

# Mass Transfer Rate in Liquid-liquid Extraction of Coal Tar Absorption Oil Using a Spray Column

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## Introduction

Coal tar, one of the byproducts from coal carbonization, contains many useful compounds to chemical industry. Absorption oil (AO) which is one of the fractions (b.p.=470~550 K) contains nitrogen heterocyclic compounds (nitrogen compounds) such as quinoline (Q), isoquinoline (IQ), indole (I), etc., and the other compounds such as naphthalene (N), 1-methylnaphthalene (1MN), 2-methylnaphthalene (2MN), biphenyl (BP), dibenzofuran (DBF) etc. Generally, these compounds in the absorption oil are separated by reactive extraction with acid and/or base. However, this process is relatively costly because of difficulty of recovering the acid and base and of corrosion of the equipment by the acid and base. For this separation, the ordinary solvent extraction with aqueous methanol (MeOH aq.) has been suggested as alternatives to the reactive extraction and the various kinds of efforts are devoted to apply this ordinary technique to the absorption separation [1-9]. In this study, the relationship of the mass transfer coefficient in a spray column[7], one of the simplest practical extractors, is examined for designing the separation process.

## 1. Experimental

The equipment used in this work was the same as in previous work [7]. The column was made of Pyrex glass and behavior in the column could be observed. The distributor of dispersed phase had 8 needle nozzles. The distributor of continuous phase had outlets on the side of cylinder to pretend falling drops entering in it. First, AO and MeOH aq. were prepared in the tanks. AO, dispersed phase, was fed to the top of the column, and MeOH aq., continuous phase, to the bottom and these two phases were contacted countercurrently. The flow rates were kept constant by checking the flow indicators. The level of interface between accumulated dispersed phase and continuous phase at the bottom was maintained by adjusting the drain valve of dispersed raffinate phase. The inlet and outlet flow rates were determined by weighing the mass collected in about 10 minutes. The holdup of dispersed phase in the column was determined by measuring the rise of the interface when the feed of AO and drain were stopped at the same time. Each phase was analyzed by GC-17AAF Ver.3 (Shimadzu Corp.) to determine the composition. The density of each phase was measured by pycnometers. The principal experimental conditions are summarized in **Table 1**. The liquid-liquid equilibrium relationships were also measured for estimating the mass transfer coefficients, beforehand.

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**Table 1:** Experimental conditions

No.	feed: absorption oil	solvent: aqueous methanol	
	$R_t$ [ $\text{kg} \cdot \text{h}^{-1} \cdot \text{m}^{-2}$ ]	$y_{w,b}$ [-]	$E_b$ [ $\text{kg} \cdot \text{h}^{-1} \cdot \text{m}^{-2}$ ]
1	$8 \times 10^2 \sim 3 \times 10^3$	$0.5 \pm 0.001$	$2 \times 10^3 \pm 500$
2	$9 \times 10^2 \sim 4 \times 10^3$	$0.7 \pm 0.004$	$2 \times 10^3 \pm 400$
3	$1 \times 10^3 \sim 5 \times 10^3$	$0.5 \pm 0.005$	$3 \times 10^3 \pm 600$
4	$4 \times 10^2 \sim 5 \times 10^3$	$0.7 \pm 0.007$	$3 \times 10^3 \pm 200$

## 2. Results and Discussion

### 2.1 Fundamental relationship

The yield,  $Y_i$ , and the separation selectivity relative to 2MN of component  $i$ ,  $\beta_{i/2MN}$ , were defined as follows:

$$Y_i = E_i \cdot y_{i,v} / (R_t \cdot x_{i,v}) \quad (1)$$

$$\beta_{i/2MN} = y_{i,v} / y_{2MN,v} / (x_{i,b} / x_{2MN,b}) \quad (2)$$

The overall mass transfer coefficient of the continuous phase,  $K_{c,i}$  was calculated by,

$$d(E \cdot y) / dz = -K_{c,i} \cdot a \cdot (y_i^* - y_i) \quad (3)$$

where  $y_i^*$  was estimated from  $x_i$  with the equilibrium relationship and  $a$  was estimated from  $d_p$ .

### 2.2 Yield

The yields calculated by Eq.(1) are plotted versus  $R_t$  in **Fig.1**.  $Y_i$  with the solvent of  $y_{w,b}=0.3$  were higher than those with  $y_{w,b}=0.5$ . In this study,  $Y_i$  of nitrogen compounds were higher than those of other compounds and these groups of compounds could be separated into each other group. The maximum yield of nitrogen compounds came up to around  $R_t=2 \times 10^3 \text{ kg} \cdot \text{h}^{-1} \cdot \text{m}^{-2}$ .

### 2.3 Separation selectivity

The separation selectivity calculated by Eq.(2) are plotted versus  $R_t$  in **Fig.2**.  $\beta_{i/2MN}$  did not change so much as  $R_t$  changed.  $\beta_{i/2MN}$  of nitrogen compounds with the solvent of  $y_{w,b}=0.5$  were higher than those with  $y_{w,b}=0.3$ .

### 2.4 Mass transfer rate

**Fig. 3** gives the effect of the superficial mass flow rate of dispersed raffinate phase,  $R_t$ , on the overall volumetric mass transfer coefficient,  $K_{c,i} \cdot a$ , obtained by Eq.(3).  $K_{c,i} \cdot a$  increased with increasing  $R_t$ , mainly because the specific interfacial area,  $a$ , increased with  $R_t$ . However, at Run No.4 condition,  $K_{c,i} \cdot a$  did not change with increasing  $R_t$ . This is because at these conditions, the axial dispersion of the raffinate phase was significant.

In general, the relationship among the overall and local height per transfer units can be represented as follows:

$$H.T.U_{OC,i} = H.T.U_{c,i} + \frac{E_b}{R_t} \cdot H.T.U_{d,i} \quad (4)$$

The intercept and slope in the plot of  $H.T.U_{OC,i}$  against  $\frac{E_b}{R_t}$  are, thus,  $H.T.U_{c,i}$  and  $H.T.U_{d,i}$ , respectively, provided these local  $H.T.U.s$  are constant independently of  $\frac{E_b}{R_t}$ ,  $E_b$ , and  $R_t$ . This plot with the results in this study is presented in **Fig.4**. The result showed that in the range of this

study  $H.T.U_{oc,i}$  increased as  $\overline{m}_i \cdot E_b / R_i$  increased. However, some data did not show the tendency especially in the range of higher  $\overline{m}_i \cdot E_b / R_i$ . From this result, in this separation process, the continuous phase was more dominant than the dispersed phase in the range of  $\overline{m}_i \cdot E_b / R_i$  lower than 2.

### Conclusion

In this study, the experiment was carried out favorably under the conditions. The better relationship of the mass transfer coefficient was obtained by estimating  $H.T.U$ , which is one of the factors to calculate the height of the columns. It was known that the continuous phase was dominant in the separation process and from this study since the dispersed phase has a little resistance, the dominant of the continuous phase had been confirmed. More consideration need to be done since some data were not in the relationship obtained.

### Acknowledgement

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### Nomenclature

$a$	=	specific interfacial area	$[m^2 \cdot m^{-3}]$
$d_p$	=	drop diameter of the dispersed phase	$[m^2 \cdot m^{-3}]$
$E$	=	superficial mass flow rate of the continuous phase	$[kg \cdot h^{-1} \cdot m^{-2}]$
$H$	=	holdup	$[-]$
$K_c$	=	overall mass transfer coefficient of the continuous phase	$[kg \cdot h^{-1} \cdot m^{-2}]$
$R$	=	superficial mass flow rate of the dispersed phase	$[kg \cdot h^{-1} \cdot m^{-2}]$
$V$	=	volume in the column	$[m^3]$
$x$	=	mass fraction in the dispersed phase	$[-]$
$y$	=	mass fraction in the continuous phase	$[-]$
$Y$	=	yield	$[-]$
$Z$	=	effective contact height of column	$[m]$
$z$	=	distance from top of the column	$[m]$
$\beta_{i/2MN}$	=	separation selectivity of component $i$ relative to 2MN	$[-]$

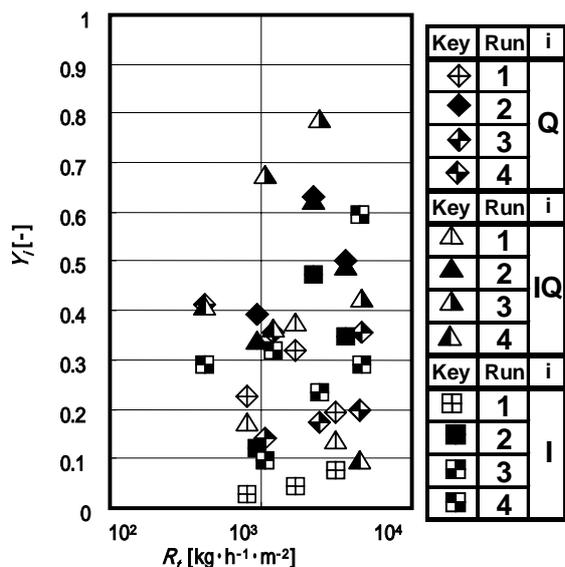


Fig.1 change of yields against  $R_i$

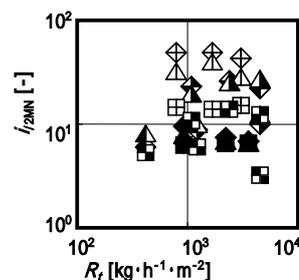


Fig.2 change of  $i/2MN$  against  $R_i$

(the keys are same as Fig.1)

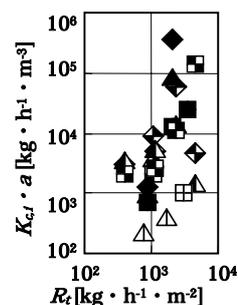


Fig.3 change of  $K_{c,i} \cdot a$  against  $R_i$

(the keys are same as Fig.1)

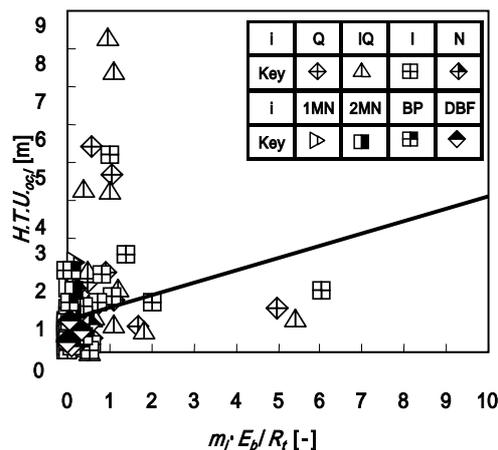


Fig.4 change of  $H.T.U_{oc,i}$  against  $m_i \cdot E_b / R_i$

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