

## Integrated facility for the use of oranges as a source for power, chemicals and juice

A. Criado, M. Martín\*

*Departamento de Ingeniería Química. Universidad de Salamanca. Pza. Caídos 1-5, 37008 Salamanca, Spain*

Received: July 9, 2019; revised: August 16, 2019

In this work, we evaluate the integration opportunities within the juice production industry. The process begins with the oranges reception and juice production. The peel is further processed for the production of limonene. Next, the waste is digested to provide for the thermal and electrical energy required for the facility using a gas turbine. The process results in a highly integrated facility that allows implementing the concept of circular economy within the fruit industry. However, even though the facility is profitable, with a benefit after taxes of 56%, if higher added value products are to be produced such as limonene, the utilities required by the process cannot be produced from the residues affecting the sustainability of the process.

**Key words:** Biogas, anaerobic digestion, manure, power production, mathematical optimization

### INTRODUCTION

The concept of circular economy is a current trends towards a more efficient production system where waste is reused and recycled as raw material. In particular, the food industry is characterized by the production of large amounts of waste in the processing of the raw material. To improve the efficiency of these plants integrated design as a chemical complex will allow reusing the waste into valuable products and energy [1]. In the case of the fruit industry, in the production of juice together with the main product waste is produced [2]. This residue is also part of the municipal solid waste that is being treated for energy production. But before that final use, it is important to notice that the peels contain added value products. In particular, citrus peels contain compounds such as pectin, limonene [3-5] that can and should be recovered before waste treatment. Anaerobic digestion (AD) has been deemed as an efficient process to treat waste [6]. The main products from the AD of waste consist of biogas, composed of CO<sub>2</sub> and CH<sub>4</sub> that can be an interesting source for chemicals via dry reforming [7], power [8] and a digestate rich in nutrients that can be recovered in different forms [9]. Therefore, a facility for the production of juice can become a highly integrated chemical complex.

So far, the evaluation of the use and products from the fruit industry has been addressed separately. AD of waste has been studied towards evaluating the yield to biogas [10]. The recovery of added value products has been experimentally studied as well as the pectin recovery [3]. Only lately the last part of the process, from the waste towards limonene and p-Cymene including the possible use of the waste to produce power via

gasification has been considered [11] However, the large water content and the need to define the synergies for the entire process, from the fruit towards the multiproduct suggest the use of a systematic approach

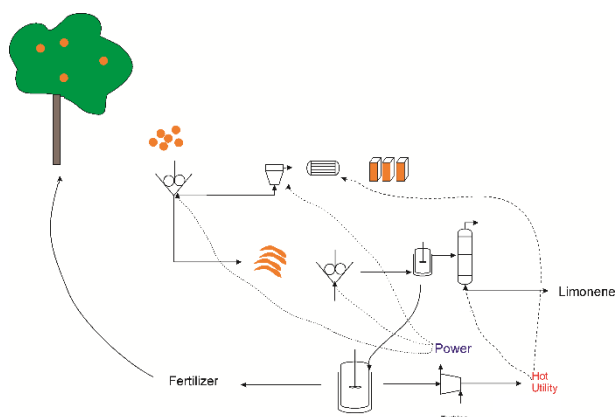
In this work we develop an integrated facility for the production of juices, limonene and power from oranges using a mathematical optimization formulation in an integrated facility that uses the exhaust gases from the gas turbine to produce steam that eventually produces power in a steam turbine. The rest of the paper is organized as follows: in section 2 we provide a brief description of the process; in section 3, the different units are described and the modelling assumptions are presented; in section 4, the results of the optimal operation of the facility are shown together with an economic evaluation; and finally, in section 5, we draw some conclusions.

### PROCESS DESCRIPTION

The process consists of fruit processing, limonene extraction, biogas production, biogas purification (biomethane generation), gas turbine (Brayton cycle) and the use of the flue gas to provide the energy within the process, see Figure 1.

Sweet oranges are washed using 2 L of water per kg of oranges and squeezed. It is a mechanical step where juice and the peels are obtained separately. The pulp can be left in the juice or removed by filtration. This pulp, if removed, can be used within the peel residue in the AD step. The juice must be thermally treated in two steps to remove microorganisms and denaturalize enzymes. The first stage consists of heating up the juice 368 K for 10-30 s. After that, and before bottling it up, the juice is heated up again for 15 s at 368 K [12] The peels constitute 40-60% of the orange.

\* To whom all correspondence should be sent:  
E-mail: mariano.m3@usal.es



**Fig. 1.** Flowsheet for the integrated production of juices, limonene and power.

Limonene, d-limonene, can be recovered from the peel by extraction. The peel must be milled to increase the surface area. Next, n-hexane is used to extract it, so that  $0.55 \text{ dm}^3$  are recovered per  $\text{m}^3$  of hexane used [13]. The peel residue is separated by filtration. Finally, the hexane is recovered by distillation. The distillation column works under vacuum to avoid decomposition of the limonene.

The peel residue is fed to a bioreactor where it is anaerobically digested to produce biogas and a decomposed substrate (digestate). The biogas is composed of methane, carbon dioxide, nitrogen, hydrogen sulphide, ammonia and moisture. It is sent to purification to remove first the  $\text{H}_2\text{S}$  in a fixed-bed reactor and later the  $\text{CO}_2$  (and traces of  $\text{NH}_3$ ), using Pressure Swing Adsorption (PSA). Once the biogas is mainly methane, it is used in the Brayton cycle, where the biomethane is compressed, burned with air and expanded in a turbine, generating power.

The digestate can be processed to recover the nutrients following different technologies. Struvite production is recommended if it is to be transported long distances, but if the facility is allocated close to the harvesting point of the oranges it can be just recovered as a cake.

### MODELLING ISSUES

The models for each of the units involved in the process, are formulated using mass and energy balances, experimental yields, thermodynamics, chemical and vapor-liquid equilibria to evaluate their performance. Details of the modelling approach can be seen in [14]. The process model is written in terms of total mass flows, component mass flows, component mass fractions, temperatures and pressures of the streams in the network. Table 1 summarizes the modelling approach for each of the units involved in the process.

**Table 1.** Summary of modelling approach for the integrated facility

Unit	Modelling approach
Washing	M&E Balances Exp. Data
Grinding	M&E Balances Exp. Data on Power
Juice treatment	M&E Balances Exp. Data on T and time
Peel milling	M&E Balances Exp. Data on power [18]
Limonene extraction	M&E Balances Exp. Data on extraction [13]
n-Hexane recovery	Fenske eqs M&E Balances [16]
AD	M&E Balances [8]
$\text{H}_2\text{S}$ Removal	M&E Balances [8]
$\text{NH}_3/\text{CO}_2$ Removal	M&E Balances [8]
Brayton cycle	Thermodynamics M&E Balances [8]

The facility is modelled and optimized for the optimal operation producing the power and hot utilities required for the operation of the entire integrated process maximizing the profit obtained from the juice, the limonene and minimizing the input of utilities. The orange peels after pretreatment for limonene recovery shows the composition given by Table 2.

**Table 2.** Orange peels composition for digestion [17]

Vbiogas ( $\text{m}^3/\text{kg}$ )	0.35
WDM	0.21
WVS	0.85
WN	0.002
WNorg	0.010
WP	0.002
WK	0.009
RCN	15

WDM: Dry matter weight percentage; WVS: Volatile weight percentage; WC: Carbon weight percentage; WN Inorganic nitrogen weight percentage; WNorg: organic nitrogen weight percentage; WP: Phosphorous weight percentage; WK: Potassium weight percentage; RCN: Carbon to Nitrogen ratio

The investment cost of the facility is based on the factorial method [15] where the cost for the different units has been estimated based on [16] updating of the units when required. For the production cost, again, the factorial method is used. The cost of the oranges for juice is taken to be  $0.1 \text{ €/kg}$  [19], the cost of steam  $0.019 \text{ €/kg}$  [20] and that of cooling water  $0.00057 \text{ €/kg}$  [21] while the fresh hexane is taken to be  $1.5 \text{ €/kg}$  [22]

### RESULTS

The mass and energy balances result in the values reported in Table 3 where the major ratios are reported for a facility that processes  $5 \text{ kg/s}$  of oranges. In the results it is possible to see that the facility can provide its own power for running the

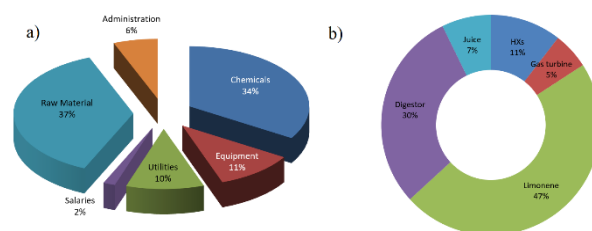
grinding, milling and centrifuges. However, the extraction of limonene requires a large amount of energy for the recovery of the solvent, resulting in a deficit that must be provided from external resources. The thermal energy available is enough for the production of the juice and its pasteurization, though, but not enough for the facility to be self-sustained. Fertilizers and limonene are also produced a part from the juice that can be further used to obtain credit and/or reduce the needs of nutrients in the following growing period.

**Table 3.** Major results of the flowsheet operation

Production of Juice	0.5 kg /kg of Oranges
Peels	0.5 kg /kg of Oranges
Hexane added	0.1209 kg/kg of peel
Biogas produced	0.0464 kg /kg of peel
Fertilizer	0.1696 kg dry /kg of peel
NPK index	0.17/0.1/0.24
Limonene produced	0.00924 kg /kg Peel
Air excess (Gas turbine)	21%
Power produced	280 kJ <sub>c</sub> /kg of Peel
Power consumed	3.6 kJ <sub>c</sub> /kg Peel (Milling) [18]
Thermal Energy available	350 kJ/kg of Peel
Thermal Energy required	150 kJ / kg peel ( Pasteurization) 5400 kJ /kg peel ( Solvent recovery)

The economic analysis can be summarized in the following values. For a facility that processes 5kg/s of oranges (155kt/yr), the integrated facility investment cost adds up to 59 M€ with a production cost of 41.7 M€/yr to produce 77760 m<sup>3</sup> of juice per year, 519 ton/yr of limonene and 5.83 GWh of power. The facility requires steam, around 6kg/s and cooling water, 304 kg/s. the profitability of the facility highly depends on the juice cost per liter. For the products prices of 1 €/L of juice, the limonene costs at 15 €/kg and 0.06 €/kWh obtained from the excess of power, without considering any credit out of the digestate, the benefit after taxes is 56%. Out of the digestate additional income can be obtained depending on the market and its form at the processing cost of each alternative [9] unless is internally used for the next harvest in which case it is expected to decrease the price of the oranges. Figure 2 shows the breakdown of the production cost (a), and the investment cost (b). almost 40% of the production cost goes to the raw material, another 34% corresponds to chemicals while utilities and equipment amortization reach 10% each. In terms of the investment, the section for the production of limonene represents 50% of the cost of the facility, while that of the waste treatment adds up to 30%.

Juice production and pasteurization reaches 10%, since it includes the HXs.



**Fig. 2.** Breakdown of the investment (a) and production costs (b).

## CONCLUSION

In this work the concept of an integrated facility for the production of juices, limonene and power from oranges is evaluated using a mathematical optimization formulation. Waste is used to produce utilities for the operation of the plant, power and heat, required by the facility including juice pasteurization and limonene extraction.

The facility can provide its own electricity, as well as thermal energy for juice production, but the production of limonene requires a large amount of energy for solvent recovery. The process is economically interesting, with a benefit after taxes of 56%, but it is highly energy intense. More efficient limonene production paths and recovery technologies must be addressed for a cleaner production process. Furthermore, scale up studies are required to evaluate the effect of the exploitation size on its economics.

**Acknowledgements:** The authors acknowledge the funding received from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 778168 and PSEM3 group. AC also acknowledges MINECO for an introduction research fellowship.

## REFERENCES

1. T.H. Kwan, D. Pleissner, K.Y. Lau, J. Venus, A. Pommeret, C.S.K. Lin, *Biores. Technol.*, **198**, 292, (2015).
2. V. Negro, G. Mancini, B. Ruggeri, D. Fino, *Biores. Technol.*, **214**, 806 (2016).
3. M. Pourbafrani, G. Forgacs, I. Sarvari Horvath, C. Niklasson, M.J. Taherzadeh, *Biores. Technol.*, **101**, 4246 (2010).
4. G. Stella Mary, P. Sugumaran, S. Niveditha, B. Ramalakshmi, P. Ravichandran, S. Seshadri, *Int. J. Recycl. Org. Waste. Agricult.*, **5**, 43 (2016).
5. B. Satari, K. Karimi, *Res. Conserv. Recycl.* **129**, 153 (2018).
6. H. Fehrenbach, J. Giegerich, G. Reinhardt, J. Schmitz, U. Sayer, M. Gretz. Criteria for a Sustainable Use of Bioenergy on a Global Scale. Germany: Federal

- Environment Agency; 2008; 245 (prepared by the Institute for Energy and Environmental Research (IFEU), Heidelberg).
7. B. Hernández, M. Martín *Ind Eng. Chem. Res.*, **55** (23), 6677 (2016).
  8. E. León, M. Martín, *Energ. Conv. Manag.*, **114**, 89 (2016).
  9. E. Martín, A. Sampat, M. Martín, V. Zavala, *Chem. Eng. Res Des.*, **131**, 160 (2018).
  10. A. Koppa, P. Pullammanappallil, *Energy*, **60**, 62 (2013).
  11. J.A. Dávila, M. Rosenberg, C.A. Cardona, *Waste Biomass Valor.*, **6**, 253 (2015).
  12. J.A. Siles, F. Vargas, M.C. Gutiérrez, A. Chica, M.A. Martín, *Biores. Technol.*, **211**, 173 (2016).
  13. R. Wikandari, H. Nguyen, R. Millati, N. Claes, M.J. Taherzadeh, *BioMed research international*, <http://dx.doi.org/10.1155/2015/494182> (2015)
  14. M. Martín, *Industrial chemical process. Analysis and design*. Elsevier. Oxford (2016).
  15. R. Sinnott, G. Towler G, *Chemical Engineering Design*. Oxford: Elsevier Ltd. (2009).
  16. Matche. 2014. Index of process equipment. <http://www.matche.com/equipcost/EquipmentIndex.html>, Last accessed 2019
  17. M.A. Martín, J.A. Siles, A.F. Chica, A. Martín, *Biores. Technol.*, **101**, 8993 (2010)
  18. G.D. Saravacos, A.F. Kostaropoulos. *Handbook of Food Processing Equipment*. Spriger. Berlin. 2002.
  19. <https://www.lasprovincias.es/economia/agricultura/precio-naranja-valencia-2018-20181112165256-nt.html>
  20. S. Pérez Uresti, M. Martín, A. Jiménez Gutierrez, *Applied Energy*, **250**, 1120 (2019.)
  21. M. Martín, I.E. Grossmann, *AIChE J.*, **57**(12), 3408 (2011).
  22. <http://product.lookchem.com/item/899/N-hexane-price.html>