

## Advances in Steady-State Process Modeling of Oil Shale Retorting

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### **Abstract**

Steady-state process modeling of oil shale retorting using simulation software Aspen Plus is progressed. More components are included to allow all significant mineral and organic reactions to be represented. Secondary thermal cracking reactions are added to predict effects on product distribution and composition. Stoichiometric reactor blocks are replaced with kinetic reactor block models that predict the extents of conversions as a function of temperature, composition, and volume. As an example, a model is developed for the retorting of Green River oil shale in the Union B process. The effect of key operating temperatures on the quantity of lost oil yield due to thermal cracking in the hot recycle gas is predicted.

### **Introduction**

Process simulators are used to develop models of chemical processes that consist of unit operations connected by streams. By ensuring mass and heat balances for each unit operation as well as the overall process, the simulators can calculate unknown stream flow rates, temperatures, pressures and compositions.

Most simulators contain built-in models for familiar unit operations and a databank or library of component parameters and methods to calculate thermodynamic properties. Simulators can be used to design new plants or to analyze and improve the operation of existing plants.

Aspen Plus V7.1 (AP) from AspenTech is a steady-state process simulation software package that includes the breadth of features needed to simulate oil shale retorting processes. AP is able to calculate phase equilibria between gases and liquids for streams that may also contain solids. The built-in libraries include unit operations, components and property methods for both hydrocarbon and mineral processing. The program allows the user to create oil shale-specific components such as kerogen and char and specify parameters used to calculate their properties. AP also allows the particle size distribution of the solids to be

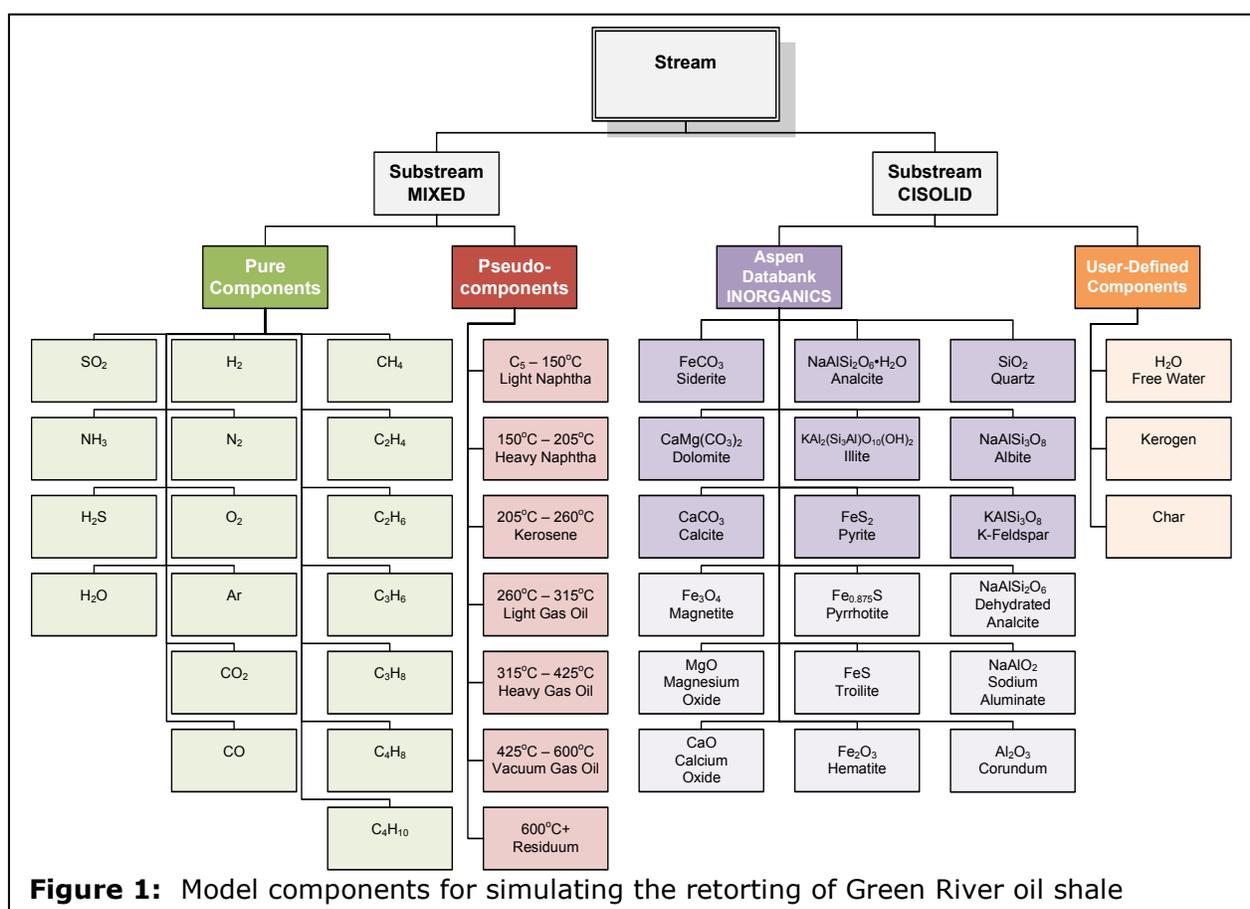
tracked and manipulated within the process.

### **Components**

Simulation of oil shale processes requires several different types of model components. Figure 1 shows the components used in a model of a Green River oil shale retorting process. Streams in AP are split into at least two substreams. One substream (MIXED) includes all liquid and gas components. Only components in the MIXED substream participate in phase equilibrium calculations. At least one other substream includes the solid components. Components in solid substreams do not participate in phase equilibrium calculations.

Components in the MIXED substream can be further divided into pure components that have a single molecular structure and pseudo-components that represent a group of compounds that make up an oil fraction with a range of boiling temperature. Property parameters for pure components are available from the AP databanks. Property parameters for pseudo-components are generated by AP using user-supplied boiling temperature curve and specific gravity of an oil or blend of oils.

AP has two types of solids substreams: NC and CISOLID. Components in the NC sol-



**Figure 1:** Model components for simulating the retorting of Green River oil shale

ids substream are not required to have a constant molecular weight throughout the process because all properties and conversions are on a mass basis. The elemental composition of an organic NC component such as kerogen may be tracked in the component attribute ULTANAL. Despite this capability, our current paper uses only the CISOLID solids substream which requires that all solid components have a constant elemental composition and molecular weight throughout the process. In AP V7.1, CISOLID components can participate in reactions in kinetic reactors (RCSTR, RPLUG) but NC components are treated as inert.

Components in the CISOLID substream are further divided into components whose property parameters are available in the AP databank and those that are defined by the user. User-defined components include kerogen, char and free water. Kerogen is the organic matter in raw oil shale. The

composition and properties of kerogen vary somewhat from deposit to deposit, but can also vary within a deposit. Therefore the properties of kerogen will be different for different oil shales. Char is the organic matter that remains in the spent shale after the kerogen is pyrolyzed. Char also has no fixed composition but depends on the original kerogen and the pyrolysis conditions.

Free water is the water held in the pore space within an oil shale particle. The water has the same thermodynamic properties as liquid water. A user-defined solid component is used to track the liquid water that may be trapped inside the particle even at elevated temperature.

### **Properties of User-Defined Components**

Components available in the AP databank do not need any additional information from the user to estimate component prop-

erties. All properties of pseudocomponents are also generated by the program. The user can override the estimates if better data are available.

All user-defined CISOLID solids components require the user to specify for each component at least the molecular weight, solids heat capacity, and solids density. If the component participates in chemical reactions then the standard heat of formation of the component must also be specified.

For organic components kerogen and char, the correlation (Boie, 1952) for coal given by Equation 1 can be used to estimate the gross heat of combustion at standard conditions (298.15 K and 1 atm) from the elemental composition.

$$\Delta_c H_{298}^o [\text{kJ/kg}] = 351.7w_C + 1162w_H + 104.7w_N - 111.0dw_O + 27w_S - 439.6 \quad (1)$$

The elemental composition is then used to derive the standard heat of formation from the heat of combustion using Equation 2.

$$\Delta_f H_{298}^o [\text{kJ/kg}] = \Delta_c H_{298}^o [\text{kJ/kg}] - (141.8w_H + 32.78w_C + 9.26w_S - 2.42w_N) \quad (2)$$

Average molecular formulas (Singleton *et al.*, 1986) are used to calculate molecular weight, elemental mass percentages, heat of combustion and heat of formation of kerogen and char for Green River oil shale. Results are given in Table 1.

The standard heat of formation of free water is the same as liquid water.

Polynomial equations (Camp, 1987) give the heat capacities of kerogen and char of Green River oil shale over a range of temperature. Camp's polynomials match the AP solids heat capacity model CPSDIP given by Equation (3). Note that AP requires a molar heat capacity for CISOLID components while Camp's correlations are on a mass basis. AP also includes extrapolation rules in case the temperature exceeds the limits. Converted coefficients and temperature limits supplied to AP are given in Table 2. Coefficients for the heat capacity of free water are also given in Table 2.

$$C_p [\text{kJ/kmolK}] = c_1 + c_2 T + c_3 T^2 + c_4 T^3 + c_5 T^4 \quad (3)$$

where  $T_{\min} \leq T[\text{K}] \leq T_{\max}$

### Properties of Pseudo-components

Properties of assay shale oil from Green River oil shale (Miknis, 1988) including true boiling point temperature (TBP), specific gravity and elemental composition are plotted in Figure 2.

AP uses the assay curves to derive property parameters for pseudocomponents. The number and cut point temperatures of the pseudocomponents are specified by the user. We chose to specify cut point temperatures for seven pseudocomponents. The molecular weight and elemental com-

**Table 1:** Standard heats of formation of user-defined organic components for Green River oil shale

	Kerogen	Char
Formula from Singleton <i>et al.</i> 1986	CH <sub>1.5</sub> N <sub>0.025</sub> O <sub>0.05</sub> S <sub>0.005</sub>	CH <sub>0.42</sub> N <sub>0.056</sub> O <sub>0.02</sub> S <sub>0.008</sub>
Molecular weight, kg/kmol	14.833	13.795
Elemental composition, wt%		
Carbon	80.972	87.066
Hydrogen	10.193	3.069
Nitrogen	2.361	5.686
Oxygen	5.393	2.320
Sulfur	1.081	1.860
Gross heat of combustion, kJ/kg	39549	34042
Standard heat of formation, kJ/kg	-1489.7	1115.9
Standard heat of formation, kJ/kmol	-22097	15394

**Table 2:** Heat capacity polynomial coefficients for user-defined solids components for Green River oil shale

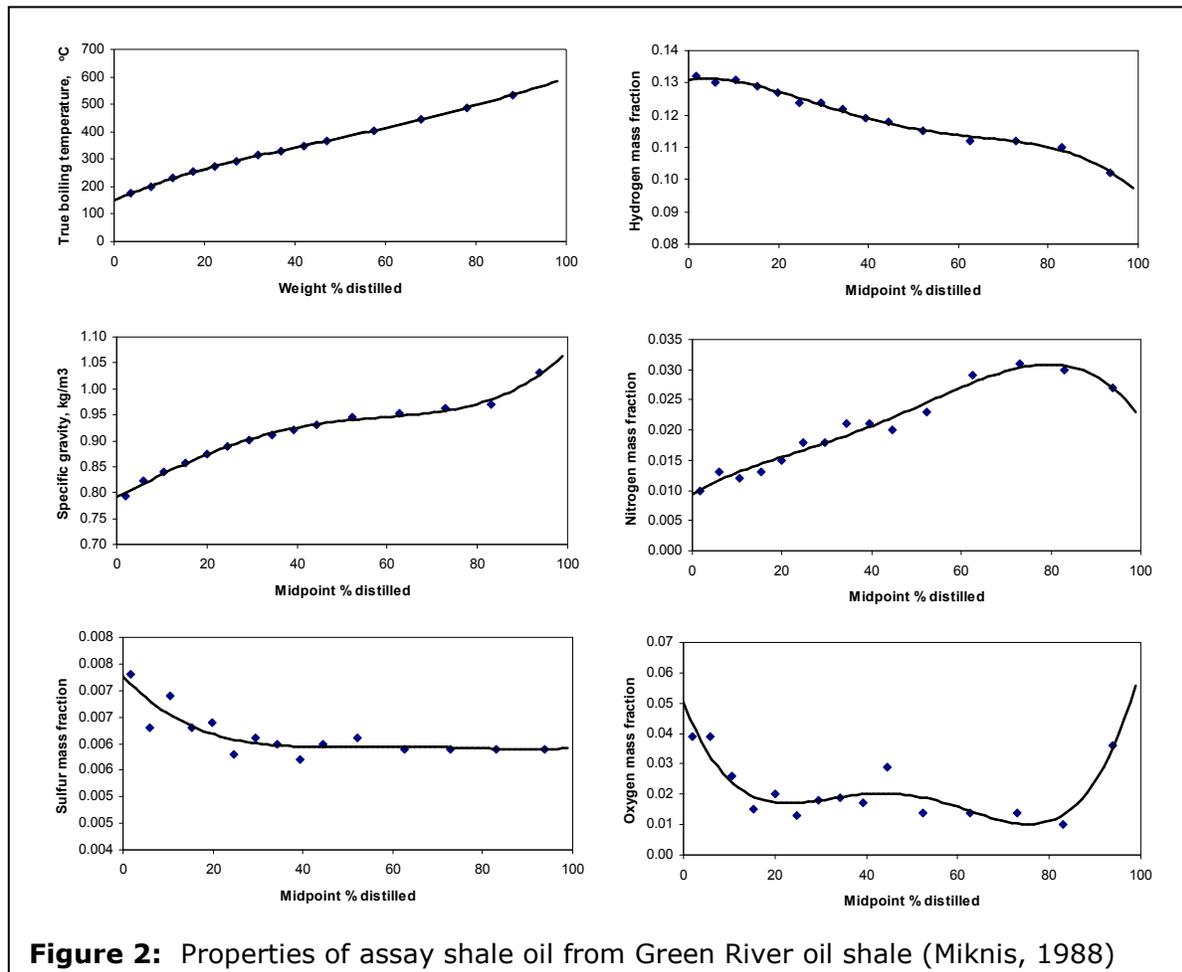
	Kerogen	Char	Free Water
MW, kg/kmol	14.833	13.795	18.015
C <sub>1</sub>	$3.311 \cdot 10^0$	$-1.626 \cdot 10^0$	$5.084 \cdot 10^1$
C <sub>2</sub>	$7.793 \cdot 10^{-2}$	$5.943 \cdot 10^{-2}$	$2.131 \cdot 10^{-1}$
C <sub>3</sub>	$-2.453 \cdot 10^{-5}$	$-2.464 \cdot 10^{-5}$	$-6.314 \cdot 10^{-4}$
C <sub>4</sub>	0	0	$6.487 \cdot 10^{-7}$
C <sub>5</sub>	0	0	0
T <sub>min</sub> , K	273	273	273
T <sub>max</sub> , K	750	1000	623

position of each pseudocomponent generated by AP is given in Table 3.

### Composition of Green River Oil Shale

A mineral composition of Green River oil shale (Brons and Siskin, 1989) is given in Table 4. All mineral components listed are

available from the AP V7.1 INORGANIC databank with a couple of exceptions. Databank SOLIDS component muscovite is use to represent illite. Dawsonite is not available in either the INORGANIC or SOLIDS databank. A user-defined component could have been created for dawsonite, but due to its low significance we



**Figure 2:** Properties of assay shale oil from Green River oil shale (Miknis, 1988)

**Table 3:** Generated properties of pseudo-components for Green River shale oil

Component	Formula	Mol. Weight g/gmol	Lower Boiling Temperature °C	Upper Boiling Temperature °C
Light naphtha	C <sub>8.5</sub> H <sub>16.4</sub> N <sub>0.10</sub> O <sub>0.45</sub> S <sub>0.03</sub>	128.3	C <sub>5</sub>	150
Heavy naphtha	C <sub>10.1</sub> H <sub>19.0</sub> N <sub>0.18</sub> O <sub>0.53</sub> S <sub>0.03</sub>	152.0	150	205
Kerosene	C <sub>13.1</sub> H <sub>23.4</sub> N <sub>0.22</sub> O <sub>0.26</sub> S <sub>0.04</sub>	188.9	205	260
Light gas oil	C <sub>15.9</sub> H <sub>27.6</sub> N <sub>0.37</sub> O <sub>0.34</sub> S <sub>0.04</sub>	231.4	260	315
Heavy gas oil	C <sub>21.2</sub> H <sub>34.4</sub> N <sub>0.59</sub> O <sub>0.53</sub> S <sub>0.06</sub>	308.5	315	425
Vacuum gas oil	C <sub>33.6</sub> H <sub>51.6</sub> N <sub>1.3</sub> O <sub>0.48</sub> S <sub>0.09</sub>	483.9	425	600
Residuum	C <sub>40.7</sub> H <sub>56.8</sub> N <sub>1.4</sub> O <sub>2.5</sub> S <sub>0.11</sub>	609.5	600	700

**Table 4:** Composition of Green River oil shale (Brons *et al.*, 1989)

Component	Formula	Mol. Weight g/gmol	wt% dry basis
Siderite	FeCO <sub>3</sub>	115.9	2.4
Dolomite	CaMg(CO <sub>3</sub> ) <sub>2</sub>	184.4	22.8
Calcite	CaCO <sub>3</sub>	100.1	14.1
Illite	K(Al <sub>2</sub> )(Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>2</sub>	398.3	10.9
Analcime	NaAlSi <sub>2</sub> O <sub>6</sub> ·H <sub>2</sub> O	220.2	0.9
Dawsonite	NaAlCO <sub>3</sub> (OH) <sub>2</sub>	144.0	0.6
Pyrite	FeS <sub>2</sub>	120.0	1.6
Quartz	SiO <sub>2</sub>	60.1	13.2
Albite	NaAlSi <sub>3</sub> O <sub>8</sub>	262.2	13.7
Kerogen			19.8
Total			100.0

choose to ignore dawsonite in the current model.

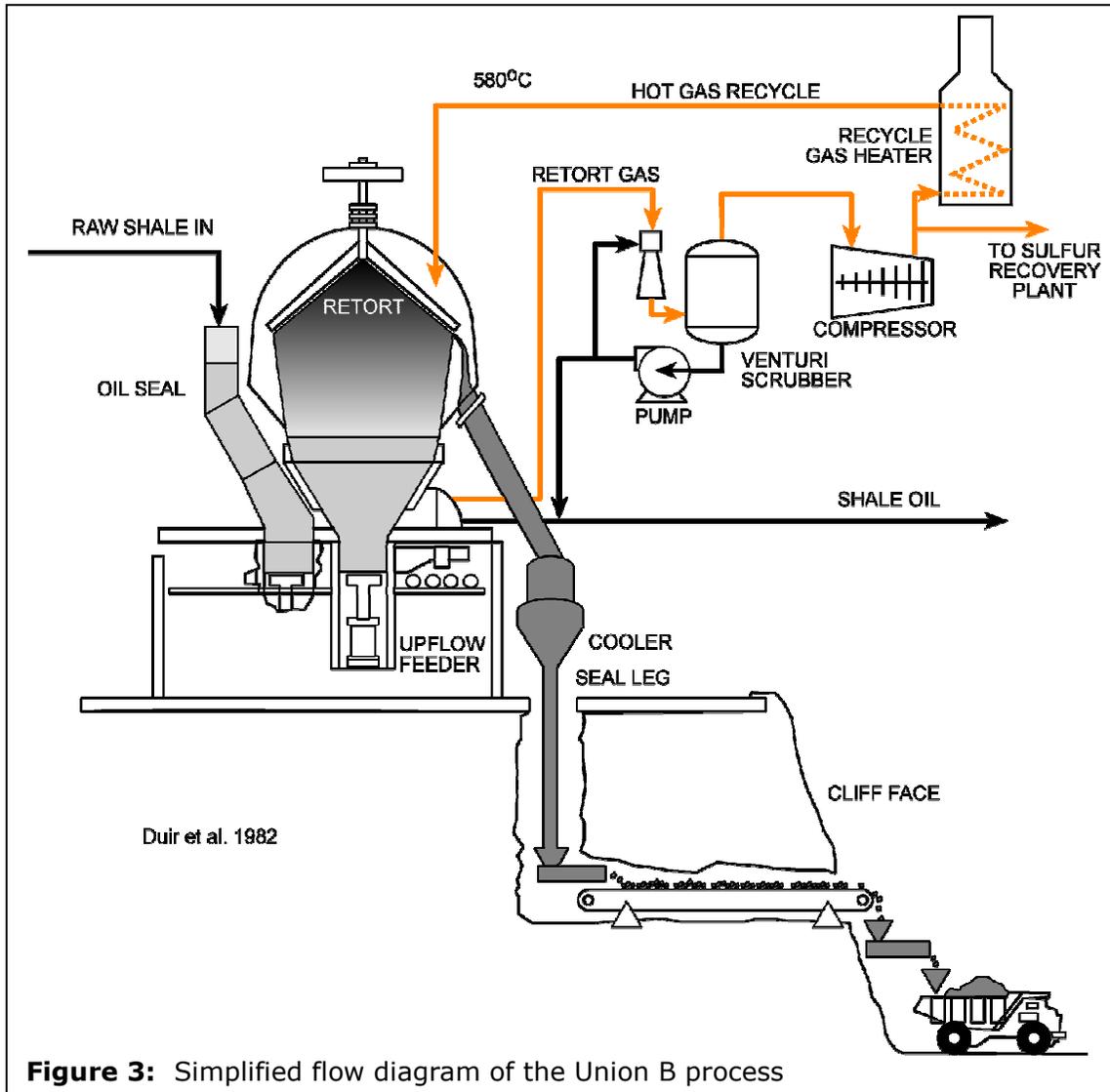
### Model Example

To illustrate the use of a process simulator to model oil shale retorting, a model of the Union B process is given. The Union B retort, developed by Unocal, was operated near Parachute Colorado from 1986 to 1991 producing about 4.5 million barrels of shale oil.

Figure 3 gives a simplified flow diagram of the Union B process. Crushed and screened oil shale is fed to the bottom of the retort through an oil seal. An upflow feeder pushes the shale up through the conical retort vessel where it is heated to about 490°C (914°F) to convert the kerogen in the oil shale to gas, oil vapor and char. The spent shale spills over the top of the retort to a cooler and discharge seal.

The oil shale is heated by hot recycle gas that enters at the top of the retort and flows counter-current to the shale. Gas and oil vapor evolved from the shale mixes with the hot gas. As the gas is cooled the oil condenses. Oil and gas are separated at the bottom of the retort. An oil level is maintained at the bottom of the retort to form the liquid seal. Gas is removed and cooled in a venturi scrubber where more oil is collected. The scrubber gas passes through a compressor before it is split into product and recycle streams. The recycle gas stream passes through a heater before returning to the top of the retort at over 500°C (932°F).

Most of the publicly available information on the Union B process was written before operation began in 1986 (Dhondt *et al.*, 1981; Barnet, 1982; Duir *et al.* 1983). Progress reported (Reeg *et al.* 1990) after a few years of operation but before the



**Figure 3:** Simplified flow diagram of the Union B process

plant's final shutdown in 1991, gives little information on the actual product quality or operating conditions except that the hot recycle gas temperature was increased from the design temperature of 540°C (1004°F) to 580°C (1076°F) by 1990.

A mathematical model (Braun and Lewis, 1985) was developed for the Union B retort. The differences between the Braun and Lewis model and our AP model are listed in Table 5. Both models include thermal cracking of light oil in the hot recycle gas stream between the heater and the top of the retort.

The AP simulation flowsheet is given in Figure 4. Free water, kerogen and mineral

are mixed in a mixer block FEEDMIXR to make the oil shale feed to the retort. The oil shale exchanges heat with the retort

**Table 5:** Differences between the Union B model (Braun and Lewis, 1985) and this work

Braun and Lewis (1985)	This work
<ul style="list-style-type: none"> <li>Few mineral components.</li> <li>Dynamic retort</li> <li>Detailed retort model with reaction kinetics, heat transfer and pressure drop</li> </ul>	<ul style="list-style-type: none"> <li>More mineral components and reactions.</li> <li>Steady-state</li> <li>Several unit operation blocks to represent retort</li> </ul>



**Table 6:** Mineral Reactions for pyrolysis of Green River oil shale

Reactant	Reaction Equation	Peak °C
Analcite	$\text{NaAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O} \rightarrow \text{NaAlSi}_2\text{O}_6 + \text{H}_2\text{O}$	150-400
Dawsonite	$\text{NaAlCO}_3(\text{OH})_2 \rightarrow \text{NaAlO}_2 + \text{CO}_2 + \text{H}_2\text{O}$	300, 440
Pyrite	$0.875\text{FeS}_2 + 0.75\text{H}_2 \rightarrow \text{Fe}_{0.875}\text{S} + 0.75\text{H}_2\text{S}$	450-550
Siderite	$3\text{FeCO}_3 \rightarrow \text{Fe}_3\text{O}_4 + \text{CO} + 2\text{CO}_2$	500-600
Magnetite	$\text{Fe}_3\text{O}_4 + \text{H}_2\text{S} \rightarrow \text{FeS} + \text{Fe}_2\text{O}_3 + \text{H}_2\text{O}$	
Illite	$\text{K}(\text{Al}_2)(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2 \rightarrow \text{KAlSi}_3\text{O}_8 + \text{Al}_2\text{O}_3 + \text{H}_2\text{O}$	550, 900
Dolomite	$\text{CaMg}(\text{CO}_3)_2 \rightarrow \text{CaCO}_3 + \text{MgO} + \text{CO}_2$	790
Calcite	$\text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2$	860-1010

INORGANIC databank and are shown in Figure 1.

The stoichiometry for the pyrolysis of kerogen in Green River oil shale to gas, oil and char given in Table 7, which is based on the simplified pyrolysis stoichiometry given by Singleton *et al.* (1986). The distribution of the whole oil to pseudocomponents is based on the true boiling point curve from Miknis (1988).

Stoichiometry from laboratory assays is assumed to be acceptable for the kerogen pyrolysis in the Union B retort because the oil shale heating rate and maximum shale temperature are similar.

The stoichiometry and kinetics of oil cracking in the reactor block CRACKING is based on Model D of Bissell *et al.* (1985). The stoichiometry for thermal cracking of the three lightest oil pseudocomponents is given in Table 8. Distribution among C<sub>2</sub>, C<sub>3</sub> and C<sub>4</sub> in the gas is based on Burnham (1980) and distribution between alkenes and alkanes is based on Voge and Good (1949). While stoichiometry and kinetics are also available for the heavier oil pseudocomponents, it is not given here because these components are for all intents and purposes removed from the recycle gas in the scrubber.

Additional components could have been created for the cokes produced from the thermal cracking of the oil,

which will have a different composition from that of the char; however, we chose to use the existing component char for coke.

Model D of Bissell *et al.* assumes a first-order reaction rate for the cracking of each oil fraction. While the activation energy E<sub>1</sub>

**Table 7:** Stoichiometry of kerogen pyrolysis for Green River oil shale (Singleton *et al.*, 1986)

Component	Molecular weight g/gmol	wt% of kerogen	moles per mole kerogen
Kerogen	14.83	-	-1.00000
		100.000	
Methane	16.04	1.399	0.01293
Hydrogen	2.02	0.297	0.02189
Carbon monoxide	28.01	0.564	0.00298
Carbon dioxide	44.01	3.542	0.01194
Hydrogen sulfide	34.08	0.229	0.00099
Water	18.01	1.208	0.00995
Ethene	28.05	0.304	0.00161
Ethane	30.07	0.894	0.00441
Propene	42.08	0.582	0.00205
Propane	44.10	0.659	0.00222
Butene	56.11	0.519	0.00137
Butane	58.12	0.519	0.00132
Gas subtotal		10.714	
Light naphtha	128.3	0.985	0.00114
Heavy naphtha	152.0	5.018	0.00490
Kerosene	188.9	7.267	0.00571
Light gas oil	231.4	9.001	0.00577
Heavy gas oil	308.5	20.626	0.00992
Vacuum gas oil	483.9	22.814	0.00699
Residuum	609.5	3.036	0.00074
Oil subtotal		68.746	
Char	13.80	20.539	0.22085

**Table 8:** Stoichiometry for thermal cracking of light naphtha, heavy naphtha and kerosene

	MW g/gmol	Light naphtha		Heavy naphtha		Kerosene	
		wt% of light naphtha	moles per mole light naphtha	wt% of heavy naphtha	moles per mole heavy naphtha	wt% of kerosene	moles per mole kerosene
KERO	188.9					-100.0	-1.00000
HNAP	152.0			-100.0	-1.00000	11.6	0.14383
LNAP	128.3	-100.0	-1.00000	23.1	0.27411	11.6	0.17038
CH <sub>4</sub>	16.04	5.8	0.46521	4.5	0.42352	4.5	0.52652
H <sub>2</sub>	2.02	1.9	1.20483	1.5	1.09686	1.5	1.36360
CO	28.01	3.4	0.15356	2.6	0.13980	2.6	0.17379
C <sub>2</sub> H <sub>4</sub>	28.05	11.2	0.51132	8.6	0.46550	8.6	0.57871
C <sub>2</sub> H <sub>6</sub>	30.07	8.1	0.34454	6.2	0.31366	6.2	0.38994
C <sub>3</sub> H <sub>6</sub>	42.08	10.7	0.32540	8.2	0.29624	8.2	0.36828
C <sub>3</sub> H <sub>8</sub>	44.10	4.7	0.13767	3.6	0.12533	3.6	0.15581
C <sub>4</sub> H <sub>8</sub>	56.11	14.1	0.32353	10.9	0.29454	10.9	0.36616
C <sub>4</sub> H <sub>10</sub>	58.12	2.2	0.04897	1.7	0.04459	1.7	0.05543
Coke	13.80	37.9	3.52600	29.1	3.21003	29.1	3.99066

is the same for all fractions, the pre-exponential frequency factor  $f_1$  increases with the average molecular weight of the oil fraction. The cracking kinetic model is given by Equations 4 and 5.

$$\frac{dy_i}{dt} = A_i k y_i \quad (4)$$

$$k = f_1 e^{-E_1/T} \quad (5)$$

The parameters that fit the cracking model to experimental data are  $f_1 = 1.04 \cdot 10^8 \text{ s}^{-1}$  and  $E_1 = 19588 \text{ K}$ . The relative rate factor  $A_i$  for each pseudocomponent was found by interpolating between molecular weights and relative rate factors given by Bissell *et al.* Results are given in Table 9. For first-order reactions with concentration on a mass fraction basis, AP requires the units

for the pre-exponential frequency factor to be  $\text{kmol/s}\cdot\text{m}^3$ . The converted pre-exponential factors for AP are also given in Table 9.

The temperature of the CISOLID streams leaving separators blocks DEHYDSEP and PYROSEP are set to 350°C (662°F) and 490°C (914°F) respectively. An AP Design Spec is used to vary the MIXED stream temperature leaving the separator block DEHYDSEP to reduce the heat duty of the block to zero. The MIXED stream leaving the separator block PYROSEP is set to 350°C (662°F). A second Design Spec varies the flow of recycle gas from splitter block GASSPLIT to heater block GASHTR to reduce the heat duty of the separator block PYROSEP to zero.

**Table 9:** Thermal cracking of oil fractions

Oil fraction	Average molecular weight g/gmol	Relative cracking rate factor $A_i$	Pre-exponential factor $\text{kmol/s}\cdot\text{m}^3$
Light naphtha	128.28	2.9	$9.03 \cdot 10^5$
Heavy naphtha	151.97	4.5	$1.17 \cdot 10^6$
Kerosene	188.92	7.6	$1.59 \cdot 10^6$

## Results

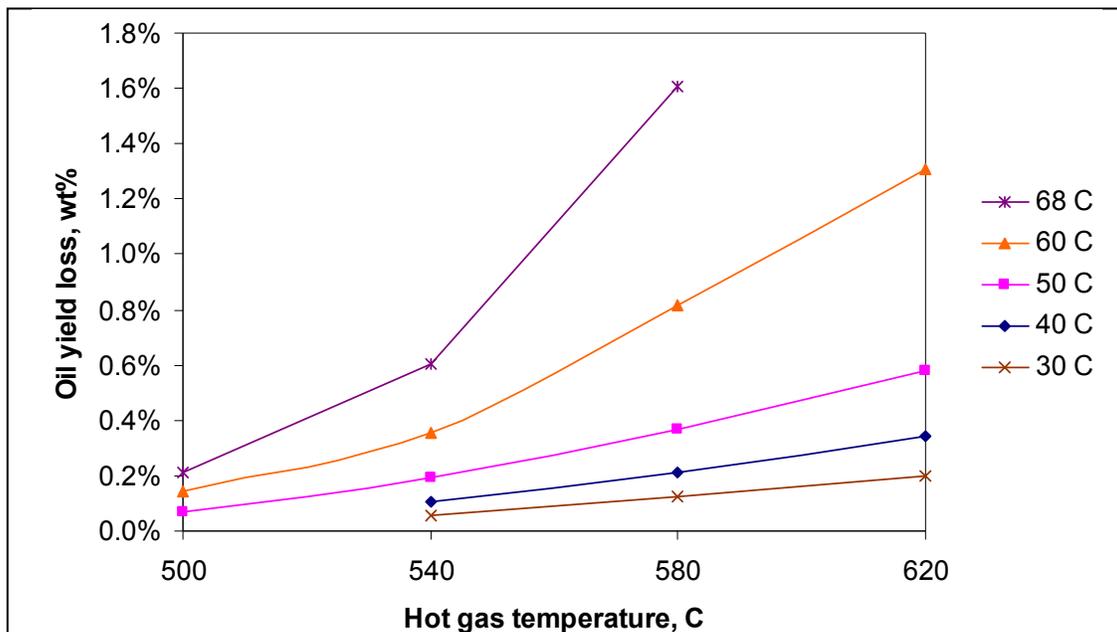
The AP Union B process model was solved for the same base case considered by Braun and Lewis (1985) with a 500°C (932°F) hot gas recycle temperature and 68°C (154°F) scrubber temperature. The volume of the thermal cracking reactor was specified to give 10 seconds residence time for the base case. Like Braun and Lewis, we found that cracking of 3% of the oil in the recycle stream resulted in about 0.2% reduction in overall oil production.

Next the model was run a number of times while changing key temperatures. The volume of the thermal cracking reactor was kept the same for all cases. Figure 5 shows the predicted effect of the hot gas temperature and the scrubber temperature on the oil yield loss due to thermal cracking in the hot recycle gas.

As the hot gas temperature was increased, the hot gas recycle flow rate was able to be decreased. However the overall effect was that oil loss due to thermal cracking increased as the temperature of the hot recycle gas was increased.

The temperature of the scrubber has a significant effect on the composition of the recycle gas. As the scrubber temperature is increased, the concentration of water in the recycle gas increases. We thought the increase in water concentration may decrease the oil concentration in the hot gas stream and reduce thermal cracking. However the model predicts that the concentration of oil in the recycle gas also increases with scrubber temperature and the predicted net result is that oil yield loss due to thermal cracking increases as the scrubber temperature is increased.

As pointed out by Braun and Lewis, minimizing oil yield loss by decreasing the hot recycle gas temperature and the scrubber temperature requires additional compressor power, fuel for the heater and cooling water for the condensers. Figure 6 shows the effect of the hot gas temperature and scrubber temperature on the heating and cooling duty for the 7950 m<sup>3</sup> oil/d (50,000 bbl/d) plant considered by Braun and Lewis. At the lower hot recycle gas temperature, the gas flow rate is higher and more cooling at the scrubber requires more heating by the heater.



**Figure 5:** Predicted effect of hot recycle gas temperature and scrubber temperature on lost oil yield due to thermal cracking of oil in hot recycle gas

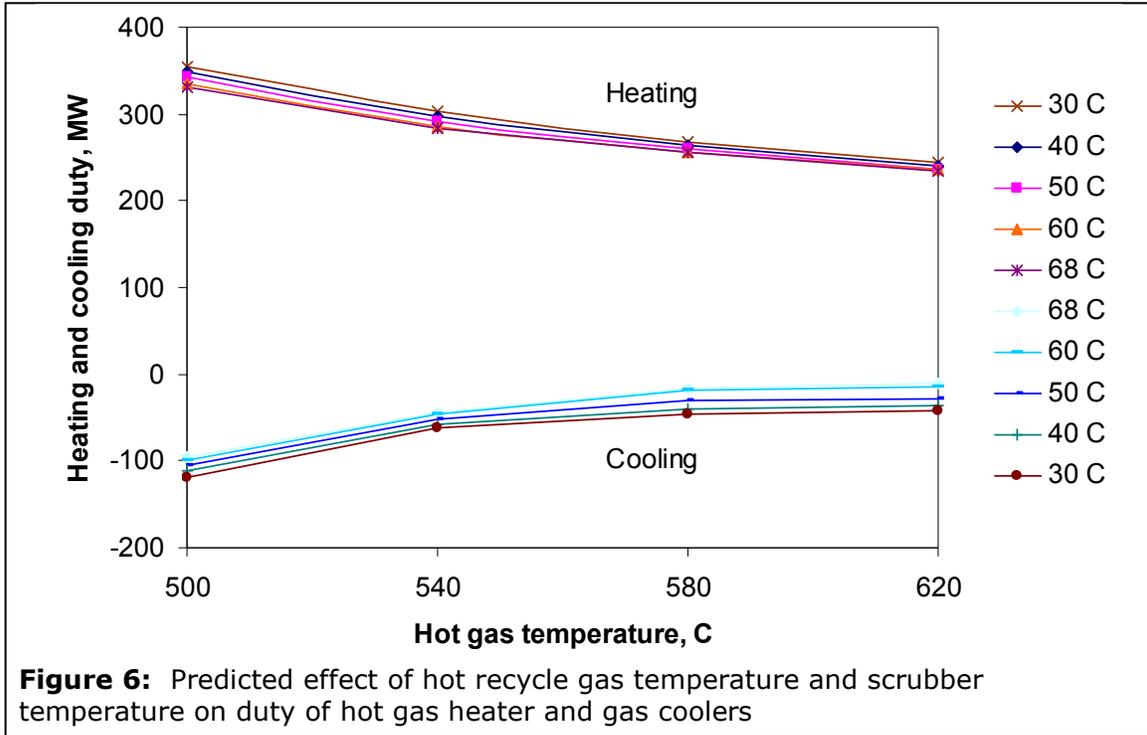
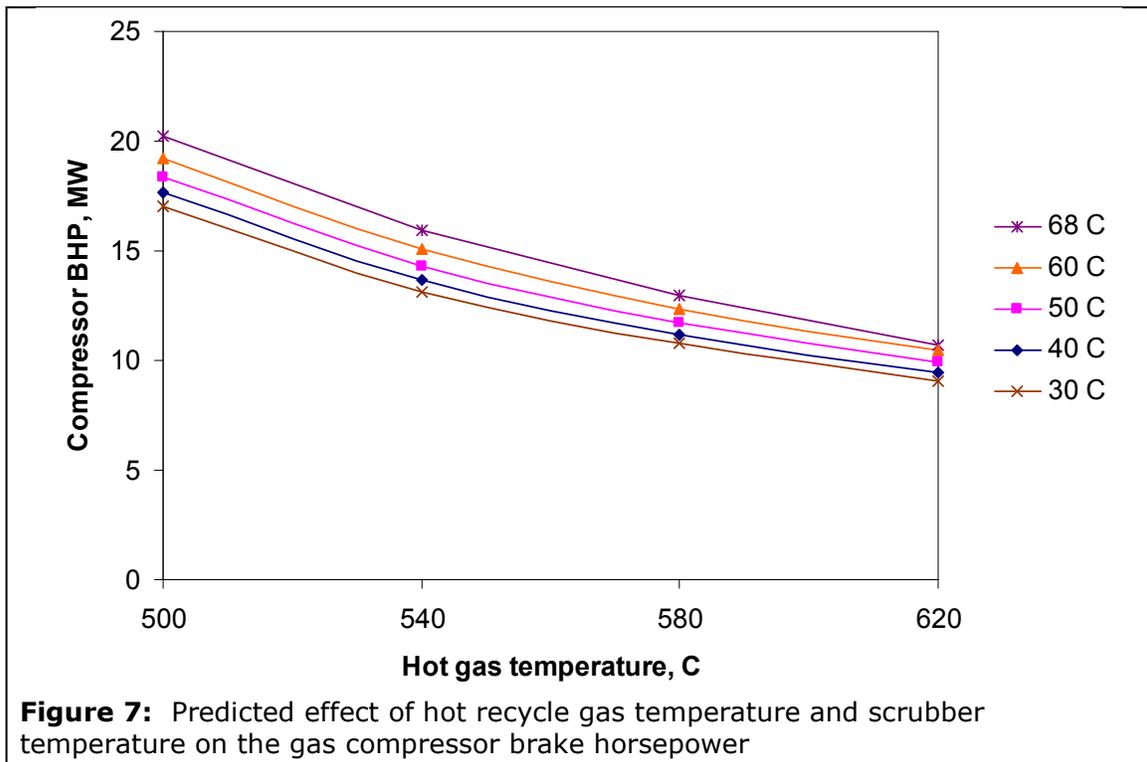


Figure 7 shows the effect of the hot recycle gas temperature and scrubber temperature on the compressor power. The compressor power decreases as the temperature of the

hot recycle gas increases due to the reduction in the gas flow rate. Lower scrubber temperature reduces the compressor power.



## Conclusions

We have shown that a general purpose process simulator is a useful tool for the development of an oil shale conversion process. Robust models can be developed relatively quickly. Most components and their properties are available in the simulator databank. No grouping of components is required. User-defined components can be created for oil shale specific components that are not in the built-in databank. The required properties of user-defined components, reaction stoichiometry and reaction kinetics are available for Green River oil shale in the public literature. Built-in unit operations are sufficient for simple steady-state models.

## Nomenclature

$A_i$	relative cracking rate factor, -
$c_i$	$i^{\text{th}}$ polynomial coefficient in Equation 3, kJ/kmol-K <sup>i</sup>
$C_p$	component heat capacity, kJ/kmol-K
$E_1$	activation energy, K
$f_1$	frequency factor, s <sup>-1</sup>
$\Delta_c H_{298}$	gross heat of combustion at 298.15 K and 1 atm, kJ/kg
$\Delta_f H_{298}$	standard heat of formation at 298.15 K and 1 atm, kJ/kg
$k$	rate constant, s <sup>-1</sup>
$t$	time, s
$T$	stream temperature, K
$T_{\min}, T_{\max}$	minimum and maximum temperatures of Eqn. 3 applicability, K
$w_i$	mass percent of element $i$ , %
$y_i$	weight fraction of component $i$

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PROCOM Consultants P/L is a small but dedicated Australian company that provides specialist engineering services to a range of process industries. The directors, employees and associates of PROCOM have significant knowledge and experience in all aspects of executing an oil shale development project.

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