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Determination of Absorption Capacity and Mass Transfer Coefficient of Several Oils as Solvents for Biomass Gasification Cleaning System

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Abstract: With the aim to find an absorbing liquid suitable for the gas cleaning system in the application of gasification unit in a remote area, three types of oil were tested using a bubble column and a wetted wall column. Air streams containing toluene vapour with flowrates of 13.6 mL·min^{-1} or 27.6 mL·min^{-1} were bubbled through a 50 mL static oil in a glass tube at a temperature of 30 ^{o}C or 60 ^{o}C. In experiments using the wetted wall column, air streams containing toluene were contacted with a falling thin film of oil on the outer wall of a column with a diameter of 6.4 cm and two different contacting heights of 60 cm or 80 cm. Toluene concentrations in the air stream were adjusted in the range of 700-3000 ppm corresponding to a typical tars concentration in the producer gas. The phase equilibrium of toluene was represented as values of *1/H* of 326, 220 and 182 respectively for lubrication oil, palm oil and sunflower oil (*H* is Henry's constant with the toluene concentrations in g·L^{-1} for liquid phase and g·Nm^{-3} for gas phase). From experiments using the bubble column, it was found that the overall mass transfer coefficient ($K_G a$) was in the order of 10^{-3} cm^3·min^{-1} and the overall liquid phase mass transfer coefficient ($K_L a$) was about 10^{-3} cm^3·min^{-1}. Although lubrication oil had a slightly better absorption capacity than the other two tested oils, it had a lower mass trasfer coefficient than that of palm oil. All three proposed oils had a much better absorption capacity and absorption rate than that of water used conventionally as a scrubbing liquid in a small biomass gasification plant.

Key words: Henry's constant, absorption of VOC, scrubbing oil, tars removal.

Nomenclature

a	Interfacial area (cm^2)
$C_{G,i}$	Solute concentration in gas phase at the inlet stream (mg·m^{-3}) and (mg·L^{-1})
$C_{G,o}$	Solute concentration in gas phase at the outlet stream (mg·m^{-3})
$C_G^{\#}$	Solute concentration in gas phase at interface (mg·m^{-3})
C_L	Solute concentration in liquid phase (mg·m^{-3})
$C_{L,i}$	Solute concentration in liquid phase at the inlet stream (mg·m^{-3})
$C_L^{\#}$	Solute concentration in liquid phase at interface (mg·m^{-3})
$C_{L,eq.}$	Equilibrium solute concentration in liquid phase (mg·L^{-1})
∂_L	Liquid film thickness (mm)
H	Henry's constant (mg·L^{-1}/mg·L^{-1})
L	Height of wetted wall column tube (cm)
L_f	Height of liquid film (cm)
k_G	Gas phase mass transfer coefficient (cm·s^{-1})
k_L	Liquid phase mass transfer coefficient (cm·s^{-1})
$K_G a$	Overall gas phase mass transfer coefficient (cm^3·min^{-1})
$K_L a$	Overall liquid phase mass transfer coefficient (cm·s^{-1})
N	Mass transfer (mol·s^{-1})
r	Radius of small inner column (cm)
Re	Reynolds number
Q_G	Gas flowrate (mL·min^{-1})
Q_L	Liquid flowrate (mL·min^{-1})

1. Introduction

Producer gas from biomass gasification process

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contains particulates and tars which may cause severe operational problems in the gas application to an internal combustion engine (IC-engine). In this type of application, the producer gas must only have particulates contents of less than 50 mg·Nm^{-3} and tars content of less than 100 mg·Nm^{-3} [1]. The conventional water scrubbing system has been reported to give a clean gaseous fuel, but several reports also mention that some tars still escape through this type of cleaning system and enter diesel-engine resulting in serious operation and maintenance problems. Such a problem in the gasification field unit which is coupled with a 60 kW diesel-generator set also is found [2]. Thus a gas cleaning system is an important part in integrating a gasification unit to an IC-engine.

A typical composition of tar from the biomass gasification plant is listed in Table 1 [3]. Tars component escapes from the conventional cleaning system may consist of organic compounds having low boiling points, such as toluene and benzene. These compounds are difficult to be removed completely using scrubbing water [4]. There are many reports related to the absorption of volatile organic compounds (VOC) from air stream concerning with environmental aspects. Effective solvents are usually organic compounds having following specifications [5]:

- a high capacity to absorb VOC;
- a low vapor pressure to reduce loss during regeneration by means of stripping and to prevent unwanted air pollution by absorbent;

Table 1 Typical composition of biomass tar [3].

Compound	Mass fraction (%)
Toluene	24
Other one ring aromatic hydrocarbons	22
Naphthalene	15
Other two ring aromatic hydrocarbons	13
Heterocyclic compounds	10
Phenolic compounds	7
Three ring aromatic hydrocarbons	6
Four ring aromatic hydrocarbons	1
Others	2

- no toxicity and no fire or explosion risks;
- a low viscosity and a high diffusion coefficient which control absorption kinetic.

Unfortunately, those organic solvents reported in literature are relatively expensive to be implemented for a biomass gasification plant operated usually in a remote area. For this application, solvent have to be available at low price locally and may be regenerated or reused in order not to produce another type of waste.

The aim of the experimental study is the selection of a better absorbing liquid for the gas cleaning system in a biomass gasification plant. The low boiling point tars components were represented using toluene. In this study, crude palm oil or waste engine lubrication oil as a solvent have been considered to use. Sunflower oil and water were included for a comparison purpose. The absorption experiments were carried out upto pressumable equilibrium conditions between the gas and liquid phases, i.e. when the solute concentration in the gas phase at the outlet approached that in the inlet. The absorption capacities of proposed solvents are represented using Henry's constant and the mass transfer rate are also examined.

2. Theoretical Background

The absorption experiments were conducted in a bubble column and a thin falling film column similarly used by many researchers [6-9]. The gas phase flowed continuously, while the absorbing liquid was operated batchwise.

2.1 Mass Balance in Absorption

If the solute concentrations in the gas phase can be measured both in the inlet and the outlet streams, the solute concentration in the liquid phase can be calculated using the following mass balance (Eq. (1)):

$$C_L(t) = \frac{Q_G}{V_L}\left(C_{G,i}t - \int_0^t C_{G,o}(t)dt\right) \quad (1)$$

The solute concentration in the liquid phase will approach a limit value of the saturation concentration ($C_{L,eq.}$). While the outlet concentration in the gas phase

($C_{G,o}$) will approach that of the inlet ($C_{G,i}$). When the equilibrium condition is achieved, the solute concentration in the gas phase at interface ($C_G^{\#}$) is related to that in the liquid phase using Henry's constant (H), as shown in Eq. (2):

$$H = \frac{C_G^{\#}}{C_L^{\#}} \quad (2)$$

Furthermore at the equilibrium condition, the solute concentration at interface is assumed almost the same as that at the outlet stream ($C_{G,o}$).

If the absorption process does not approach the equilibrium condition yet, the values of Henry's constant can also be calculated using a mass balance of the evolution of solute concentration in liquid phase (with assumption that perfectly mixes in liquid phase). Henry's constant and overall mass transfer coefficient in gas phase (K_Ga) also can be evaluated simultaneously as proposed by Mackay et al. [8], as shown in Eq. (3).

$$C_L = \frac{(K_Ga+Q_G)}{K_GaH} C_{G,o} - \frac{Q_G}{K_GaH} C_{G,i} \quad (3)$$

Similar to the above expression, but for the case of liquid phase mass transfer limitation, Henry's constant and overall mass transfer coefficient in liquid phase (K_La) can be calculated by using the following Eq. (4):

$$C_L = \left(\frac{1}{H} + \frac{Q_G}{K_La}\right) C_{G,o} - \frac{Q_G}{K_La} C_{G,i} \quad (4)$$

Values of H, K_Ga and K_La are then evaluated from the slope and intersept of the above two equations.

2.2 Wetted Wall Column

The wetted wall column has additional advantages over the bubble column, such as: (a) fixed and measurable surface area and (b) possibility for measurements of film thickness. If the absorbing liquid flowrate and its surface velocity (representing as the maximum velocity of flowing liquid) can be observed visually, then the liquid film thickness (∂_L) can be calculated using the following Eq. (5) [5]:

$$\partial_L = \frac{3}{4}\frac{Q_L}{V_{max}\pi r} \quad (5)$$

Mass transfer for absorption in gas phase at interface in the wetted wall column can be written as shown in Eq. (6):

$$N = k_Ga\left(C_G^{\neq} - C_G\right) \quad (6)$$

where: $a = 2\pi(r + \partial_L)L_f$. Mass transfer rate ($N$) can be calculated using the differential mass balance of solute along the wetted wall. Assumption is made that the solute concentration in the gas phase at interface ($C_G^{\neq}$) equals with that at the outlet gas stream ($C_{G,O}$). Differential mass balance of solute in the gas phase can be written for calculation of mass transfer rate with gas phase limitation (k_G), as shown in Eq. (7) [10].

$$k_G = \frac{Q_G}{2\pi(r+\partial_L)L_f} \ln\frac{C_{G,i}}{C_{G,o}} \quad (7)$$

Similarlly the case of liquid mass transfer limitation (k_L) is shown in Eq. (8).

$$k_L = \frac{Q_G}{2\pi(r+\partial_L)L_f} \ln\frac{C_{L,i}}{C_{L,s}} \quad (8)$$

From the definitions of overall mass transfer coefficients, the relation between the overall gas phase side mass transfer coefficient (K_Ga) and the film mass transfer coefficients (k_La) and (k_Ga) can be written in Eq. (9) [11].

$$\frac{1}{K_Ga} = \left(\frac{1}{k_Ga} + \frac{H}{k_La}\right) \quad (9)$$

Overall liquid side mass transfer (K_La) can be calculated by Eq. (10).

$$\frac{1}{K_La} = \left(\frac{1}{k_GaH} + \frac{1}{k_La}\right) \quad (10)$$

3. Material and Method

The experimental setup consists of three sections (Fig. 1): (a) a gas mixing section to prepare the gas stream containing solute; (b) an absorption section with the main unit of a bubble column or a wetted wall column; (c) an analysis section. The bubble column was made of a glass tube with a diameter of 6 mm and a height of 60 mm. Absorption experiments were carried out at a gas flowrate of 13.6 mL·min^{-1} or 27.6 mL·min^{-1}. The oil temperature was kept in constant at 30 ^{o}C or at 60 ^{o}C.

The wetted wall column is a vertical flexy tube with a diameter of 6.4 cm and a height of 100 cm. The height of thr mass transfer was adjusted to be 60 cm or 88 cm. A moving slice liquid distributor was made of glass for adjusting a gab between the inner cylinder

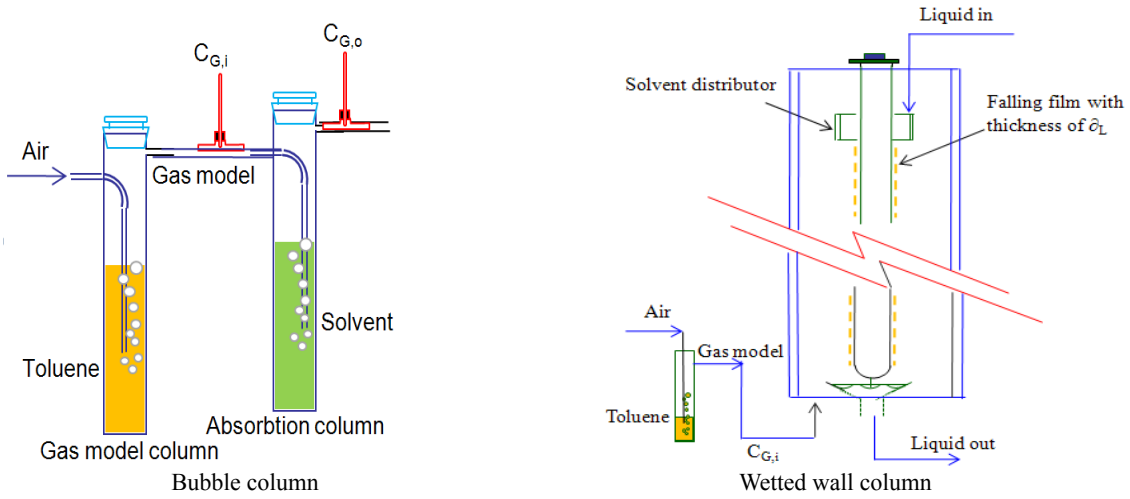


Fig. 1 Experimental setup.

Table 2 Main variables in absorption experiment.

Variable	Bubble column			Wetted wall column		
Volume of oil (mL)	50			250		
Gas flowrate (mL·min^{-1})	13.6 or 27.6			1214		
Liquid circulation flowrate (mL·min^{-1})	-	-	-	16.94	19.70	23.00
$C_{G,i}$ (mg·L^{-1})	2.38*	2.45*	2.25*	76	71	66
$C_{L,eq.}$ (mg·L^{-1})	618	660	502	22283	16261	12441
Saturation time (min)	720			330		

* for bubble column at the gas flowrate 27.6 mL·min^{-1}.

and the oil distributor. With this device, the film thickness (∂_L) was adjusted to be about 0.7 mm. This distributor was necessary to ensure the uniform liquid film on the outer surface of the column.

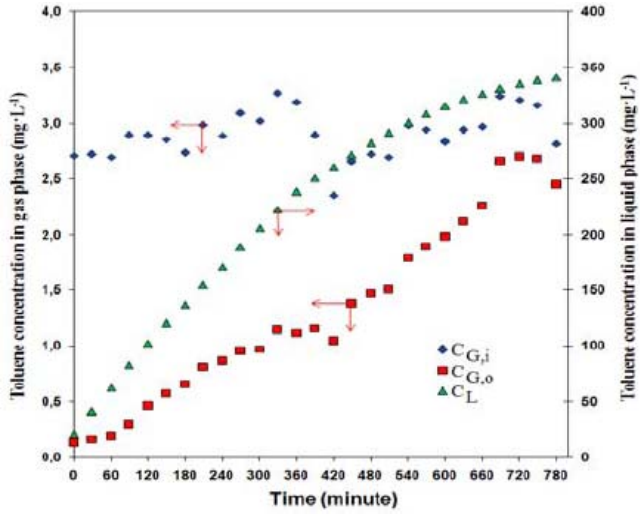
The gas flow was supplied by an oil free compressor, and the oil was continuously fed to the wetted wall column using a peristaltic pump. Air was mixed with toluene vapor in a thermostated bottle, so that toluene concentrations were about 70 ppm. The flowrate of the gas was adjusted of 1214 mL·min^{-1} (at 24 ^{o}C) and the oil flow rate was varied at 16.94 mL·min^{-1}, 19.70 mL·min^{-1} or 23.00 mL·min^{-1} (roughly corresponding to $1.5 < Re < 4.4$). At this condition, the laminar flow of thin film liquid could be obtained. The summary of experimental condition was listed in Table 2.

Analytical grade toluene $C_6H_5CH_3$, pure palm oil, sunflower oil and engine lubrication oil were obtained from a local chemicals supplier in Tokyo. Concentrations of toluene in the gas phase were measured by taking sample 1 mL and analyzed using a gas chromatography *Shimadzu GC-8APF* (with a separation column having a diameter of 3.2 mm and a length of 2.1 m and filled with 5% *shin carbon*).

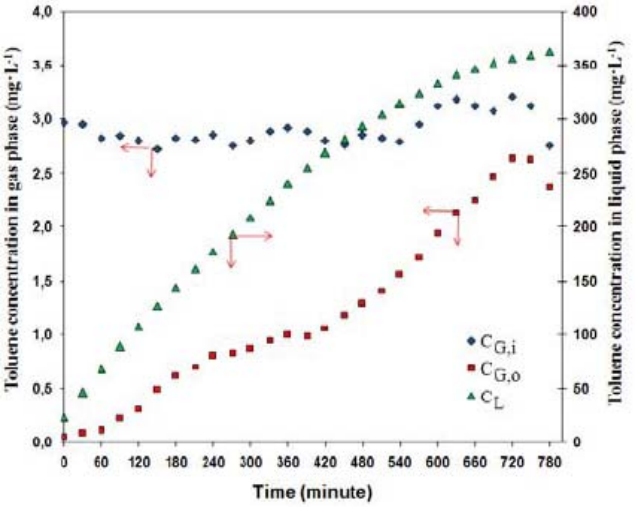
4. Results and Discussion

4.1 Absorption Capacity

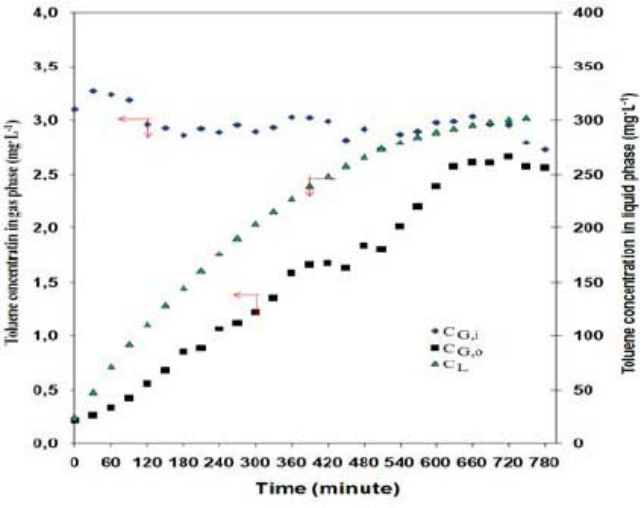
Examples of experimental data of toluene concentration in the gas phase at the inlet and outlet stream and calculated concentration of toluene in the liquid phase for experiments in the bubble column and the wetted wall column are presented in Figs. 2 and 3, respectively. From these figures, the progress of absorption could be examined. The concentration and time to attain saturation of toluene in the liquid phase clearly depended on the type of absorbing oil. As expected, all three types of oil had a much higher absorption capacity than water. Lubrication oil had the



Palm oil

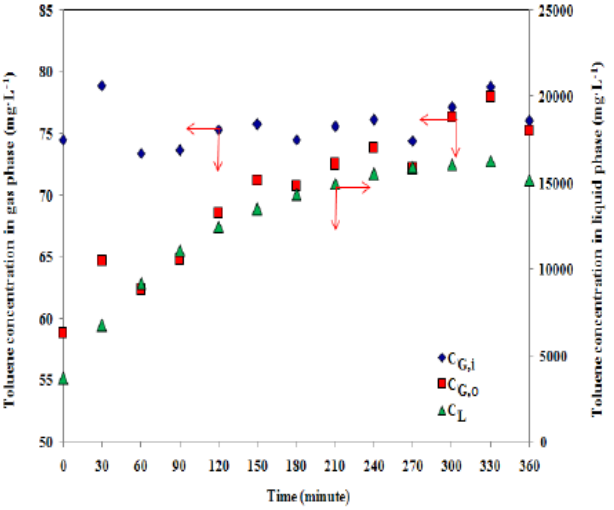


Lubrication oil

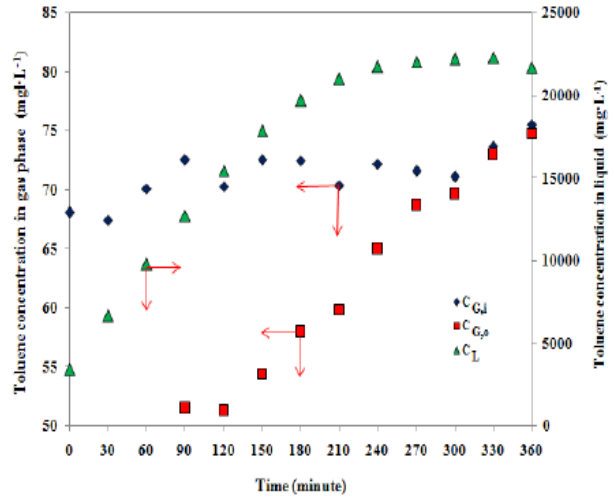


Sunflower oil

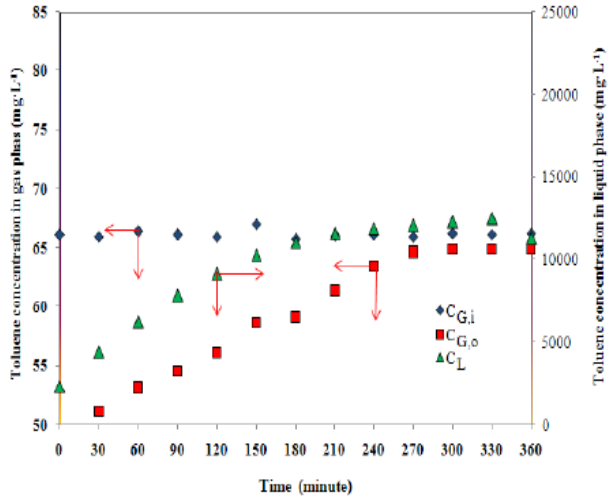
Fig. 2 Toluene concentration in gas and liquid phases from the bubble column experiment: Q_G = 13.6 mL·min^{-1} and T = 30 °C.



Palm oil



Lubrication oil



Sunflower oil

Fig. 3 Toluene concentration in gas and liquid phases from the wetted wall column experiment: Q_L = 16.94 mL·min^{-1} and L = 60 cm.

highest absorption capacity than other oils.

Henry's constant based on experiments in the bubble column and calculated using various equations as mentioned in Section 2 are presented in Table 3 and Table 4. Since Henry's constants calculated using Eqs. (2) and (3) were found to depend on the gas flowrate, the equilibrium condition between the gas and liquid phases apparently did not attain yet. While those calculated using Eqs. (3) and (4) were more or less constant. Thus the use of Eqs. (3) and (4) gave reliable results on absorption capacity even without attaining the equilibrium conditions.

Values of Henry's constant from experiments in the wetted wall column are presented in Table 5. As expected, the absorption capacities (values of *1/H*) were found to be independent of column height and the liquid flow rate. The lubrication oil was found to have a highest toluene absorption capacity among the tested oils. Meanwhile the absorption capacities of all oils were higher than that of water, as shown in Table 6. The differences in absorption capacity among oils and water might be due to their chemical properties, i.e. lubrication oil is aromatics; palm oil and sunflower oils are glyceride compounds containing O-element; water is a polar solvent. Compared to several commercial organic solvents [5, 9], all three proposed oils as absorbing liquid showed a satisfactory capacity for toluene absorption, as shown in Table 6.

The Henry's constants obtained in this experimental study were also used in the thermodynamic on vapour-

Table 3 Toluene concentration at equilibrium in gas and liquid phases in bubble column, Q_G = 13.6 mL·min^{-1}.

	Palm oil		Lubrication engine oil		Sunflower oil		Water	
	30 °C	60 °C	30 °C	60 °C	30 °C	60 °C	30 °C	60 °C
$C_{G,i}$ (mg·m^{-3})	2976	3307	2815	2956	3123	2791	2830	2905
$C_{G,o}$ (mg·m^{-3})	2380	2751	2449	2737	2249	2112	2773	2773
C_L (mg·L^{-1})	341	308	348	333	289	266	19	16
1/H (Eq. (2))	145	135	155	158	121	97	7	6
1/H (Eq. (3))	219	241	298	382	241	199	11	6
1/H (Eq. (4))	219	241	298	382	241	199	11	6

Table 4 Toluene concentration at equilibrium in gas and liquid phases in bubble column, Q_G = 27.6 mL·min^{-1}.

	Palm oil		Lubrication engine oil		Sunflower oil		Water	
	30 °C	60 °C	30 °C	60 °C	30 °C	60 °C	30 °C	60 °C
$C_{G,i}$ (mg·m^{-3})	2976	3307	2815	2956	3123	2791	2830	2905
$C_{G,o}$ (mg·m^{-3})	2380	2751	2449	2737	2249	2112	2773	2773
C_L (mg·L^{-1})	618	544	660	613	502	463	27	22
1/H (Eq. (2))	255	224	321	283	190	165	10	8
1/H (Eq. (3))	198	180	310	432	193	146	5	4
1/H (Eq. (4))	198	180	310	432	193	146	5	4

Table 5 Toluene concentration at equilibrium in gas and liquid phases in wetted wall column, Q_G = 1214 mL·min^{-1}; with the height of column (*L*) as parameter.

Liquid flow rate, Q_L (mL·min^{-1})	Film ∂_L (mm) (Eq. (5))	Palm oil *L* (cm)		Lubrication oil *L* (cm)		Sunflower oil *L* (cm)	
		60	88	60	88	60	88
16.94	0.515	220	219	321	327	187	183
19.70	0.851	218	222	327	328	165	197
23.00	0.625	220	221	327	327	176	185
Average, *1/H* (Eq. (2))		219	221	325	327	176	188
Average, *1/H* (Eq. (2))		220		326		182	

liquid equilibrium using UNIFAC-FV [12]. As vegetable oils are mixtures of saturated and unsaturated triglycerides, palm oil and sunflower oil as pseudo-triacyglyceride were represented:

$[(CH_2COO)_2CHCOO](CH=CH)_m(CH_2)_n(CH_3)_3$

The term in the square brackets represents the triglyceride functional-group. Parameters *m* and *n* are used to reproduce the molecular weight and the degree of unsaturation of the original oil. It was found that UNIFAC-FV could be used for the prediction of Henry's constant in the cases which were provided with a slight adjustment on two additional parameters in Free-Volume model, named C_1 and *b* related to solvent characteristic.

4.2 Mass Transfer Coefficient

Overall mass tranfer coefficients, K_G and K_L from experiments in the bubble column were calculated by Eqs. (3) and (4), while those from experiments in the wetted wall column were calculated by Eqs. (9) and (10). Results of calculations are presented in Table 7 and Table 8, respectively. Mass tranfer coefficients (k_G and k_L) from experiments in the wetted wall column were calculated by Eqs. (7) and (8) and were also presented in Fig. 4.

Mass transfer coefficients in liquid phase were found much smaller than those in the gas phase. These fenomena usually occur in the absorption of slightly soluble solute in liquid such toluene-water and toluene-oils in this experiment. In the experiments using the bubble column, values of K_Ga decreased with increasing temperature and flowrate. At higher temperature and flowrate, there was probably some evaporation of solvent which resulted an inhibition to

Table 6 Henry's constant for various solvents and solutes.

Solute	Solvent	*1/H* (*H* = mol·L^{-1}/mol·L^{-1}), 25 °C		Experiment
		Bubble column	Wetted wall column	
Toluene	Palm oil	209	220	Our experiments
	Engine lubrication oil	304	326	
	Sunflower oil	220	182	
	Water	5	-	
		4	-	
	di(2-ethylhexyl) adipate	2821	-	[9]
	n-Hexadecane	990	-	
	Polyethylene glycol 400	1645	-	
Acetone	Silicone oil	52	-	[5]

Table 7 Mass transfer coefficients of toluene in oils in bubble column.

Q_G (mL·min^{-1})	*T* (°C)	Palm oil	Lub. oil	Sunflower oil	Water
		K_Ga (cm^3·min^{-1}) (Eq. (4))			
13.6	30	9.33×10^{-1}	6.17×10^{-1}	5.22×10^{-1}	0.10×10^{-1}
	60	2.56×10^{-1}	4.11×10^{-1}	2.20×10^{-1}	0.12×10^{-1}
27.6	30	2.86×10^{-1}	2.75×10^{-1}	1.53×10^{-1}	0.23×10^{-1}
	60	1.21×10^{-1}	8.76×10^{-1}	1.16×10^{-1}	0.23×10^{-1}
		K_La (cm^3·min^{-1}) (Eq. (5))			
13.6	30	4.27×10^{-3}	2.07×10^{-3}	2.17×10^{-3}	0.89×10^{-3}
	60	4.15×10^{-3}	1.07×10^{-3}	5.05×10^{-3}	2.12×10^{-3}
27.6	30	1.45×10^{-3}	0.89×10^{-3}	0.79×10^{-3}	4.34×10^{-3}
	60	0.67×10^{-3}	0.20×10^{-3}	0.80×10^{-3}	9.31×10^{-3}

Table 8 Mass transfer coefficients of toluene in oils in wetted wall column.

Q_L (mL·min^{-1})	Palm oil, L (cm)		Lub. oil, L (cm)		Sunflower oil, L (cm)	
	60	88	60	88	60	88
			K_Ga (cm^3·s^{-1}) (Eq. (9))			
16.94	13.68	13.30	11.85	13.32	19.52	26.74
19.70	27.09	33.25	22.73	31.95	28.88	39.13
23.20	34.31	45.10	29.00	39.82	36.05	49.36
			K_La (cm^3·s^{-1}) (Eq. (10))			
16.94	0.06	0.06	0.04	0.04	0.11	0.15
19.70	0.12	0.15	0.07	0.10	0.16	0.21
23.20	0.16	0.20	0.09	0.12	0.20	0.27

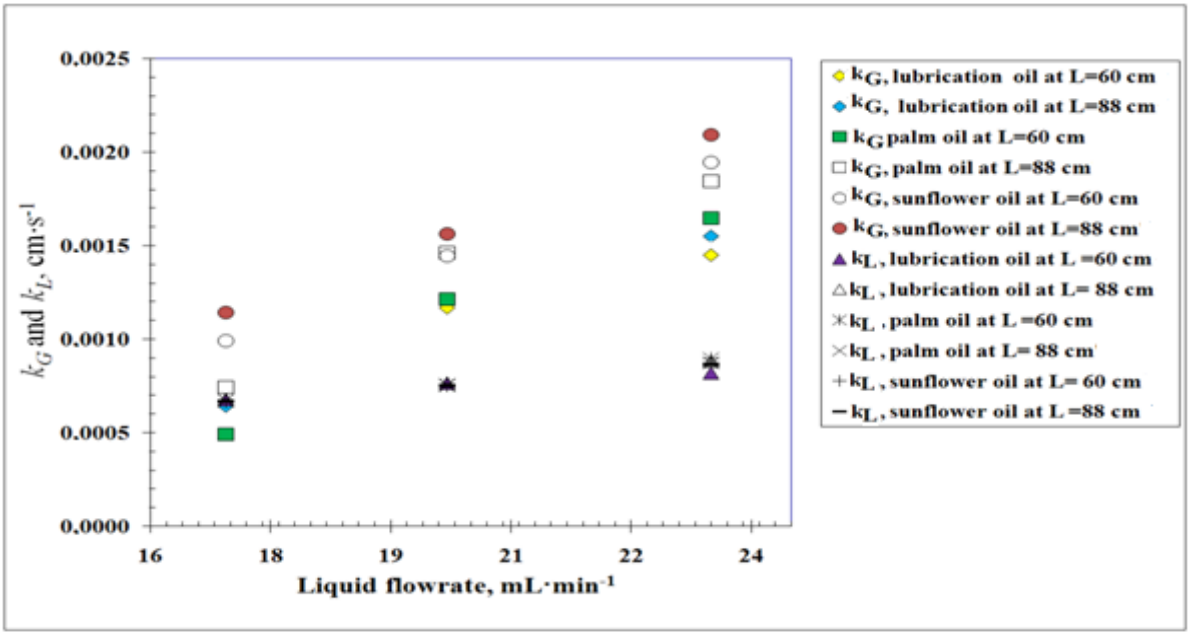


Fig. 4 Mass transfer coefficients from experiments in wetted wall column (calculated using Eqs. (7) and (8)).

absorption at the interface. On the other hand, K_La decreased with increasing temperature due probably to decreasing solvent viscosity.

From experiments in the wetted wall column, liquid phase mass transfer was also found to give more contribution, similar to those from experiment using the bubble column. In the wetted wall column, both K_Ga and K_La depended on the liquid flow rate (Table 8). The effect of types of solvent was clearly observed on the values of k_G, but not on the values of k_L (Fig. 4). The increase in the rate of mass transfer with increasing the thickness and height of film were probably due to formation of wavy film at a high liquid flowrate.

Although lubrication oil seems to be the best solvent for toluene absorption, it has a lower K_La and k_L (Table 8 and Fig. 4). However, both k_L and K_La depend on the design of the column used to perform the experiments and regarding Eqs. (7) and (8), k_G will certainly be higher than k_L. The low mass transfer rate of toluene in lubrication oil probably is due to its higher viscosity which in turn strongly influences the rate of diffusion and the hydrodynamic.

5. Conclusions

For just the selection of absorbing liquid based on its capacity, the simple bubble column can be used satisfactorily. Without waiting for the equilibrium, the absorption capacity can be evaluated from the transient data using Eq. (3) or Eq. (4).

As a more slightly complex apparatus, the wetted wall column can be used for evaluating both absorption capacity and mass transfer coefficient with a better results than the bubble column. Data from experiments in this wetted wall column may be used further for evaluating the diffussivity of toluene in oils. Results from both experiments using bubble and wetted wall columns show that the mass tranfer of toluene in air-oils system is limited by diffusion in liquid film. The value of mass transfer coefficient in the gas phase was in the order of 10^3 higher than that in the liquid phase. Alhough lubrication oil had the highest absorption capacity, it had the lowest mass transfer rate among the proposed oils. So the preference in the use of lubrication oil and palm oil is more or less similar, so the choice will depend on their availablity locally. Instead of fresh or new oils, waste lubrication oil or crude palm oil has been considered to be used in the cleaning system for the implementation of biomass gasification in remote area, such as in oil-palm plantation.

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