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# An optimisation-based framework for the conceptual design of reaction-separation processes

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ABSTRACT

Prior to detailed process design, it is vital to first generate a good flowsheet that meets particular objective. This is particularly the case in bio-based materials and products, where, given a range of chemistries, the synthesis problem is not about the best way to make a particular product but rather the best way to convert a specific feedstock. In order to do so, an optimisation-based framework, which can be used to identify the optimal configuration of a process network that consists of both reactions and separation systems to achieve maximum economic potential, is presented in this paper. A process superstructure, which includes the concept of master reaction stages and subsidiary separation stages, is introduced to facilitate the theory. The problem is formulated as a generalised mixed integer linear programming (MILP) model which accounts for the simultaneous selection of products and identification of the process configuration. The solution of the optimisation problem includes the best possible economic performance, identification of active reactions, reaction ordering and separation sequences along with the corresponding flowsheet of the optimal process. The economic criterion takes account of raw materials costs, product values and separation related costs. Two bio-based chemical case studies are presented to illustrate the applicability of the proposed methodology.



## 1. Introduction

Polymers have been one of the most important bulk chemicals across the world since World War II. Global demand for polymers in 2013 was 250 million tonnes, of which the UK's plastic demand accounts for approximately 1.5% (PlasticsEurope, 2014). Polymers are consumed by various sectors which are dominated by packaging, construction and transport. As the largest polymer consumer in the UK, the plastic packaging sector was forecasted to have a 14.2% market growth between 2014 and 2018 (BP and R, 2014). The UK is not only a large polymer consumer, but also an innovative and experienced plastic producer. The UK plastics industry processes 4.8 million tonnes of raw materials to produce 2.5 million tonnes of polymers annually, with an annual turnover of £19 billion in 2012. In general, the plastic sector accounts for about 7.5% of the chemicals industry in the UK and provides approximately 180,000 job opportunities (UKT and I, 2012).

However, widely used polymers such as polycarbonate (Fiege et al., 2000), polyethylene and polyvinyl chloride are still produced from oil/gas-based feedstocks. Although developments in the technology of unconventional production of oil/gas, improvements in energy efficiency of vehicles all around the world and other demand/supply factors have contributed to the drop of the oil price for the last two years (Krauss, 2015), petrochemical derived polymers are still facing the issues of increasing greenhouse gas emissions and waste accumulation (Gandini, 2008). Currently, global companies such as P&G are devoted to exploring consumer driven processes and products with lower environmental impacts. Therefore, sustainability issues associated with the conventional polymer industry may lead to its diminished

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**Nomenclature**

*Indices descriptions*

| | |
|---|---|
| $i$ | components |
| $j$ | reactions |
| $k$ | master stages |
| $l$ | separation stages |
| $p$ | discretised split fraction points |

*Binary variables*

| | |
|---|---|
| $b_k^{next}$ | =1 if all the flow from reaction stage $k$ goes directly to the next stage; 0 otherwise |
| $b_k^R$ | =1 if all the flow from reaction stage $k$ goes to direct removal; 0 otherwise |
| $b_k^{sepin}$ | =1 if all the flow from reaction stage $k$ goes to the separation; 0 otherwise |
| $EX_{i,k,l}^T$ | =1 if top removal of separation stage $l$ of master stage $k$ contains only pure component i; 0 otherwise |
| $EX_{i,k,l}^B$ | =1 if bottom removal of separation stage $l$ of master stage $k$ contains only pure component i; 0 otherwise |
| $EXQ_{i,k}^R$ | =1 if stream $Q_{i,k}^R$ contains only pure component i; 0 otherwise |
| $SA_{k,l}$ | =1 if separation stage $l$ in master stage $k$ is active; 0 otherwise |
| $w_{i,k,l}$ | =1 if component $i$ is the light key at separation stage $l$ of master stage $k$; 0 otherwise |
| $x_{k,l,l'}^T$ | =1 if the stream from stage $l$ to stage $l'$ is active at the top of stage $l$ in master stage $k$; 0 otherwise |
| $x_{k,l,l'}^B$ | =1 if the stream from stage $l$ to stage $l'$ is active at the bottom of stage $l$ in master stage $k$; 0 otherwise |
| $y_{j,k}$ | =1 if reaction $j$ occurs in master reaction stage $k$; 0 otherwise |
| $Yb_{i,k,l}$ | =1 if component $i$ is chosen as the heavy key at separation stage $l$ and in master stage $k$; 0 otherwise |
| $\lambda_{k,l,p}^{TR}$ | =1 if value of point p is chosen for top removal split fraction at separation stage $l$ of master stage $k$; 0 otherwise |
| $\lambda_{k,l,p}^{Tout}$ | =1 if value of point p is chosen for top out split fraction at separation stage $l$ of master stage $k$; 0 otherwise |
| $\lambda_{k,l,l',p}^{Ts}$ | =1 if value of point p is chosen for top split fraction for stream goes from separation stage $l$ to $l'$ at master stage $k$; 0 otherwise |
| $\lambda_{k,l,p}^{BR}$ | =1 if value of point p is chosen for bottom removal split fraction at separation stage $l$ of master stage $k$; 0 otherwise |
| $\lambda_{k,l,p}^{Bout}$ | =1 if value of point p is chosen for bottom out split fraction at separation stage $l$ of master stage $k$; 0 otherwise |
| $\lambda_{k,l,l',p}^{Bs}$ | =1 if value of point p is chosen for bottom split fraction for stream goes from separation stage $l$ to $l'$ at master stage $k$; 0 otherwise |
| $NQ_{i,k}^R$ | =1 if component i exists in flow $Q_{i,k}^R$; 0 otherwise |
| $N_{i,k,l}^{in}$ | =1 if component $i$ exists at the inlet of separation stage $l$ of master stage $k$; 0 otherwise |
| $N_{i,k,l}^T$ | =1 if component $i$ exists at the top stream of separation stage $l$ of master stage $k$; 0 otherwise |
| $N_{i,k,l}^B$ | =1 if component $i$ exists at the bottom stream of separation stage $l$ of master stage $k$; 0 otherwise |
| $NQ_{i,k}^{in}$ | =1 if component $i$ exists at the inlet stream of reaction stage $k$; 0 otherwise |

*Continuous variables*

| | |
|---|---|
| $A_{i,k}$ | flowrate of component $i$ added to the master mixing stage $k$ |
| $Cost_{k,l}$ | separation cost of each stage $l$ of master stage $k$ |
| $CostT_k$ | total separation cost of master stage $k$ |
| $DX_{i',i'',k,l}$ | dummy variable for linearisation of separation cost equation |
| $EP$ | economic potential of the process |
| $f_{i,k,l}^{in}$ | flowrate of component $i$ entering separation stage $l$ |
| $f_{i,k,l}^T$ | flowrate of component $i$ leaving the top of separation stage $l$ |
| $f_{i,k,l,l'}^{Ts}$ | flowrate of component $i$ leaving the top of separation stage $l$ and going to stage $l'$, where $l<l'$ |
| $f_{i,k,l}^{Tout}$ | flowrate of component $i$ leaving the top of separation stage $l$ and going straight to the next reaction stage |
| $f_{i,k,l}^{TR}$ | flowrate of component $i$ leaving the top of separation stage $l$ and going straight to removal as product |
| $f_{i,k,l}^B$ | flowrate of component $i$ leaving the bottom of separation stage $l$ |
| $f_{i,k,l,l'}^{Bs}$ | flowrate of component $i$ leaving the bottom of separation stage $l$ and going to stage $l'$, where $l<l'$ |
| $f_{i,k,l}^{Bout}$ | flowrate of component $i$ leaving the bottom of separation stage $k$ and going straight to the next reaction stage |
| $f_{i,k,l}^{BR}$ | flowrate of component $i$ leaving the bottom of separation stage $l$ and going straight to removal as product |
| $Q_{i,k}^{in}$ | flowrate of component $i$ entering master reaction stage $k$ |
| $Q_{i,k}^{out}$ | flowrate of component $i$ leaving master reaction stage $k$ |
| $Q_{i,k}^{sepin}$ | flowrate of component $i$ entering separation at stage $k$ |
| $Q_{i,k}^{sepout}$ | flowrate of component $i$ leaving separation stages of $k$ and goes directly to the mixing point at stage $k+1$ |
| $Q_{i,k}^R$ | flowrate of component $i$ goes to direct removal at stage $k$ |
| $Q_{i,k}^{next}$ | flowrate of component $i$ goes directly to the mixing point at stage $k+1$ |
| $Q_{i,k}$ | flowrate of component $i$ entering mixing point at stage $k$ |
| $R_{i,k}$ | flowrate of component i removed from the master separation stage $k$ |
| $R_{i,k}^{sep}$ | total amount of $i$ out from separation going straight to the removal |
| $U_{i',k,l}$ | dummy variable for linearisation of separation cost equation |
| $VQ_{i,k}^R$ | economic value of component $i$ in flow $Q_{i,k}^R$ |
| $V_{i,k,l}^{TR}$ | value of top removal of component $i$ at separation stage $l$ of master stage $k$ |