

The I.G. High-velocity Gas Recycle Process

The following additional information about this method, in which the heat of reaction is removed by recycling cooled residual gas, at a very high rate, was obtained from Mr. Michael who was responsible for its development.

The physical state of the catalyst is very important. In the first place it must be very hard or otherwise it breaks up in the rapid gas stream and the resistance of the bed becomes uneven, with the result that part of the

catalyst is only exposed to a slow gas stream and so becomes overheated. Hence sintered carbonyl iron catalysts were normally used but precipitated catalysts will do equally well if they are sintered. Irregular pieces, of size about 10 mm. were used and with these, good gas distribution can always be attained by using a large excess pressure for the recirculating gas. It was realized, however, that if regular shaped pieces could be used the excess pressure might be reduced to 1/5th of the present value, with a corresponding saving of power.

The design of the reactors must be arranged so that the end where the gas enters is streamlined. This was not done in a 4 cu.m. reactor made by I.G. and turbulence occurred in such a way that in certain places the gas flow was not large and hot spots developed. If another large reactor had to be built for this purpose, one with about six alternate layers of catalyst and cooling tubes would be used and with such a design a recirculation ratio of 25 would be sufficient instead of 100. Figure 6 is a sketch of such a plant and was obtained from the I.G. Farben at Ludwigshafen-Oppau. Another interesting type of reactor that has been used for 5 litres of catalyst had a high-speed fan at the top and a central tube through the catalyst bed, and the fan was arranged to circulate gas up through the central tube and down through the catalyst bed. Both fan and motor were enclosed in the pressure resisting casing of the reactor. In this way an internal recirculation could be carried out in addition to the main external cooling recirculation, and the temperature distribution in the reactor was thereby improved. Dr. Michael thought it might also be a good idea to arrange the catalyst in an annular space between two perforated walls and pass the gas through, radially, from the inside to the outside. With such an arrangement catalyst could be removed continuously from the bottom, reactivated, and put in again at the top.

Starting with synthesis gas with $H_2/CO = 0.8$ to 1.2, Dr. Michael said that the high-velocity gas recycle process will give an output of 800 kg./cu.m. catalyst/h. at a conversion of $U_{CO} + H_2 = 75 - 80\%$ and at a temperature of $300 - 350^\circ C$. The reaction goes in such a way as to produce four moles carbon dioxide to one of water, as the oxygen products. Of the gas converted, 20% goes to form methane and the composition of the products higher than methane is given in Table 45.

Table 45.

Products from High-velocity Gas Recycle Process

Products higher than methane		Gasol Fraction	
	%		%
Gasol, C ₂ - C ₄	24-30	Xtylene	20
Benzin	50	Propylene	35
Middle Oil	15	n - Butylene	10
Higher Oil	1-2	Isobutylene	20
		Propane + Butane	10-20

The benzin has an octane number of 65 and the much higher figures frequently quoted refer to the product obtained by refining the benzin with platinum or bleaching earth. The higher oil is an inferior lubricating oil. Very little wax is formed, but what little there is, is branched chain and resembles ceresin.

The life of the catalyst should to some 5 months on the large scale, but on the smaller scale, with internal circulation as described above, lives of 10 months have been obtained.

At lower temperatures, 220°C, for example, there is less methane formed but the quality of the benzin is worse. More wax is also formed, and the lower the temperature the higher the percentage of oxygen in the wax. At those lower temperatures the catalyst requires treatment with hydrogen at 420°C. every three days or so, as it gets covered with oxygen-containing residues.

It was stated that when a distribution curve is plotted for the products of this process, showing yield of each individual chain length plotted against chain length, the yield falls off steadily as the chain length increases, approaching zero asymptotically for large chain lengths, and has no maximum and minimum at C₂-C₃ and C₄ respectively, as in the case for the products of the normal synthesis. This is very surprising.

The final position with regard to the high-velocity gas recycle process was that it was generally acknowledged to be too expensive for the production of benzin. To get heavier

products more active catalysts were required but precipitated catalysts were found to be much too soft to stand up to the high recycle conditions.

In several respects, the conditions are similar to those that hold good when the fluidized bed technique is used, and Dr. Michael had actually carried out experiments with a fluidized bed in the reactor with internal circulation. Results were very good at first, but the particles of catalyst soon stuck together and the method ceased to work. During the initial period the rate of production of products was ten to twenty times the normal rate.

As is well-known, the catalysts used in the work with high-velocity gas recirculation were made by sintering carbonyl iron. It was thought to be useful therefore to find out how I.G. Farben produced their carbonyl iron, and the following information was obtained. Type A iron is produced by decomposing pure iron carbonyl vapour at 250°C. The particles formed contain 1.2% carbon and are of sizes ranging from 3 to 10 microns. Type E iron is prepared by adding 10% ammonia to the carbonyl and then decomposing it as before. The amount of carbon is less than in type A but the powder contains a little nitrogen. All the carbon in these preparations can be removed by hydrogen at 400.