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 ENGELHARD MINERALS CORP \*US 4483-691-A  
 13.02.84-US-579842 (+US-430200) (20.11.84) C01b-03/40 C10j-  
 03/16 C10k-03/02

H(9-C) N(1-C2, 2-E, 2-F2)

160

Coal gasification liq. by/product conversion - to synthetic natural gas via partial oxidn. over monolith catalyst contg. platinum and palladium

#### ADVANTAGE

Conversion of the environmentally-damaging liq. by-prod. is simpler and cheaper than by conventional partial oxidn. or steam reforming. The monolithic catalyst is small, with low pressure drop, and brings overall economies, in spite of using Pt metals. Operation is possible at lower  $O_2/C$  ratios and lower temps. than in non-catalytic oxidn., without carbon deposition.

C84-130713

#### CLAIMED PROCESS

In a coal gasification process, coal is reacted with steam and  $O_2$  to give

(a) a gasifier synthesis gas, which is then methanated to provide a synthetic natural gas (SNG), and

(b) a liq. by-prod., which is treated by:

(i) mixing with  $H_2O$  and  $O_2$  and preheating to initiate reaction in stage (ii), but not above  $1200^\circ F$ ;

(ii) feeding the mixt., with  $H_2O/C = 0.5-5$  and  $O_2/C = 0.15$  to  $0.4$ , to a monolithic catalyst body, with many gas flow passages and contg. dispersed Pt and Pd, for oxidn. and cracking, to give mainly  $CH_4$ ,  $H_2$ ,  $CO$ ,  $CO_2$ ,  $H_2O$  and  $C_2-C_4$  hydrocarbons;

(iii) removing  $CO_2$  and  $H_2O$  from the effluent; and

(iv) methanating the synthesis gas which remains, giving more SNG.

#### SECOND CATALYTIC STAGE

Before being sent to treatment stage (iii), the effluent from stage (ii) is opt. passed to: (iia) steam reforming on a catalyst pref. contg. Pt and Rh, for further hydrocarbon conversion.

#### PROCESS

In stage (ii), the feed, preheated pref. to  $800-1200^\circ F$ , is passed at a rate of about 100,000 vols./catalyst vol. x h, or more. The temp. in the monolith is pref.  $1400-2000^\circ F$ , part at least being at least  $250^\circ F$  above the ignition temp. of the inlet stream.

Some hydrocarbons are oxidised, and remaining  $C_5+$  hydrocarbons are cracked;

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pref. at least about 50 wt. % of the feed is converted to 1C cpds. In optional stage (iia), the stage (ii) effluent passes directly to the catalyst at 2,000-20,000 vols./vol. catalyst x h, and the overall conversion to 1C cpds. rises to at least 98 wt. %. The pressure in both catalyst zones is, pref. 50-1500 psig.

#### CATALYSTS

In stage (ii), the monolithic support is a ceramic or stainless steel honeycomb, with a support coating, pref. of a stabilised, high surface area transitional alumina. The active component pref. comprises Pd (pref. 10-90, more esp. 40-60 wt. %), and Pt (to 100%), opt. with Rh.

The stage (ii) catalyst may comprise  $Al_2O_3$  pellets or extrudates (surface area 10-200  $m^2/g$ ), stabilised with La and Ba oxides and supporting a mixt. of Rh (pref. 10-90 wt. %, esp. 20-40 wt. %) and Pt (to 100%). (12pp1492MHDwgNo 0/2).

84-127535/21 H04 ENGH 30.09.82  
 ENGELHARD MINERALS CORP \*AU 8319-728-A  
 30.09.82-US-430452 (+US-430147) (05.04.84) C01b-03/32  
 Auto-thermal reforming by partial oxidn. and steam reforming -  
 using monolithic platinum-gp.-metal-contg. partial oxidn. catalyst

H(4-C1, 4-C2, 4-F2C) N(2-E, 2-F2)

004

C84-053889 Full Priorities: 30.09.82.(5) - US-430147,  
 430200, 430320, 430451, 430452.

CLAIMED PROCESS

Prod'n. of synthesis gas is effected in two stages:

- (i) catalytic partial oxidn. of a feed mixt. comprising hydrocarbon feed stream, H<sub>2</sub>O and O<sub>2</sub>-contg. gas, the preheated feed mixt. being contacted with a monolithic (honeycomb-type) catalyst (I) comprising Pd and Pt (and opt. Rh) on a refractory metal oxide, and  
 (ii) catalytic steam reforming of the first-stage effluent over a Pt-Rh steam reforming catalyst.

Specifically, the feed mixt. to stage (i) is controlled to give an H<sub>2</sub>O:C ratio of 0.5-5 and an O<sub>2</sub>:C ratio of 0.2-0.8. The step is carried out at 1-142 atm. and at such temps. that at least part of (I) is at at least 121°C above the ignition temp. of the inlet stream, providing cracking of any unoxidised 5C+ hydrocarbons to light (4C or below) hydro-

carbons.

USES/ADVANTAGES

The specifically claimed embodiments relate to integrated processes utilising the H<sub>2</sub>-rich product gas for the prodn. of (A) ammonia, (B) methanol, (C) SNG and (D) liq. hydrocarbons. The low pressure drop and the high (at least 100,000) volumetric throughput of stage (i) allows reduced size and volume of catalyst to be used, and the use of Pt-gp. metals allows very low catalytic metal loadings to be used. Operation is at relatively low H<sub>2</sub>O:C and O<sub>2</sub>:C ratios, without catalyst fouling by C deposition.

PREFERRED

The stage (i) catalyst comprises 10-90 (pref. 25-75, esp. 40-60) wt. % each of Pd and Pt, and the stage (ii) catalyst 10-90 (pref. 60-80) wt. % Pt and 10-90 (pref. 20-40) wt. % Rh. The GHSV in stage (ii) is pref. 2,000-20,000.

The O<sub>2</sub>:C ratio is 0.35-0.65 (0.5-0.6) in embodiments (A) and (B), 0.15-0.4 in embodiment (C) and 0.4-0.65 in embodiment (D).

In embodiment (C), the feed is the liq. AU8319728-A+

hydrocarbon by-product of a coal gasification process, and the H<sub>2</sub>-rich gas product of step (ii) is mixed with the gasifier effluent for methanisation to SNG.

Feed for embodiment (D) is either natural gas or methane; liq. by-products may be recycled to form part of the feed.

The preheat temp. is generally 427-760°C and stage (i) is generally operated at 954-1316°C (427-649 and 760-1093°C respectively for embodiment (C)). (104pp1639MHDwgNo 0/7).