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CANMET CO-PROCESSING: A STATUS REPORT

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Synthetic Fuels Research Laboratory

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by

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ABSTRACT

CANMET is developing a process to simultaneously liquefy coal and upgrade bitumen, heavy oil or petroleum residues. This coprocessing concept is similar to a direct hydroliquefaction process in which the normal coal derived recycle oil is replaced with an externally supplied bitumen or other solvent. Alternatively, it can be viewed as an extension of the CANMET hydrocracking process where much greater coal concentrations are employed in the feed. The net result is a single stage process which combines aspects of both coal liquefaction and heavy oil hydrocracking technologies.

At the Synthetic Fuels Research Laboratory of CANMET a continuous bench-scale coprocessing unit has been used to demonstrate the feasibility of the concept using Forestburg subbitumnious C coal from Alberta and Cold Lake vacuum bottoms as solvent. The same coal has also been processed with an anthracene oil solvent to allow direct experimental comparison of product qualities and yields between coprocessing and liquefaction.

The characteristics of coprocessing were investigated by changing the major process variables over wide operating ranges including the base case of simple hydrocracking of the solvent. A number of significant trends were observed especially with respect to the effect of coal concentration in the slurry feed. These results are discussed including the nature of potential coal-solvent synergism during coprocessing.

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COITRAITEMENT CANMET: UN RAPPORT D'ÉTAPE

de

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RÉSUMÉ

CANMEt procède à la mise au point d'un procédé susceptible de permettre la liquéfaction du charbon tout en améliorant le bitume, le pétrole lourd ou les résidus de pétrole. Ce concept de cotraitement ressemble à un procédé d'hydroliquéfaction directe où le pétrole régulier recyclé tiré du charbon est remplacé par un bitume ou un autre solvant provenant de l'extérieur. on peut également le charge d'alimentation. le résultat net prend la forme d'un procédé à étape unique qui combine certains aspects de la technologie de la liquéfaction du charbon et de la technologie de l'hydrocraquage du pétrole lourd.

Au Laboratoire de recherche sur les carburants synthétiques de CANMET, on a utilisé une installation de cotraitement en continu, à l'échelle du banc d'essai, pour démontrer la faisabilité du concept en utilisant du charbon subbitumineux C Forestburg, en provenance de l'Alberta, et des fonds à vide de cold Lake comme solvant. L même charbon a également été traité avec un solvant de pétrole anthracénique pour permettre une comparaison expérimentale directe des qualités de produit et des rendements fournis par le cotraitement et la liquéfaction.

Les caractéristiques du cotraitement ont été étudiées en modifiant les grandes variables du procédé, sur d'importantes gammes d'exploitation, y compris le cas de base du simple hydrocraquage du solvant. Un certain nombre de tendances significatives ont été observées, tout particuliérement en ce qui concerne l'incidence de la concentration de charbon dans la charge d'alimentation semi-liquide. Ces résultats sont examinés, notamment la nature du potentiel synergétique charbon-solvant durant le cotraitement.

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INTRODUCTION

The close proximity of large reserves of low-rank coals and bitumens or heavy oils in Canada presents a special upgrading opportunity. CANMET has expended many years of research and development to bring its hydrocracking process to the commercialization stage. A logical extension is to investigate its potential for coprocessing coal and heavy oil resulting in a process having advantages over the individual upgrading of each feedstock. Therefore, a research program was initiated to develop a CANMET coprocessing technology. Autoclave and continuous bench-scale facilities were installed at CANMET's Energy Research Laboratories. Preliminary research beginning since the fall of 1981 has demonstrated the feasibility of coprocessing (1-4). Figure 1 shows the research and development plan for the next 5 years. Currently a 0.5 t/d (4 bbl/day) pilot plant is being constructed to investigate longer term process operability. It is expected to be operational by the beginning of 1986. Consideration is being given to the construction of a 200 bbl/day process development unit flexible enough to run in either the hydrocarcking or the coprocessing modes. The CANMENT hydrocracking process is being commercialized by Petro-Canada and Partec Lavalin. A 5,000 bbl/day demonstration plant is under construction at Montreal, Quebec. It is expected to be operational by the end of 1985. Operation of this plant in the coprocessing mode is projected for 1990.

When discussing coprocessing, there is a need to know how it compares with the individual upgrading of coal or oil feedstocks. Using results generated from the coprocessing of an Alberta subbituminous coal (Forestburg subbituminous C) and Cold Lake vacuum bottoms a comparison of each process was made. The same experimental unit (1 kg/h bench scale) was used to allow a more representative comparison between hydrocracking of the vacuum bottoms only (based on the CANMET hydrocracking process), liquefaction of the coal (using hydrogenated anthracene oil solvent) and coprocessing.

Also of major importance is the nature or characteristics of coprocessing as a separate processing route. For example, in the published literature there is reference to a "synergism" during coprocessing (5-7). This paper presents an overview of the characteristics of CANMET coprocessing with special emphasis on the effects of coal concentration in the slurry feed and the nature of this synergism.

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EXPERIMENTAL

FEEDSTOCKS

Forrestburg subbituminous C coal from Luscar Ltd. was used for all experiments. The nut-size coal was sent to Ontario Research Foundation for grinding (with dry ice) to $-74 + 37 \mu m$. Table 1 gives the characteristics of the ground coal.

Cold Lake vacuum bottoms from the Strathcona refinery of Imperial Oil Ltd. were used for the coprocessing and hydrocracking runs. The bottoms are nominally the +454°C cut. Table 2 gives the characteristics of this feedstock.

For the coal liquefaction experiments, the slurrying solvent was hydrogenated anthracene oil obtained from Crowly Tar Products Inc. It was hydrogenated by the Sandwell Beak Research Group using their process development unit. Table 3 shows the characteristics of the hydrogenated anthracene oil.

Feed slurries were prepared by mixing the desired amounts of coal, and vacuum bottoms or anthracene oil under atmospheric pressure in a stirred feed tank. A disposable catalyst (CANMET additive) was used and the amount of catalyst was kept constant on a moisture and ash free (maf) slurry feed basis for all experiments. CONTINUOUS-FLOW EXPERIMENTS

Figure 2 shows a schematic diagram of the CANMET continuous-flow coprocessing unit used for this work. The gas feed system consists of two identical pressure and flow controlled lines for hydrogen and carbon monoxide. The slurry feed system consists of a slurry feed tank, recirculation pump and a high pressure metering pump. The slurry feed tank is mounted on a scale to monitor and record the exact amount of slurry feed to the reactor. The recirculation pump provides additional mixing action and generates the necessary net positive suction head for the metering pump. The four-headed metering piston pump delivers the slurry feed at a preset rate to the reactor. Both the reacting gases and the slurry are fed to the bottom of a 1-L stirred tank reactor. The reactor is fitted with an overflow tube through which the product flows to either an off-specification receiver or a sample receiver. The vapours from either receiver pass through a water cooled condenser where condensible light ends are separated. The effluent gas then passes through a scrubber and an automatic high pressure let-down system. The volume of exit gas is metered before it is discharged to vent.

Both the whole slurry product and the light ends are discharged directly into pressurized vessels which are weighed before depressurization and product workup. PRODUCT WORKUP

Samples of the outlet gas were analyzed by gas chromatography to determine hydrogen, hydrocarbon gases (C_1-C_4) , hydrogen sulphide, and carbon oxides contents. Water was separated from the light ends by freezing and the organic light ends were then mixed with the whole slurry product. A sample of this mixture was distilled using a spinning band distillation unit into distillate (-525°C product) and residue (+525°C product) by a combination of atmospheric and vacuum distillation. The distillate was analyzed for:

- specific gravity by PARR densimeter;
- boiling range by g.c. simulated distillation according to ASTM D2287;
- hydrogen, carbon and nitrogen using a CHN 240 Perkin Elmer analyzer. Sulfur was determined by X-ray fluorescence, Tech. Chemical Analyser Model 100;
- PONA analysis by GC-MS using a GC/MS Finnigan 4000 instrument with INCOS data system;
- aromaticity by H.n.m.r. using a Varian CFT-20 pulse Fourier Transform instrument (Brown Ladner treatment).

The residue was analyzed to obtain:

- elemental composition; C, H, N, S and O contents
- the content of oils, asphaltenes and preasphaltenes by sequential soxhlet extraction
- ash content by ASTM D-482
- metal content by Jarrell Ash Model 850AA spectrometer
- The results presented were obtained from a total of 30 experimental runs. In

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26 runs, the material balance was better than $\pm 5\%$ and in 4 runs it was better than $\pm 7\%$.

COAL LIQUEFACTION, COPROCESSING AND HYDROCRACKING AS UPGRADING ALTERNATIVES

In comparing the alternative processes, it is necessary to define the basis for the comparison and to note terms and definitions that do not coincide with more common usage in coal liquefaction or hydrocracking technology. Figure 3 presents a schematic diagram of the basic process flows used for this comparison. Hydrocracking is represented by the coprocessing schematic less the separate coal feed. However, a small amount of coal is added to the bitumen as a processing additive in the CANMET hydrocracking process, which was selected for this comparison. The bench-scale continuous-flow experimental unit was used in different operating modes at the same process conditions. These conditions were close to the optimum for coprocessing. No attempt was made to optimize the liquefaction or hydrocracking processes since the basis for the comparison was to fix the operating conditions. Figure 3 shows a basic advantage of coprocessing over liquefaction in that no recycle oil stream is needed.

Since the comparison is basically between three stirred tank reactors of equal volume operating at the same space velocity, all calculated process data are referenced to each reactor input on a maf slurry feed basis. This is different from standard coal liquefaction practice where the net maf coal input is usually used as a basis for process comparisons. Also, when comparing pitch conversions between coprocessing and hydrocracking it should be noted that maf coal in the feed is considered as pitch (i.e. +525°C ash-free material) for the liquefaction and coprocessing cases. Normally, the CANMET hydrocracking process operates in the +85 wt % pitch conversion range. The process conditions chosen for this comparison resulted in a much lower pitch conversion.

PROCESS COMPARISON

Table 4 compares the experimental results from each process on the basis of 100 g maf slurry fed to the reactor. The liquefaction yields were calculated by subtracting the necessary recycle oil stream from the total oil yield (less the naphtha fraction) assuming that the hydrogenated anthracene oil solvent in this one-pass operation behaved as a steady state recycle solvent. On this basis

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coprocessing results in about 4-5 times more total oil yield (distillate + residual oil) than liquefaction but slightly less than hydrocracking. Due to the different solvents used in liquefaction and coprocessing, it is not surprising to see a much larger residue yield (+525°C ash-free product) for coprocessing. Under more severe operating conditions, the residue yield for hydrocracking can be reduced with corresponding increases in oil and gas yields. The higher gas yield for coprocessing compared with hydrocracking is probably due to a higher concentration of catalytically active mineral matter in the slurry feed in the coprocessing mode.

Similar coal conversions for liquefaction and coprocessing indicate that Cold Lake vacuum bottoms perform as well as the hydrogenated anthracene oil solvent, at least, under the process conditions used in this comparison. Other work has shown Cold Lake vacuum bottoms to be as good as or better hydrogen donor than tetralin in the absence of gaseous hydrogen (8).

The hydrogen consumption values show the expected trend with coprocessing slightly higher than hydrocracking due to the presence of much more coal. For liquefaction, a hydrogen consumption of 2.7 wt % on maf slurry feed (8.2 wt % on maf coal) was measured. This was higher than would be expected for a direct hydroliquefaction process with a total oil yield of 16.5 wt % on maf slurry feed (50 wt % on maf coal). However, the trend shown for hydrogen utilization efficiency from liquefaction to coprocessing to hydrocracking is typical. In fact, the hydrogen utilization efficiency for CANMET coprocessing is higher than that reported for most integrated two-stage coal liquefaction processes (9-11). DISTILLATE YIELDS AND CHARACTERISTICS

Total distillate yields for the three processes and their naphtha, light gas oil (LGO) and heavy gas oil (HGO) fractions are shown in Figure 4. Overall, there is a decreasing trend in distillate yields from hydrocracking, to coprocessing, to liquefaction. As a percentage of the total distillate yield, there is little difference in the naphtha, LGO and HGO fractions when comparing hydrocracking with coprocessing. However, coal liquefaction shows a smaller LGO yield and larger HGO yield as a percentage of the total distillate yield. This is probably due to the conversion of most of the coal into high molecular weight products with little upgrading of the hydrogenated anthracene oil solvent.

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Due to the use of a hydrogenated anthracene oil solvent and a single-pass operation, care must be taken in making yield comparisons with liquefaction. However, the total distillate yield for liquefaction of about 14.5 wt % (44.0 wt % on maf coal basis) is in the range reported for single-stage direct hydroliquefaction processes of this type. The overall trends shown in Figure 4 are correct, but the absolute values shown for the distillate fraction yields for liquefaction would have to be verified by running long enough to generate a true steady state recycle solvent and products.

Figure 5 shows some of the characteristics of the whole distillate (IBP to 525°C) from each process. Because of the nature of single-pass operation only the trends for the liquefaction data should be considered and not the absolute values. The trends in aromaticity, H/C atomic ratio and sulfur content are a strong function of the nature of the slurry feedstock in each case. Using H/C atomic ratio change between feedstock and distillate as an indication of hydrogenation shows a 36% increase for coprocessing versus 18% for hydrocracking and only 7% for liquefaction. The higher value for coprocessing could be due to a slight catalytic effect of the coal mineral matter. Sulfur removals (to gases plus residue) for hydrocracking and coprocessing are equal at about 75 wt %. For liquefation the distillate sulfur content is much lower because of the low sulfur content of the starting feedstock (0.17 wt %). As expected, aromaticities are higher for liquefaction due to the nature of the anthracene oil solvent employed. RESIDUE YIELDS

Figure 6 shows a comparison of the residue yields for the three processes. Residual oil values follow the expected trend based on the pitch or residue contents of the feedstocks. However, the preasphaltenes yield for hydrocracking is significantly low, although the sum of asphaltenes plus preasphaltenes for coprocessing and hydrocracking is almost equal. Again, this is due to the nature of the feedstocks. Vacuum bottoms contain very little preasphaltenes. In coprocessing, the liquefaction of coal through the preasphaltenes-asphaltenes route would result in higher preasphaltenes content, primarily derived from the coal portion of the feedstock. Due to the presence of large amounts of coal during

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coprocessing and liquefaction, much larger amounts of THF insolubles in the residues can be expected compared with hydrocracking. However, coprocessing does not generate significantly larger amounts of THF insolubles compared with liquefaction. Also, the ash content of the residue is about 11% for coprocessing versus 27% for liquefaction.

CANMET COPROCESSING CHARACTERISTICS

The coprocessing of Forestburg subbituminous C coal and Cold Lake vacuum bottoms was investigated over a wide range of operating conditions. A series of experiments was carried out using a slurry of the coal and the heavy oil with a fixed coal concentration. The gross performance was examined at various processing severities.

YIELDS

Figure 7 shows the distillate (C_5 to $-525^{\circ}C$ product), residue (+525°C product) and hydrocarbon gas yields as a function of pitch conversion. Distillate yields as high as 75.4 wt % (based on maf slurry feed) were obtained at a pitch conversion of 87.4 wt %.*. The linear increase of distillate yield is accompanied by an equivalent linear decrease in the residue yield. The increase in the gas yield with pitch conversion is relatively small, indicating that at the highest level of severity,most of the hydrocracking resulted in the production of liquid rather than gaseous products.

Figure 8 shows the yields of distillate fractions as a function of pitch conversion. The overall increase in distillate yield at higher severity is due to an increase of light and middle distillates since the heavy gas oil II fraction decreases at higher severity. Control of the product slate by controlling the severity of coprocessing seems feasible if the market value of the desired products will tolerate a slight penalty in the overall yield obtained at lower severities.

* Pitch Conversion = 1 - +525°C fraction in product, including coal x 100 +525°C fraction in feed, including coal

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DISTILLATE CHARACTERISTICS

Figure 9 shows the specific gravity and H/C atomic ratio of the whole distillate product as a function of pitch conversion. A very distinct drop in the specific gravity with increasing pitch conversion can be observed. The specific gravity changed from 0.953 to 0.893 which corresponds to a 10 unit increase in the "API gravity over the range of pitch conversions tested. The decrease in the specific gravity of the distillate product relative to that of the feed ranges from 0.085 to 0.145. This corresponds to an "API uplift in the range of 12 to 22 "API. Two-stage coprocessing was reported to give "API uplifts in the range of 6 to 12.4" API (6). Despite the scatter in the H/C atomic ratio data, linear regression shows an overall increase as expected from the drop in the specific gravity.

Figure 10 shows the distillate sulfur content. Comparing the sulfur content of the slurry feed with that of the distillate shows that desulfurization occurs to a significant extent even at low severity. The sulfur removal continues to increase at higher severities.

COAL CONVERSION

Coal conversions from 51 wt % to 87 wt % were obtained depending on the severity. This level of conversion is similar to that obtained using classical coal liquefaction solvents. It was observed that pitch conversion was more sensitive to changes in temperature and space velocity than coal conversion. Figure 11 shows that the primary dissolution of coal during coprocessing can be accomplished to a large extent at low temperatures. The activation energy for secondary upgrading is higher, therefore higher temperatures are required for significant secondary upgrading to take place. Figure 12 shows the effect of space velocity on coal conversion and pitch conversion. The less significant effect of space velocity on coal conversion compared with pitch conversion is not unexpected. It is known from short contact time studies in coal liquefaction that high coal conversions can be obtained in the first few minutes of reaction (12). This would support a two-stage approach for coprocessing where most of the primary coal dissolution could be accomplished in a separate reactor at high space velocity and where secondary upgrading would be allowed to take place in a second-stage reactor at lower space velocity and using appropriate catalysts. Optimization of the overall liquid yields would be accomplished by controlling the temperatures and the space velocities of both reactors. Preliminary bench-scale tests at CANMET indicate that the two-stage approach is feasible, although much more research is needed to determine overall process performance improvement over the single-stage approach (13).

YIELDS OF RESIDUE COMPONENTS

Soxhlet extractions were carried out on all the residue products in order to estimate the individual yields of residual oils, asphaltenes, preasphaltenes and THF insolubles. Figure 13 shows the variation of the yields of each component with process severity. At high pitch conversions, the sum of yields of residue components, i.e., the overall residue yield based on maf slurry feed is much lower than the original coal concentration in the slurry feed. This provides direct evidence that the coal portion of the feedstock contributes to the distillate product. Figure 13 demonstrates that at all levels of pitch conversion, residual oils constitute a major portion of the nondistillate product. It also indicates that preasphaltenes do not build up to a significant extent with increasing severity. Due to their high reactivity, preasphaltenes quickly react progressively to form asphaltenes or oils. The THF insolubles depletion and that of the asphaltenes are equivalent. This suggests that the sensitivity of asphaltene and coal conversions to process severity is similar. COPROCESSING USING SYNTHESIS GAS

The same feedstock at the same coal concentration was tested using synthesis gas in place of hydrogen at a CO/H_2 molar ratio of 1:2. It was found that the distillate, residue and gas yields using synthesis gas were identical to those using hydrogen (14). Also, coal and pitch conversions were similar for both reducing atmospheres. A significant difference, however, was observed in the hydrogen consumption. Figure 14 shows the hydrogen equivalent consumption in both atmospheres as a function of pitch conversion. For synthesis gas, hydrogen

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equivalent consumption is calculated taking into account the hydrogen generated from carbon monoxide through the water-gas shift reaction. Under low severity conditions significantly lower hydrogen consumption is observed for synthesis gas. This difference appears to diminish at higher pitch conversions.

Another distinguishing feature associated with using synthesis gas, is the product slate. Figure 15 shows the yields of distillate fractions using synthesis gas. A comparison of Figures 8 and 15 indicates that the relative yields of distillate fractions are different for synthesis gas compared with hydrogen. At high severity, synthesis gas tends to produce more middle distillate and less naphtha. This trend is reversed at low severity. The simulated distillation data shown in Figure 16 supports this statement. The curves for distillates obtained at medium severity coincide. Examination of the specific gravity and the H/C atomic ratio of the distillates also supports this observation.

EFFECT OF COAL CONCENTRATION ON PROCESS PERFORMANCE

To assess coprocessing as an upgrading technology, it is important to investigate the effect of increasing coal concentration in the slurry feed. In this section a technical assessment provides some insight into the effect of coal concentration on the yields and qualities of the products. However, an economic evaluation is also needed to complement the technical assessment. CANMET is therefore planning a separate study for the economic assessment of coprocessing versus hydrocracking.

DISTILLATE YIELDS AND SYNERGISM

A number of reports in the literature refer to a "synergism" during coprocessing (5-7). However, careful examination reveals that the definition of this synergism is somewhat arbitrary and that the comparisons claiming the effect are not necessarily uniform. There can be a synergism related to process operability or some other non-quantitative process variable. However, for CANMET coprocessing, it is felt that the best definition from a quantitative process point of view should be based on distillate yield using the same basis for comparing different processes, i.e., distillate yield based on a wt % maf slurry feed basis. To investigate the potential synergism during CANMET coprocessing, distillate yields for 11 levels of maf coal concentration in the slurry feed were measured. All tests were carried out under identical operating conditions. Figure 17 shows the variation of the distillate yield with increasing coal concentration. There is a definite true synergism at low coal concentrations. In this region, increases in the distillate yield of up to 9 wt % were found over operating with no coal. For the range of coal concentrations of interest in coprocessing, distillate yields remain constant and equal to those for the no coal case and start to drop for higher coal concentrations. Therefore, over the range of coal concentrations of most interest, there is no observed synergism for coprocessing compared with hydrocracking of the solvent only based on distillate yield. However, there was a marked improvement in the operability of the process for coal concentrations in this range compared with the no coal case. One goal of CANMET's research program is to broaden the distillate yield peak so that a true process synergism can be obtained over a wider range of coal concentrations.

For comparison, the dashed line in Figure 17 is a tie-line connecting the yield obtained for the no coal case (on a <u>maf feed basis</u>) with the 100% coal case (coal liquefaction on a <u>maf coal feed basis</u>). This type of tie-line has been used to indicate the expected distillate yields from coprocessing assuming additive behaviour of the mixtures (5). Except at the highest coal concentrations shown, CANMET coprocessing results in distillate yields above the tie-line and could therefore be credited as showing a synergism. However, over the range of coal concentrations where a plateau in distillate yield was observed, there may also be a synergism in terms of hydrogen addition. As the coal concentration was increased in the feedstock, the H/C atomic ratio of the feed became lower. More hydrogen addition would be required to achieve the same distillate yield (and quality as indicated in the distillate characteristics section) from a lower quality feedstock.

RESIDUE AND GAS YIELDS

Figure 18 shows corresponding residue and gas yields as a function of coal concentration. As expected, the residue yields result in a rough mirror image of the distillate yields, since the hydrocarbon gas production remains relatively constant with increasing coal concentration. Since the catalyst concentration was

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fixed on a maf slurry feed basis, the increase in residue (and decrease in distillate) at higher coal concentrations may be related to the need for higher catalyst concentrations at these levels, (i.e., it may be preferable to fix the catalyst concentration on a maf coal basis).

DISTILLATE CHARACTERISTICS

Over the range of coal concentrations studied, the distillate quality remained basically constant. Figure 19 shows specific gravity, aromaticity and H/C atomic ratio. There is only a marginal decrease in the H/C atomic ratio and consequently a slight increase in the aromaticity of the distillate. Detailed compound type analysis showed that the percentage of paraffinic, olefinic, aromatic and polar compounds in the distillate did not change significantly with the addition of coal to the feedstock (15).

Figure 20 shows the sulfur content of both the distillate and the slurry feed as a function of coal concentration. An overall decrease in the sulfur content was observed with increasing coal concentration. However, this is primarily due to the feedstock progressively containing less sulfur as more coal replaces the vacuum bottoms.

COAL CONVERSION

Figure 21 presents the coal conversion data. Despite some scatter, a trend is shown as a decrease followed by a gentle increase in the coal conversion. We have no explanation for the initial decrease in coal conversion. The subsequent increase, however, indicates that at all coal concentrations, the solvent to coal ratio used was not a limiting factor. More coal was converted as more coal was introduced. Also, the slight increase in coal conversion at higher levels of coal concentration may be attributed to larger amounts of mineral matter which can have a slight catalytic effect.

CONCLUSIONS

1. The feasibility of coprocessing Forestburg subbituminous C coal and Cold Lake vacuum bottoms was demonstrated. Coprocessing of this feedstock was compared with coal liquefaction and hydrocracking under identical operating conditions. This comparision showed:

- (a) The total oil yield for coprocessing was 4.6 times greater than that for coal liquefaction and 7% less than that for hydrocracking.
- (b) Similar coal conversions were obtained for coprocessing and coal liquefaction.
- (c) The hydrogen consumption for coprocessing was 50% higher compared with coal liquefaction and 21% higher compared with hydrocracking.
- (d) The hydrogen utilization efficiency for coprocessing was 2.6 times as much as that for coal liquefaction and 30% lower than that for hydrocracking.
- (e) The hydrogen addition was 36% for coprocessing, 18% for hydrocracking and 7% for coal liquefaction.
- (f) The distillate obtained from coprocessing had lower specific gravity, higher H/C atomic ratio, lower aromaticity and higher sulfur content, compared with the distillate obtained from coal liquefaction.
- 2. The coprocessing of Forestburg coal and Cold Lake vacuum bottoms was tested over a range of operating conditions. Examination of product yields and qualities showed:
 - (a) Distillate, residue and hydrocarbon gas yields changed linearly with pitch conversion.
 - (b) The increase in the overall distillate yield with pitch conversion was mainly due to an increase in middle distillate yield.
 - (c) At a typical pitch conversion the H/C atomic ratio of the distillate product was 1.58, the API gravity was 25.7° and the sulphur content 1.56 wt %.

- (d) The level of coal conversion was similar to that reported for good coal liquefaction solvents.
- (e) Coal conversion was more sensitive to temperature variations than to space velocity variations.
- (f) The coal portion of the feedstock contributes to the distillate product yield, particularly at high severity.
- (g) Using synthesis gas in place of hydrogen reduces the hydrogen consumption at low severities and enhances the production of middle distillates at high severities.
- 3. The process performance was studied over a range of coal concentrations and this revealed the following:
 - (a) At very low coal concentrations, the distillate yields were remarkably enhanced compared with the no coal case.
 - (b) Over a considerable range of coal concentrations the distillate yield was constant and equal to that obtained in the no coal case.
 - (c) Over a large range of coal concentrations synergism was observed in terms of process operability and hydrogen addition rather than distillate yields.
 - (d) Above a critical level of coal concentration the distillate yields decrease significantly.
 - (e) Over the range of coal concentrations tested, the distillate quality was essentially constant.

REFERENCES

- Fouda, S.A., Kelly, J.F. and Rahimi, P.M. "The Coprocessing Behaviour of Cold Lake Vacuum Bottoms and an Alberta Subbituminous Coal", <u>Division Report ERP/ERL 82-53</u> (Confidential), CANMET, Energy, Mines and Resources Canada; 1982.
- Fouda, S.A. and Kelly, J.F.
 "The Coprocessing of an Alberta Subbituminous Coal and Distillate Fractions of Cold Lake Heavy Oil", <u>Division Report ERP/ERL 83-45</u> (Confidential), CANMET, Energy Mines and Resources Canada; 1983.
- 3. Rahimi, P.M., Fouda, S.A. and Kelly, J.F. "Characteristics of Distillates, Gases and Residues Obtained in the Coprocessing of Cold Lake Vacuum Bottoms and an Alberta Subbituminous Coal", <u>Division Report</u> ERP/ERL 82-54 (Confidential), CANMET, Energy, Mines and Resources Canada; 1982.
- Fouda, S.A. and Kelly, J.F.
 "Cold Lake Heavy Oil Fractions as Coprocessing Media", <u>Division Report</u> ERP/ERL 84-25 (Confidential), CANMET, Energy, Mines and Resources Canada; 1984.
- Duddy, J.E. and MacArthur, J.B. "Coal/Oil Coprocessing", AIChE, Summer National Meeting, August 21, 1984, Philadelphia, Pennsylvania, Paper No. 16b.
- Shinn, J.H., Dahlberg, A.J., Kuehler C.W. and Rosenthal, J.W. "The Chevron Co-Refining Process", Ninth EPRI Contractors' Conference on Coal Liquefaction Palo Alto, California, May 1984.
- 7. De Vaux, G.R., Johanson, E.S. and Chervenak M.C. "The Coil Process, First Generation Commercial Coal Liquefaction Plant" Alternative Energy Sources, Nejat Vezirglu, T. Editor, Volume 7, Page 3267 (1978).
- Rahimi, P.M., Price, J.T. and Kelly, J.F.
 "Are Bitumens/Heavy Oils Hydrogen Donors? Quantitative Determination of Hydrogen Donor Abilities of Bitumens and Pitch Materials using 1, 1 -Binaphthyl and Anthracene", <u>Division Report ERP/ERL 84-18</u> (Confidential), CANMET, Energy, Mines and Resources Canada; 1984.
- 9. Moniz, M.J., Pillai, R.S., Lee, J.M. and Lamb, C.W., "Process Studies of Integrated Two-Stage Liquefaction at Wilsonville" Ninth EPRI Contractors' Conference on Coal Liquefaction Palo Alto, California May 1984.

- 15 -

- Peluso, M., Schiffer, A.N., Schindler, H.D. and Chen, J.M. "Recent Developments with the Lummus Crest Integrated Two Stage Liquefaction Process" Ninth EPRI Contractors' Conference on Coal Liquefaction Palo, Alto, California, May 1984.
- 11. MacArthur, J.B., Comolli, A.G. and McLean, J.B. "HRI's Two Stage Catalytic Liquefaction Program" Ninth EPRI Contractors' Conference on Coal Liquefaction, Palo Alto, California, May 1984.
- 12. Longanbach, J.R., Droege, J.R. and Chauhan, S.P. "Short-Residence-Time Coal Liquefaction" Coal Liquefaction Fundamentals, Whitehurst, D., Editor, ACS Symposium Series No. 139, Page 164, 1980.
- 13. Ikura, M.I., Fouda, S.A. and Kelly, J.F. "The Coprocessing of an Alberta Subbituminuous Coal and Cold Lake Vacuum Bottoms Using Synthesis Gas: III - A Proposal on Two Stage Coprocessing", <u>Division Report ERP/ERL 84 -47 (Confidential)</u>, CANMET, Energy, Mines and Resources Canada; 1984.
- Ikura, M.I., Fouda, S.A. and Kelly, J.F.
 "The Coprocessing of an Alberta Subbituminuous Coal and Cold Lake Vacuum Bottoms Using Synthesis Gas: I - Gross Performance, <u>"Division Report ERP/ERL</u> 84-26 (Confidential), CANMET, Energy, Mines and Resources Canada; 1984.
- 15. Ahmed, S., Fouda, S.A., Rahimi, P.M., and Sawatzky, H. "Compound Type Studies of Coprocessing Products of Cold Lake Vacuum Bottoms and Alberta Subbituminous Coal", <u>Division Report ERP/ERL 83-22</u> (Confidential), CANMET, Energy, Mines and Resources Canada; 1983.

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CHARACTERISTICS OF FORESTBURG SUBBITUMINOUS C COAL

PROXIMATE ANALYSIS "AS RECEIVED ", w1 %

MOISTURE	19.17
ASH	7.68
VOLATILE MATTER	34.00
FIXED CARBON	39.15

ULTIMATE ANALYSIS "DRY BASIS", w1%

CARBON	64.04
HYDROGEN	3,87
SULPHUR	0.53
NITROGEN	1.65
ASH	9.50
OXYGEN BY DIFFERENCE	20.41

METAL CONTENT

Fe	2379	ppm
NI	18	ppm
V	TRACE	

CALORIFIC VALUE	VALUE	4933	cal/g
	8879	Btu/lb	

PETROGRAPHIC ANALYSIS

MEAN RO (RANDOM)	0.42	
VITRINITE	92.2	vol %
LIPTINITE	2.6	vol %
INERTINITE	3.1	vol %
MINERAL MATTER	2.1	vol %

SUPPLIED BY THE ENERGY RESEARCH UNIT, UNIVERSITY OF REGINA

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CHARACTERISTICS OF COLD LAKE VACUUM BOTTOMS

GENERAL

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SPECIFIC GRAVITY 15/15°C 1.038		nt %
CONRADSON CARBON RESIDUE	17.1 1	vt %
ASPHALTENES	23.48 v	nt %
PRE ASPHALTENES	0.2 1	nt %
ARDMATICITY (I H NMR)	34.5	*
VISCOSITY AT 80°C	249.12 p	polse
100°C	39.40	M
110°C	21.59	M.

DISTILLATION (SPINNING BAND METHOD)

IBP	420	•C
DISTILLATE (-525°C)	16.75	wt %
RESIDUE (+ 525 °C)	83.25	wt %

ELEMENTAL ANALYSIS, w1%

CARBON	° 78 .6
HYDROGEN	9.3
SULPHUR	5.5
NITROGEN	0.6
ASH	0.0
OXYGEN BY DIFFERENCE	5.9

METAL CONTENT, ppm

NI	93
V	235
F●	18

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CHARACTERISTICS OF HYDROGENATED ANTHRACENE OIL

GENERAL

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SPECIFIC GRAVITY 15/15°C	1.08
VISCOSITY AT 15 °C	. 0 • 205 poise
AT 25 °C	0.146 poise
AROMATICITY	81.01 %

DISTILLATION

IBP	243	•C
50 VOL % OFF AT	308	•C
FBP	376	•C

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ELEMENTAL ANALYSIS, w1%

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COMPARISON OF PROCESSES

BASIS : g/100 maf SLURRY FEED

CONDITIONS	LIQUEFACTION	COPROCESSING	HYDROCRACKING	
COAL (mof) CONCENTRATION TEMPERATURE PRESSURE SPACE VELOCITY H ₂ FEED RATE	33	30 SAME " "		
SOLVENT	ANTHRACENE	VACUUM BOTTOMS	VACUUM BOTTOMS	
YIELDS				
GASES (C1-C4)	1.3	5.4	3.8	
TOTAL OIL	16.5	75.9	81.9	
RESIDUE	13.2	29.6	22.5	
CONVERSIONS				
COAL (wt%)	85.6	83.9		
PITCH (wt%)	57.5	66.5	72.2	
HYDROGEN				
CONSUMPTION	2.7	4.0	3,3	
UTILIZATION EFFICIENCY (g dist./gH ₂)	6.0	15.5	22.2	

RESEARCH AND DEVELOPMENT PLAN

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	TIME FRAME				
UNIT OPERATION	85	86	87 8	<u>38 8</u>	<u>9 90</u>
Bench Scale Research (Ikg/hr) CANMET/ERL		.		•	<u>,</u>
Lab Scale Pilot Plant (4barrels/day) De CANMET/ERL	sign/Constru	ction	Operation —	•	-
Process Development Unit	Design / C	on structio	<u> </u>	•	······································
Process Developer / EMR	Design7C	0131100110	" Start-up Hydrocracking mode	I Start-up Coprocessin mode	g .
Demonstration Plant			• • • • • • • • • • • • • • • • • • • •	••	
Proc ess Developer/EMR	Oj	peration H	ydrocracking mo	de	T Coprocessing mode



FIGURE 2

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PROCESS SCHEMATICS

FIGURE 3

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FIGURE 5

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COPROCESSING YIELDS AS A FUNCTION OF SEVERITY

FIGURE 7

FIGURE 8

YIELDS OF DISTILLATE FRACTIONS AS A FUNCTION OF SEVERITY





DISTILLATE CHARACTERISTICS AS A FUNCTION OF SEVERITY











COAL AND PITCH CONVERSION VS TEMPERATURE



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COAL AND PITCH CONVERSION VS SPACE VELOCITY











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FIGURE 14

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YIELDS OF DISTILLATE FRACTIONS AS A FUNCTION OF SEVERITY - SYNTHESIS GAS



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FIGURE 16

SIMULATED DISTILLATION CURVES



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FIGURE 17

- 37 -





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FIGURE 18





FIGURE 19

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SULFUR CONTENT VS COAL CONCENTRATION

FIGURE 20



FIGURE 21

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- 41 -