ON-CHIP MICROSCALE DISTILLATION FOR ACETONE-WATER SEPARATION

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ABSTRACT

A silicon-based microscale distillation chip was fabricated consisting of a micro-channel 600 μm in width and 40 cm in length. Micropillars were incorporated in the micro-channel to direct liquid flow from the cooled to the heated end by capillarity. A feed of a 19 mol% acetone-water mixture was separated to 95+ mol% acetone in the distillate and 11 mol% in the bottom stream. This distillation corresponds to three equilibrium stages at total reflux conditions. The purity of the bottom stream could be increased by increasing the distillate flowrate, but at the expense of the distillate stream purity.

KEYWORDS: Microdistillation, microchannel, micropillars, separation

INTRODUCTION

Although there has been a great deal of work on the development of microchemical processing systems such as microreactors, only limited attention has been paid to microseparation units. The objective of this work is therefore to develop a silicon-based microscale distillation chip. In binary distillation, a liquid feed of two components with different volatilities is separated into a bottom product enriched in the less volatile component, and a distillate product enriched in the more volatile component. Due to its high efficiency and inherent capacity for purifying samples in small volume, microdistillation is potentially applicable to a number of fields such as pharmaceuticals research and development, environmental sampling and blood analysis.

The miniaturization of a distillation process is associated with a few challenges. Conventional distillation requires gravitational force to drive reflux liquid from the top of the column to the bottom. As a microchannel has narrow width, typically in the range of less than 1000 μm, gravitational force is insignificant compared to capillary force which is induced by surface tension [1]. Thus, the liquid flow has to be directed by other driving forces. Another issue is that the liquid flow in microchannel is commonly in the laminar regime, in which the liquid is not well-mixed.

Previous work has demonstrated the feasibility of microscale distillation by the introduction of an inert carrier gas. Wootton and de Mello [1] utilized helium for the vapour transport from the heating zone to the cooling zone in their micro-distillation device. The achieved separation of an equimolar mixture of acetonitrile and dimethylformamide was equivalent to 0.72 theoretical plates. Recently, Hartman et al. [2] developed a single stage microdistillation device. Segmented flow of liquid was produced by introducing nitrogen before feeding to the distillation chip. After vapour saturation, the nitrogen gas was separated from the liquid slugs by a membrane separator. An equimolar mixture of methanol in toluene and an equimolar mixture of dichloromethane in toluene were separated, resulting in 79mol% methanol and 63 mol% dichloromethane in the distillate product respectively. The drawback in using carrier gas is that no liquid reflux flows back to the reboiler, which limits the separation performance of the distillation to maximum one equilibrium stage. This is similar to differential distillation (i.e. Rayleigh distillation).

Microscale distillation without carrier gas introduction was attempted by Hibara et al. [3], who developed a micro-nano combined structure for the separation of 9.0 wt% of an ethanol-water mixture. The reported bottom stream consisted of 8.6 wt% ethanol while the distillate contained 19 wt% ethanol. Boyd et al. [4] used a localised heating method to achieve distillation, but did not present any detailed performance tests for mixture separation.

The objective of this work is to design a multi-stage microdistillation chip, which employs capillarity for liquid movement through integrated microengineered structures. Similar approaches have been presented by Seok and Hwang [5] and Sundberg et al. [6], but for mesoscale distillation and utilising foreign wick materials for liquid flow.

EXPERIMENTAL

In this work, the microscale distillation chip was fabricated on silicon wafer using conventional semi-conductor processing techniques. The chip consisted of a micro-channel 600 μm in width and 40 cm in length. To set up the temperature profile, one end of the chip was mounted on a heating block while the other end was clamped on a cooling block. The temperatures of both heating and cooling blocks were adjusted by individual temperature controllers. The feed was located at 10 cm (i.e. 1/4 of the channel length) from the heated end. The chip contained one outlet for the distillate (cooled side) and one for the bottom product (heated side). Numerous micro-pillars at the sides of the micro-channel were fabricated to guide the liquid flow from the cooled to the heated end by capillarity. It should be noted that there was no carrier gas supplied to the system.

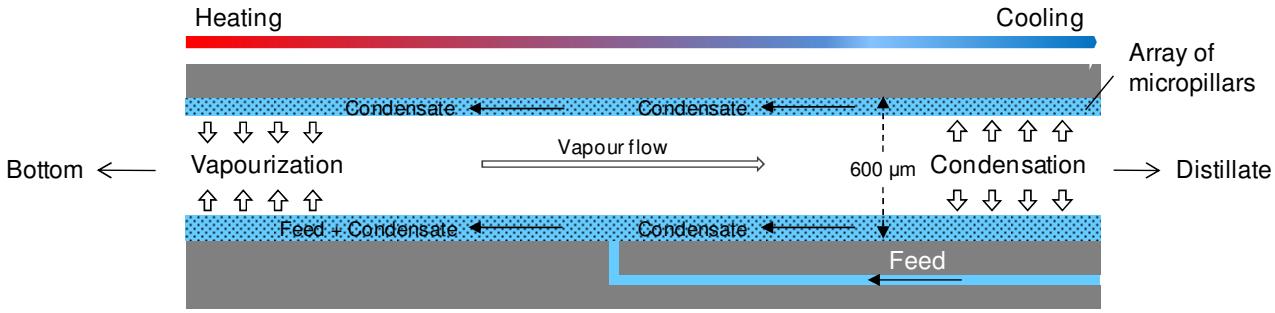

Figure 1: Operating principle of the microdistillation chip illustrated at the feed location

Throughout the experiment, liquid samples were collected from the distillate outlet at different time intervals. The samples were weighed, before being analyzed by gas chromatography using a Thermal Conductivity Detector (Agilent 6890N). The samples were measured twice and average values were calculated. The experiment was assumed to be at steady state when the concentration in the distillate remained constant. The composition of the bottom product was calculated by a simple mass balance since the flowrates and compositions of the feed and distillate were known. The volumes of the distillate and bottom were doubled-checked at the end of the experiment to confirm the closure of mass balance. Liquid flow inside the microchannel was examined by an optical microscope (Keyence VHX-600) during the experiment. An acetone-water mixture was used to evaluate the separation performance of the micro-distillation. The acetone (99% purity) was purchased from Aldrich and de-ionized water was supplied in-house.

RESULTS AND DISCUSSION

The fabricated microdistillation chip is shown in Figure 2a. The micropillars in the microchannel are shown in detail in Figure 2b. Each of the micropillars is 20 µm in diameter and the distance between them is also 20 µm. The array of micropillars on the side of the feed is wider to accommodate the additional feed flow.

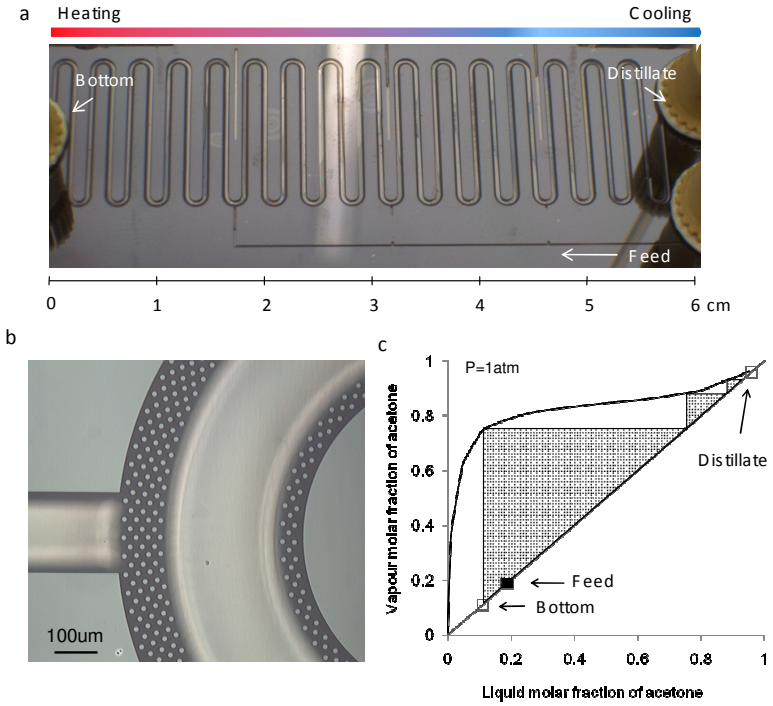

Figure 2: Characteristics of the microscale distillation chip: (a) chip configuration, (b) micro-pillars at the feed location and (c) performance of a 19 mol% acetone-water mixture separation

The separation of a 19 mol% (i.e. 50 vol%) acetone-water mixture was performed with the heating and cooling temperatures 85 ^{o}C and 42 ^{o}C, respectively. The feed flowrate was 0.15 ml/hr whereas the flowrates at bottom and distillate outlets were 0.04 and 0.11 ml/hr, respectively. The separation performance is demonstrated by the McCabe Thiele diagram (at total reflux), as shown in Figure 2c. The feed of 19 mol% acetone was separated to purer than 95 mol% acetone in the distillate and 11 mol% acetone in the bottom stream. The distillation corresponds to at least three equilibrium stag-

es. The number of equilibrium stages was estimated at total reflux conditions because the liquid and vapour flows inside the microchannel were unknown.

The effect of the distillate flowrate on the separation performance of the microdistillation chip was investigated. The experimental conditions of the rest of the parameters were unchanged. Table 1 summarizes the separation of the 19 mol% acetone-water mixture. High purity of acetone was obtained when the flowrate of distillate was lower than 0.04ml/hr, which is equivalent to approximately one-third of the feed flowrate. On the other hand, the bottom contained 11-14 mol% of acetone at these conditions. When the distillate flowrate increased, the purity of acetone in the distillate decreased and the purity of water in the bottom product increased. These observations are also commonly observed in conventional distillation columns [7]. At low distillate flowrate, the reflux stream is larger and this increases the purity of the light component at the distillate end. On the other hand, when the distillate flowrate is high, it implies a low reflux flow back to reboiler and thus resulting in a poorer separation for the distillate. There is thus a trade-off between product quantity and purity.

Table 1. Separation of 19 mol% acetone-water mixture at 0.15 ml/hr feed flowrate with varying distillate flowrates

Distillate		**Bottom**	
Flowrate (ml/hr)	Acetone composition (mol%)	Flowrate (ml/hr)	Acetone composition (mol%)
0.02	95+	0.13	14
0.04	95+	0.11	11
0.05	90	0.10	8
0.08	69	0.07	2

CONCLUSION

In this work, a multi-stage microdistillation chip has been demonstrated for the separation of acetone-water. The chip consisted of a serpentine microchannel with micropillars incorporated on both sides of the channel. The array of micropillars were necessary to aid the liquid flow from the cooling to the heating region. The experimental results showed high separation efficiency. The feed of 19 mol% acetone was separated into 95+ mol% acetone in the distillate and 11 mol% in the bottom stream, respectively. The distillation performance corresponded to at least three equilibrium stages at total reflux conditions and this result is one of the best microdistillation separations reported in the literature. The purities of the product streams were shifted by changing the distillate flowrate.

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