THE EFFECT ON PRODUCT YIELD STRUCTURE AND PRODUCT QUALITY IN THE HYDROTREATMENT OF COAL DERIVED EXTRACT PRIOR TO DEASHING IN A TWO-STAGE LIQUEFACTION PILOT PLANT OPERATION

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INTRODUCTION

A two-stage liquefaction (TSL) operation typically consists of the sequential processing of coal in three distinct steps. These are: a thermal liquefaction first stage, an intermediate deashing step, followed by a second-stage hydrotreating unit. The TSL operation provides flexibility in that a number of process options are possible, depending on the sequencing of process units, and recycle of process streams (1, 2). This paper provides a comparison of the effects of resequencing process units in a TSL operation on product yields and product quality for processing bituminous coal (Illinois No. 6, Burning Star Mine).

The configuration modes compared were the integrated TSL (ITSL) mode (Figure 1) and the reconfigured integrated TSL (RITSL) mode (Figure 2). In both modes, the reaction stages are coupled by recycling full-range hydrotreated resid and distillate solvent from the hydrotreater to the thermal liquefaction step. The RITSL mode differs from the ITSL mode in that for the RITSL mode, the vacuum-flashed product from the thermal stage is hydrotreated prior to being deashed.

PRODUCT YIELD COMPARISON

A wide range of thermal and hydrotreater reaction conditions were investigated in the ITSL mode to determine their effects on product yield structure (3). For comparison purposes, Table 1 lists a typical set of RITSL conditions relative to three sets of ITSL conditions. Thermal stage reaction conditions are comparable for the runs listed in Table 1. Catalytic stage reactor conditions for ITSL-1 are fairly comparable to RITSL, with a 10°F higher reactor temperature for ITSL-1. A higher temperature of 740°F was used at an extended catalyst age for ITSL-2. In ITSL-3, catalyst age was maintained in the 1300-1400 lb resid/lb catalyst range with catalyst addition and withdrawal, and space velocity was decreased to 0.75 lb feed/(hr·lb catalyst).

For a similar reaction severity, RITSL indicates a higher distillate yield, accompanied by a significantly higher hydrogen consumption. Hydrogen efficiency is hence lower for RITSL as compared with ITSL.

A typical set of process unit yields is given in Table 2 for the RITSL and ITSL modes. Gaseous hydrogen consumption was comparable for both modes in the thermal stage, but was significantly higher for the RITSL mode in the hydrotreater stage, despite a slightly lower hydrotreater temperature employed for RITSL mode. The thermal unit resid yield for the RITSL mode was higher than that typically observed for the ITSL mode, which may be attributed to differences in the process solvent. Resid conversion in the hydrotreater was significantly higher for the RITSL mode, resulting in a higher distillate yield. The amount of organics rejected with the ash concentrate in the CSD unit was comparable for both modes. The H/C atomic ratio of the organics was higher for the RITSL mode, which implies a slightly higher rejection, on an energy basis for the RITSL mode.

The conversion of coal to cresol solubles in the individual process units and for the TSL system are shown in Table 3, for the ITSL and RITSL configuration modes. For similar thermal unit reaction conditions, observed coal conversions for the ITSL mode (92.5±1.1) were similar to the RITSL mode (91.7±1.4). In the ITSL mode, a statistically significant increase is observed in unconverted coal yield, in the fractionation and deashing steps (-4.8±1.4), downstream of the thermal reactor. In the RITSL mode, conversion of coal in the hydrotreating unit (0.7±1.5), and downstream of the reaction stages (-1.4±0.9) is statistically insignificant. Hence, for similar thermal unit coal conversions, the two-stage coal conversion is higher for RITSL mode as compared with ITSL mode. In both configuration modes, there was pressure letdown, and absence of hydrogen atmosphere between reaction stages. In the RITSL mode, the cool-down and heat-up temperature cycles of the liquefaction extract and the hold-up time between stages is reduced in comparison to the ITSL mode. The adverse effect of interstage cooling on regressive reaction occurrence, leading to reduced coal conversion and distillate yield has been reported (4). This interstage processing difference between configuration modes may have reduced the potential for regressive reaction occurrence for the RITSL mode, explaining the observed differences in overall coal conversion.

PRODUCT QUALITY COMPARISON

"Synthetic crude blends" were prepared for the RITSL and ITSL product slates, by combining all product streams in proportion to their respective flow rates. A comparison of the elemental contents for various boiling-point ranges of the crudes is shown in Table 4.

A slightly lower naphtha yield but a higher distillate yield was obtained for the RITSL crude in comparison to the ITSL crude. The hydrogen contents for comparable fractions were much higher for the RITSL crude. This result is to be expected because of the comparatively higher hydrogen consumption observed for RITSL mode, for similar hydrocarbon gas yield. The heteroatomic content in the product fractions was also lower for the RITSL crude, indicating an overall, better quality product compared with the ITSL crude.

CATALYST REQUIREMENT COMPARISON

Batch deactivation trends for resid conversion in the hydrotreater were developed for the RITSL and ITSL modes (1), using a first-order resid conversion model (5). The trends showed an initial period of rapid deactivation, followed by slower deactivation rates, for the deashed (ITSL) extract as well as for the nondeashed (RITSL) extract. The deactivation rates were comparatively higher for the nondeashed extract. However, the nondeashed resid was observed to be more reactive (higher resid conversion rate constants) as compared to the deashed resid, over the range of catalyst age investigated.

The batch deactivation data developed for the deashed and nondeashed bituminous runs may be used to estimate catalyst requirements in a steady-state, catalyst addition/withdrawal operation. The equilibrium activity is computed by combining the equilibrium catalyst residence time distribution (RTD) function and the activity function as shown below:

$$Keq = \int_0^\infty RTD(t)K(t)dt \qquad 1)$$

The RTD function for a continuous, stirred-tank, recycle reactor (6), is given as:

$$RTD(t) = \frac{1}{\bar{t}} e^{-t/\bar{t}} \quad 2)$$

where $\bar{t}$ = catalyst hold-up/catalyst addition rate.

Integration of Equation 1 for a batch catalyst activity function has been reported (7).

The projected catalyst requirement for deashed and nondeashed bituminous extracts for steady-state operation is shown in Figure 3. The catalyst requirement is projected for a fixed level of resid conversion and for similar reactor space rates. Owing to a higher resid reactivity, the catalyst requirement for the nondeashed extract, in the temperature range of 680-730^{o}F, is projected to be lower than the catalyst requirement for the deashed extract.

CONCLUSIONS

Significant changes were observed in product yields and product quality by hydrotreating coal derived extract prior to the deashing step. At approximately comparable reaction conditions, overall coal conversion and hydrotreater resid conversion were higher for the RITSL mode. A higer distillate yield accompanied by a significantly higher hydrogen consumption was reported for the RITSL mode, yielding product fractions of higher hydrogen and lower heteroatomic content, as compared to the ITSL mode. For a fixed level of resid conversion and for similar reactor space rates, catalyst requirement is projected from batch deactivation trends to be lower for the nondeashed extract (RITSL) mode.

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Table 1

Two-Stage Reaction Conditions and Yields for RITSL and ITSL Modes

Mode Catalyst Operating Mode	RITSL Batch	ITSL-1 Batch	ITSL-2 Batch	ITSL-3 Addition/ Withdrawal
Thermal Stage				
Reactor temperature (°F)	810	810	810	810
Inlet hydrogen partial pressure (psi)	2160	2040	2040	2040
Coal space velocity [(lb/hr-ft^3(>700°F)]	27	28	28	26
Catalytic Stage				
Reactor temperature (°F)	710	720	740	720
Space velocity (lb feed/hr-lb cat)	0.9	1.0	1.0	0.75
Catalyst Age (lb resid/lb cat)	445-670*	350-400	1200-1350	1300-1400**
Two-Stage Yield***(% MAF Coal)				
C_1-C_3 Gas	6	7	6	6
C_4+ Distillate	62	59	57	61
Resid	3	4	7	3
Hydrogen Consumption	-6.1	-5.3	-5.4	-5.6
Hydrogen Efficiency (lb C_4+Dist/lb H_2 Cons.)	10.2	11.1	10.6	10.8

* Resid contains unconverted coal and coal ash components.
** Average catalyst age: catalyst addition/withdrawal rate of 1.0 lb/ton (MF) coal employed.
*** Elementally balanced yield structure.

Table 2

Process Unit Yields for RITSL and ITSL Modes

Mode	RITSL	ITSL
Thermal Unit Yield (% MAF Coal)		
C_4+ Distillate	27.0	34.1
Resid	50.0	46.5
Hydrogen Consumption	-1.7	-1.5
Hydrotreater Unit Yield (% MAF Coal)		
C_4+ Distillate	35.0	24.9
Hydrogen Consumption	-4.4	-3.8
CSD Unit (% MAF Coal)		
Organic Rejection with Mineral Ash	19.6	21.0
[Atomic (H/C) of Rejected Organics]	0.68	0.75

Table 3

Effect of Configuration Mode on Coal Conversion*

Mode	Thermal Stage Conversion	Catalytic Stage Conversion	CSD Unit "Conversion"	Two-Stage Conversion**
ITSL	92.5±1.1		-4.8±1.4	87.7±1.3
RITSL	91.7±1.4	0.7±1.5	-1.4±0.9	91.0±0.8

* Conversion of Coal to Cresol Solubles.
** Two-Stage Conversion = Sum of Process Unit Conversions.

Table 4

Product Quality Comparison for RITSL and ITSL "Synthetic Crudes"

Distillation Cut	Wt % of Crude	Elemental (Wt %)				
		C	H	N	S	O (diff)
ITSL*						
Naphtha (IBP-360°F)	18.4	85.21	12.86	845 ppm	0.36	1.50
Distillate (360°F-650°F)	45.7	86.34	10.73	0.23	0.22	2.48
Gas Oil (650°F-1000°F)	35.0	89.07	9.69	0.31	0.16	0.76
Resid (1000°F)	0.9	86.71	6.94	1.13	0.60	3.15
RITSL**						
Naphtha (IBP-360°F)	14.9	85.50	14.07	500 ppm	0.35	0.03
Distillate (360°F-650°F)	49.8	86.74	11.54	0.23	0.16	1.33
Gas Oil (650°F-1000°F)	35.3	89.48	10.44	0.06	0.02	-

*Samples obtained with hydrotreater temperature of 730°F; catalyst age of approximately 1,400 lb resid/lb catalyst.
**Samples obtained with hydrotreater temperature of 700°F; catalyst age of approximately 350 lb resid/lb catalyst.

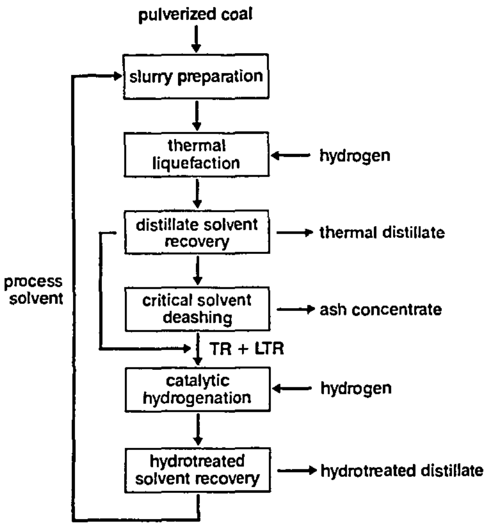

Figure 1. Block Diagram of the ITSL Configuration Mode

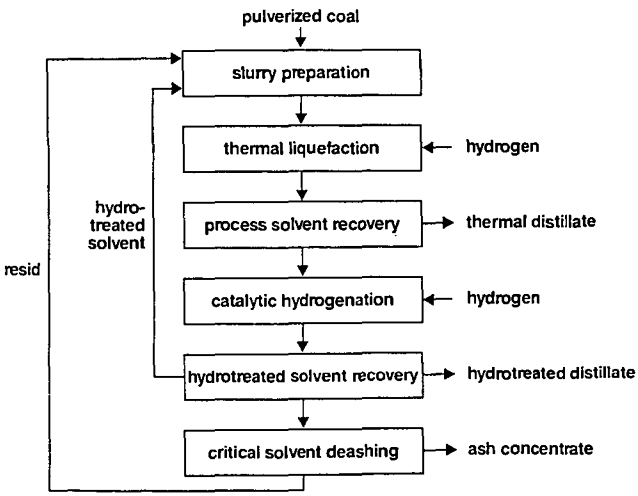

Figure 2. Block Diagram of the RITSL Configuration Mode

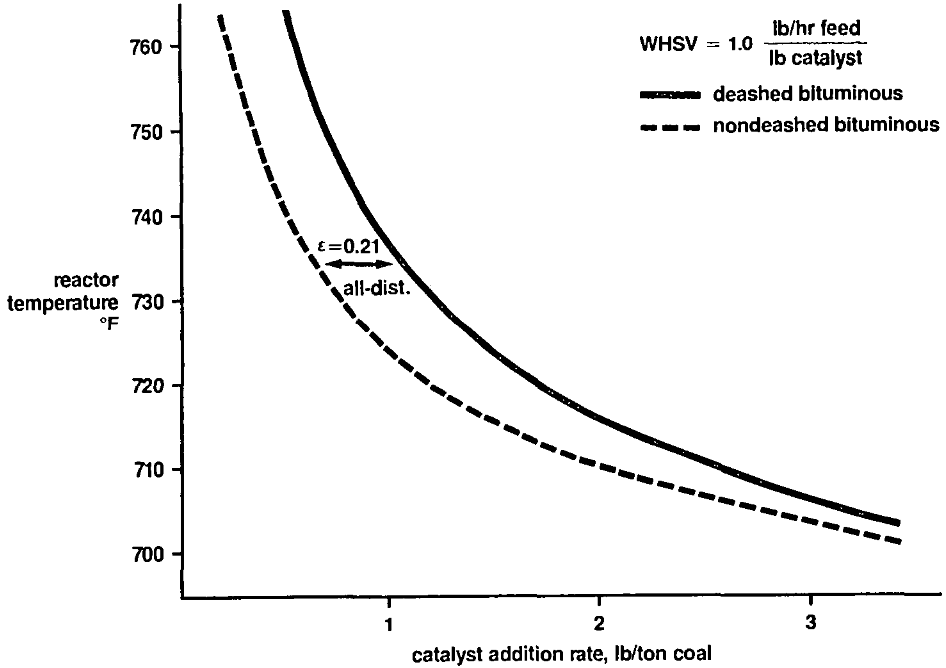

Figure 3. Projected Catalyst Requirement for RITSL and ITSL Modes