000
	p-value	5.4591e-289	1.4540e-291	0.9954	5.1929e-291	0.1061	5.1332e-288		
	Energy usage (J)	731.6405	719.7539	0.0017	0.0022	0.0012	0.0028	1.0000	1.0000
	p-value	4.4705e-112	1.1906e-114	2.6578e-05	1.7297e-10	0.0013	1.2263e-09		
Vacuum filtration	Flow of algae broth ($m^3 h^{-1}$)	0.3816	0.3754	-1.4900e-18	0.3691	-1.3548e-16	0.3631	1.0000	1.0000
	p-value	1.6400e-288	4.3681e-291	0.9859	6.0282e-291	0.1199	5.9588e-288		
	Energy usage (J)	1.6096e+03	1.5835e+03	0.0017	0.0022	0.0012	0.0028	1.0000	1.0000
	p-value	1.3942e-118	3.7134e-121	2.6578e-05	1.7297e-10	0.0013	1.2263e-09		

Table 16 Robustness of the processing unit drying in drying with U_1 = Flow in ($m^3 h^{-1}$) encoded

Drying	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Drying	Flow of algae broth ($m^3 h^{-1}$)	0.0047	0.0027	8.6367e-19	-	-	-	1	1
	p-value	1.8703e-31	4.7122e-31	0.8093	-	-	-		
	Concentration algae (kg m^{-3})	1.6000e+04	-1.0965e-12	-2.4956e-12	-	-	-	0	-1
	p-value	9.4166e-33	0.5179	0.4047	-	-	-		
	Energy usage (J)	2.0063e+05	1.1471e+05	1.3339e-07	-	-	-	1	1
	p-value	4.0464e-29	1.0195e-28	2.1532e-04	-	-	-		

Table 17 Robustness of the processing unit bead milling in disruption with U_1 = Flow in (m^3 h^{-1}) encoded

Disruption	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Bead milling	Flow of algae broth (m^3 h^{-1})	0.0696	0.0694	2.0205e-17	-	-	-	1	1
	p-value	5.5248e-31	4.5810e-31	0.8229	-	-	-		
	Concentration lipids (kg m^{-3})	11.6145	-0.9175	1.3124	-	-	-	0.8567	0.7135
	p-value	0.0011	0.1185	0.1551	-	-	-		
	Energy usage (J)	3.3000e+03	5.5317e-06	2.7564e-06	-	-	-	1.0000	1.0000
	p-value	1.3835e-32	4.0548e-15	4.6658e-14	-	-	-		

Table 18 Robustness of processing units in extraction with U_1 = flow of algae(m^3 h^{-1}), and U_2 = concentration lipids (L h^{-1}) encoded

Extraction	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Hexane	Mass lipids (kg h^{-1})	1.2928	0.7391	5.2404e-17	0.0198	-4.2711e-16	0.0113	1	1
	p-value	2.1231e-289	1.7006e-287	0.8382	1.2385e-257	0.1079	3.6817e-250		
	Energy usage (J)	6.0443e+03	1.5370e+03	0.2114	3.2081e-07	2.4578e-09	3.1530e-07	1.0000	1.0000
	p-value	1.2436e-116	4.8293e-108	2.4163e-30	0.9997	1.0000	0.9998		
Supercritical CO_2	Mass lipids (kg h^{-1})	1.4206	0.8122	-5.5431e-16	0.0218	-7.7296e-16	0.0124	1	1
	p-value	1.8448e-288	1.4777e-286	0.0913	1.0762e-256	0.0227	3.1991e-249		
	Energy usage (J)	1.9337e+04	1.1508e+04	317.9387	31.1433	0.1763	28.9687	1.0000	1.0000
	p-value	4.3532e-59	1.6277e-57	1.3765e-23	2.3570e-09	0.9723	1.3699e-06		

Table 19 Robustness of the processing unit conversion where U_1= Flow of algae(m^3 h^{-1}), and U2 = concentration algae (kg m^{-3}) or mass lipids (kg h^{-1}) encoded

Conversion	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Acidic	Flow of biodiesel (l h^{-1})	0.0016	5.3130e-20	-6.2020e-19	9.8224e-04	-6.7302e-19	-1.3938e-20	1	1
	p-value	2.8698e-292	0.6901	0.0115	6.2536e-291	0.0068	0.9409		
	Energy usage (J)	7.7440e+03	0.7248	7.8221e-04	143.7456	5.2789e-10	0.0026	1.0000	1.0000
	p-value	2.8570e-185	1.9602e-111	9.8798e-51	4.3878e-155	0.9991	3.7602e-62		
Alkaline	Flow of biodiesel (l h^{-1})	0.0016	2.2345e-20	-1.2483e-18	9.9647e-04	-5.0871e-19	7.2306e-21	1	1
	p-value	5.3962e-289	0.9113	0.0014	1.1759e-287	0.1450	0.9797		
	Energy usage (J)	6.2189e+05	3.3468e+04	2.6097e+03	-4.4639	-9.2396e-11	-2.5521	1.0000	1.0000
	p-value	6.5884e-86	1.6656e-64	4.0293e-39	0.2418	1	0.6309		
Enzymatic direct conversion	Flow of biodiesel (l h^{-1})	0.0016	9.4006e-04	-5.1614e-19	2.5203e-05	-7.7277e-19	1.4409e-05	1	1

	p-value	2.6230e-290	2.1010e-288	0.0894	1.5301e-258	0.0148	4.5487e-251		
	Energy usage (J)	6.2189e+05	3.3468e+04	2.6097e+03	-4.4639	-9.2396e-11	-2.5521	1.0000	1.0000
	p-value	6.5884e-86	1.6656e-64	4.0293e-39	0.2418	1	0.6309		
Microwave assisted dry conversion	Flow of biodiesel ($l\ h^{-1}$)	0.0452	0.0414	7.2444e-04	0.0441	7.7304e-04	0.0399	0.9999	0.9999
	p-value	5.6615e-35	5.8299e-37	0.0035	1.7404e-37	0.0021	8.3699e-34		
	Energy usage (J)	1.1148e+05	5.7634e+03	7.9607	17.9288	8.4089	22.7823	1.0000	1.0000
	p-value	1.3906e-79	7.5423e-58	0.0037	1.1672e-10	0.0025	7.0910e-10		
Supercritical methanol wet conversion	Flow of biodiesel ($l\ h^{-1}$)	2.3193e-05	1.3260e-05	3.0517e-21	-	-	-	1	1
	p-value	9.8469e-32	2.4808e-31	0.8102	-	-	-		
	Energy usage (J)	6.5515e+04	1.6901e+04	1.2966	-	-	-	1.0000	1.0000
	p-value	9.0576e-15	1.1209e-13	5.4406e-05	-	-	-		

Appendix C. Result processing units flexibility

Table 20 Flexibility of the processing units in harvesting with their U_1 and U_2

Harvesting	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Centrifugation *(concentration factor)*	Flow of algae broth ($m^3\ h^{-1}$)	0.0911	-0.9540	1.1332	-	-	-	0.9094	0.8187
	p-value	0.7808	0.0671	0.1234	-	-	-		
	Energy usage (J)	5.1000e+03	0.0027	-0.0027	-	-	-	0.9449	0.8898
	p-value	1.3394e-14	0.0374	0.0955	-	-	-		
Pressure filtration *(concentration factor)*	Flow of algae broth ($m^3\ h^{-1}$)	0.0911	-0.9540	1.1332	-	-	-	0.9094	0.8187
	p-value	0.7808	0.0671	0.1234	-	-	-		
	Energy usage (J)	2.7778e+03	0.0027	-0.0027	-	-	-	0.9449	0.8898
	p-value	4.5151e-14	0.0374	0.0955	-	-	-		
Vacuum filtration *(concentration factor)*	Flow of algae broth ($m^3\ h^{-1}$)	0.0911	-0.9540	1.1332	-	-	-	0.9094	0.8187
	p-value	0.7808	0.0671	0.1234	-	-	-		
	Energy usage (J)	6.1111e+03	0.0027	-0.0027	-	-	-	0.9449	0.8898
	p-value	9.3288e-15	0.0374	0.0955	-	-	-		
Chitosan flocculation *(concentration factor, chitosan concentration)*	Flow of algae broth ($m^3\ h^{-1}$)	0.2806	-0.4046	0.3430	-0.4376	-0.0268	0.4036	0.9339	0.9165
	p-value	1.2840e-04	1.0388e-08	1.2268e-04	2.9382e-09	0.7115	1.8549e-06		
	Energy usage (J)	2.5003e+03	0.0103	-0.0085	0.0120	0.0010	-0.0101	0.9419	0.9266
	p-value	6.2899e-108	4.9135e-09	9.8067e-05	4.5055e-10	0.5635	1.3538e-06		
Poly-glutamate flocculation *(poly-glutamate concentration)*	Flow of algae broth ($m^3\ h^{-1}$)	0.0926	-4.9156e-08	6.9116e-14	-	-	-	1.0000	1.0000
	p-value	5.4537e-33	1.5941e-20	2.3038e-08	-	-	-		
	Energy usage (J)	600.0194	2.3701e-10	-2.8541e-26	-	-	-	1.0000	1.0000
	p-value	8.7185e-33	4.6016e-08	1	-	-	-		

Table 21 Flexibility of the processing unit drying in drying with its U_1

Drying	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Drying *(concentration factor)*	Flow of algae broth ($m^3\ h^{-1}$)	-2.4809e-04	-0.0048	0.0062	-	-	-	0.8861	0.7722
	p-value	0.8960	0.0888	0.1382	-	-	-		
	Algae concentration ($kg\ m^{-3}$)	8.2000e+03	7.8000e+03	2.8042e-12	-	-	-	1	1
	p-value	2.2706e-31	2.0666e-31	0.6859	-	-	-		
	Energy usage (J)	2.6264e+05	2.4414e+04	-3.1691e+04	-	-	-	0.8861	0.7722
	p-value	0.0011	0.0888	0.1382	-	-	-		

Table 22 Robustness of the processing unit bead milling in disruption with its U_1

Disruption	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Bead milling *(Bead filling)*	Concentration accessible lipids ($kg\ m^{-3}$)	11.7053	0.1128	-0.0065	-	-	-	1.0000	1.0000
	p-value	1.1024e-10	9.7737e-07	8.3071e-04	-	-	-		
	Energy usage (J)	3.3000e+03	-1.7582e-13	2.1173e-29	-	-	-	1.0000	1.0000
	p-value	0	0	0	-	-	-		

Table 23 Robustness of the processing unit supercritical CO_2 in extraction with its U_1

Extraction	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Supercritical CO_2 *(temperature, pressure)*	Mass lipids ($kg\ h^{-1}$)	0.6028	-0.3380	0.2216	0.4477	0.1065	0.0676	0.9727	0.9655
	p-value	5.4910e-14	4.8169e-12	1.2066e-05	3.1252e-14	0.0110	0.0458		
	Energy usage (J)	2.0430e+04	579.6763	-1.3229e-04	513.3317	2.6167e-06	-1.6930e-04	1.0000	1.0000
	p-value	2.2484e-158	1.0879e-131	4.8332e-04	1.0953e-130	0.9346	3.6670e-06		

Table 24 Flexibility of the processing units in conversion with their U_1 and U_2

Conversion	Output (Y)	β_0	β_1	β_{11}	β_2	β_{22}	β_{12}	R^2	Adjusted R^2
Acidic *(flow of methanol)*	Energy usage (J)	1.5828e+04	8.5377e+03	461.6937	-	-	-	1.0000	1.0000
	p-value	5.8537e-07	1.6568e-06	0.0016	-	-	-		
Alkaline *(flow of methanol)*	Energy usage (J)	8.8406e+05	4.4281e+05	461.6937	-	-	-	1.0000	1.0000
	p-value	1.8764e-10	6.1592e-10	0.0016	-	-	-		
Enzymatic direct conversion *(flow of methanol)*	Energy usage (J)	1.6544e+06	1.3390e+06	3.1128e+05	-	-	-	1.0000	0.9999
	p-value	2.0215e-05	2.5415e-05	0.0013	-	-	-		
Microwave assisted dry conversion *(flow of methanol)*	Flow of biodiesel (l h^{-1})	0.0452	-1.6215e-04	8.6865e-05	-	-	-	0.9935	0.9870
	p-value	5.6123e-08	0.0036	0.0340	-	-	-		
	Energy usage (J)	2.1666e+05	1.0529e+05	110.3031	-	-	-	1.0000	1.0000
	p-value	1.6688e-10	5.8196e-10	0.0015	-	-	-		
Supercritical methanol wet conversion *(flow of methanol, temperature)*	Flow of biodiesel (l h^{-1})	0.0012	-2.0375e-04	7.6960e-05	-8.7275e-06	-3.3267e-04	-4.1241e-05	0.9983	0.9978
	p-value	1.9026e-36	7.9757e-25	1.0351e-12	0.0052	1.5370e-24	2.1789e-09		
	Energy usage (J)	1.0565e+05	3.7963e+04	160.8693	3.8596e+03	-26.7018	1.6352e+03	1.0000	1.0000
	p-value	3.0527e-82	1.6623e-76	4.2562e-27	1.2142e-57	1.5098e-12	1.0732e-47		

Appendix D. Response surface methodology

Creating a RSM for data can be done through different templates. In this research a central composite design (CCD) will be used, which is widely used for second order models with multivariable. The main advantage of CCD is that a full factorial experiment is not needed. In a full factorial experiment all possible states of a system are explored. Normally a CCD consist of twelve runs when there are three parameters, four runs at the centre, four runs at the corner points and four runs at the axial runs as can be seen in Figure 22. In Figure 22 the length of a_1 and b_1 are respectively 1 and $\sqrt{2}$. In this paper the axial runs are neglected and only runs within the cube are performed. Reason is that our control factors have a minimum and maximum value. With equation 2 it is easy to encode these into a range of -1 to 1. Assuming $\sqrt{2}$ is possible in the equation means that the minimum or maximum value of the control factor will be exceeded.

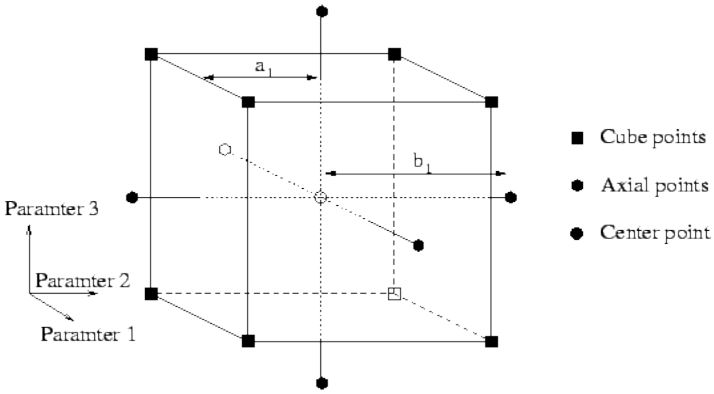

Figure 22 Central composite design for three parameters.

Every point in the CCD has an outcome Y. We can create two matrixes, an X matrix which is size [M x N] and a Y matrix which is size [M x 1]. The X matrix holds all the possible coded variables, the Y matrix the outcome of the system with these different coded values. The size of M and N are dependent on the amount of parameters. The amount of M and N can be calculated with equation 4 and 5.

$$M = 2^{|parameter|} + 1 \tag{4}$$

$$N = 1 + 2 \cdot |parameter| + \sum_{i=1}^{|parameter|} (|parameter| - i) \tag{5}$$

In which |parameter| denotes the amount of parameters. Equation 4 is just the calculation of all the corners of the cube, plus one for the point in the middle of the cube. For the length of N, one is added. This is because the first column of the X matrix only consist of the number one. This is needed for equation 6. The calculation of the coefficients (β) of equation 1 is done with equation 6.

$$\beta = (X^T X)^{-1} X^T \tag{6}$$

Appendix E. Statistics

To test if the coefficient has a significant influence on the output a student's t-distribution is applied. This test is useful to see if two distributed means have different values. This method is useful since it can be used to test if a coefficient has an influence on the mean. If the test indicates that the mean is altered, the coefficient has an influence on the output. A two-sided test is applied since both below or above the mean indicates that the control factor has influence on the output(Press et al., 1996). To apply the student's t-distribution equation 7 is used.

$$A(t|v) = I_{\frac{v}{v+t^2}}\left(\frac{v}{2},\frac{1}{2}\right) \tag{7}$$

$A(t|v)$ stance for the result of the two-sided test. Where v is the degrees of freedom. The t can be calculated with equation 8.

$$t = \frac{\beta_n}{\sqrt{var_{\beta_n}}} \tag{8}$$

β_n is the coefficient n that is tested. Var_{β_n} is the variation of the coefficient n.

To calculate the regularized incomplete beta function equation 9 is needed.

$$I_x(z,w) = \frac{1}{B(z,w)}\int_0^x t^{z-1}(1-t)^{w-1}dt \tag{9}$$

And the beta function:

$$B(z,w) = \int_0^1 t^{z-1}(1-t)^{w-1}dt \tag{10}$$

Appendix F. Relative gain for non-square multivariable systems

Normally for the calculation of a relative gain matrix the amount of inputs of the systems is equal to the outputs. However not all the systems in this research hold this property. Chang and Yu (1990) found a way to calculate the relative gain for multivariable systems where the amount of inputs and outputs is not identical.

For the calculation of the relative gain for non-square multivariable systems, or in other terms the non-square relative gain, the steady state gain matrix is needed. Since our system is already in steady state, the relative gain is only needed to be calculated. This is done with equation 11.

$$G = \begin{bmatrix} \lambda_{11}^N & \cdots & \lambda_{1j}^N \\ \vdots & \ddots & \vdots \\ \lambda_{i1}^N & \cdots & \lambda_{ij}^N \end{bmatrix} \tag{11}$$

In which G is the steady state gain matrix. λ_{ij}^N is the open-loop gain over the closed-loop gain as seen in equation 12.

$$\lambda_{ij}^N = \frac{open - loop\ gain}{closed - loop\ gain} = \frac{\left[\frac{\delta y_i}{\delta u_j}\right]_{u_k, k\neq j}}{\left[\frac{\delta y_i}{\delta u_j}\right]_{y_k, k\neq i}} \tag{12}$$

The open-loop gain is the change of y_i if u_i is increased with δu_i while all the other inputs remain constant.

The closed loop is derived where an change of δu_i causes $y_{k,k\neq i}$ to increase. The system will try to keep $y_{k,k\neq i}$ constant by changing $u_{k,k\neq j}$. Therefore a new $u_{k,k\neq j}$ is derived which will change y_i with δy_i.

For the calculation of the NRG equation 13 is used.

$$\Lambda^N = G \otimes (G^+)^T \tag{13}$$

In which $\otimes$ denotes element-by-element multiplication of the matrixes. Λ^N is our normalised relative gain. For the calculation of the NRG the inverse of G is needed. The matrix G is a non-square matrix so therefore calculation the inverse is done with the Moore-Penrose procedure. This creates a pseudo-inverse matrix of G stated as G^+ in equation 13.