Rapid Hydropyrolysis of Softening Coal Particles—A Modeling Study

Part II: Model Predictions and Comparison with Experimental Results

The mathematical model described in Part I is applied to predict CH_4 and tar yields in rapid hydropyrolysis of softening coal particles. Predicted effects of pressure and particle size agree with trends previously measured in a screen heater apparatus.

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SCOPE

Previous experimental results with Pittsburgh Seam bituminous coal indicate that mass transfer plays an important role in rapid hydropyrolysis. The measured effects of particle size and pressure on CH_4 and tar yields could not be adequately described with available kinetic models owing to their not accounting for the drastic changes in physical properties associated with softening and resolidification during hydropyrolysis of softening coals. A new hydropyrolysis model which does account for the property changes of softening coals has been formulated and presented in Part I. In the present paper the model is tested by comparing its predictions with experimental results in order to verify its underlying mechanistic concept.

CONCLUSIONS AND SIGNIFICANCE

Mathematical representation of the global chemical reactions and mass transfer leads to predictions for particle size and pressure effects on CH_4 and tar yields. The agreement of predictions with measurements, e.g., of the different effects of total pressure and H_2 partial pressure on the yields, is better than previous models for softening coal (Anthony et al., 1976). This success can be attributed to the underlying mechanistic concept of diffusional transport of H_2 in a material whose transient physical properties range from those of a fluid during the plastic period to those of a porous solid after resolidification. Accordingly, the coal's transient fluidity is significant, as penetration of H_2 into the particle is hindered during the plastic period due to the low H_2 diffusivities in the plastic-liquid state. Diffusion is comparatively faster in the resolidified coke after pyrolysis gas evolution is nearly completed.

The present experimental data base is sparse. Areas of particular interest for further experimental investigation and model validation are the rheological properties of softened coal under rapid hydropyrolysis conditions and the separate effects of inert gas pressure, H_2 pressure, and particle size on individual product yields (e.g., CH_4 and tar).

The results of this study are pertinent to coal (hydro-) pyrolysis and gasification modeling and to addressing such questions as how to maximize individual product yields or how to minimize agglomeration of softening coal in conversion processes.

INTRODUCTION

A mathematical model for rapid hydropyrolysis of softening coal particles is presented in Part I. The model includes a representation of H_2 and tar transport, and is based on results of previous experimental studies in a screen heater apparatus. A simple chemical reaction scheme for competitive hydrogenation and crosslinking of the reactive hydrogenatable species is combined with a description of H_2 transport in terms of appearance and disappearance of plasticizing material. H_2-consuming reactions are treated as

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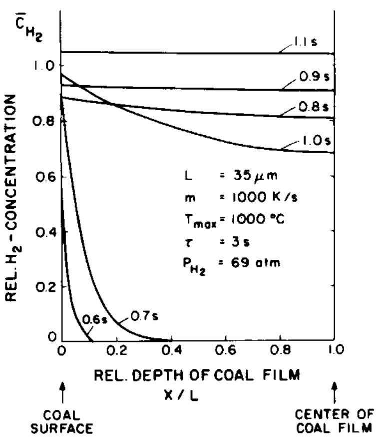

Figure 1. Profiles of H_2 concentration (relative to Henry's law equilibrium concentration at maximum temperature) in the coal film during the plastic period ($t \leq 1.0$ s) and after resolidification ($t = 1.1$ s).
Coal film thickness, $2L = 70\ \mu m$
Nominal heating, cooling rate, $m = 1{,}000$ K/s
Maximum temperature, $T_{max} = 1{,}000°C$
H_2 pressure, $P_{H_2} = 69$ atm (6.99 MPa)

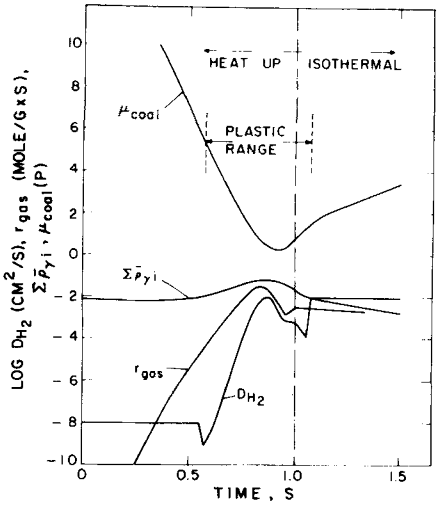

Figure 2. Model predictions of H_2 diffusivity D_{H_2}, pyrolysis gas formation rate r_{gas}, relative metaplast mass concentration $\Sigma\bar{\rho}_{\gamma i}$, and coal mass viscosity μ_c, during first 1.5 s of rapid hydropyrolysis. (Reaction conditions as in Fig. 1.)

gas-liquid reactions during the plastic period and as gas-solid reactions after resolidification of the coal particle. The model represents the limiting case of small particles which in the screen heater apparatus form a thin layer or film during the coal's transient plastic behaviour. The resulting coal film is assumed to be spatially isothermal, and transport of plasticizing material yielding tar is considered to be governed solely by an extraparticle resistance, according to an evaporation-diffusion transport mechanism.

By use of correlations and parameter values from the literature the model is able to predict effects of particle size and pressure on the yields of CH_4 and tar which, of all the hydropyrolysis products, exhibit the highest sensitivity to pressure and particle size (Suuberg et al., 1980). Furthermore, physical properties such as the transient viscosity and H_2 diffusivity during hydropyrolysis can be predicted.

This paper persents model predictions and compares them with experimental data on product yields and weight loss from hydropyrolysis of highly softening Pittsburgh Seam bituminous coal (Anthony, 1974; Anthony et al, 1976; Suuberg, 1977; Suuberg et al., 1980).

BASE CASE

Results reported in this section are for a typical set of conditions in a screen heater apparatus (Anthony et al., 1976; Suuberg et al., 1980): coal film thickness, 70 μm; H_2 pressure, 69 atm (6.99 MPa); holding time, 3 s; nominal heating and cooling rate, 1,000 K/s. Figure 1 shows H_2 concentration profiles inside the coal film during the plastic period (times indicated ≤1.0 s) and after resolidification (1.1 s), as given by integration of the H_2 continuity equation. In Figure 2, some of the predicted time-dependent quantities are plotted vs. time during the first 1.5 s of hydropyrolysis. H_2 concentrations are divided by Henry's law equilibrium concentration at maximum temperature, the depth of the coal film x by the half-thickness of the film L, and the metaplast mass concentration by the initial coal density.

According to the definition used, coal softening is predicted to occur when the relative metaplast concentration $\Sigma\bar{\rho}_{\gamma i}$ reaches the defined value of 10^{-2} g/cm^3 for the critical metaplast concentration, i.e., at 0.575 s when the temperature is 575°C. The computation shows that the plastic period lasts until 1.075 s, i.e., shortly after the final temperature of 1,000°C is reached, when metaplast concentration decreases below the critical value. Coal mass viscosity, correlated with metaplast concentration and temperature, exhibits a minimum during the plastic range, with a value of about 10 P(1 Pa·s)—Figure 2. Measurements of Pittsburgh Seam bituminous coal fluidity in a Giesler plastometer (Lloyd et al., 1980) and extrapolation of the heating rate effect on fluidity (van Krevelen et al., 1956) indicate that the predicted low minimum coal viscosity may well be realistic for the hydropyrolysis conditions considered in this calculation. Figure 2 also reflects the definition used for the onset of softening and resolidification. The definition, based on a critical metaplast concentration, leads to different predicted values for coal viscosity at the onset and termination of plasticity, due to the incorporated effect of temperature on coal viscosity.

The predicted variation of coal-mass viscosity, μ_c, and pyrolysis gas formation rate, r_{gas}, leads to the transient behavior of H_2 diffusivity during the plastic period. H_2 diffusivity is approximated by a low constant value before softening of the coal occurs and a larger constant value during the isothermal period after resolidification, both representative of gas phase diffusion in porous solids with different pore structures. During the plastic range, however, H_2 diffusivity exhibits drastic variations resulting from changes in the coal's physical properties. When softening begins, the pores of the reacting coal close. This effect combined with the high viscosity of the plastic coal mass results in an initial reduction in the H_2 diffusivity. However, with decreasing coal viscosity and increasing pyrolysis gas formation, to which mixing of the fluid coal is attributed, H_2 diffusivity increases to values which should be representative of a dissolved gas diffusing in a stirred liquid. Toward the end of the plastic period, when most of the metaplast has either evaporated or decomposed, the coal mass becomes more viscous and therefore H_2 diffusivity again decreases. The slight minimum in pyrolysis gas formation rate at about 0.95 s, which is due to difference in rate parameters for the individual gas species formed, is reflected in the H_2 diffusivity curve.

Hydrogen concentration profiles in Figure 1 can be largely interpreted in terms of the transient diffusivity for H_2 inside the coal film. As initially no H_2 is present inside the coal, steep concentration profiles result from the slow penetration of H_2 during stages of low diffusivity, i.e., before softening and at the beginning of the plastic period (0.6 and 0.7 s). Essentially no H_2 is present in the coal at that time. When H_2 diffusivities reach their maximum values (between 0.8 and 0.9 s) due to low coal viscosities and mixing associated with

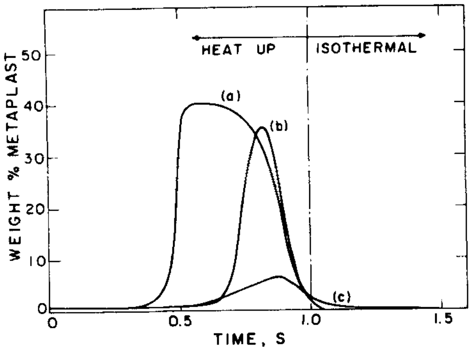

Figure 3. Effect of metaplast formation rate parameters k_{2_0} and E_{A2} on mass concentration of metaplast, predicted by the present model. (Reaction conditions as in Fig. 1.)

(a) $k_{2_0} = 10^{13}$ s^{-1}; E_{A2} = 40,000 cal/mol (Unger and Suuberg, 1981)

(b) $k_{2_0} = 1.67 \times 10^{11}$ s^{-1}, E_{A2} = 47,000 cal/mol (Chermin and van Krevelen, 1957)

(c) $k_{2_0} = 1.23 \times 10^{3}$ s^{-1}; E_{A2} = 14,900 cal/mol (Zacharias, 1979, generally used in this study)

high pyrolysis gas formation rates, concentration profiles are smoothed and the coal mass becomes more easily accessible to externally introduced H_2. The steeper concentration profile at 1.0 s reflects the decrease in H_2 diffusivity toward the end of the plastic period. After resolidification (at 1.1 s), the assumed high porosity of the coke leads to large values for effective pore diffusivities resulting in only minor intraparticle concentration gradients.

Time variations in the H_2 concentration at the coal surface ($x/L = 0$) reflect the temperature dependence of the Henry's law equilibrium concentration leading to increased H_2 concentrations with time during heatup. In addition, evolution and countercurrent flow of pyrolysis gas and CH_4 from hydrogenation affect the gas phase mole fraction of H_2 and thus the corresponding equilibrium concentration of H_2 in the liquid phase. The effect is strongest during the period of high pyrolysis gas formation rates (between 0.8 and 0.9 s). As a result, the gas phase mole fraction of H_2 is relatively low at 0.8 s, namely 0.940, as compared to 0.995 at 0.7 s. The combined effects of temperature and gas phase mole fraction of H_2 lead to approximately the same values for H_2 concentration at the coal surface at 0.7 s and 0.8 s. Thus, within this modeling concept, high pyrolysis gas formation rates not only increase the accessibility of the coal mass due to mixing effects in the plastic coal but also cause extraparticle H_2 concentration gradients, lowering the H_2 gas phase mole fraction at the coal's surface, although the latter effect is only of minor importance, in agreement with Sprouse's (1980) analysis for spherical coal particles.

The effect of metaplast formation rate parameters k_{2_0} and E_{A2} on mass concentration of metaplast is shown in Figure 3. Three different parameter sets lead to different magnitudes and appearance times for the concentration maximum although the overall tar yield is not affected considerably. Curve (*a*) results from parameters suggested by Unger and Suuberg (1981) for metaplast formation during rapid pyrolysis of bituminous coal; parameters for curve (*b*) are taken from a theoretical study of softening coal pyrolysis by Chermin and van Krevelen (1957). Curve (*c*) represents the base case of this modeling study, with parameter values from Zacharias' (1979) model of tar formation and evaporation during rapid pyrolysis of bituminous coal. According to the metaplast balance equations, more rapid formation leads to its earlier appearance, thus an earlier onset of plasticity, and to a higher accumulation of metaplast in the coal. Variations in rate parameters for metaplast decomposition affect the balance in an analogous way.

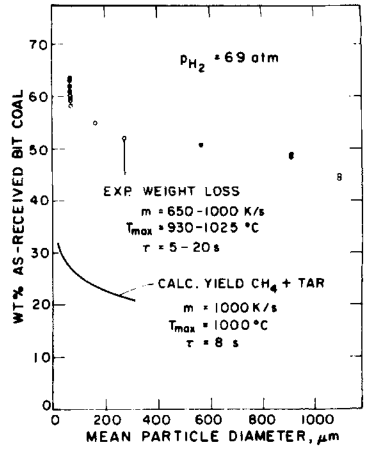

Figure 4. Effect of mean particle diameter on weight loss measured for hydropyrolysis of Pittsburgh Seam bituminous coal (○ Anthony et al., 1976; • Suuberg et al., 1980), and on yields of CH_4 plus tar predicted by the present model (*Calc. Yield* curve). (Nomenclature as in Fig. 1.)

In evaluating rate parameters for metaplast formation, decomposition, and transport, lack of experimental information about plastic behavior under rapid hydropyrolysis conditions emerges as a major shortcoming. Experiments which allow visual observation of the coal particles under the pressure and heating rates of interest here have been carried out so far only with a different, possibly less highly softening coal (Arendt, 1980). The plastic period, denoted period of tar evolution, was found in Arendt's experiments to last from 490–500°C to 900°C, for a heating rate of 950 K/s, He pressure of 90 atm (9.12 MPa), and the German coal Leopold Floez Hagen. Although the duration of the plastic period is not too different from predictions presented here for Pittsburgh Seam bituminous coal hydropyrolysis, even minor variations can be critical to assessing model reliability. The only means available so far to help evaluate parameters in the metaplast balance equations are experimental data on tar yields during rapid hydropyrolysis (Suuberg et al., 1980) and pyrolysis (Suuberg et al., 1979). This means that any model predictions for the time variation of metaplast concentrations during hydropyrolysis can be tested only indirectly. However as the modeling studies by Zacharias (1979) and Unger and Suuberg (1981) show, the same experimental tar yields can be fitted by models with different sets of internally self-consistent reaction and transport parameters.

EFFECT OF PARTICLE DIAMETER

Model predictions of the effect of particle diameter, equivalent to a coal film thickness, on CH_4 plus tar yields are shown in Figure 4, together with measured weight loss data from rapid hydropyrolysis of Pittsburgh Seam bituminous coal (Anthony et al., 1976; Suuberg et al., 1980). The abscissa gives the average size of the original coal particles for the experimental data and the film thickness for the calculated data. The calculated curve represents very well the measured trend for weight loss, for example, the difference being about 6–8 wt.% in each case for 70 and 275 μm particles (or film thickness). The gap between the calculated curve and the data points can be ascribed to pyrolysis products which are not, or only slightly, affected by variations in particle diameter, such as H_2O, CO, CO_2, etc. (Suuberg et al., 1980). Calculations are not extended to film thickness beyond 300 μm in order to avoid a regime where intraparticle heat or metaplast mass transfer limitations become significant. Model predictions for the individual CH_4 and tar yields are compared in Figure 5 with the sparse data available on individual product yields under screen heater hy-

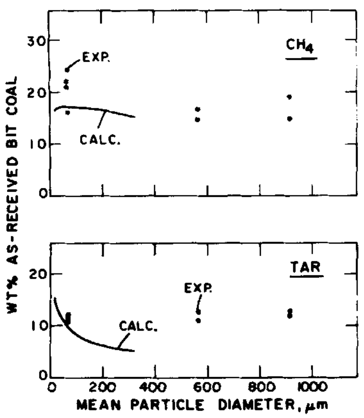

Figure 5. Effect of mean particle diameter on CH_4 and tar yields. (Reaction conditions as in Fig. 4.)

• Suuberg, 1977; *Calc.* curves: predictions from the present model.

dropyrolysis conditions (Suuberg, 1977). The experimental CH_4 yields only represent CH_4 formed in hydrogenation reactions through uptake of external H_2 and do not include CH_4 formed in pyrolysis reactions. The shape of the calculated curves, which in the case of tar reflects a greater particle size effect than the experimental data, can be attributed largely to two distinct phenomena.

First, the decrease in tar yield with increasing coal film thickness is due to the decrease in surface-to-volume ratio for thicker films. As the evaporation rate of metaplast (yielding the final product tar) is assumed to be proportional to the surface-to-volume ratio, thicker coal films with smaller external surface favor decomposition reactions of metaplast versus evaporation. This external surface effect may be overpredicted by the present model because in practice variations in original coal particle diameter may lead to lesser variations in thickness of the coalesced coal film. However, the effect is an interesting consequence of the evaporation-diffusion metaplast transport concept. Because in the hydrogenation concept of this model it is assumed that the vaporizable metaplast may be hydrogenated to form CH_4, variations in metaplast evaporation rate also affect CH_4 yields. In the screen heater experiment the amount of metaplast evaported from the coal becomes unavailable for CH_4 formation by hydrogenation, due to the rapid quenching in that experimental system.

The second phenomenon leading to an effect of particle diameter or coal film thickness on CH_4 yield is the limited accessibility of the softened coal mass during the plastic period, as indicated by the H_2 concentration profiles in Figure 1. Given the relatively low H_2 diffusivities in the viscous coal mass, in thicker films (or larger individual particles) a larger fraction of the coal is inaccessible to H_2. As a consequence, crosslinking of the reactive species is enhanced and less CH_4 is formed, according to the competitive reaction scheme used in the model.

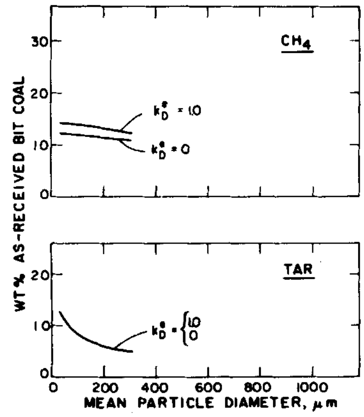

Figure 6. Predicted dependence of CH_4 and tar yields on mean particle diameter, as affected by variation of the eddy-diffusion parameter k^e_D. (Reaction conditions as in Fig. 1.)

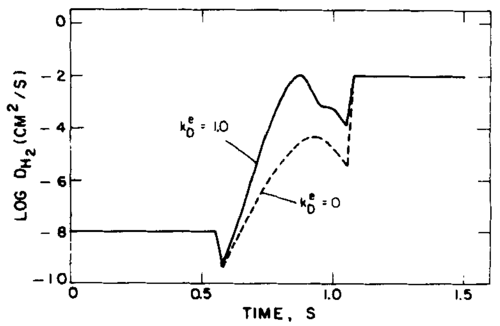

Figure 7. Predicted H_2 diffusivity, D_{H_2}, as affected by variation of the eddy-diffusion parameter k^e_D, for 70 μm coal film. (Other reaction conditions as in Fig. 1.)

As for the effect of limited accessibility of the plastic coal to H_2, the important parameter is the effective diffusivity of H_2, which represents molecular diffusion and an eddy contribution due to the pyrolysis gas stirring the plastic coal mass. Neither the molecular nor the eddy contribution can be predicted on theoretical grounds for the conditions of interest here. Therefore, the sensitivity of model predictions to variations in H_2 diffusivity was studied. Results are shown in Figure 6, in terms of calculated CH_4 and tar yields vs. coal film thickness for two different values of the eddy-diffusion parameter, k^e_D, and the same conditions as given in Figure 1. In the case of CH_4, increasing k^e_D from 0 to 1.0 leads to increased yields, although the shape of the curves remains unchanged. The difference between the curves reflects the difference in H_2 diffusivity and thus accessibility of the plastic coal to H_2. The variation in the eddy-diffusion parameter k^e_D does not have a significant effect on calculated yields of tar. Enhanced H_2 penetration in the plastic coal does indeed lead to increased metaplast formation, but the overall effect on metaplast evaporation rates and thus tar yield is small, as shown in Figure 6. The effect of k^e_D on CH_4 yield can also be visualized by the model predictions for H_2 diffusivities, shown in Figure 7 for a coal film thickness of 70 μm. The lower, broken-line curve represents the case where H_2 diffusion is assumed to occur by a purely molecular mechanism. Therefore the resulting diffusivities can be seen as molecular diffusivities of a gas (H_2) dissolved in a liquid. Given the high temperatures (500–1,000°C) and the complex chemical composition of the solvent, no information is available for an a priori estimate of comparable diffusivities. However, from existing data on diffusion of dissolved gases in ordinary liquids and molten polymers at moderate temperatures, the range of the predicted molecular diffusivities $10^{-8} - 10^{-4}$ cm²/s) appears realistic. A value of 1.0 for k^e_d leads to a strong increase in D_{H_2} during the plastic period, which reflects mixing and stirring of the plastic coal mass by the formation and expansion of bubbles of evolving pyrolysis gas. The resulting continuous-line upper curve in Figure 7 represents the base case of this modeling study discussed in the context of Figure 2.

EFFECT OF H_2 PRESSURE

The effect of H_2 pressure on calculated yields of CH_4 plus tar attained in 3 s at a final temperature of 1,000°C is shown in Figure

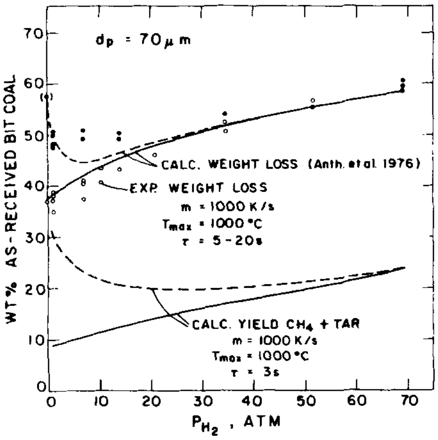

Figure 8. Effect of H_2 pressure on CH_4 plus tar yield as predicted by this model, and on weight loss during rapid hydropyrolysis of Pittsburgh Seam bituminous coal as measured and predicted by Anthony et al., 1976. (Nomenclature as in Fig. 1.)

● and - - - curves: pure H_2
O and —— curves: H_2-He mixture
Total pressure, 69 atm (6.99 MPa)
Particle diameter d_p, 70 μm

8 for two cases, pure H_2 atmosphere (broken-line curve) and an atmosphere of H_2 and He at a total pressure of 69 atm (6.99 MPa; solid curve). Also shown are experimental weight loss data for Pittsburgh Seam bituminous coal and predicted weight loss curves for that coal and the same two cases (pure H_2, H_2/He), given by Anthony et al. (1976). The latter are based on their weight loss model in which no intraparticle mass transfer was considered and extraparticle mass transfer of reactive volatiles was lumped into one single temperature-independent transport coefficient. The model developed in the present study incorporates a more sophisticated description of intra- and extraparticle transport processes for H_2 and of extraparticle metaplast transport. As in the case of particle diameter effects, the CH_4 plus tar yields curves predicted in this study represent well the measured trend for weight loss, the difference between data points and CH_4 plus tar yield curves being ascribable to coal pyrolysis products not or only slightly affected by H_2 pressure.

The individual curves for predicted CH_4 and tar yields are given in Figure 9 for the two cases of a pure H_2 and a H_2/He atmosphere. No experimental screen heater data showing the effect of H_2 pressure on individual product yields are available to test the trends predicted by the present model. However the curves shown in Figure 9 are instructive in the model's basic mass transfer concept. In the case of pure H_2, CH_4 yields increase with increasing H_2 pressure, as hydrogenation of the reactive species is favored over crosslinking at higher H_2 concentration, according to the competitive reaction scheme. At the same time, increased H_2 pressure leads to lower gas phase diffusivities for metaplast and to lower yields of tar. Summation of CH_4 and tar yields gives the upper curve with the characteristic minimum which reflects the combined physical and chemical effects of H_2 pressure. In H_2/He mixtures with constant total pressure of 69 atm (6.99 MPa), an increase in H_2 partial pressure leads as well to an enhanced formation of CH_4, as the concentration of H_2 contacting the decomposing coal increases. However, tar yields remain constant, as the extraparticle transport resistance remains constant with variations in H_2 partial pressure. This behavior is in agreement with the finding by Suuberg et al. (1980) that tar yield under conditions of rapid volatiles quenching is the same for 69 atm of H_2 as for 69 atm of He. Inclusion of intraparticle metaplast transport would smooth the predicted pressure effect on tar yields if at low ambient gas pressure, i.e., low extraparticle transport resistance, intraparticle transport becomes more and more rate-limiting.

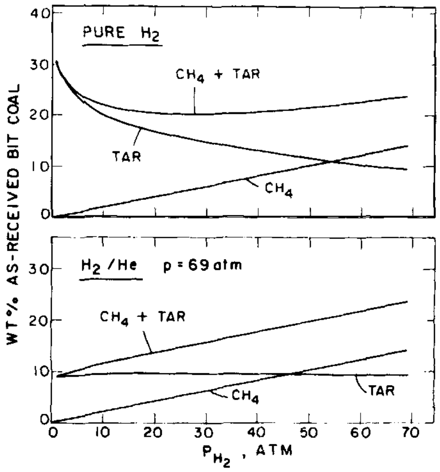

Figure 9. Effect of H_2 pressure on CH_4 and tar yields as predicted by this model, in pure H_2 and in H_2-He mixtures with total pressure 69 atm (6.99 MPa). (Reaction conditions as in Fig. 8.)

The enhancing effect of H_2 pressure on metaplast formation rate, as included in the model, is too small to be seen in the integral tar yields in Figure 9. However, it can be seen in the duration of the plastic period, which becomes longer with increasing H_2 pressure, and in the increasing maximum fluidity, Table 1. Maximum fluidity is given as a relative quantity, with reference to the maximum fluidity at 1 atm (101 kPa) of pure H_2. The predicted trends indicate that there is both a physical effect of inert gas pressure and a chemical effect of H_2 pressure, in agreement with the coal plasticity data under low heating rate conditions in the literature.

Anthony (1974) compared the similar effects of increasing H_2 partial pressure and decreasing particle diameter on weight loss during rapid hydropyrolysis. From his data with Pittsburgh Seam bituminous coal, he determined what H_2 partial pressure for 70 μm coal particles, under a total pressure of H_2 and He of 69 atm (Figure 8), would give the same weight loss as was measured for different particle diameters under 69 atm of H_2 and otherwise identical reaction conditions (Figure 4). He expressed the results as a crossplot of so-called "effective" H_2 pressure against coal particle diameter, shown in Figure 10. As a reference, the effective H_2 pressure of 69 atm was defined for 70 μm particles. This defi-

TABLE 1. EFFECT OF H_2 PRESSURE ON MAXIMUM FLUIDITY (RECIPROCAL VISCOSITY) AND DURATION OF PLASTIC PERIOD DURING HYDROPYROLYSIS

Model Prediction for:

Coal film thickness, 70 μm Nominal heating rate, 1,000 K/s	Max. temp., 1,000°C Holding time, 3 s

	Relative Max. Fluidity		Duration Plastic Period, s	
p_{H_2} atm*	Pure H_2	H_2/He P_{tot} = 69 atm	Pure H_2	H_2/He P_{tot} = 69 atm
1	1.0	4.6	0.400	0.475
10	3.6	5.9	0.475	0.500
35	5.8	6.6	0.500	0.500
69	6.6	6.6	0.525	0.525

SI Conversion: kPa = atm × 101.325.

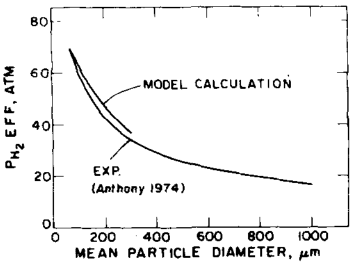

Figure 10. "Effective" H_2 pressure as a function of particle size. *Exp.* curve constructed from crossplot of data of Anthony, 1974 (○ in Figs. 4 and 8); *Model* curve constructed from crossplot of *Calc. Yield* CH_4 + *Tar* curves in Figs. 4 and 8.

nition is somewhat arbitrary, implying that the 70 μm particles saw the full 69 atm H_2, which can be doubted with respect to the measured particle size effect (Figure 4). In addition, it is not consistent with the H_2 concentration profiles shown in Figure 1 for a 70 μm coal film. Therefore, not the absolute values but their trend should be of interest. Anthony's interpretation was that diffusion is a limiting factor for getting sufficient H_2 in contact with the reactive hydrogenable species. This interpretation led him to view the quantity resulting from the crossplot as an effective H_2 pressure.

If the predicted yields of CH_4 plus tar are crossplotted in the same way as Anthony's weight loss data (i.e., from Figures 4 and 8), the resulting curve is close to the experimental one, as shown in Figure 10. The good agreement of the curves indicates that the present model's mass transfer concept is consistent with the limited experimental data on weight loss. However, there is an important difference from Anthony's qualitative interpretation of his crossplot. Anthony considered the effect of particle diameter to be solely due to diffusion limitations affecting H_2 concentration and thus limiting contact of H_2 with the reactive species. The concept developed in this study considers not only this limited accessibility of the reactive hydrogenatable species to H_2 but also incorporates an effect of external particle surface area on tar transport away from the coal.

CONCLUSIONS

This study was intended to contribute to a better understanding of the complex processes involved in rapid hydropyrolysis of single coal particles. The understanding helps in developing models for practical reactor design, where the overall process is more complex than in the single-particle case and thus where simplifications and omission of insignificant details are often needed. The type of modeling study presented here can be used to detect such insignificant details or address specific questions such as how to optimize feed particle size to achieve a desired product distribution or how to minimize agglomeration of softening coal under different process operating conditions.

At present, the data base for model validation is sparse. However, as a result of the present calculations, previously measured effects of H_2 pressure and particle size on CH_4 and tar yields can be interpreted in terms of the plastic period being significant for intraparticle H_2 transport limitation due to the expected low diffusivities in the highly viscous coal melt. Modeling transport of the plasticizing products (metaplast) of coal pyrolysis as evaporation-diffusion from a liquid surface predicts trends which agree with experimentally observed effects of pressure on both tar yield and coal fluidity.

Areas where the experimental data base should be enlarged can be pinpointed. Especially needed are measurements of the coal's plastic behavior, i.e., its rheological properties under the conditions of interest here. Such work is now under way at M.I.T. Furthermore, additional measurements of individual product yields (e.g., CH_4 and tar) as affected by inert gas pressure, H_2 pressure, and particle size would be invaluable for further testing the model and thus elucidating the mechanisms involved in hydropyrolysis of softening coal.

Some aspects of the model's mechanistic concept should be improved when additional experimental results become available. For example, the description of the eddy contribution to H_2 diffusivity in the plastic coal is a purely empirical approach. An ongoing theoretical study of bubble dynamics due to pyrolysis gas evolution will help to elucidate this phenomenon as well as the swelling behavior of the coal, which was not represented in the model. Also warranting further study is the model description of the softening and resolidification processes. The effect of H_2 on the rates of metaplast reactions, which is seen here as an enhancing effect on only the formation reaction, may also include the secondary decomposition reaction. Of particular interest is the question of how significant is the external surface-to-volume ratio of the coal with respect to metaplast transport, viewed here as evaportion-diffusion. In the present calculations, the effect probably is overpredicted since a comparable particle size effect on tar yield would occur in pyrolysis, but has not been found. Further verification is also needed for the assumption, introduced here as a working hypothesis, that metaplast as part of the hydrogenatable species is vaporizable.

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