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MODELING THE FLUID DYNAMICS OF THE H-COAL REACTOR

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ABSTRACT

A key feature of the H-Coal process is the use of an ebullated bed for achieving good mixing between slurried coal, hydrogen, and extruded catalyst particles. The H-Coal reactor can be modeled as a four-phase fluid bed system containing liquid, fines, gases, and catalyst particles. In order to study the hydrodynamic behavior of this gas/slurry/catalyst system, a cold flow model was constructed consisting of a glass reactor 15.24 cm in ID and 6 m long. A recycle cup located in the upper part of the reactor is used for the partial separation of gas from the slurry before part of the slurry is recycled to the bottom of the reactor. The layout and dimensions of this unit are very similar to a 3 T/D coal liquefaction process development unit (PDU) successfully operated by Hydrocarbon Research, Incorporated (HRI). Using kerosene, pulverized coal char, and nitrogen, it has been found that at ambient conditions the bed expansion of cylindrical catalyst extrudates 1.6 mm in diameter and 4.8 mm in length closely simulates the behavior of the H-Coal reactor. Operating conditions which favor the distribution of the gas phase in the stable ideal bubbly regime are also described.

INTRODUCTION

The H-Coal process, developed by Hydrocarbon Research, Incorporated (HRI), involves the direct catalytic hydroliquefaction of coal to low-sulfur boiler fuel or synthetic crude oil (1,2). The process has been demonstrated by HRI in their process development unit (PDU) at Trenton, New Jersey. This unit is capable of processing up to three tons of dried coal per day. A 200-600 ton/day pilot plant is under construction at Catlettsburg, Kentucky (2).

In the H-Coal process, coal is dried, pulverized to approximately 100-mesh size, and slurried with coal-derived oil. This slurry is charged with hydrogen to an ebullated bed reactor containing a hydrogenation catalyst (3). The reactor operates between 427-482°C and 136-218 atm.

The unique feature of HRI's H-Coal process is the use of an ebullated bed as the means of achieving good mixing between the catalyst particles, the slurried coal, and the hydrogen gas. The catalyst is in extruded form and is similar to those used in petroleum hydrotreating processes. Typically, catalyst dimensions are 1.6 mm in diameter and 4.8 mm in length.

A schematic diagram of the reactor used in the process development unit is shown in Figure 1. A key feature of the H-Coal reactor is a large capacity internal circulation pump (ebullating pump) which takes suction from a point in the reactor near the top of the fluidized bed. The discharge from the ebullating pump is mixed with the fresh feed slurry and makeup gas and is recycled to the reactor through a specially designed distributor. The ratio of the ebullating flow to fresh feed flow may be as high as 10:1.

In the ebullating bed the upward flow of the slurried coal and gases forces the catalyst bed to expand. The expansion of the bed allows fine particles such as coal and ash to pass through the bed and out with the liquid stream, leaving the catalyst within the bed.

Downstream of the PDU reactor, the product slurry is separated into various streams, as shown in Figure 2. The product pressure is let down at about reactor temperature to atmospheric pressure in a flash drum. During the flash, a portion of the hydrocarbon liquids is flash-vaporized and fed to the atmospheric distillation tower. The bottoms from the flash drum are directed either to a hydroclone or to a vacuum distillation tower. The feed slurry oil used to disperse the coal particles may come from the atmospheric tower bottoms, the hydroclone overflow, or the vacuum distillation overhead. The weight ratio of slurry oil to solids in the feed is typically about 1.5-2.0. The composition of the recycled slurry oil varies depending on the mode of operation (fuel oil versus synthetic crude), catalyst age, and the properties of the processed coal.

The variation in feed composition and slurry concentration causes significant variation in the feed viscosity. These viscosity changes affect bed expansion and lead to a range of operating conditions, as shown below (4):

% Expansion of Catalyst Bed	63-80
Reactor Slurry Composition:	
% Resid	13-18
% Solids (Coal and Ash)	9-22
Slurry Properties (at Reactor Conditions):	
Viscosity, CP	0.2-0.5
Specific Gravity, g/cc	0.6-0.7
Gas Velocity, cm/sec	1.2-4.9
Slurry Velocity, cm/sec	2.1-5.5

From the physical description of the H-Coal reactor, it becomes evident that bed behavior is a complex function of process variables and gas, liquid, and solids properties. Several models have been proposed in the literature for describing the behavior of three-phase fluidization as a function of these variables. However, these models have been tested only for air/water fluidized beds. Only limited data exist for liquids other than water, and no data exist for slurries which are of primary importance to the H-Coal process. In order to fill these voids, Amoco Oil has contracted with DOE to study ebullating beds. The ultimate objective is the development of an improved mathematical model which will accurately describe the behavior of the ebullating beds in the H-Coal reactor.

The contract is divided into three main tasks: 1) review of literature; 2) construction of a cold flow model to simulate the H-Coal reactor; and 3) data collection and analysis. This paper reports on the first two tasks, which have been completed, and gives some of the preliminary results from the third area. Particular attention is given to analyzing the data in terms of the three-phase models proposed in the literature.

LITERATURE

According to Darton and Harrison (5), a three-phase fluidization model should fulfill the following criteria:

1) As the volume fraction of gas approaches zero, the model should describe liquid/solid fluidization behavior.
2) As the volume fraction of solids approaches zero, the model should reduce to a suitable gas/liquid model.

3) The sum of the volume fraction of the various phases should add up to one.

4) The model should predict whether the bed will expand or contract upon the addition of gas.

In addition to the first criteria, understanding the liquid/solid fluidization is also important in predicting the H-Coal reactor bed height when there is an unexpected interruption in the hydrogen flow. Numerous correlations for predicting the bed height in liquid fluidized systems have been reported. These are summarized in Table I. The most widely used method of correlating liquid/solid fluidization data is that of Richardson and Zaki (6). Their analysis recognizes the similarities between sedimentation and fluidization. They observed that the settling velocity of a suspension relative to a fixed horizontal plane was equal to the upward liquid superficial velocity needed to maintain the suspension at the same concentration.

As shown in Table I, the correlation relates the liquid volume fraction (ϵ_l) due to bed expansion to the ratio of the superficial liquid velocity (U_l) to the terminal velocity of a single particle (U_t):

$$\epsilon_l^n = U_l/U_t \qquad (1)$$

For spherical particles:

$$n = f(Re_t, d/D) \qquad (2)$$

where: $Re_t = d_p \rho_l U_t/\mu_l$, Reynolds number
d_p = particle diameter
ρ_l = liquid density
μ_l = liquid viscosity
D = bed diameter

The functional form of Equation 2 for different ranges of Reynolds number is given in Table I. It should be noted that for values of Re_t less than 0.2 or greater than 500, the exponent n in Equation 1 is independent of liquid viscosity.

For non-spherical particles, the exponent n in Equation 2 also becomes a function of particle shape. For turbulent Reynolds numbers (> 500), Richardson and Zaki found:

$$n = 2.7\, K^{0.18} \tag{3}$$

where: $$K = (\pi/6) d_s^3 / d_p^3 \tag{4}$$

The constant K is the particle shape factor proposed by Heywood (7). In Equation 4, d_s is the diameter of a sphere with the same volume as the particle, and d_p is the diameter of a circle of the same area as the projected particle when lying in its most stable position.

The addition of gas to a liquid fluidized bed increases the complexity of the system considerably. Kim, Baker, and Bergougnou (11) and Darton and Harrison (5) reported that for the range of superficial gas and liquid flow rates of interest to the H-Coal process, two gas/liquid flow regimes exist in the catalyst bed: 1) the ideal bubbly or bubble disintegration regime, in which the bubbles rise as a uniform, steady cloud with little interaction; and 2) the churn turbulent or bubble coalescing regime, which is a transition between ideal bubbly flow and fully developed slug flow. The churn turbulent regime is dominated by bubble coalescence. Hence, the bubble size is larger than in the ideal bubbly regime, bubble wake effects become important, and the flow is unsteady.

The effect of catalyst particle size on the rate of bubble coalescence has been pointed out by Kim, et al. (11) and by Ostergaard (12,13). Small particles promote the churn turbulent (bubble coalescence) flow regime and reduce gas holdup. Large particles, on the other hand, tend to favor the bubble disintegrating (ideal bubbly) flow regime and increase gas holdup.

Numerous investigators (17,18,19,20,21) have found evidence to support this effect of catalyst particle size. The experimental conditions used by these investigators are listed in Table II. From these studies it follows that the critical particle size between coalescing and non-coalescing beds is between 3 and 4 mm for the systems studied.

In a related phenomenon, Turner (14) was the first to report the contraction of some liquid fluidized beds upon the addition of gas. Kim, et al. (11) proposed that this contraction will occur only in beds in the bubble-coalescing flow regime. According to Stewart and Davidson (15), contraction occurs because of the formation of liquid wakes behind the bubbles which move through the bed at the bubble velocity. This action reduces the interstitial liquid velocity in the rest of the bed, thus causing its contraction. Ostergaard has also offered a similar explanation.

A summary of additional experimental studies on three-phase fluidized beds is shown in Table III. Many of these investigators have correlated their data in various forms, as shown in Table IV. It should be emphasized that these correlations should be used only for the specific system and range of variables from which they are developed.

The correlations presented in Table IV were developed through empirical observation or dimensional analysis. Semi-theoretical models have also been proposed to correlate three-phase fluidization data. These models take into consideration details of the bed structure, such as bubble rise velocity or bubble wake volume. The latter are often described by empirical relations.

Ostergaard (16) has proposed a model based on the assumption that the bed consists of three phases: gas, wake, and a liquid/solid particulate phase. It is assumed that the wake phase moves through the bed at the bubble velocity. The model consists of seven empirical equations which must be solved by an iterative procedure for the volume fractions of the various phases. Ostergaard (16) reports qualitative agreement between his proposed model and experimental data available in the literature. However, the model assumes that the porosity of the wake phase is equal to that of the liquid/solid particulate phase, and thus cannot predict a bed contraction.

Darton and Harrison (5) have also proposed a semi-empirical model. The model is based on the assumption that the bubble wakes are particle-free and that the liquid flux in the bubble wakes is given by $\bar{k}U_g$, where $\bar{k}$ is the mean value of the liquid wake volume/bubble volume ratio. Darton and Harrison develop an empirical correlation for $\bar{k}$ as a function of U_l and U_g:

$$1 + \bar{k} = 1.4(U_l/U_g)^{0.33} \qquad (5)$$

The superficial liquid velocity and the liquid holdup in the particulate phase are given by $(U_l - \bar{k}U_g)/(1 - \epsilon_g - \bar{k}\epsilon_g)$ and $(\epsilon_l - \bar{k}\epsilon_g)/(1 - \epsilon_g - \bar{k}\epsilon_g)$, respectively. These expressions are then introduced

into the Richardson-Zaki correlation to obtain:

$$\epsilon_l = \left(\frac{U_l}{U_t} - \frac{\bar{k}U_g}{U_t}\right)^{1/n} (1 - \epsilon_g - \bar{k}\epsilon_g)^{1-1/n} + \bar{k}\epsilon_g \qquad (6)$$

This equation and the fact that the fractional holdups must sum to unity give two equations with three unknowns.

In order to obtain a third relation, Darton and Harrison use the drift flux approach proposed by Wallis (31) for describing gas/liquid flow regime. Physically the drift flux represents the volumetric flux of gas relative to a surface moving at the average gas plus liquid velocity. The drift flux (V_{CD}) is defined as follows:

$$V_{CD} = U_S \epsilon_g (1 - \epsilon_g) \qquad (7)$$

where U_S is the gas liquid slip velocity defined below:

$$U_S = \frac{U_g}{\epsilon_g} - \frac{U_l}{\epsilon_l} \qquad (8)$$

Combining Equations 7 and 8, the following relationship is obtained:

$$V_{CD} = U_g(1 - \epsilon_g) - \frac{U_l \epsilon_g}{\epsilon_l}(1 - \epsilon_g) \qquad (9)$$

Darton and Harrison analyzed the fluidization data of Michelsen and Ostergaard (23) in terms of Equation 9. A plot of V_{CD} vs. ϵ_g revealed two flow regimes: the ideal bubbly and the churn turbulent. This plot is reproduced in Figure 3. As can be seen from the figure, the data from the two flow regimes fall along different lines. Typical transition data are indicated by the dashed lines between the two

regimes. The data for the ideal bubbly regime fall on the line given by:

$$V_{CD} = 180\ \epsilon_g (mm/sec) \tag{10}$$

The condition for bed contraction can be found by adding ϵ_g to both sides of Equation 9 and then taking the derivative of $(\epsilon_l + \epsilon_g)$ with respect to U_g as $\epsilon_g \to 0$. The condition for contraction is:

$$(1 + \bar{k})(1 - \epsilon_l + \frac{\epsilon_l}{n}) < \epsilon_l \frac{\bar{k}}{n} \frac{U_b}{U_l} \tag{11}$$

Epstein (32) has proposed a similar criterion for bed contraction.

Bhatia and Epstein (33) have also proposed a generalized wake model. Following Ostergaard (16), they also propose three phases in the bed: gas, wake, and the liquid particulate phase. The solids concentration in the wake phase can be varied between zero and that in the particulate liquid/solid fluidized phase by means of an adjustable parameter, X_k. The value of X_k ranges between 0 and 1. The wake volume is related to that of the gas by an empirical function. The particulate liquid/solid phase is described by a Richardson-Zaki type model. The model also takes into consideration the possible existence of two gas/liquid flow regimes in the bed. Bhatia and Epstein develop eight equations for the eight unknowns in the system. These equations are listed in Table V. The equations must be solved by an iterative procedure.

The authors test this model vs. the data of Michelsen and Ostergaard (23) and data of their own. Their data are for the fluidization of spheres ranging in size from 0.25 to 3.0 mm. The solid density ranged from 2.5 to 11.1 mg/m^3. The gas used was air, and the liquids were water (1 cp),

aqueous glycerol (2.1 cp), or aqueous polyethylene glycol (63 cp). The experiments were carried out in 20 and 50 mm columns. Best agreement between the model and data was obtained when $X_k = 0$ (solids-free wake).

This model, along with the contraction criteria proposed later by Epstein (32), also meets the four criteria suggested by Darton and Harrison. In many respects Bhatia and Epstein's wake model is very similar to the model proposed by Darton and Harrison.

EXPERIMENTAL

From the literature search it becomes evident that although significant contributions have been made in the field of gas/liquid/solid systems, no data exist for slurry systems directly applicable to the H-Coal process. For this reason, equipment was constructed in Amoco Oil which is almost on scale with the 3 T/D coal liquefaction process development unit (PDU) operated by HRI for several years. Data were obtained at ambient conditions with gas/slurry/catalyst systems having similar physical properties to the H-Coal liquids at reactor conditions. Equipment details and properties of the liquid/slurry used will be described in this section.

Equipment

A schematic diagram of the H-Coal fluid dynamics unit is shown in Figure 4. Design and construction of this unit were coordinated by the systems design group of Amoco Oil. Systems design incorporates elements of process design, mechanical design, instrumentation, automation, and computerization. Details on the work done in each area are given in Reference 34.

The process flow of the H-Coal fluid dynamics unit resembles that of the process development unit (PDU) built by HRI. The reactor consists of a 15.2 cm ID vessel 6 m in length. The slurry is prepared in a 227 liter tank. Other major vessels include: one 378 liter feed tank, and a 227 liter gas/liquid separator with a mist eliminator.

Three pumps are needed for the operation of the unit: 1) a transfer mixing pump for facilitating the slurry preparation and the transfer of the slurry to the feed tank; 2) a slurry feed pump for supplying the slurry to the reactor; and 3) a slurry recycle pump for internal slurry circulation in the reactor vessel. Under typical testing operating conditions, the slurry feed rate amounts to 11 liters per minute, while the slurry recycle pump supplies 57 liters per minute. Gas and liquid overflow from a 2.54 cm ID pipe located at the same level as the top of the recycle cup. The gas is separated from the liquid in the separator, D-3. Entrained liquid droplets accumulate in a demister located on top of the separator. The gas passes through Cooler E-3, and it can either be vented to the atmosphere through Valve PV-1 or can be directed to a gas recycle compressor. This flexibility is necessary because of the need to use gases such as Freon-12 and helium. The gas combines with the total liquid stream before it returns to the reactor. Some details on selected equipment items are given below.

Reactor Components.--The reactor is constructed from four glass sections 152 cm in length and 15.24 cm in diameter. The glass sections are connected through flanges to five metal spool pieces. A schematic diagram of the assembled reactor is shown in Figure 5.

The spool pieces have entries for sample taps, pressure taps, and thermowells to monitor the system. A drawing of a spool piece is shown

in Figure 6. As shown in this figure, the system is designed to insert two pressure taps through each spool piece. Thus, a total of eight ΔP measurements is feasible with this design. The lines are maintained clear by bleeding a low gas rate through each tap.

A sample probe can also be inserted into the reactor through each spool piece. A drawing of the sample probe assembly is shown in Figure 7. The probe is inserted into the reactor through a full-bore ball valve. Samples of the slurry are used to monitor the coal fines in slurry concentration along the reactor.

The reactor inlet distributor is based on a proprietary design supplied by Hydrocarbon Research, Incorporated (HRI). The distributor was modeled after the one used by HRI on the PDU. Establishing the proper distance between the bubble cup and the reactor bottom is the key for eliminating plugging problems.

The recycle cup design is also a proprietary design supplied by HRI. It is also similar to the one used by HRI in the PDU. The recycle cup is connected to the downcomer with a section of flexible pipe. The recycle cup is spaced inside the glass with Teflon tabs to keep the cup from scratching the reactor. The cup is supported from the top spool piece with four thin steel pipes.

Gamma-Ray Equipment.--An elevator is designed so that a gamma-ray source and detector can travel vertically along the reactor to monitor the fluid dynamics of the system. The source and detector are supported on a Unistrut frame. The frame runs in Unistrut guides with Unistrut trolley wheels. The framework is adjustable so two detector/source combinations can be mounted up to 91 cm apart.

The gamma-ray system attached to the elevator used for scanning the reactor was obtained from Harshaw and K-Ray. The scintillator detection

system was manufactured by Harshaw. A NaI scintillation crystal closely coupled to the photomultiplier tube (PMT) and preamplifier is mounted on the elevator. Other components used are: a stabilized amplifier/single-channel analyzer, a high-voltage source for the PMT, and a rate meter. The scan output is monitored using a Texas Instruments recorder on the panel board. Ten millicuries of Cs-137 from K-Ray is used as the gamma-ray source. Using the elevator system, the reactor can be scanned continuously from the bottom spool piece to the recycle cup.

An Ortec scintillation detection system and a Cs-137 source are used to scan the internal liquid recycle line at the bottom of the reactor. The components used in this system are the same as in the Harshaw system except that the amplifier/single-channel analyzer does not have the stabilizer option. A vertical length of pipe is scanned. The system is used to monitor the quantity of entrained gas in the recycle liquid. The output is recorded on a Leeds and Northrup recorder.

Compressor and Pumps.--A Corken reciprocating compressor, Model D-290K9, with a 5-HP motor was chosen for recycling gas. This compressor has a water-cooled aftercooler and a continuous pressure unloader. It is designed to pump 102 liters per minute at 1 atm inlet and 4 atm outlet pressures. The compressor discharges into a capacity tank to damp out flow rate fluctuations.

March magnetic drive pumps, Model TE-7S-MD, were chosen for the feed, slurry recycle, and transfer pumps. These centrifugal pumps are magnetically coupled to the motor so there are no problems with liquids or gas leaking through a dynamic seal. Because of the magnetic coupling design of this pump, it will not uncouple from the motor, so pump stoppage

is not a problem. At a head of 1.4 atm, the pump will deliver about 7.6 liters per minute.

Other mechanical design details are reported in Reference 34.

Control Components.--A Honeywell TDC-2000 microprocessor-based system is used for controlling slurry feed and slurry recycle flows, gas flows, gas/liquid separator level, and reactor pressure. Dual-pen analog recording is provided for hard copy of these process variables. Digital displays of variables, set points, and valve positions are also provided.

Flow Measurement.--A Nusonics Model 8000 meter was chosen to monitor slurry feed flow rate. The difference in frequency of sonic beams propagated axially to the flow is correlated with flow rate. A MicroMotion flow meter is used to measure slurry recycle flow rate. The deflection of this U-shaped flow tube due to the Coriolis-type acceleration of the flowing liquid is correlated with the mass flow rate. This flow meter was selected because tests show that entrained gases which are well dispersed do not affect flow measurement and entrained gases are expected in the recycle line. A Nusonics Model 6180 concentration analyzer was chosen for continuous monitoring of the coal concentration in the slurry feed. The gas flow is monitored with a Honeywell Model 41105 differential pressure transmitter with an integral orifice assembly. The DP across the orifice is dependent on the gas velocity.

Pressure.--Pressure drop measurements along reactor sections are made to determine the average ebullated bed three-phase volume fraction. Bournes Model 5020 differential pressure transmitters with high speed of response were chosen for this function. Analog recorders used are:

a three-pen Gould/Brush Model 2600 with 100 mm scales for the critical high-speed measurements; and two three-pen Texas Instruments Servo/Riters with overlapping pens on a 25 cm grid. All three recorders feature multiple chart speeds for maximum flexibility.

A small, continuous bleed of feed gas through each 3.2 mm ID pressure tap impulse line is used to keep the lines free of catalyst or slurry. Should the lines become plugged, however, a high-pressure gas surge can be initiated manually via three-way solenoid valves in the purge lines.

Originally, eight DP measurements were made with the taps spaced about 61 cm apart. However, the first series of pilot plant tests indicated that recorder pen sweeps were too large to obtain meaningful data. The data fluctuations were due to the vibration of the DP impulse lines within the reactor. A liquid-filled impulse line without purge flow was tried, but was unsuccessful because of catalyst and slurry plugging.

Thermowell taps in each spool piece are now being utilized for the differential pressure measurements. This eliminated the impulse line vibration problem, but reduced the number of differential pressure measurements to four. The DP across each 1.52 m reactor section is now being monitored.

Temperature.--Process and coolant temperatures are displayed digitally via a 1° resolution Doric indicator and recorded on a Honeywell Model 112 24-point recorder.

Computer Hardware Configuration.--The general hardware configuration for the computerization of the unit is shown in Figure 8. Selected equipment items are described below.

ModComp II Computer--The computer is a general-purpose 16-bit processor with 128K bytes of memory. It employs a dual moving head disc with 5196K bytes of storage capacity. This computer is equipped with asynchronous ports for serial devices such as CRT's and teletypes and with ditigal I/O for interfacing with the parallel data bus.

Tektronix 4662 Plotter--This device is used to generate user plots offline for numerous applications. It is driven by the Tektronix Plot 10 software package, which contains the following features: automatic coordinate transformations (linear, logarithmic, and polar); virtual plotting; relative plotting; segmented line drawing; character generation allowing unlimited control of character size and slant; scaling and rotating of relative virtual vectors. Besides being used for graphic representation of test results, the plotter is also used for generating graphic displays similar to those produced by the Intelligence Systems Corporation (ISC) color graphic CRT, thus allowing hard copy of CRT displays. Examples of this feature are reported in Reference 34.

ISC 8001G Color Graphic CRT--This peripheral is employed for real-time display of unit data. The CRT has eight colors, which may be used as foreground or background colors, with which graphic displays are generated of the pilot plant. When these displays are output to the CRT, real-time data--which are scanned every minute--are inserted at appropriate locations on the screen. An additional useful feature is a software refresh of data in real time. In this way the data are kept current and trends are easy to spot. Also, this refresh feature can be used effectively when lining out the unit.

Computer Software Configuration.--The ModComp II computer used for monitoring this unit is running under MAX II/III, which is a vendor-supplied executive system. In addition to this ModComp-supplied operating system, several tasks have been added to the system environment which implement some very useful features.

All of the "system" programs provide a base for application programs specific to this unit. A generalized flow chart of the applications software configuration is shown in Figure 9. All of the programs are written primarily in Fortran with subroutine calls to Assembly language subroutines. An exception to this is Program TSC, which operates the traveling scintillation counter. This program is written with a mixture of Fortran and Assembly language. A brief description of each major applications program is given in Reference 34.

Physical Properties of Liquids and Solids

The objective of the experimental program is to establish the hydrodynamic properties of the H-Coal reactor. Typical operating conditions of the H-Coal reactor and properties of the liquids at actual operating conditions were given in an earlier section. In this paper, kerosene/coal char slurries will be used which at about room temperature have properties similar to those of the H-Coal slurries in the H-Coal reactor. The properties of the kerosene are shown in Table VI. The surface tension of the kerosene was measured as a function of temperature. A Fischer Scientific surface tensiometer (Model 20) was used. The results indicate that surface tension varies linearly with temperature over the temperature range studied.