960564) - 1
IPOS Copy

TS 0012 SIN

MANUFACTURING ETHYLENE OXIDE

The present invention relates to a process of manufacturing ethylene oxide.

USA patent specification No. 4 904 807 discloses a process of manufacturing ethylene oxide comprising allowing to react in a reactor ethylene and free oxygen to ethylene oxide in the presence of methane, carbon dioxide and argon; withdrawing from the reactor a reactor effluent; removing ethylene oxide from the reactor effluent to obtain a recycle stream; removing carbon dioxide and argon from the recycle stream to obtain a treated recycle stream; supplying the treated recycle stream to the reactor; and supplying additional reactants and methane to the reactor.

Additional reactants, ethylene and free oxygen-containing gas, are supplied to the reactor to replace the reactants which reacted to ethylene oxide. Methane is supplied to maintain the methane concentration at such a level that its presence is beneficial to the efficiency of the reaction as described in USA patent specification No. 3 119 837.

Carbon dioxide is a by-product of the reaction and part of the carbon dioxide is removed to maintain the concentration of carbon dioxide below a predetermined level.

Argon enters into the process with the free oxygen-containing gas. In order to maintain the argon concentration below a predetermined level, part of this contaminant is removed from the recycle stream downstream of the removal of ethylene oxide. In addition trace amounts of nitrogen enter into the process with the free oxygen-containing gas.

In the known process removing argon comprises

separating a side stream from the recycle stream, passing the side stream to the feed inlet of a membrane unit, removing an argon-containing stream from the permeate outlet of the membrane unit, removing a retentate stream from the retentate outlet of the membrane unit, and adding the retentate stream to the recycle stream.

As the flow through the membrane is large, the area of the membrane is large as well.

It is an object of the present invention to reduce the area of the membrane in the membrane unit.

To this end the process of manufacturing ethylene oxide according to the present invention comprises allowing to react in a reactor ethylene and free oxygen to ethylene oxide in the presence of methane, carbon dioxide and argon; withdrawing from the reactor a reactor effluent; removing ethylene oxide from the reactor effluent to obtain a recycle stream; removing carbon dioxide and argon from the recycle stream to obtain a treated recycle stream; supplying the treated recycle stream to the reactor; and supplying additional reactants and methane to the reactor, wherein removing argon comprises separating a side stream from the recycle stream, passing the side stream to the feed inlet of a membrane unit, removing an argon-containing stream from the permeate outlet of the membrane unit, removing a retentate stream from the retentate outlet of the membrane unit, adding the retentate stream to the recycle stream, and purging the permeate side of the membrane unit with inert gas.

The invention will now be described by way of example in more detail with reference to the enclosed Figure showing schematically a line-up of the process of the present invention.

A mixture including ethylene, free oxygen-containing gas, argon, carbon dioxide and methane is supplied through conduit 1 to a reactor 2. In the reactor 2

ethylene and free oxygen are allowed to react to ethylene oxide in the presence of methane and contaminants such as carbon dioxide and argon. The reactor 2 contains a suitable catalyst. The catalyst, the reaction conditions and the concentration of the reactants are known per se, and as they are not essential to the present invention they will not be discussed.

From the reactor 2 a reactor effluent is withdrawn through conduit 4. The reactor effluent includes ethylene oxide, carbon dioxide, methane and argon. Carbon dioxide is a by-product of the reaction, a further by-product is ethane. Argon enters into the process with the free oxygen-containing gas, in addition trace amounts of nitrogen enter into the process with the free oxygen-containing gas.

Ethylene oxide is removed from the reactor effluent in an ethylene oxide removal unit 6, and from the ethylene oxide removal unit 6 are removed an ethylene oxide-containing stream through conduit 8 and a recycle stream through conduit 9. The ethylene oxide removal unit is known per se and will not be discussed.

The other contaminants are removed downstream of the ethylene oxide removal unit. For the present invention we are not concerned with the removal of nitrogen and ethane.

Downstream of the ethylene oxide removal unit 6 the recycle stream is compressed in compressor 11 to compensate for the pressure drops in the treating units.

The compressed recycle stream in conduit 12 is divided into a side stream 16 and a main stream. The main stream is divided into two streams 18 and 19. Carbon dioxide is removed from stream 19 in carbon dioxide removal unit 22. From the carbon dioxide removal unit 22 are withdrawn a carbon dioxide-containing stream through conduit 23 and a treated stream having a reduced carbon dioxide content through conduit 24. The carbon dioxide

removal unit is known per se and will not be discussed. The flow rate of the side stream 16 is between 1 and 5% of the flow rate of the recycle stream, the flow rate of stream 19 is between 10 and 40% of the flow rate of the recycle stream, and the flow rate of stream 18 is the balance.

Removing argon from the recycle stream comprises passing the side stream 16 to the feed inlet 29 of a membrane unit 30, removing an argon-containing stream from the permeate outlet 32, and removing a treated stream from the retentate outlet 33. The membrane unit 30 comprises a hollow fibre membrane schematically represented by dashed line 35, the hollow fibres include polysulfone. To purge the retentate side of the membrane unit 30, inert gas is supplied to the membrane unit 30 through conduit 36, the inert gas flows counter-currently through the retentate side of the membrane unit 30.

The treated stream 24 from the carbon dioxide removal unit 22 and the treated stream 33 from the membrane unit 30 are combined with the untreated stream 18 and supplied through conduit 40 and conduit 1 into the reactor 2. Additional reactants, ethylene and free oxygen-containing gas, and methane are supplied to the reactor 2 through conduit 42 which opens into conduit 40.

To illustrate the effect of purging the performance of the membrane unit 30 in case the permeate side is not purged is compared with the performance in case the permeate side is purged.

Table 1 gives an example of the compositions and the flow rates of the components in the feed to a membrane unit used in the process without purging, and in the retentate and the permeate leaving the membrane unit. The membrane unit includes a hollow fibre membrane having an area of 1 630 m^2, the hollow fibres include polysulfone.

Table 1. Compositions and flow rates of the feed, permeate and retentate of the membrane unit without

purging (not according to the present invention).

	Feed		Permeate		Retentate	
	%vol	kmol/h	%vol	kmol/h	%vol	kmol/h
Nitrogen	1.22	4.54	0.81	0.14	1.24	4.40
Argon	9.30	34.6	11.88	2.09	9.17	32.51
Oxygen	4.16	15.48	7.70	1.36	3.98	14.12
Methane	50.24	186.90	38.36	6.75	50.83	180.15
Ethylene	27.20	101.19	22.72	4.00	27.42	97.19
Ethane	0.95	3.53	0.63	0.11	0.97	3.42
Carbon dioxide	6.93	25.78	17.90	3.15	6.39	22.63
total	100.00	372.02	100.00	17.60	100.00	354.42

Table 2 gives the compositions and the flow rates of the components in the feed to a membrane unit used in the process of the present invention, and in the retentate and the permeate leaving the membrane unit. The membrane unit includes a hollow fibre membrane having an area of 1 310 m^2, the hollow fibres include polysulfone. The purge gas is methane and the flow rate of the purge gas is 7.44 kmol methane per hour, which corresponds to 2 mol% of the feed flow rate.

Table 2. Compositions and flow rates of the feed, permeate and retentate of the membrane unit with purging (according to the present invention).

	Feed		Permeate		Retentate	
	%vol	kmol/h	%vol	kmol/h	%vol	kmol/h
Nitrogen	1.34	4.99	0.59	0.14	1.36	4.85
Argon	9.28	34.52	8.85	2.09	9.11	32.44
Oxygen	4.15	15.44	7.19	1.70	3.86	13.74
Methane	50.12	186.46	50.09	11.82	51.17	174.64
Ethylene	27.14	100.97	15.54	3.67	27.35	97.30
Ethane	1.06	3.94	0.47	0.11	1.08	3.83
Carbon dioxide	6.91	25.71	17.28	4.08	6.07	21.63
total	100.00	372.02	100.01	23.60	100.00	348.42

Comparing the performances of the two membrane units, it is clear that with purging the same amount of argon passes through a membrane with a significantly smaller area, and that a smaller amount of ethylene passes through the membrane.

The flow rate of the purge gas is suitably in the range of from 1 to 10 mol% of the feed flow rate. The amount of ethylene permeating through the membrane decreases with increasing purge gas flow rate, however, above 10 mol% the decrease is too small to justify such a large purge gas flow rate.

In the process described with reference to the Figure the main stream is divided into streams 18 and 19, and carbon dioxide is removed from stream 19. Alternatively, carbon dioxide can be removed from the main stream, so that dividing the main stream can be omitted.

The membrane material can be any material that allows selective permeation of argon. Suitably the membrane is a dense membrane of a polymeric material such as polydimethyl siloxane or polysulfone.

Examples of suitable inert gases are methane, nitrogen, carbon dioxide, or mixtures of these gases.

C L A I M S

1. A process of manufacturing ethylene oxide comprising allowing to react in a reactor ethylene and free oxygen to ethylene oxide in the presence of methane, carbon dioxide and argon; withdrawing from the reactor a reactor effluent; removing ethylene oxide from the reactor effluent to obtain a recycle stream; removing carbon dioxide and argon from the recycle stream to obtain a treated recycle stream; supplying the treated recycle stream to the reactor; and supplying additional reactants and methane to the reactor, wherein removing argon comprises separating a side stream from the recycle stream, passing the side stream to the feed inlet of a membrane unit, removing an argon-containing stream from the permeate outlet of the membrane unit, removing a retentate stream from the retentate outlet of the membrane unit, adding the retentate stream to the recycle stream, and purging the permeate side of the membrane unit with inert gas.
2. Process as claimed in claim 2, wherein to purge the permeate side of the membrane, the inert gas is passed counter-currently along the membrane.
3. Process as claimed in claim 1 or 2, wherein the flow rate of the purge gas is in the range of from 1 to 10 mol% of the feed flow rate.

