# THERMAL HYDROCRACKING OF ATHABASCA BITUMEN: CORRELATION OF REACTOR VOIDAGE IN VERTICAL TWO-PHASE FLOW 

A.M. Shah, B.B. Pruden and J.M. Denis

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THERMAL HYDROCRACKING OF ATHABASCA BITUMEN:
CORRELATION OF REACTOR VOIDAGE
IN VERTICAL TWO-PHASE FLOW
by

A.M. Shah*, B.B. Pruden** and J.M. Denis***

## ABSTRACT

Pilot-plant experiments were carried out to determine the reactor voidage in vertical two-phase flow of hydrogen and Athabasca bitumen at various operating conditions of temperature ( $330-430^{\circ} \mathrm{C}$ ), pressure ( $3.55-13.89 \mathrm{MPa}$ ), liquid feed rate ( $2100-12,500 \mathrm{~g} / \mathrm{h}$ ) and recycle gas rate ( $0.028-0.085 \mathrm{~m}^{3} / \mathrm{h}$ at system pressure and $20^{\circ} \mathrm{C}$ ). These conditions are within the range at which thermal hydrocracking takes place.

The linear relationship, $\varepsilon=m V_{G}$, between the reactor voidage, $\varepsilon$, and the superficial gas velocity, $V_{G}$, was found to apply in all cases. The value of the slope, $m$, depended on the reactor geometry and experimental conditions. The variation of $m$ with pressure and temperature was best represent-
 effect on $m$ for the system at 13.89 MPa and $400^{\circ} \mathrm{C}$ in a tubular reactor. However, the voidage increased with increased $V_{L}$ at constant $V_{G}$ in an annular reactor.

Investigation of vertical two-phase flow in a tubular reactor of first, kerosene and hydrogen at room temperature and different pressures, i.e., 3.55-17.34 MPa, and second, water and nitrogen at room temperature and atmospheric pressure also indicated a linear relation between the voidage and the superficial gas velocity. The superficial liquid velocity did not have any effect on the voidage in either case and the pressure dependency for the kerosene-hydrogen system was similar to that observed for the hydrogenbitumen system.

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# L'HYDROCRAQUAGE THERMIQUE DU BITUME DE L'ATHABASCA: CORRELATION DE LA FRACTION DE VIDE DU REACTEUR dans l'ecoulement vertical en deux phases 

## par

A. M. Shah*, B.B. Pruden** et J.M. Denis***

RESUME

Des expériences en usine pilote ont été effectuées afin de déterminer la fraction de vide du réacteur dans l'écoulement vertical en deux phases de I'hydrogène et du bitume de l'Athabasca à différentes conditions de température $\left(330-430^{\circ} \mathrm{C}\right)$, de pression ( $3,55-13,89 \mathrm{MPa}$ ), de vitesse d'alimentation en liquide ( $2100-12500 \mathrm{~g} / \mathrm{h}$ ) et du taux de circulation du gaz de recyclage ( $0,028-0.035 \mathrm{~m}^{3} / \mathrm{h}$ à la pression du système et à $20^{\circ} \mathrm{C}$ ). Ces conditions se trouvent dans la gamme où se produit l'hydrocraquage thermique.

Les auteurs ont déterminé que le rapport linēaire ( $\varepsilon=\mathrm{mV}_{\mathrm{G}}$ ) entre la fraction de vide du réacteur ( $\varepsilon$ ) et la vitesse du gaz dans I'appareil vide ( $V_{G}$ ) s'appliquait dans tous les cas. La valeur de la pente (m) dépendait de la forme du réacteur et des conditions d'expérience. La variation de m selon la pression et la température a été représentée le mieux par $1^{\prime}$ expression $m \alpha P^{0}, 18 \mathrm{~T}^{2,11}$. La vitesse du liquide dans $I^{\prime}$ appareil vide ( $V_{L}$ ) n'a pas eu d'effet important sur m pour le système à $13,89 \mathrm{MPa}$ et $400^{\circ} \mathrm{C}$ dans un réacteur tubulaire. Toutefois, la fraction de vide a augmenté avec $I^{\prime}$ accroissement de $V_{G}$ à $V_{L}$ constant dans un réacteur annulaire.

La recherche d'écoulement vertical en deux phases dans un réacteur tubulaire, tout d'abord dans le cas du kérosène et de l'hydrogène à température ambiante et sous différentes pressions, c'est-à-dire 3,55-17,34 MPa, puis dans celui de l'eau et de l'azote à température ambiante et sous pression atmosphérique, les auteurs ont également établi une relation linéaire entre la fraction de vide et la vitesse du gaz dans l'appareil vide. La vitesse du liquide dans $l^{\prime}$ appareil vide n'a eu aucun effet sur la fraction
 système kérogène-hydrogène était semblable à celle observée dans le cas du système hỳdrogène-bitume.

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## INTRODUCTION

Research has been in progress for a number of years at the Canada Centre for Mineral and Energy Technology (CANMET) for the purpose of developing a hydrogenation process to upgrade low grade petroleum reserves of Canada. CANMET scientists (1) have developed a thermal hydrocracking process to upgrade Athabasca bitumen which increases distillate yield by 10 to $15 \%$ and thus offers an excellent alternative method to delayed coking and flexicoking in the first stage refining of the bitumen to distillable hydrocarbons. This first stage refining or upgrading step is necessary because Athabasca bitumen normally contains about $50 \%$ pitch, i.e., the fraction boiling above $525^{\circ} \mathrm{C}$. Research is being continued to further develop the process and to determine optimum operating conditions for the maximum utilization of non-renewable resources. The objective of the present investigation was to determine the correlation of the reactor voidage, i.e., the fraction of the reactor volume occupied by vapours or gas holdup, with thermal hydrocracking processing parameters to assist in the engineering of commercial-scale plants. This is in accordance with the goals of the Energy Research Program of the Canada Centre for Mineral and Energy Technology of the Department of Energy, Mines and Resources and its policy of ensuring the effective use of Canada's mineral and energy resources.

In the thermal hydrocracking process, liquid bitumen feed and re-cycle-hydrogen gas enter at the bottom of the reactor and move up. The twophase flow in the vertical reactor is complicated by vaporization of feed and product, heat and mass transfer, and hydrocracking and hydrogenation reactions. As has been observed in a glass column with other gas-liquid systems (2), the flow pattern in vertical reactors is such that bullet-shaped bubbles are separated by slugs of liquid which may contain small bubbles. Visual observations are difficult to make during the thermal hydrocracking reaction which occurs at elevated temperatures and pressures. Two-phase flow in vertical as well as horizontal pipes without accompanying chemical reactions has been investigated in the past by several workers. However, no publications have been issued concerning the voidage in a hydrogen-bitumen or a hydrogen-oil system at high pressure and temperature. A knowledge of the voidage or gas holdup in vertical two-phase flow serves a practical purpose in facilitating calcula-
tion of the average gas and liquid residence times, and in determining the average bubble rise velocity. This project was undertaken for these reasons to determine the voidage under various thermal hydrocracking operating conditions of temperature $\left(330-430^{\circ} \mathrm{C}\right)$, pressure ( $3.55-13.89 \mathrm{MPa}$ ), superficial liquid velocity ( $0.075-0.436 \mathrm{~cm} / \mathrm{s}$ ) and superficial gas velocity ( $0.3-4.7$ $\mathrm{cm} / \mathrm{s}$ ). In addition, two-phase flow in the systems of kerosene/hydrogen and water/nitrogen was investigated to verify the experimental procedure and establish the effect of pressure with less complex systems which could be compared with published data.

Previous Work
Two-phase gas-liquid flow is encountered in many petroleum and chemical industries. Extensive studies have been done on two-phase gasliquid flows in horizontal (3-7) and inclined pipes (8-10) and various correlations for the pressure drop, friction factor, gas holdup and liquid holdup and flow pattern maps have been developed and critically evaluated (11-13). Attention is focussed in this section on two-phase gas-liquid flow in vertical pipes. Dumitrescu (14), and Davies and Taylor (2) studied the mechanics of bubble flow and showed that the bubble rise velocity in stagnant liquid in vertical tubes is given by $C(g D)^{\frac{1}{2}}$, where $g$ is the gravitational acceleration and $D$ is the tube diameter. A value of 0.35 for the constant, $C$, was suggested from potential flow theory and it was verified from tube emptying experiments. Since then, extensive studies have been carried out on two-phase vertical flow (14-22). These investigations were carried out either in stagnant liquids or under fully developed flow conditions at or near atmospheric pressure and were concerned with characterization of two-phase flow, determination of slug shapes and sizes and the prediction of voidage, pressure drop and rise velocities. Most of the studies on two-phase vertical flow were with air and water or air and light-oil systems without chemical reactions. Two-phase flow as such is a complicated phenomenon and there is no sound theoretical basis for the prediction of gas holdup or pressure drop. However, numerous correlations have been developed based on experimental data for particular systems. Nicklin ( 17,18 ) derived an expression for the rise velocity of slugs, $U_{s}$, in steady two-phase flow in vertical tubes at or near atmospheric pressure:

$$
\begin{equation*}
\mathrm{U}_{\mathrm{S}}=\mathrm{C}_{1}\left(\mathrm{~V}_{\mathrm{G}}+\mathrm{V}_{\mathrm{L}}\right)+\mathrm{C}_{2}(\mathrm{gD})^{\frac{1}{2}} \tag{array}
\end{equation*}
$$

The reactor voidage (gas holdup), $\varepsilon$, is calculated from the rising velocity of slugs, $\mathrm{U}_{\mathrm{S}}$, and the superficial gas velocity, $\mathrm{V}_{\mathrm{G}}$, using Eq 2:

$$
\begin{equation*}
\varepsilon=\frac{\mathrm{V}_{\mathrm{G}}}{\mathrm{U}_{\mathrm{S}}} \tag{Eq2}
\end{equation*}
$$

where $V_{G}$ and $V_{L}$ are superficial gas and liquid velocities respectively. The second term on the right hand side of Eq 1 corresponds to a rise velocity of a single bubble in stagnant liquid and $C_{2}$ has a value of 0.35 as suggested earlier. A value of 1.2 for constant, $C_{1}$, was suggested for liquid-slug Reynolds numbers greater than $8000 . C_{1}$ approached a value of 2.0 as superficial liquid velocity decreased. Griffith and Wallis (16) suggested a similar equation with $C_{1}=1.0$ and $C_{2}$ consisting of two constants - one dependent on bubble Reynolds numbers and the other on liquid Reynolds numbers. Brown and Govier (19) investigated two-phase flow in vertical tubes for various systems. Taking into consideration the liquid film thickness and liquid viscosities, they suggested different equations for constants, $C_{1}$ and $C_{2}$, for fully developed slug flow.

Recently Bhaga and Weber (21) carried out detailed theoretical analysis of two- and three-phase flow in vertical tubes and proposed a more general method of predicting the reactor voidage. They also compared the previous correlations for two-phase flow and showed these correlations were special cases of the general theory proposed by them.

Zuber (23) and Bankoff (24) proposed a variable-density singlefluid model for two-phase one-component flow of steam and water assuming power-law distributions for the radial void fraction and velocity and derived an expression for gas holdup:

$$
\begin{equation*}
\frac{1}{X}=1-\frac{\rho_{L}}{\rho_{G}} \quad\left(1-\frac{K}{\varepsilon}\right) \tag{Eq3}
\end{equation*}
$$

where $\mathrm{X}=$ quality, mass vapour/mass fluid flowing, $\rho_{L}=$ density of liquid and $\rho_{G}=$ density of vapour. The flow parameter, $K$, was a function of the void fraction and velocity distribution exponents. The pressure dependency for K was represented by Eq 4 in the pressure range of 0.1 to 14 MPa :

$$
\begin{equation*}
\mathrm{K}=0.71+0.0145 \mathrm{P} \tag{Eq4}
\end{equation*}
$$

where $P$ was in units of MPa . A closer examination of their data and equations 3 and 4 indicated that at constant gas flow rate the void fraction increased with increased pressure, and at constant quality the voidage decreased with increased pressure. The equations suggested by Bankoff were based on his data in the turbulent flow region. Hughmark (25) proposed a general correlation for $K$ based on Reynolds number, Froude number and liquid volume fraction, and he showed the applicability of the method for the horizontal, as well as vertical, two-phase flow of various two-component systems. The flow parameter K ranged from 0.15 and 0.98 depending on experimental conditions.

Correlation for the bubble-rise velocity of gas kicks in an annulus has been developed by Rader et a1. (26). Some studies have been made on twophase flow with concurrent heat transfer (27-29) and mass transfer (30-31).

## EXPERTMENTAL

The dimensions of different types of reactors employed in the present study are given in Table 1. Reactors A and B had thermowells located axially to facilitate measuring the reaction temperature and each behaved like an annular reactor. Reactor $C$ had thermocouples inserted through the reactor walls, and reactor $D$ was a Plexiglas model having the same dimensions and configuration as reactor $C$ but without thermocouples, and was only used for the vertical two-phase flow investigation of water and nitrogen at room temperature and atmospheric pressure. The details of experimental runs are given in Table 2.

## Apparatus and Procedure

The pilot plant and the hydrocracking process have been described in detail elsewhere $(32,33)$. A simplified schematic diagram is shown in Fig. 1. Liquid bitumen feed and recycle-hydrogen gas, both under pressure, entered the preheaters and subsequently flowed to the bottom of a liquidphase reactor. The reactor temperature was controlled by four equallyspaced electrical heaters. As the oil was cracked, the vaporized portion along with hydrogen passed through the reactor liquid to the hot separator.

The liquid overflow from the reactor also entered the hot separator where it was collected. This liquid product was withdrawn continuously through a metering valve to a heavy-oil tank at atmospheric pressure. The uncondensed vapours and gases passed out from the top of the hot separator, were cooled, and a light oil product was collected in a cold receiver maintained at $20^{\circ} \mathrm{C}$. The uncondensed gases from the cold receiver were oil-scrubbed to remove hydrogen sulphide and some hydrocarbon gases. The scrubbed hydrogen was joined by fresh hydrogen and recycled to the reactor via a recycle pump. The scrubber maintained the recycle-gas purity at $85 \% \mathrm{H}_{2}$. The off-gas flows were measured by wet-test meters. The recycle-gas flow to the reactor was measured by a calibrated orifice meter.

The usual procedure in two-phase flow without chemical reaction at atmospheric pressure has been to determine the voidage by tube-emptying experiments (17). Because the present experiments were conducted at elevated temperatures and pressures, a different procedure was adopted. At the start of a run the system was pressurized with pure hydrogen and tested for leaks. With gas circulating, the reactor and the hot separator were brought up to 300 and $250^{\circ} \mathrm{C}$ respectively. At these temperatures the feed flow was started and set at the desired rate and the preheaters were switched on. The reactor temperature was then slowly increased to the desired level. When the liquid feed rate, the recycle-gas flow and the reactor temperature were steady, the gas flow was stopped and the time required to fill the reactor was noted. When the reactor was full, liquid overflowed to the hot separator, evidenced by an increase in its weight as recorded by the weighing system. Using data on the time required to fill the reactor, liquid feed rate, reactor volume and liquid density, the voidage was calculated.

The density of bitumen feed at reactor temperature was computed from the density at $15.6{ }^{\circ} \mathrm{C}$ using an expression for the thermal expansion given elsewhere (34). The densities computed by this method compared well with those estimated from a graph relating the specific gravities of petroleum fluids to temperatures (35). The experiments were repeated for a series of recycle-gas rates, temperatures, pressures and liquid feed rates. For each recyc1e-gas rate, two or three determinations were made for the voidage and an average value was calculated. The deviation in the individual determination from the average value was less than $5 \%$. The absolute deviation in
the voidage was less than 0.015 . Since the main interest in these experiments was in determining voidage, no attempts were made to measure and analyze gaseous and liquid products.

A similar procedure was followed in the investigation of the twophase flow of kerosene and hydrogen at room temperature and a series of pressures. The vertical two-phase flow of water and nitrogen was investigated only in a laboratory Plexiglas model reactor at room temperatures and atmospheric pressure.

## RESULTS AND DISCUSSION

The experimental results are presented and discussed in the following four sections:

1. vertical two-phase flow of water and nitrogen
2. vertical two-phase flow of kerosene and hydrogen
3. vertical two-phase flow concurrent with the thermal hydrocracking of Athabasca bitumen
4. general discussion

## 1. Vertical Two-Phase Flow of Water and Nitrogen

The vertical two-phase flow of water and nitrogen at atmospheric pressure and room temperature was investigated in a Plexiglas reactor (D, Table 1) to determine reliability of the method of measuring the voidage and to visualize the type of flow. Observations showed that, as the liquid and gas entered the bottom of the reactor and moved up, some of the small bubbles grew in size (as a result of coalescence) and assumed the characteristic bullet shape of Taylor bubbles (2). These bubbles were separated by slugs of liquid which contained small bubbles. This bubble phenomenon was observed over the entire range of gas velocities investigated.

The relationship between $\varepsilon$ and $V_{G}$ was found to be linear (Fig. 2) and, at superficial liquid velocities of 0.11 and $0.22 \mathrm{~cm} / \mathrm{s}$, it appears that $V_{L}$ had no appreciable effect on this linear relationship. The observed voidage compared well with the one predicted by the Nicklin equations (Eq 1 and 2) using $C_{1}=2.0$ at low superficial liquid velocity ( $V_{L}<30 \mathrm{~cm} / \mathrm{s}$ ). The differences between the observed voidages and those predicted were on average less
than 12\%. Equation 1 was derived for liquid-slug Reynolds numbers greater than 8000 and it has been suggested that Eq 1 is not as accurate at low liquid-slug Reynolds numbers. For this work, liquid slug Reynolds numbers were less than 800 and this may explain the discrepancy. Equation 1 does not indicate a linear relationship between $\varepsilon$ and $V_{G}$. However, the observed linearity in data from this work can be explained by the negligible contribution from the first term $\left\{2\left(\mathrm{~V}_{\mathrm{G}}+\mathrm{V}_{\mathrm{L}}\right)\right.$ \} at low $\mathrm{V}_{\mathrm{G}}$ and $\mathrm{V}_{\mathrm{L}}$. Elimination of this term results in a linear relationship. In the plot of $\varepsilon$ versus $V_{G}$, the inverse of the slope is the bubble rise velocity relative to the reactor wall. From Fig. 2, this is $27.3 \mathrm{~cm} / \mathrm{s}$ which compares well with the predicted value of $24.0 \mathrm{~cm} / \mathrm{s}$ from Eq 1 with $\mathrm{C}_{1}=2.0$.

This study showed that the reactor voidage could be determined by measuring the time required to fill up the reactor after the gas flow was stopped, and that reliable results, consistent with other published results, could be obtained.

## 2. Vertical Two-Phase Flow of Kerosene and Hydrogen

Vertical two-phase flow of kerosene and hydrogen at room temperature and elevated pressures was investigated in the pilot plant to determine the effect of pressure and $V_{L}$ on the $\varepsilon$ versus $V_{G}$ relationship in the absence of a chemical reaction. A tubular reactor, $C$, (Table 1) was used. The reactor, hot separator, cold receiver and all the lines were maintained at room temperature. Kerosene overflowed to the hot separator when the gas flow was stopped and liquid flow maintained. No light oil product was formed as there was no reaction and the cold receiver did not accumulate any kero. sene.

Figure 3 shows that, for superficial liquid velocities of 0.138 and $0.264 \mathrm{~cm} / \mathrm{s}, \mathrm{V}_{\mathrm{L}}$ has no appreciable effect on $\varepsilon$ for several values of $\mathrm{V}_{\mathrm{G}}$. The computed curve for $\varepsilon$ using Nicklin equations (Eq 1 and 2) with $C_{1}=2.0$ and $\mathrm{V}_{\mathrm{L}}=0.264 \mathrm{~cm} / \mathrm{s}$ is shown by a dotted line. The experimental voidages are higher than those predicted by Nicklin's method. Figure 4 shows voidage as a function of $V_{G}$ for a series of pressures, and indicates that, for a given $V_{G}$, increasing the pressure increases the voidage. Data of Fig. 4 were replotted in Fig. 5 where voidage is represented as a function of inlet
quality, $X=$ mass rate of gas/mass rate of gas plus liquid. Figure 5 shows that, at constant inlet quality, increasing the pressure decreases the voidage. Bankoff (24) observed similar behaviour in two-phase flow of steam and water in horizontal pipes. However, his correlations were based on data at high inlet qualities and high Reynolds numbers, i.e., in the turbulent region, and did not corroborate with data in this work. Hughmark (25) correlated the flow parameter, $K$, of the Bankoff equation (Eq 3) with another parameter, Z, and showed applicability of the method for horizontal as well as vertical two-phase flow of various systems.

The parameter, $Z$, is comprised of Reynolds number ( $\mathrm{N}_{\mathrm{Re}}$ ), Froude number ( $\mathrm{N}_{\mathrm{Fr}}$ ) and input liquid volume fraction $\left(\mathrm{Y}_{\mathrm{L}}\right)$ as shown in Eq 5:

$$
\begin{equation*}
\mathrm{Z}=\left(\mathrm{N}_{\mathrm{Re}}\right)^{1 / 6}\left(\mathrm{~N}_{\mathrm{Fr}}\right)^{1 / 8} /\left(\mathrm{Y}_{\mathrm{L}}\right)^{1 / 4} \tag{array}
\end{equation*}
$$

$\mathrm{N}_{\mathrm{Re}}$ and $\mathrm{N}_{\mathrm{Fr}}$ are defined by equations 6 and 7:

$$
\begin{align*}
& \mathrm{N}_{\mathrm{Re}}=\mathrm{D}\left(\mathrm{~V}_{\mathrm{G}} \rho_{\mathrm{G}}+\mathrm{V}_{\mathrm{L}} \rho_{\mathrm{L}}\right) /\left\{\mu_{\mathrm{G}} \varepsilon+\mu_{\mathrm{L}}(1-\varepsilon)\right\}  \tag{Eq6}\\
& \mathrm{N}_{\mathrm{Fr}}=\left(\mathrm{V}_{\mathrm{G}}+\mathrm{V}_{\mathrm{L}}\right)^{2} /(\mathrm{gD}) \tag{Eq.7}
\end{align*}
$$

where $\mu_{G}$ and $\mu_{L}$ are gas and liquid viscosities, $g$ is the gravitational acceleration and $D$ is the diameter. Rearranging Eq 3, the flow parameter K is expressed as:

$$
\begin{equation*}
K=\varepsilon\left(\frac{V_{G}+V_{L}}{V_{G}}\right) \tag{array}
\end{equation*}
$$

The drawback of the Hughmark method is that it involves a trial-and-error procedure since $\varepsilon$ appears in both Eq 5 and 8 . The present data for a kerosene-hydrogen system at 13.89 MPa are beyond the range of the Hughmark correlation. Accordingly his method is not suitable for predicting voidage in vertical two-phase flow of kerosene and hydrogen in the present work.
3. Vertical Two-Phase Flow Concurrent with the

Thermal Hydrocracking of Athabasca Bitumen
Voidage during thermal hydrocracking of Athabasca bitumen in annular as well as tubular reactors has been determined. The bubble rise velocity depends on the reactor geometry in addition to the gas and liquid properties. In the case of a tubular reactor, bubbles are bullet-shaped ( $2,14,18$ ), and in the case of an annular reactor, they are bend hot-dog- or doughnut-shaped (26). The superficial velocities, $V_{G}$ and $V_{L}$, are based on volumes at the reactor temperature and pressure.

The results of voidage measurements for the thermal hydrocracking reaction at $400^{\circ} \mathrm{C}$ in annular reactors ( $A$ and $B$ ) are shown in Fig. 6 to 8. Figure 6 shows the effect of pressure as parameter at constant $V_{L}$ and varying $V_{G}$. Figure 7 illustrates the effect of $V_{L}$ on $\varepsilon$ at constant pressure and $V_{G}$. Increasing $V_{L}$ from $0.075 \mathrm{~cm} / \mathrm{s}$ to $0.436 \mathrm{~cm} / \mathrm{s}$ increased $\varepsilon$ from 0.325 to 0.366 . This effect of superficial liquid velocity on $\varepsilon$ is confirmed in Fig. 8, again for an annular reactor.

A series of experiments was conducted on the thermal hydrocracking of Athabasca bitumen in a tubular reactor (C) under various conditions. Figure 9 shows that $V_{L}$ had no appreciable effect on $\varepsilon$ for a range of $V_{G}$ at $400^{\circ} \mathrm{C}$ and 13.89 MPa . However, the range of $\mathrm{V}_{\mathrm{L}}$ was smaller than for the annular reactors. The effect of pressure on the voidage at $330^{\circ} \mathrm{C}$, given in Fig. 10, shows that increasing the pressure increased the voidage. Figure 11 shows the effect of the reaction temperature on the voidage for a range of $V_{G}$ at 13.89 MPa pressure. Figure 12 is a cross-plot of voidage as a function of temperature at a constant recycle-gas flow rate of $0.0425 \mathrm{~m}^{3} / \mathrm{h}\left(1.5 \mathrm{ft}^{3} / \mathrm{h}\right)$ at the system pressure and $20^{\circ} \mathrm{C}$. The voidage increased with increased temperature.

From the experimental results, the constant, $m$, of the linear relation $\varepsilon=\mathrm{mV}_{\mathrm{G}}$ has been evaluated by the method of least squares. The computed values of $m$ and $1 / \mathrm{m}$ for different experimental conditions are given in Table 3. The reciprocal of $m$ is the average rise velocity of bubbles with respect to the reactor wall since $1 / m=V_{G} / \varepsilon=U_{S}$ from Eq 2. Examining Table 3, it is found that $U_{S}$ decreased with increased pressure for kerosene-hydrogen as well as for bitumen-hydrogen system. $U_{S}$ also decreased with increased temperature for bitumen-hydrogen system.

Assuming the power-law dependency of $m$ on the temperature and pressure ( $m=a P^{b} T^{c}$ ), the exponents $b$ and $c$ were determined statistically from the slopes of the plots of log magainst log $P$ at constant $T$ (Fig. 13) and $\log \mathrm{m}$ against $\log \mathrm{T}$ at constant P (Fig. 14), respectively. The pressure dependency exponent, $b$, was found to be 0.18 for the thermal hydrocracking reaction in an annular reactor, type A , at $400^{\circ} \mathrm{C}$ as well as in a tubular reactor, type C , at $330^{\circ} \mathrm{C}$ and also for the vertical two-phase flow of kerosene and hydrogen in a tubular reactor, type $C$, at room temperature. The tempera-ture-dependency exponent $c$ determined from the thermal hydrocracking reaction in a tubular reactor, type C , at 13.89 MPa was found to be 2.11. Assuming the same temperature dependency for the hydrocracking reaction in an annulus, the general expressions for the reactor voidage during the thermal hydrocracking of Athabasca bitumen can be formulated as follows:

$$
\begin{align*}
& \varepsilon=1.54 \times 10^{-7} \mathrm{P}^{0.18} \mathrm{~T}^{2.11} \mathrm{~V}_{\mathrm{G}} \quad(\text { reactors } \mathrm{B} \text { and } \mathrm{C})  \tag{Eq9}\\
& \varepsilon=1.845 \times 10^{-7} \mathrm{P}^{0.18} \mathrm{~T}^{2.11} \mathrm{~V}_{\mathrm{G}}(\text { reactor } \mathrm{A}) \tag{Eq10}
\end{align*}
$$

where $P$ is pressure in MPa, $T$ is temperature in ${ }^{\circ} C$ and $V_{G}$ is the superficial gas velocity at reactor temperature and pressure in $\mathrm{cm} / \mathrm{s}$. The expression for the two-phase vertical flow of kerosene and hydrogen at room temperature in a tubular reactor, type C, is:

$$
\begin{equation*}
\varepsilon=0.0448 \mathrm{P}^{0.18} \mathrm{~V}_{\mathrm{G}} \tag{Eq11}
\end{equation*}
$$

These expressions are derived from the experimental data with liquid hourly space velocity (LHSV) of 2.0 and are valid for other liquid hourly space velocities for tubular reactors as the superficial liquid velocities appeared to have no effect on the voidage, but have to be modified for annular reactors where the voidage increases with increased superficial liquid velocities.

A more general correlation is obtained when the reactor voidage is plotted as a function of the dimensionless superficial gas velocity $V_{G}^{*}=$ $\mathrm{V}_{\mathrm{G}} /(\mathrm{gD})^{\frac{1}{2}}$. Experimental results of thermal hydrocracking of Athabasca bitumen at $13.89 \mathrm{MPa}, 400^{\circ} \mathrm{C}$ and an LHSV of 2.0 in three different reactors, i.e., tubular reactor $C$ as well as annular reactors $A$ and $B$, plotted in this fashion
are shown in Fig. 15. This graphical presentation shows that it is the smaller dimension, i.e., diameter, rather than the larger length of the reactor which is important in the correlation of the reactor voidage.

The reactor voidage depends upon the reactor geometry and numerous correlations for air-water flows in various geometrics such as tube bundes, rectangular channels, circular pipes (27) and annuli (26) have been developed. Slight positive deviations observed in Fig. 15 at low $V_{G}{ }^{*}$ are for a short annular reactor where bubble swarms may not have had sufficient time to coalesce. For a given $\mathrm{V}_{\mathrm{G}}{ }^{*}$, bubble swarms would have higher $\varepsilon$ associated with them than bullet-shaped slugs. However, for long reactors, it appears that reactor geometry has no significant effect on the voidage correlation when proper diameter is taken into account.

## 4. General Discussion

The effect of $V_{G}$ on the voidage was as expected from Eq 1 and as observed by several workers (15-24). The voidage increased with $V_{G}$ in all experimental cases and a linear relationship was found at elevated temperatures and pressures.

The effect of pressure on the voidage was surprising. Intuitively, pressure should have little effect on the voidage; however, an increase in $\varepsilon$ was observed with increased pressure for the kerosene-hydrogen as well as for the bitumen-hydrogen system. This type of behaviour has been reported for two-phase flow of steam and water $(23,24)$ and for gas-solid fluidization (36). As the pressure increases, the density of gas increases which reduces the bubble rise velocity and thus increases the voidage. Decrease in the bubble rise velocity with increased gas density has been reported elsewhere $(20,26)$. Another possible explanation is the combined effect of pressure and temperature on bitumen viscosity and this is not known. It is reported that the viscosity of various oils increases at elevated pressures $(37,38)$. As observed previously $(19,26)$ increased liquid viscosity reduces the bubble rise velocity and thus increases the voidage. Both of these statements appear to explain the observed phenomenon in kerosene-hydrogen systems. Since the pressure dependency exponent was the same (0.18) for the kerosene-hydrogen as for the bitumen-hydrogen system, this indicates
that it is the pressure effect on the gas density rather than the liquid viscosity which affects the voidage.

For the two-phase flow of gas and liquid without chemical reaction, one would expect from $E q 1$ that $\varepsilon$ should decrease with increased $V_{L}$. No appreciable effect of $V_{L}$ was observed for the kerosene-hydrogen and waternitrogen systems. This was understandable because of the small values of $V_{L}$ and low liquid-slug Reynolds number of $<800$ encountered. For the bitumenhydrogen system in a tubular reactor also, this insensitivity of $\varepsilon$ is explained by consideration of the small range of $V_{L}$. However, the voidage increased with increased $V_{L}$ for the hydrogen-bitumen system in an annular reactor (Fig. 7 and 8). This is the reverse of what one would expect, and is explained by taking into consideration the hydrocracking reaction. $A s V_{L}$ is increased, the density of gas entering the reactor is also increased because of vaporization of high molecular weight bitumen. Furthermore, at high $V_{L}$ and the same $P, T$ and $V_{G}$, the conversion is reduced making viscosity of the reactor £luid higher. The increased gas density and liquid viscosity would decrease bubble rise velocity and thus increase the voidage at high $V_{L}$.

The expected effect of temperature on $\varepsilon$ for two-phase flow without chemical reaction is to decrease $\varepsilon$ with increased $T$ (20), but for the hydrocracking of bitumen, $\varepsilon$ increased with increased $T$. The thermal hydrocracking reaction is highly temperature-dependent. As mentioned earlier, as $T$ is increased, the amount of feed vaporized also increases significantly for the same $V_{L}, V_{G}$ and $P$. Also, the light-oil product formed is vaporized. This leads to a considerable increase in gas density. The molar volumetric flows at the bottom and top of the reactor are the same. However, increased density of gas reduces the bubble rise velocity and thus increases $\varepsilon$ with $T$.

During the hydrocracking reaction, the liquid and gas properties (viscosity, density, etc.) change appreciably and it is not possible at present to determine these parameters under experimental conditions. Because of this, $\varepsilon$ or $m$ could not be correlated in terms of Reynolds number, Froude number and liquid volume fractions. Surface effects have been found significant for two-phase gas-liquid flow without chemical reaction in tubular reactors of a diameter less than $1.25 \mathrm{~cm}(16-19)$ and surface effects may be important in the case of bitumen-hydrogen systems. Because there are no published results on voidage during thermal hydrocracking of Athabasca
bitumen, there was no basis for comparing the results obtained in the present study.

CONCLUSIONS

The reactor voidage during the thermal hydrocracking of Athabasca bitumen in vertical reactors has been successfully correlated with the system variables. The linear relationship $\varepsilon=m{ }_{G}$ has been observed in all cases. The correlation constant, $m$, was found to be proportional to $\mathrm{p}^{0.18} \mathrm{x}$ $\mathrm{T}^{2.11}$ for the thermal hydrocracking reaction. Whereas the reactor voidage increased with increased superficial liquid velocity for the reaction in an annular reactor, it was independent of the superficial liquid velocity for the hydrocracking reaction for the two-phase flow of kerosene and hydrogen, and for the water-nitrogen flow in tubular reactors. The pressure dependency exponent for the vertical two-phase flow of kerosene and hydrogen was the same (0.18) as that observed for the hydrocracking reaction. Present correlations provide means of estimating the reactor voidage under prevailing thermal hydrocracking reaction conditions and hence of determining the actual residence time and true space velocity of gas and liquid in the reactor.

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## Details of Experimental Reactors

| Reactor | Material | Reactor <br> ID <br> cm | Reactor <br> Length <br> m | Thermowell <br> OD <br> cm | Reactor <br> Volume <br> $\ell$ | $D^{*}$ <br> cm |
| :---: | :--- | :---: | :---: | :---: | :---: | :---: |
| A | S.Stee1 | 3.81 | 3.96 | 1.27 | 4.03 | 2.54 |
| B | S.Stee1 | 7.62 | 0.91 | 0.95 | 4.05 | 6.67 |
| C | S.Steel | 3.81 | 3.96 | - | 4.52 | 3.81 |
| D | Plexiglas | 3.81 | 3.96 | - | 4.52 | 3.81 |

* D = inside diameter of tubular reactor or hydraulic diameter of annular reactor.

TABLE 2

Details of Experimental Runs

| Run No. | Reactor | $\begin{gathered} \text { Pressure, } \\ \text { MPa } \end{gathered}$ | Temperature, ${ }^{\circ} \mathrm{C}$ | Liquid Velocity* $\mathrm{V}_{\mathrm{L}}, \mathrm{cm} / \mathrm{s}$ | $\begin{aligned} & \text { Gas Velocity* } \\ & V_{G}, \mathrm{~cm} / \mathrm{s} \end{aligned}$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| 1 | D | Water-Nitrogen System |  |  | 0.30-1.79 |
|  |  | 0.101 | Room | 0.22, 0.11 |  |
|  |  | Kerosene-Hydrogen System |  |  |  |
| 2 | c | 13.89 | Room | 0.138, 0.264 | 0.34-1.38 |
| 3 | c | 17.34 | Room | 0.275 | 1.00-1.72 |
| 4 | C | 7.00 | Room | 0.275 | 1.03-2.41 |
| 5 | C | 3.55 | Room | 0.275 | 0.69-2.41 |
|  | Thermal Hydrocracking of Athabasca Bitumen |  |  |  |  |
| 6 | A | 13.89 | 400 | 0.28 | 2.00-4.66 |
| 7 | A | 7.00 | 400 | 0.28 | 1.84-4.44 |
| 8 | A | 13.89 | 400 | 0.075-0.436 | 4.05 |
| 9 | B | 13.89 | 400 | 0.081 | 0.37-1.36 |
| 10 | B | 13.89 | 400 | 0.038 | 0.34-1.25 |
| 11 | C | 13.89 | 400 | 0.139, 0.278 | 1.00-3.16 |
| 12 | C | 13.89 | 430 | 0.284 | 0.90-3.30 |
| 13 | c | 13.89 | 360 | 0.271 | 2.20-3.63 |
| 14 | c | 13.89 | 330 | 0.267 | 1.42-3.51 |
| 15 | C | 7.00 | 330 | 0.267 | 2.85-3.51 |
| 16 | C | 3.55 | 330 | 0.267 | 2.90-3.97 |

$* V_{\text {L }}$ and $V_{G}$ are superficial liquid and gas velocities at reactor temperature
and pressure

TABLE 3

Correlation Constants (m) and Bubble Rise Velocities (1/m)

| Run No. | Reactor | Pressure, $\mathrm{MPa}$ | $\begin{gathered} \text { Temperature, } \\ { }^{o_{C}} \end{gathered}$ | $\stackrel{\mathrm{m}}{\mathrm{~s} / \mathrm{cm}}$ | $\begin{gathered} 1 / \mathrm{m} \\ \mathrm{~cm} / \mathrm{s} \end{gathered}$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| 1 | D | Water-Nitrogen System |  | 0.0367 | 27.26 |
|  |  | 0.101 | room |  |  |
|  |  | erosene-Hyd | gen System |  |  |
| 2 | C | 13.89 | room | 0.0727 | 13.76 |
| 3 | C | 17.34 | room | 0.0806 | 12.41 |
| 4 | C | 7.00 | room | 0.0658 | 15.20 |
| 5 | C | 3.55 | room | 0.0604 | 16.56 |
|  |  |  |  |  |  |
|  | Thermal Hydrocracking of Athabasca Bitumen |  |  |  |  |
| 6 | A | 13.89 | 400 | 0.0911 | 10.98 |
| 7 | A | 7.00 | 400 | 0.0812 | 12.32 |
| 8 | A | 13.89 | 400 | - | - |
| 9 | B | 13.89 | 400 | 0.0766 | 13.06 |
| 10 | B | 13.89 | 400 | 0.0635 | 15.75 |
| 11 | C | 13.89 | 400 | 0.0768 | 13.02 |
| 12 | C | 13.89 | 430 | 0.0915 | 10.93 |
| 13 | C | 13.89 | 360 | 0.0662 | 15.11 |
| 14 | C | 13.89 | 330 | 0.0511 | 19.57 |
| 15 | C | 7.00 | 330 | 0.0450 | 22.39 |
| 16 | C | 3.55 | 330 | 0.0402 | 24.88 |




FIGURE 2 - Voidage as a Function of Superficial Gas Velocity, Water-nitrogen System at Room Temperature and 0.10 MPa


FIGURE 3 - Voidage as a Function of Superficial Gas Velocity, Kerosene-hydrogen System at Room Temperature


FIGURE 4 - Voidage as a Function of Superficial Gas Velocity, Kerosene-hydrogen System at Room Temperature


FIGURE 5 - Voidage as a Function of Inlet Quality, Kerosene-hydrogen System at Room Temperature


FIGURE 6 - Voidage as a Function of Superficial Gas Velocity, Thermal Hydrocracking of Athabasca Bitumen at $400^{\circ} \mathrm{C}$ and $\mathrm{V}_{\mathrm{L}}=0.28 \mathrm{~cm} / \mathrm{s}$


FIGURE 7 - Voidage as a Function of Superficial Liquid Velocity, Thermal Hydrocracking of Athabasca Bitumen at 13.89 MPa and $400^{\circ} \mathrm{C}$


FIGURE 8 - Voidage as a Function of Superficial Gas
Velocity, Thermal Hydrocracking of Athabasca Bitumen at 13.89 MPa and $400^{\circ} \mathrm{C}$


FIGURE 9 - Voidage as a Function of Superficial Gas Velocity, Thermal Hydrocracking of Athabasca Bitumen


FIGURE $10-\frac{\text { Voidage as a Function of Superficial Gas }}{\text { Velocity, Thermal Hydrocracking of }}$


FIGURE 11 - Voidage as a Function of Superficial Gas Velocity, Thermal Hydrocracking of Athabasca Bitumen at 13.89 MPa


FIGURE 12 - Voidage as a Function of Temperature, Thermal Hydrocracking of Athabasca Bitumen at 13.89 MPa and Recycle Gas Rate of $0.0425 \mathrm{~m}^{3} / \mathrm{h} \mathrm{@} \mathrm{P} \mathrm{and} 20^{\circ} \mathrm{C}$


[^2]

FIGURE $14-\frac{\text { Plot of } \log \mathrm{m} \text { against } \log \mathrm{T} \text {, Thermal }}{\frac{\text { Hydrocracking of Athabasca Bitumen }}{\text { at } 13.89 \mathrm{MPa} \text { and } 400^{\circ} \mathrm{C}}}$


FIGURE 15 - Voidage as a Function of Dimensionless Superficial Gas Velocity, Thermal Hydrocracking of Athabasca Bitumen

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[^2]:    FIGURE 13 - Plots of $\log \mathrm{m}$ against $\log P$
    1 - Thermal hydrocracking of Athabasca bitumen at $400^{\circ} \mathrm{C}$ (Reactor A)
    2 - Kerosene-hydrogen system at room temperature (Reactor C)
    3 - Thermal hydrocracking of Athabasca bitumen at $330^{\circ} \mathrm{C}$ (Reactor C )

