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A new double flash process and heat integration for better energy utilization of toluene disproportionation

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When a chemical complex has similar demands for both benzene and xylenes, toluene disproportionation is an attractive process to produce both benzene and p-xylene. Nevertheless, the conventional production process is heavily energy intensive with low economic benefits. In the conventional process, the high temperature reaction effluent of 458 °C is cooled to 36 °C for hydrogen separation. The flash liquid is then heated again and enters the stripper. The repeated effluent cooling and heating results in a low efficiency of energy utilization. A new double flash toluene disproportionation process for better energy utilization is presented in this study to avoid the repeated cooling and heating of the reaction effluent. The new and conventional processes are carefully compared on the basis of heat integration and economic evaluation. The new process reduces the hot utility by 13.27%, and the cold utility by 12.30%. Additionally, the investment in a heat exchanger network is decreased, while the income from products is increased.

1. Introduction

TDP (Toluene disproportionation process) is an important chemical production system to directly produce aromatic product xylenes. Xylenes consist of three isomers of dimethylbenzene, which are distinguished as ortho- (o-), meta- (m-), and para- (p-), as shown as Fig. 1. The configuration of p-xylene (p-xylene) is the principal precursor to terephthalic acid and dimethyl terephthalate, both of which are monomers used in the production of polyethylene terephthalate plastic bottles and polyester clothing. Ninety eight percent of p-xylene production, and half of all xylene, is consumed in this way. o-xylene (O-xylene) is an important precursor to phthalic anhydride. The demand for isophthalic acid is relatively modest, so m-xylene (m-xylene) is rarely sought, and thus, the m-xylene is often converted to o-xylene and p-xylene. The toluene disproportionation process involves three parts: disproportionation reaction, resultant separation, and heat recovery. The high reaction temperature and complex separation process makes it highly energy intensive. Process improvement and heat integration are essential to minimize the utility requirements to reduce the production costs.

Many studies have been conducted to improve the energy performance of process systems to reduce their hot and cold utility requirements [1]. Those methods can be generally classified into three categories: pinch analysis, mathematical programming and meta-heuristic optimization methods [2]. Pinch analysis is based on the first and second laws of thermodynamics and provides insights into understanding energy flows and ensuring the best possible design and operation [3]. Pinch analysis has also been widely used to form new graphical techniques to target the utility requirements of process units [4], total sites [5], or even industrial parks [6]. This has been reviewed by Morar and Agachi [7]. Recently, CGCC (column grand composite curve) was used to assess the thermodynamic feasibility of implementing a heat pump system and self-heat-recuperation technology into a conventional distillation column for energy savings on the NGL (natural gas liquid) recovery process [8]. “R-curve”, “R-ratio vs. the TAC (total annual cost) curve” and “R-ratio vs. the Emissions curve” were proposed on the basis of the graphical method to give a sense of how to improve the operation cost and emission values of any operation or under design total site [9]. These graphical techniques give a visual sense of the heat flow in processes.

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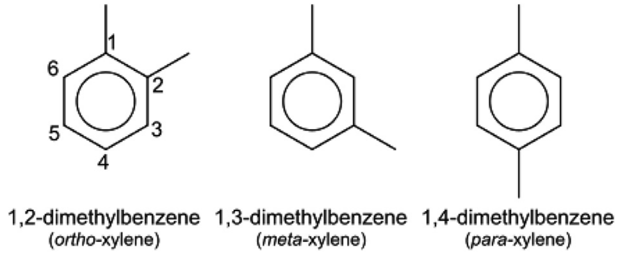

Fig. 1. The xylene isomers configuration.

Mathematical programming is widely used to obtain the minimum TAC, energy requirement or emission of processes. A transhipment model was first introduced to minimize the utility requirements of HENs (heat exchanger networks) [10], and the transhipment model was then used to design HENs [11]. Recently, new mathematical models were presented to address heat integration that involved variable stream data. An alternative model, based on an implicit temperature ordering and the transhipment model, was proposed to target the energy requirement of non-isothermal systems with variable inlet and outlet temperatures [12]. Two novel formulations for the optimization of the heat integration of chemical processes with variable stream data and non-linear process constraints were proposed, and the Multi-M model was presented [13]. Rigorous and complex MINLP (mixed integer nonlinear programming) models were also exploited for heat integration. Two optimization stages (network structure optimization and investment optimization) were presented and solved with the MILP (mixed integer linear programming) iteration method to retrofit HENs for both small-scale and industrial-scale problems [14]. A multi-objective MINLP problem, in which LCA (life cycle assessment) principles were used to quantify the environmental impact, were presented to address the optimal design of HENs, considering economic and environmental concerns [15]. The optimization problems of process systems results most often in MINLP models because of their inherent complexity. Meta-heuristic optimization methods, such as the commonly-used Genetic Algorithms [16] and Simulated Annealing [17] methods, were then introduced to solve complex and large-scale systems. However, those heuristic methods still often fail to solve industrial scale problems due to high nonlinearity.

According to the onion model, improvement of chemical reactions and separation processes leads to better energy performance of process systems than only optimizing heat recovery processes. This is also indicated by the essence of energy utilization in chemical processes. Therefore, process changes, integration of processes and HENs have been attracting more attention. Combining distillation models and HEN models in crude distillation units was proposed to reduce utility requirements, and the reduced model, the statistical model and the rigorous model were compared to each other to obtain an easier engineering application [18]. An approach with simultaneous considerations of material and energy performances was proposed to analyse and evaluate the three processes of crude distillation units to provide insights for designers to screen a suitable process and target energy requirement [19]. Based on HEN simulation, an MINLP model was formulated for a diesel hydrotreating unit to simultaneously optimize mass and energy [20]. Process and utility system models were combined to optimize materials and energy simultaneously in a total site [21]. This study is motivated to the process retrofit of the toluene disproportionation process. Utility requirements are then targeted, and a HEN design is also proposed to evaluate the energy performance and economics of process retrofit.

In the past decade, many researchers have explored the toluene disproportionation process. Since p-xylene is in greatest need among xylene isomers, the major challenges of this process are to improve the selectivity of p-xylene and separate p-xylene from its isomers while keeping energy consumption at reasonable level. Odedairo et al. investigated the xylene selectivity over an array of zeolites in toluene disproportionation and methylation, indicating that xylene selectivity is related to the size of channel [22]. Lobão et al. focused on the kinetics of toluene disproportionation in operation ranges that include real industrial operation conditions [23]. Mitra and Kunzru studied the p-xylene productivity for the disproportionation of toluene in micro-structured reactors, only to find that the increased p-xylene productivity is obtained at the cost of increased recycle cost [24]. The separation of p-xylene is industrially performed by crystallization or adsorption. Lima and Grossmann employed mathematical programming techniques to synthesize and optimize the crystallization processes, and the superstructure proposed demonstrated enough flexibility for further study [25]. Lim proposed an eight-zone SMB (simulated-moving-bed) operation at the industrial scale to improve the purity and productivity of p-xylene considering flushing of the stream trapped in bed lines [26]. Membrane separation has also attracted a lot of attentions. A ZSM-5 membrane synthesized on a stainless steel porous tube was introduced by Tarditi, which presented good thermal stability [27]. Yeong studied the effect of temperature, p-xylene feed partial pressure and p-xylene composition on the separate performance of the silicate-membrane [28]. Some authors have combined the reaction and separation of toluene disproportionation process. These publications were motivated by new catalytic mechanisms and reactors. Stitt compared the reaction distillation for toluene disproportionation with the conventional process and suggested that toluene disproportionation may be not a fruitful development opportunity for reactive distillation due to economic considerations [29].

Our previous studies used CGCC and GCC (grand composite curve) to propose heat integration of fractionating systems in p-xylene plants [30] and presented a mathematical model to integrate the distillations and HEN for a toluene disproportionation plant, in which the feed temperatures of distillation columns are variable [31]. These studies only focused on heat recovery in the toluene disproportionation process and did not involve process changes. In this study, we investigate the energy efficiency of the conventional toluene disproportionation process with pinch technology and develop a new double flash retrofit for a TDP. The conventional process and the new double flash retrofit process are both heat integrated and compared based on energy targets and economic evaluation. The Aspen Energy Analyser [32] is then applied to develop HENs with efficiency and accuracy.

2. Conventional process description and analysis

2.1. The description of conventional TDP

The conventional TDP is shown as Fig. 2. The feedstock of fresh toluene is mixed with recycled hydrogen and toluene. The mixture is heated to 450 °C before being fed into the disproportionation reactor, where toluene is partly transformed into benzene and xylene and some side reactions occur. In this study, the chemical reactions of the toluene disproportionation process are listed in Fig. 3. These chemical reactions were also widely applied to investigate the disproportionation reaction of toluene. The third chemical reaction indicated in Fig. 3 was often used to replace the slight side reactions of aromatic cracking in the toluene disproportionation process [33].