Performance Analysis of Isopropanol-Acetone-Hydrogen Chemical Heat Pump

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Abstract: This paper presents an experimental system of isopropanol-acetone-hydrogen chemical heat pump (IAH CHP) for the recovery of low-grade (<100°C) industrial waste heat. Using coefficient of performance (COP) and exergy efficiency as evaluation indices, the system performance is systematically evaluated based on the experimental data. The performance of the IAH CHP is found to be greatly affected by the endothermic and exothermic temperatures, and the flow rates of working fluid. The system performance reaches the optimum at about 360 K and 1.40 ml/s in the actual experimental system. And good heat insulation can reduce the heat loss and energy consumption.

Keywords: IAH CHP, COP, exergy, temperature, flow rate.

1. INTRODUCTION

With the continuous decline of the fossil fuels and to prevent the environment from deterioration, it appeals to develop alternative energy systems to recover low-grade waste heat. Most of the waste heat released to the environment cannot be reutilized because of its low temperature, discontinuousness and the high cost for recovery. Heat pump, including mechanical heat pump, sorption heat pump and chemical heat pump, can transform low-grade heat that cannot be or is at least very difficult to be reused into high-grade heat, so that part of the waste heat is recovered. Mechanical heat pumps are widely used in daily life, but its shortcomings like high cost, limited temperature-upgrading range, and low efficiency make it hard to be applied for low-grade industrial waste heat recovery (Ito *et al*., 1991; Morten *et al*., 2012). Although sorption heat pump has a wide temperature-upgrading range, its large pressure shift leads to high operation cost (Hasan *et al*., 2008; Kitikiatsophon *et al*., 2004). Xu *et al*. (2017) established a small scale prototype of the IAH-CHP to evaluate the technical and economic feasibility of the Isopropanol-AcetoneHydrogen chemical heat pump based on a lab-scale prototype. The results proved that the IAH-CHP system is efficient in recovering the low-level waste heat.

Chemical heat pump (CHP), in which chemical reaction plays a key role in thermal storage (Hamdan *et al*., 2013) and transforming low-grade heat into high-grade heat and also in the conversion between chemical energy and heat, is proposed to overcome the limitations of mechanical and sorption heat pumps (Hasan *et al*., 2008). Some CHPs are introduced and classified in different ways (Ito *et al*., 1991). By adopting an appropriate reaction pair, the CHP may have a different energy density and temperature-upgrading range (Spoelstra *et al*., 2002; Kim *et al*., 1992; Prevost *et al*., 1980; KlinSoda *et al*., 2007; Gandia *et al*., 1992; Jonas *et al*. 2015). In 1980, a CHP system using Isopropanol-Acetone-Hydrogen (IAH) as working pair was firstly proposed; since then, the possibility of using the dehydrogenation of isopropanol in a heat-driven CHP has been widely studied (Kitikiatsophon *et al*., 2004; Gandia *et al*., 1994; Ito *et al*., 1991; Taneda *et al*., 1994; Yamashita M., Saito Y., 1993; Taneda *et al*., 1995; Cho *et al*., 2007; Cho *et al*., 2007).

As one of the most promising CHPs, the IAH CHP was studied mainly through simulation and experiment. A dynamic computer simulation of a micro-CHP system (Anikeev *et al*., 1993; Cunningham *et al*., 1996) demonstrated that the dynamics of micro-CHP can be successfully simulated and an economic analysis can be readily incorporated as part of the dynamic simulation. Xu *et al*. (2014) proposed an IAH-CHP system with two exothermic reactors in serial connection and found that the COP and exergy efficiency increased by 7.6% and 10.3%, respectively, relative to the conventional one that has only one high-temperature equilibrium reactor. Guo and Huai (2012) designed a multi-parameter optimization recuperator in

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an IAH CHP system based on the entransy dissipation theory; they believed that the optimization design approach of heat exchanger based on entransy dissipation theory was very effective at both component and system levels. Xin *et al*. (2013) proposed a reactive distillation column for liquid phase dehydrogenation of isopropanol, and found that the liquid phase dehydrogenation of isopropanol using reactive distillation needed less energy and was much more efficient.

The performance of the IAH CHP was found to be greatly affected by some parameters. The increase of reflux ratio, exothermic reaction temperature and endothermic reaction temperature leads to the decrease of enthalpy efficiency and exergy efficiency of IAH heat pump. (Guo *et al*., 2014; Xin *et al*., 2014) Theoretical analysis and experimental study of IAH CHP system are of paramount importance to the industrial use of IAH CHP. Comparison of the effects of ultrasound and mechanical agitation on the isopropanol dehydrogenation was investigated (Xu *et al*., 2015), the results indicated that the superiority of the IAH-CHP system with ultrasound was present even if more than 50% of the power of the ultrasound equipment was lost.

Nevertheless, previous studies of IAH CHP mainly focused on the dehydrogenation of isopropanol and hydrogenation of acetone by either simulation or experiment. In this paper, we conduct a comprehensive analysis of the system operating performance. In order to get the optimal operation conditions, we build an experimental system with a newly designed reactive distillation column and an exothermic reactor. Based on the experimental results, we propose some principles for the design and optimum operation of IAH CHP.

2. IAH CHP SYSTEM

A schematic of the IAH CHP experimental system is shown in Figure **1**. The test rig mainly consists of seven sections, including an endothermic reactor, a distillation column, a pre-heater, an exothermic reactor, a regenerator, a pump and data acquisition system. The endothermic reactor is a tank using an electric heater with a temperature controller to mimic the waste heat. The regenerator is a shell-and-tube heat exchanger. Raney nickel catalyst is filled in the reactors and distillation column to accelerate the reaction. All the components and line are made of stainless-steel and insulated to reduce the heat loss.

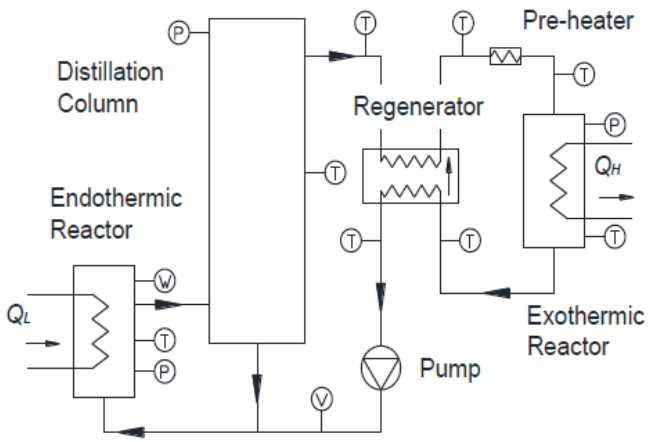


Figure 1: Schematic of the IAH CHP and the data acquisition system. P: pressure sensor, T: thermocouple, V: flow meter, W: voltage & current meter.

Prevost and Bugarel (1980) proposed that the dehydrogenation of isopropanol and hydrogenation reaction of acetone as follows:

$$(CH_3)_2 CHOH(l) \rightarrow (CH_3)_2 CO(g) + H_2(g)$$
$$\Delta H_L = 100.4 kJ \cdot mol^{-1} \quad (1)$$

$$(CH_3)_2 CHOH(l) \rightarrow (CH_3)_2 CHOH(g)$$
$$\Delta H = 45.4 kJ \cdot mol^{-1} \quad (2)$$

$$(CH_3)_2 CO(g) + H_2(g) \rightarrow (CH_3)_2 CHOH(g)$$
$$\Delta H_H = -55.0 kJ \cdot mol^{-1} \quad (3)$$

where ΔH_L represents the endothermic reaction enthalpy.

The dehydrogenation of isopropanol takes place in the endothermic reactor and absorbs heat at temperature T_L to yield acetone and hydrogen. The products including acetone, hydrogen and unreacted isopropanol, are fed into the distillation column in which isopropanol is separated and fed back to the endothermic reactor. The mixture of acetone and hydrogen flows to the regenerator and the exothermic reactor after being heated up to the reaction temperature in the pre-heater which only works when the system starts.

The hydrogenation of acetone takes place in exothermic reactor at temperature T_H and releases heat. The product isopropanol is then introduced to the regenerator and subsequently pumped back to the endothermic reactor to close a cycle.

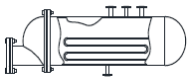
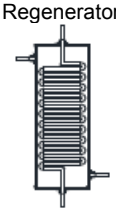
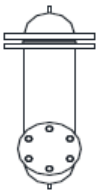
In the experiment, some cases are set to study the characteristics of the IAH CHP cycle: the endothermic temperature is set from 343K to 393K and the flow rate is set from 1.2ml/s to 393ml/s. In each test case, data is recorded after stable running of the system for 15 minutes. Then we can get the COP and exergy data to evaluate the performance.

3. FIDELITY ANALYSIS

The IAH CHP system was instrumented to measure the parameters of interest to evaluate the performance, such as temperature, pressure, and flow rate; locations of the monitoring points are shown in Figure **1**. Temperature is measured with K-type thermocouples with an accuracy of ±0.5 K. There are 9 temperature monitoring points in total, positioned in the endothermic and exothermic reactor, at the inlet and outlet of the regenerator and the pre-heater, on the surface of the column, endothermic and exothermic reactor, and the environment, respectively. The energy consumed by the electric heater and pump is measured with the electrical voltage and current meter with an accuracy of 0.5%. The pressure in the endothermic reactor, distillation column and the exothermic reactor is measured with pressure sensors with an accuracy of 0.5%. The flow rate is measured with a flow meter with an accuracy of 4.0%. And two sampling holes are set for chromatographic analysis at the inlet and outlet of the exothermic reactor.

And the components used in the system were shown in the Table **1**:

Table 1: System Components Parameters (Exemplified with the Endothermic Temperature 85℃ and Flow Rate 1.47ml/s)

Component	Energy Flow	Fidelity %	Efficiency %
Endothermic reactor 	Input: $Q_{in} = U \cdot I$ Output: $Q_{out} = Q_C + h \cdot A \cdot \Delta T$	90.75	82.9
Regenerator 	Input: $Q_{in} = Q_C$ Output: $Q_{out} = Q_C' + h \cdot A \cdot \Delta T$	94.55	78.6
Exothermic reactor 	Input: $Q_{in} = Q_C$ Output: $Q_{out} = Q_C' + h \cdot A \cdot \Delta T$		93.3
Pump 	Input: $Q_{in} = U \cdot I + Q_C$ Output: $Q_{out} = E_H + h \cdot A \cdot \Delta T$		98.2

Using the Coleman and Steele method (Coleman *et al*., 1989) and the ANSI/ASME standard (ANSI/ASME, 1986), we can obtain the measurement error of the COP and exergy. The relative uncertainty of COP and exergy are calculated as 8.36% and 6.52%, respectively.

Taking the fidelity of the endothermic reactor as an example, the energy flow is shown as Figure **2**. The energy input into the component is supplied by the electric heater. The energy output includes the heat loss through the surface and the chemical energy from the dehydrogenation. The energy absorbed and released can be obtained as follows:

$$Q_{in} = U \cdot I \tag{4}$$

$$Q_{out} = Q_C + h \cdot A \cdot \Delta T \tag{5}$$

$$Q_C = v \cdot w \cdot \Delta H_L \tag{6}$$

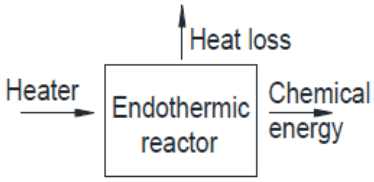


Figure 2: Energy flow of component (exemplified with endothermic reactor).

Here, *U* and *I* represent the voltage and current, respectively; *h* represents the heat transfer coefficient; *A* represents the heat transfer area; ΔT represents the temperature difference; Q_C represents the chemical energy that was absorbed in the reaction, and *v* and *w* represent the flow rate and content, respectively.

With the meteorological chromatographic analysis we get that the fidelity is 90.75%. So we think that the result of the experiment is reliable. Similarly, the other

components' system fidelity can be obtained and are shown in Table **1**.

4. THERMODYNAMIC ANALYSIS

In this study, the following assumptions were handled in the energy and exergy analysis (Acar *et al*., 2017):

1. Steady state conditions were taken into account for all the components,
2. Pressure losses in the pipelines were neglected, and the heat loss is not considered.
3. Enthalpy of liquid is a function of temperature only, independent on the pressure.
4. Different phases in the fluid are always in phase-equilibrium state
5. No side reaction and other productions in the reaction.
6. The catalytic effect does not change during the life cycle.
7. The purity of the rectification is 100%.
8. The reference state is 101.325 kPa, and 298.15 K.

Under these assumptions, the governing energy equations for IAH CHP were obtained as follows.

Heat absorbed:

$$Q_L = \Delta H_L + \Delta H_{vAcl} + \Delta H_{vIsl}[y_I/(1-y_I)] \quad (7)$$

where Q_L represents the waste heat, ΔH_{rd} represents the reaction heat, ΔH_{vAcl} represents vaporization heat of acetone, ΔH_{vIsl} represents vaporization heat of unreacted isopropanol and y_I represents the isopropanol content.

Heat released:

$$Q_H = v \cdot \alpha_H \cdot \Delta H_H \quad (8)$$

where Q_H represents the heat released, v represents the flow rate of the acetone, α_H represents the reaction rate of the hydrogenation, and ΔH_H represents the reaction heat.

The coefficient of performance (COP) can be expressed as:

$$COP = \frac{Q_H}{Q_L + W_t} = \frac{\Delta H_H}{\Delta H_L + \Delta H_{vAcl} + \Delta H_{visl}[y_I/(1-y_I)] + W_t} \quad (9)$$

where W_t represents the energy consumed by the pump can be got from the power meter.

Exergy (E_X) represents the amount of energy present in a system that is available for useful work. Exergy efficiency (η) is used to evaluate the efficiency of a process by taking the second law of thermodynamics into account.

The exergy is defined as:

$$E_{X,Q} = \left(1 - \frac{T_0}{T}\right) \cdot \delta Q \quad (10)$$

$$\eta = \frac{E_{X,out}}{E_{x,in}} = \frac{\left(1 - \frac{T_0}{T_H}\right) \cdot \delta Q_H}{\left(1 - \frac{T_0}{T_L}\right) \cdot \delta Q_L + W} \quad (11)$$

where T_0 represents the environmental temperature.

And the parameters of different state can be obtained as follows.

Vaporization heat of isopropanol or acetone:

$$\Delta H_{v2} = \Delta H_{v1}[(1 - T_2/T_{cr})/(1 - T_1/T_{cr})]^n \quad (12)$$

n=0.380 for acetone, n=0.405 for isopropanol:

$T\left(^oC\right)$	ΔH_{vAcl}	ΔH_{vIsl}	T_{cr} (K)
56.4	29.10		iP：508.3
25.0	30.99	45.39	Ace：508.1
82.4		39.85	

Specific heat of isopropanol and acetone in liquid phase:

$$C_{P(iP)} = 500.641 - 3.756T + 1.2*10^{-2}T^2 - 1.067*10^{-5}T^3 \quad (13)$$

$$C_{P(AC)} = 231.767 - 1.159T + 0.4*10^{-2}T^2 - 3.2*10^{-6}T^3 \quad (14)$$

Specific heat of isopropanol and acetone in gas phase:

$$C_{P(iP)} = 32.43 + 1.885*10^{-1}T + 6.406*10^{-5}T^2 - 9.261*10^{-8}T^3 \quad (15)$$

$$C_{P(AC)} = 6.301 + 2.606*10^{-1}T - 1.253*10^{-4}T^2 - 2.038*10^{-8}T^3 \quad (16)$$

$$C_{P(H)} = 27.14 + 9.274 * 10^{-3} T - 1.381 * 10^{-5} T^2 + 7.645 * 10^{-9} T \quad (17)$$

The reaction heat calculated by the temperature as the parameter:

$$\Delta H_L = 82261.638 - 241.734T + 1.30314 * T^2 - 2.6713 * 10^{-3} T^3 + 1.866941 * 10^{-6} T^4 \quad (18)$$

$$\Delta H_H = -53139.911 - 1.011T - 4.0687 * 10^{-2} T^2 + 6.7723 * 10^{-5} T^3 - 3.015875 * 10^{-8} T^4 \quad (19)$$

5. RESULTS AND DISCUSSION

At a given running condition, we studied the heat production, COP and exergy efficiency as a function of T_L, T_H and v.

Firstly, the relationship between temperature and COP, also between temperature and exergy efficiency were calculated, as shown in Figures **3** and **4**. Then some experiments were carried out to compare the actual results with the theoretical analysis. At a given endothermic temperature or flow rate, the influences of exothermic temperature and flow rate on heat production, COP and exergy efficiency were studied respectively and the variations are shown in Figures **5-8**.

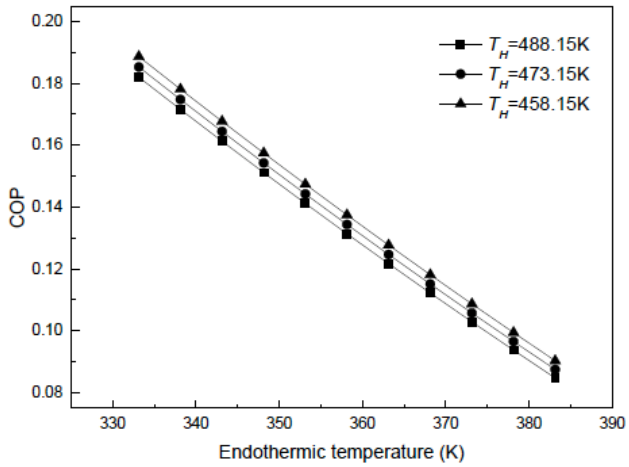


Figure 3: COP with various endothermic temperatures.

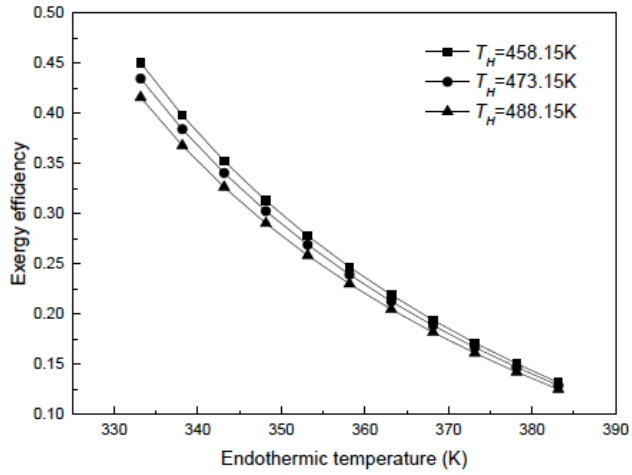


Figure 4: Exergy efficiency with various endothermic temperatures.

As shown in Figure **3**, COP decreases as the endothermic reaction and exothermic reaction temperature increases. The cause is not only the effect of temperature on reaction rate, but also the changes of the pressure. The higher the endothermic temperature, the larger the isopropanol vapor enthalpy is and the higher the vapor pressure is. The vapor in the endothermic reactor consumes more energy from the heater as the pressure increases. And also higher endothermic temperature drives more mixture into the distillation column including the unreacted isopropanol. The increased uncreated isopropanol absorbs the heat but no dehydrogenation to convert the energy. All these lead to larger Q_L; therefore, the COP decreases. But in the subsequent experimental analysis, the variations are different from which are shown in Figures **5** and **6**. As the endothermic temperature increases, exothermic temperature, heat production, COP and exergy efficiency all increase and reach the maximum when the endothermic temperature is about 360K, then decrease as the endothermic temperature further increases. The main reason for this difference is the distillation column in the experimental system. There is a decrease of temperature due to the heat loss from the distillation column, which is not considered in numerical calculations. Endothermic reactions take

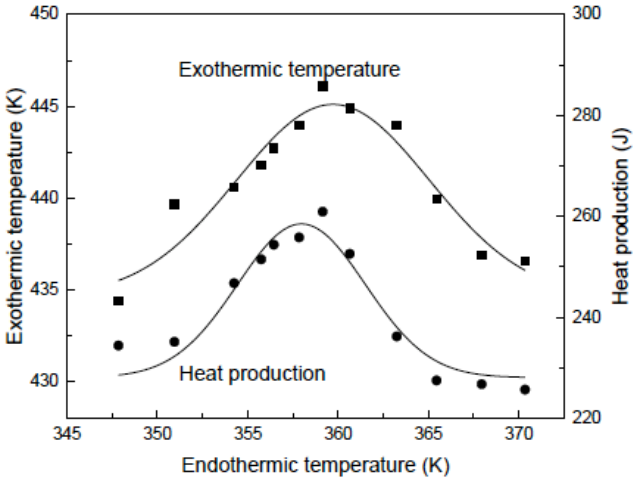


Figure 5: Exothermic temperature and heat production with various endothermic temperatures.

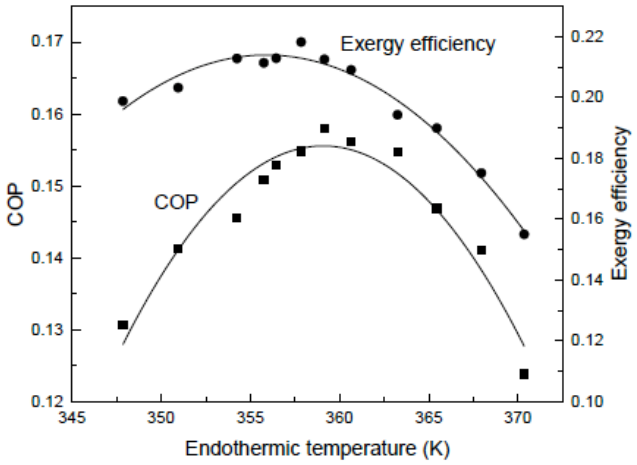


Figure 6: COP and exergy efficiency with various endothermic temperatures.

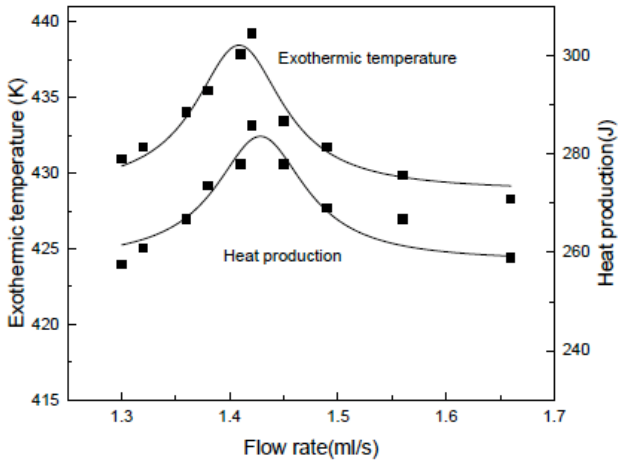


Figure 7: Exothermic temperature and heat production with various flow rates.

place mainly at 353K, but the temperature in the distillation column is lower than that in the endothermic reactor. It needs more heat to keep the separation efficiency. So the exothermic temperature, heat production and COP increases before the temperature rises to 360K. When T_L is above 360K, the temperature in the distillation column is above the boiling point of isopropanol. The vapor pressure and isopropanol increases with the increase of T_L which leads to larger Q_L and lower COP. Therefore, the higher the temperature, the lower the COP, the exothermic temperature and the heat production is.

Figures **7** and **8** display the influence of flow rate at a given endothermic temperature. It has a very similar effect to the influence of endothermic temperature as discussed above. The exothermic temperature, heat production, COP and exergy efficiency all reach the maximum point at a flow rate of 1.4 ml/s. The flow rate increases by adjusting heating power and the pressure of the distillation column. Under the condition of keeping the purity of the production, the endothermic temperature, exothermic temperature, heat production, COP and exergy efficiency would increase with the flow rate. But in the actual experiment process, the purity would decrease after the flow rate reaches about 1.40 ml/s. With the decrease of the purity, more isopropanol flows to the exothermic reactor and consumes more energy, leading to lower Q_H. Therefore, the exothermic temperature, heat production, COP and exergy efficiency would decrease with the flow rate.

CONCLUSIONS

Based on the experiment results and earlier discussion, the following conclusions can be drawn from this study:

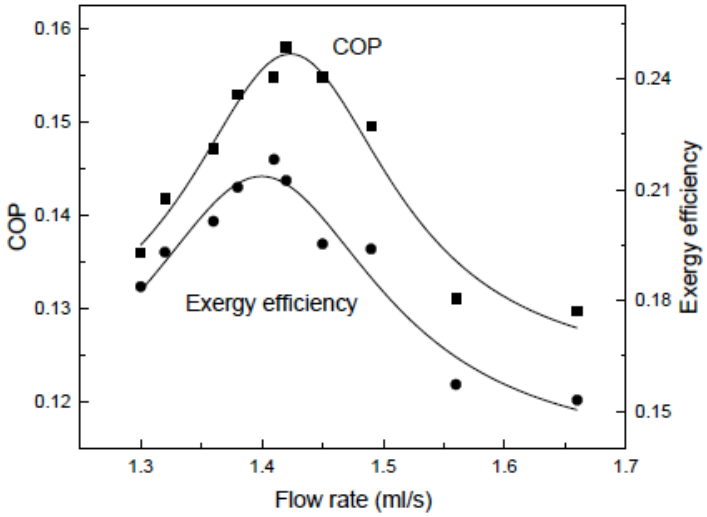


Figure 8: COP and exergy efficiency with various flow rates.

1. The endothermic temperature and flow rate has significant effects on hydrogen production and the separation of isopropanol. The system performance reaches the optimum at temperature of about 360 K and flow rate of 1.40 ml/s in the actual experimental system.

2. The design and heat insulation of the distillation column is important to the performance of the system. A better design leads to a higher reaction rate and better separation of isopropanol. Good heat insulation can reduce the heat loss and energy consumption.

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