Abstract—Studies on gas solid mass transfer using Supercritical fluid CO2 (SC-CO2) in a packed bed of palm kernels was investigated at operating conditions of temperature 50 °C and 70 °C and pressures ranges from 27.6 MPa, 34.5 MPa, 41.4 MPa and 48.3 MPa. The development of mass transfer models requires knowledge of three properties: the diffusion coefficient of the solute, the viscosity and density of the Supercritical fluids (SCF). Matematical model with respect to the dimensionless number of Sherwood (Sh), Schmidt (Sc) and Reynolds (Re) was developed. It was found that the model developed was found to be in good agreement with the experimental data within the system studied.

Keywords—Mass Transfer, Palm Kernel, Supercritical fluid.

I. INTRODUCTION

MATHEMATICAL modeling of complex phenomena, such as the extraction of natural materials, is important from economic point of view. It is important to develop models for the extraction process when the extraction operations are optimized for commercial applications. However, such predictions require the establishment of model which can predicts phase behavior, equilibrium, solubility, adsorption, desorption and others. Relationships, as well as models for equipment design should take into consideration the effect of fluid flow, mass and heat transfer and also the phase contacting mechanisms.

The development of mass transfer models require an understanding of three important properties namely, the diffusion coefficient of the solute, the viscosity and the density of the supercritical fluid (SCF) phase. These properties are important in the correlation of mass transfer coefficients [1].

There is still different in opinions regarding the determination of the correlations model for the mass transfer coefficient of supercritical fluid flowing inside the packed bed columns. According to Lim et al. [1], the mass transfer correlations between a fluid and solid, in a packed bed of solids can be described in the form of Equation (1):

\[ Sh = f(Re, Sc, Gr) \]  (1)

Where:

- \( Sh \) = Sherwood Number (related to mass transfer).
- \( Re \) = Reynolds Number (fluid flow).
- \( Sc \) = Schmidt Number (related to diffusivity).
- \( Gr \) = Grashof Number (related to heat transfer).

On the other hand, as pointed by Debenedetti and Reid [2], the buoyant effects is important consideration factor in supercritical fluids because the fluids could show extremely small variations in the kinematics viscosities for high densities or low dynamic viscosities. For the same Reynolds number, the effect of buoyant forces in supercritical fluids is two times greater in the order of magnitude compared to in normal liquids. However, when natural convection is the controlling factor, the effect of Reynolds number is no longer significant. Then, the general correlation expression for the mass transfer relationship is given by Equation (2),

\[ Sh = f(Sc, Gr) \]  (2)

Nevertheless, for a large Schmidt number (usually in a liquid-solid system) Karabelas et al. [3] proposed the correlation relationship in a natural convection as given by Equation (3):

\[ Sh = a(Sc Gr)^b \]  (3)

But, Lim et al. [1], pointed out that, if forced convection is the controlling factor, then the Grashof number is insignificant and the general expression is given by Equation (4).

\[ Sh = f(Re, Sc) \]  (4)

According to Damronglerd et al. [4], some studies suggested that data of Sherwood, Schmidt, and Reynolds are generally correlated in the form as in Equation (5):

\[ Sh = c Re^{1/3} Sc^{1/3} \]  (5)

In this study, the Grashof number is considered insignificant because all pressure applied in this study were above the critical pressure of carbon dioxide (CO2). Moreover, according to Lim et al. [1], above critical pressure, forced convection dominated. These conditions generally, would be associated with greater velocities than the natural convection. The changed in fluid density as the fluid is heated up was small and always almost negligible. Therefore, no buoyant effects could be produced. The changed in fluid density, however, was much dependent on pressure rather than temperature. A study by Eggers and Sievers [5], on the scaling up of a packed bed of evening primrose seed by using supercritical carbon dioxide (SC-CO2) extraction method,
pointed out that only three dimensionless numbers namely, the Sherwood (Sh), Schmidt (Sc) and Reynolds (Re) numbers were considered essential. Therefore, the mass transfer correlation model in this study, followed the general expression in Equation (6):

\[ \text{Sh} = f(\text{Re}, \text{Sc}) \]  

(6)

II. EXPERIMENTAL SET UP

The laboratory scale supercritical fluid (SC-CO\(_2\)) extraction system model ISCO, Inc., Lincoln, NE. U.S.A. was used in the study. The SC-CO\(_2\) extraction system comprises: a carbon dioxide cylinder, with 99.99 % purity of CO\(_2\), a chiller, to liquefied CO\(_2\) gas; a high pressure syringe pump, with maximum operating pressure of 68.95 MPa, and an extractor, with size 22.7 cm by 21.2 cm by 24.4 cm equipped with a 2.5 ml stainless steel extraction vessel. In addition, the system also comprised of a heated capillary restrictor for reducing analyte deposition, with outside diameter 50 \(\mu\)m and maximum operating temperature of 150 °C; and a 30 ml vial for collection of the analyte. Fig. 1 shows the schematic diagram of SC-CO\(_2\) extraction process for Palm Kernel Oil (PKO) extraction.

III. RESULTS AND DISCUSSION

The individual dimensionless numbers of mass transfer, Sherwood number (Sh), diffusivity (Schmidt number (Sc), and fluid flow Reynolds number (Re) developed in this study were statistically validated. The data of Sh, Sc and Re were tested to establish a correlation model (equation) which related to the ratio of Sherwood to Schmidt or (Sherwood/Schmidt\(^{1/3}\)) versus Reynolds number. The data of Sh, Sc and Re obtained from the experiments, were plotted to observe a trend of the Reynolds number (Re) versus a ratio of Sh to Sc to power of 1/3. The power “1/3” was introduced merely to modify the ratio of (Sh to Sc) to correct for temperature effect. These relationships are as shown in Fig. 2, Fig. 3 and Fig. 4 by a linear relationship. By reformulated these linear relationships mathematically, a general correlation/model was established.

The correlation of Sh/Sc\(^{1/3}\) versus Re shows that the bigger the Reynolds Number the higher would be the mass transfer rate since the ratio of (Sh/Sc\(^{1/3}\)) is related to the mass transfer. The high mass transfer rate with Reynolds number may be due to the large density differences that occur as palm kernel oil (PKO) dissolves in the SC-CO\(_2\).
Thus, from Fig. 2, Fig. 3 and Fig. 4, the three correlation models of the mass transfer for palm kernel oil extracted by Supercritical Carbon Dioxide (SC-CO$_2$) extraction method are summarized as in Equation (7) to Equation (9).

\[
\text{Sh} = 0.980 \text{Re Sc}^{1/3} - 1.925 \text{Sc}^{1/3} \quad (7)
\]
\[
\text{Sh} = 3.521 \text{Re Sc}^{1/3} - 11.679 \text{Sc}^{1/3} \quad (8)
\]
\[
\text{Sh} = 4.126 \text{Re Sc}^{1/3} - 11.553 \text{Sc}^{1/3} \quad (9)
\]

However, according to Damronglerd et al. [4], the second term of the above correlation equations can be ignored since it represents the contribution of molecular diffusion, which usually is small and negligible. The empirical correlation models of the mass transfer are reduced to Equation (10) to Equation (12).

\[
\text{Sh} = 0.980 \text{Re Sc}^{1/3} \quad (10)
\]
\[
\text{Sh} = 3.521 \text{Re Sc}^{1/3} \quad (11)
\]
\[
\text{Sh} = 4.126 \text{Re Sc}^{1/3} \quad (12)
\]

From the three correlation equations, Equation (10), is the best-fitted equation for correlating the observed (experimental) data of the Sherwood, Schmidt and Reynolds numbers over the entire range of pressures ranging from 27.6 MPa to 48.3 MPa as shown in Table I.

### TABLE I
VALIDATED EMPIRICAL MODELS OF MASS TRANSFER CORRELATIONS BASED ON DIMENSIONLESS NUMBERS OF SHERWOOD (SH), SCHMIDT (SC) AND REYNOLDS (RE) FOR SUPERCRITICAL CARBON DIOXIDE (SC-CO$_2$) EXTRACTION OF PALM KERNEL OIL (PKO) AT DIFFERENT TEMPERATURES AND PRESSURES

<table>
<thead>
<tr>
<th>Operating Parameters</th>
<th>Observed</th>
<th>Predicted</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (°C)</td>
<td>Pressure (MPa)</td>
<td>Re $\times 10^3$</td>
</tr>
<tr>
<td>50 °C</td>
<td>27.6</td>
<td>3.45</td>
</tr>
<tr>
<td>60 °C</td>
<td>41.4</td>
<td>4.67</td>
</tr>
<tr>
<td>70 °C</td>
<td>48.3</td>
<td>4.79</td>
</tr>
</tbody>
</table>

This model (Equation 10) is statistically validated as evidence by a good coefficient of correlation ($r^2$) more than 0.9. Since Equation (11) and Equation (12) in this study do not show a best-fitted correlation, thus, the equations were not applied to validate the Sherwood number and the mass transfer coefficient (K) of the palm kernel oil instead; Equation (10) was used to verify the Sherwood number and the mass transfer coefficient (K) throughout these experiments.

Another validation analysis was performed to establish the correlation between the observed and predicted values of the Sherwood number (Sh) which relates the mass transfer for palm kernel oil (PKO) by using the Supercritical Carbon Dioxide (SC-CO$_2$) extraction method. Table II shows a comparison between observed (experimental) and predicted (model) values of Sherwood number (Sh) for palm kernel oil (PKO) extracted from overall palm kernels in a packed bed of supercritical extractor at a constant consecutive temperatures of 50 °C, 60 °C and 70 °C and variation of pressures ranging from 27.6 MPa to 48.3 MPa.

### TABLE II
OBSERVED AND PREDICTED VALUES OF DIMENSIONLESS NUMBERS THE SHERWOOD (SH), THE SCHMIDT (SC) AND THE REYNOLDS (RE), FOR EXTRACTION OF PALM KERNEL OIL

<table>
<thead>
<tr>
<th>Operating Parameters</th>
<th>Temperature (°C)</th>
<th>Pressure (MPa)</th>
<th>Re $\times 10^3$</th>
<th>Sc $\times 10^{-1}$</th>
<th>Sh $\times 10^1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>50 °C</td>
<td>27.6</td>
<td>3.42</td>
<td>3.32</td>
<td>1.70</td>
<td>2.93</td>
</tr>
<tr>
<td>60 °C</td>
<td>41.4</td>
<td>4.67</td>
<td>3.72</td>
<td>1.77</td>
<td>3.29</td>
</tr>
<tr>
<td>70 °C</td>
<td>48.3</td>
<td>4.79</td>
<td>3.84</td>
<td>2.17</td>
<td>3.41</td>
</tr>
</tbody>
</table>

This model (Equation 10) is statistically validated as evidence by a good coefficient of correlation ($r^2$) more than 0.9. Since Equation (11) and Equation (12) in this study do not show a best-fitted correlation, thus, the equations were not applied to validate the Sherwood number and the mass transfer coefficient (K) of the palm kernel oil instead; Equation (10) was used to verify the Sherwood number and
The statistical analysis conducted as shown in Table III demonstrated that the Sherwood number (Sh) was found to be strongly correlated between the observed and predicted data with the correlation of determination ($r^2$) above 0.8 at the significant level ($\alpha$) of 0.05.

### IV. Conclusion

Extraction of palm kernel oil in a packed bed of palm kernels was conducted at supercritical conditions of a variation of temperatures and pressures of 50 °C, 60 °C and 70 °C; and 27.6 MPa, 34.5 MPa, 41.4 MPa and 48.3 MPa respectively. It was found that the best correlation model or equation of the mass transfer relating to diffusivity and fluids conditions generated by the empirical modeling process was $Sh = 0.980 \text{ReSc}^{1/3}$. The best-fitted correlation model was obtained at a constant temperature of 50 °C over the entire range of pressures from 27.6 MPa, 34.5 MPa, 41.4 MPa and 48.3 MPa.

### REFERENCES


Norhuda Ismail is a senior lecturer at the Faculty of Chemical Engineering, Universiti Teknologi MARA, Malaysia. She obtained her B. (Eng), in Chemical at Universiti Teknologi Malaysia in 1990, and her M.Sc. in Environment at Universiti Putra, Malaysia and her PhD in Chemical Process at the Universiti Sains, Malaysia. Her research interests are in mass transfer, environmental engineering, and separation processes particularly in the field of supercritical fluids. Norhuda was employed as a Quality Assurance Executive in the process industries for several years. She has research collaborated with several oil and gas industries such as AMOCO, Petroleum Nasional Malaysia (PETRONAS), BASF-PETRONAS, MTBE and also other industries such as Malaysia Institute of Nuclear Technology (MINT) and Malaysia Airlines System (MAS), in areas pertaining to Chemical Engineering. She is a member of Board of Engineers Malaysia since 1994.

Mohd Omar Abdul Kadir is currently a professor in the School of Industrial Technology, Universiti Sains Malaysia (USM). He obtained his Bachelor and Master Degree in Chemical Engineering at Mississippi State University, USA, and his PhD at MTU, USA. He joined USM in 1989 and is a professional Chemical Engineer active both in engineering research of Supercritical Fluid Technology as well as in the area of industrial wastewater treatment plant design and built, environmental audit and hazardous waste handling. He has a patent in supercritical fluid technology in palm kernel oil extraction and is also an active member of the Japanese Supercritical Research Group of Tohoku University. He is widely recognized in the area of industrial wastewater treatment. He is a consultant to many local and international companies in the same area. He has over 150 papers in refereed journals, proceedings, presented at international and local conferences. He is a committee member of the National Air Quality Committee, member of American Institute of Chemical Engineers (AIChE), U.S.A. since 1993, member of Specialist Group on Small Wastewater Treatment Plants (IAWQ), United Kingdom and others.