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<p>(54) Title: CATALYTIC PROCESS FOR THE PREPARATION OF LIGHT OLEFINS FROM METHANOL IN A FLUIDISED BED REACTOR</p>		
<p>(57) Abstract</p> <p>Methanol is converted in light molecular olefins C₂-C₄ with 93-100% degree of transformation and more than 90% selectivity in which more than 80% are ethylene and propylene upon a microspherical catalyst based on SAPO-34 zeolite, with continuous reaction-regeneration in a fluidized bed reactor-regenerator system. Ethylene/propylene ratio is changed in relatively large limits, 0.69-1.36, by the modification of reaction temperature and space velocity of the feed.</p>		

Catalytic Process for the Preparation of Light Olefins from Methanol in a Fluidised Bed Reactor.

Background of the Invention

Field of the Invention

- 5 This invention relates to a process for conversion of methanol to olefins using SAPO-34 zeolite in a fluidised bed reactor with continuous regeneration of catalyst.

Background Art of the Invention

Light olefins, namely ethylene and propylene, are important raw materials for production of polymers.

- 10 Industrial ethylene and propylene are obtained by steam cracking of C₂-C₄ paraffins and petroleum fractions in a so called hydrocarbons pyrolysis process. The continuous rise of the requirement for olefins and the oil reserve shortage in the future makes of interest research for new olefin manufacture technologies from non-petroleum raw materials.

- 15 One of the more attractive methods for C₂-C₄ olefin production is based on catalytic conversion of methanol, because methanol is manufactured in advanced technologies with very high capacity up to 800,000 mt/year for a single line and has wide raw material availability, like natural gases, including methane, coal, and renewable biomass.

- 20 Methanol conversion into light olefins with industrially acceptable yields was possible only after the synthesis of high silica zeolite ZSM-5 by Mobil Oil's researchers (US Patent 3.702.886). After this, many other types of zeolites were tested in the reaction of methanol to olefins like ZSM-34 (US Patent 4079096), Mordenite (Ro Patent 87685, US Patent 3244766), Offretite (US Patent 4079096) arseno-silicates (Ger. Off. 2830830), boro-silicates (Ger. Off. 2830787). Methanol conversion to olefins is claimed
25 also in many patents on synthetic alumino-silicates, like US Patents 4062905, 3979472, 3911041 and Ger. Off. 2755299, 2615150.

These catalysts exhibit low selectivities in olefins and must be periodically regenerated with air at 470-570 °C.

- 30 Numerous methods for modification of zeolites and reaction conditions have been proposed for olefin selectivity rising and increasing the active cycle of the catalysts. Interesting results were obtained by zeolite silification (US Patent 4100219, 4145315),

increasing Si/Al atomic ratio by aluminum extraction (US Patent 4447669, Ger. Off. 2935863), ionic exchange or impregnation with Cs, Ba, Pb, Tl (US Patent 4066714, B, Mg (US Patent 4049573, Ger. Off. 3300892), Hf, Zr (US Patent 4481376, Ger. Off. 3300982), dilution of the catalyst with inert materials (US Patent 4025572), partial
5 deactivation with steam (UK Patent 2127036) or HF (US Patent 4486617). Good results have given complex treatments with Mg-Mg or Mg-Sb (RO Patent 87413). Some reaction parameters were also modified, for instance under atmospheric pressure utilization (US Patent 4025575), steam dilution of feed (US Patent 4083889) or dilution with air (US Patent 4433189), oxygen (US Patent 4049735) and aldehyde (US Patent
10 4374295).

The synthesis by Union Carbide Corporation's researchers of Si-Al-P zeolites, named SAPO-zeolites (US Patent 4310440, 4440871), has opened new perspectives for methanol conversion to olefins, MTO-process. As the Chinese researchers have demonstrated for the first time, on SAPO-34 zeolite it obtained up to 89% C₂-C₄ olefins
15 at practical total conversion of methanol 57-59%, being ethylene and ethylene/propylene molar ratio 2.24-2.31 (Applied Catalysis, Vol. 40, Nr. 1-2, 1988, p.316). Due to the catalyst, the coking active cycle is only 1-2 hours.

The use of SAPO-34 zeolite synthesised as in US Patent 4440871 gave rise to contradictory literature data concerning thermal and steam stability and olefin
20 selectivity. It must be also emphasised that the synthesis of the zeolite is carried out using expensive materials, like aluminum isopropoxyde, tetraethylammonium hydroxide or quinuclidine. Neutralisation of the reaction mixture with NaOH has complicated the technology for SAPO-34 manufacture, due to the necessity of ammonium ion exchange and a supplementary calcination step.

25 Thermal analysis (Gr.Pop et al., Progress in Catalysis, Bucharest, 1, 1993, p.1) showed that SAPO-34 zeolites with crystals smaller than 4 micrometer have a good thermal and steam stability.

MTO-process was materialised in tubular reactors with fixed bed catalyst (US Patent 4590320) and in fluidised bed reactors with catalyst regeneration in fluidised beds.
30 Vaporised methanol feed mixed with the zeolite catalyst is charged to the bottom of the riser contact zone to form a suspension for flow upwardly through the riser (US Patent 4328384). The reactors with fixed bed catalyst have many disadvantages in methanol reaction to olefins, because the removal of the reaction heat is difficult and frequent catalyst regeneration diminishes production capacity. Increasing the coke deposits on
35 the catalyst in the active cycle changes continuously the reaction products composition.

Fluidised bed reactors and continuous regeneration of the catalyst eliminate these disadvantages but in a riser reaction the optimal reaction conditions cannot be realised.

Kinetic studies in fluidised bed reaction have shown a maximum ethylene formation at short reaction time of about two seconds. (C. Tsakiris et al., Proc. IFAC Symposium 5 DYCORD 92 College Park, Maryland, April 1992), which is not obtainable in a riser reactor. In a riser type reactor two important reaction parameters, contact time and temperature, which determine product selectivities cannot be well controlled.

All synthesised zeolite including SAPO-34 has low selectivity in methanol conversion to olefins. By zeolite modifications its selectivity can be increased. For example 10 PCT/US96/19673 teaches a process in which SAPO-34 zeolite is modified by ion exchange with Ni and then give a catalyst with 30% more selectivity in methanol conversion into ethylene and propylene.

In US 5095163, SAPO-34 catalyst is hydrothermally treated 3-50 hours, in air or steam, at 650-775°C. The catalyst acidity decreases from about 5.2 meq. NH₃ / cc to 1.8 meq. 15 NH₃ / cc and selectivity to C₂ + C₃ olefins in methanol conversion increase from 63 to 80%. US 4873390 uses a selective coking of the SAPO-34 zeolite up to 5-30% coke in the catalyst, to improve the methanol transformation in ethylene from 6.04% to 46.65% and in propylene from 15.43% to 50.60%. Maximum ethylene + propylene selectivity obtained is 83.7%.

20 Summary of the Invention

According to the present invention a method for the preparation of light olefins from methanol conversion using a microspherical catalyst including SAPO-34 zeolite is provided. The method includes the step of contacting methanol with a fluidised or moving bed of said catalyst wherein said SAPO-34 zeolite is prepared from alumina, 25 silica and a preformed template tetraethylammonium phosphate.

The Invention eliminates these difficulties since the catalyst is obtained from cheap raw materials like industrial alumina and aqueous silica sol and the template, tetraethylammonium phosphate, is prepared "in situ" from ethyl bromide and triethylamine. The methanol conversion and continuous catalyst regeneration are 30 conducted in fluidised bed reactors, without riser, methanol feed being injected in a dense bed catalyst.

The claimed process may find applications in the industry to obtain, by a new more economic route, ethylene and propylene, basic raw materials for petrochemistry and synthetic fuels.

Detailed Description of the Invention

5 The catalyst synthesis made only with cheap, industrial raw material, namely triethylamine, ethyl bromide, concentrated phosphoric acid, more than 70% weight, hydrated alumina and silica sol, all with very low, under 0.01%, Na content. Concentrated silica sol can be stabilised with ammonia. In accordance with the invention, by hydrothermal treatment of Si-Al-P amorphous gel, the active H form of
10 SAPO-34 zeolite, is obtained in relatively short zeolitisation time. After the calcination at 350-580 °C to remove the organic template, the obtained zeolite is used as a catalyst. The zeolite is atomised at 400-450 °C in a silica matrix as microspheres. The composition of amorphous gel and the reaction condition in the crystallisation, calcination and atomisation steps ensure that there is obtained an active and selective
15 catalyst for methanol conversion to olefins, with a granulation curve suitable in a fluidised bed process and with good thermal and mechanical resistance.

The process of methanol conversion to light olefins, mainly ethylene and propylene, is realised in a fluidised bed, including a reactor - regenerator system, with continuous circulation of the coked catalyst from reactor to regenerator and the regenerated
20 catalyst from regenerator to reactor. The methanol feed and regeneration air are injected into the dense bed of the catalyst. This system assures constant temperature in the catalyst beds and contact time of about two seconds. The reactor and regenerator risers only hinder the fluxes reversing. By steam or nitrogen purging in the risers, the catalyst is purified by the methanol and hydrocarbons adsorbed in the reactor and oxygen
25 adsorbed in regenerator. So the loss of methanol by burning in the regenerator is avoided. Also is avoided the burning of the methanol in the reactor by the oxygen adsorbed on the catalyst in the regenerator. The purging of the catalyst assures very low carbon oxides in the reaction products with a supplementary reduction of the costs for olefins separation. To keep constant the catalyst activity, a small amount is removed
30 from reactor or regenerator in parallel with adding an equal quantity of fresh catalyst. The reactor and regenerator have interior devices for taking-over the heat reactions of methanol conversion and coke burning.

The following examples illustrate, but do not limit, the present invention.

Examples

Example 1

By known method is prepared a tetraethylammonium phosphate aqueous solution 25%, from triethylamine, ethylbromide and phosphoric acid 73%.

Hydrated alumina 65% Al₂O₃ with 40% bayerite is suspended in demineralised water and is charged, with stirring, in a 3500 I autoclave over tetraethylammonium phosphate solution and then is added the 28% SiO₂ silica sol stabilised with ammonia. The pH of resulting suspension is fixed at 6.3-6.5 with phosphoric acid.

Molar ratio of the components in the suspension is P₂O₅: Al₂O₃: SiO₂: TEAOH 1:1.5:0.37:1.1.

Zeolitisation is carried out in six successive steps: the first step of the crystallisation begins with 15% of the whole suspension at 198-205 °C. After 20 hours the autoclave is cooled to 30-40 °C and a new quantity of suspension is added. The zeolitisation process is resumed under the same conditions. The operation is repeated five times. The entire zeolitisation process, including interstep cooling, is about 100 hours.

Analytical control, by XRD technique, of the product obtained shows more than 90% SAPO-34 zeolite and about 7% unreacted bayerite.

In table 1 are shown the characteristic bands in the XRD-spectrum of the SAPO-34 zeolite obtained, and SAPO-34 spectrum reported in US Patent No. 4440871, for comparison.

Table 1. Characteristic bands in XRD-spectrum (Cu lamp, Cu k_α = 1.5418)

US Patent No. 4440871			Sample, Example 1		
2 θ	d, Å	100 Xi/I _o	2 θ	d, Å	100 Xi/I _o
9.45-9.65	9.36-9.17	81-100	9.63	9.18	100
12.8-13.05	6.92-6.78	8-20	12.87	6.88	18
13.95-14.20	6.35-6.24	8-23	14.17	6.25	21
16.0-16.2	5.54-5.47	25-54	16.16	5.4	47

17.85-18.15	4.97-4.89	11-26	18.3	4.89	22
19	4.67	0-2	-	-	-
20.55-20.9	4.32-4.25	44-100	20.67	4.3	97
22.05-22.50	4.03-3.95	0-5	22.33	3.98	4
23.0-23.15	3.87-3.84	2-10	23.15	3.84	8
24.95-25.4	3.57-3.51	12-87	25.38	3.52	26
25.8-26.0	3.45-3.43	14-24	25.97	3.43	17
27.5-27.7	3.243-3.220	1-4	27.68	3.22	4
28.05-28.4	3.181-3.143	1-12	28.4	3.14	4
29.2-29.6	3.058-3.018	3-9	29.15	3.06	4
30.5-30.7	2.931-2.912	19-75	30.67	2.91	29
31.05-34.4	2.880-2.849	15-28	31.25	2.86	22
32.2-32.4	2.780-2.763	1-5	32.42	2.76	3
33.4-33.85	2.683-2.648	0-6	33.66	2.66	4
34.35-34.65	2.611-2.589	4-15	34.48	2.6	7
36.0-36.5	2.495-2.462	2-11	36.33	2.47	4
38.8-38.9	3.321-2.315	0-2	38.8	2.32	2
39.6-39.7	2.276-2.070	2-4	39.76	2.27	5

The crystallite dimensions are between 1 and 3 micrometer (Figure 1). The zeolite is stable with regard to calcination and in air steam, as shown in Figure 2.

In the zeolitisation phase there results a zeolite suspension with 16.7% solid which is separated with 6.7 l/m² hours filtration rate. After washing with demineralised water and
 5 air drying there results a paste of zeolite with 57% humidity.

The humid paste of zeolite is mixed with 28% SiO₂ Silica sol stabilised with ammonia in weight ratio zeolite: SiO₂ 60-40, fixed at pH 6.3 with nitric acid 40% and atomised under pressure with 400-450 °C hot air at entrance and 175-180 °C at exit. Injection pressure is 4-4.5 bars and the productivity of atomiser is 50 kg/hour dry catalyst.

5 Finally, the catalyst is calcined with a heating rate of 100 °C/min. at two constant level, three hours at 350-400 °C and ten hours at 580 °C. The cooling time of the catalyst is 4 hours. All the raw materials used for catalyst preparation have a Na content under 0.01%. The microspherical catalyst obtained has good flow property and granulation curve as shown in Figure 3.

10 Figure 4 is a schematic flow chart of the reaction - regeneration fluid bed catalyst system for methanol conversion to olefins, MTO process, part of invention. With reference to Figure 4, the reactor R1 is filled with 100 l catalyst and the regenerator R2 with 30 l catalyst. By fluidisation the dense fluid bed catalyst 2-2' and upper interface 3-3' takes form. The temperature of the dense fluid bed catalyst in R1 is fixed at 440°C
 15 and in R2 at 480-610 °C. The temperature in R1 and R2 are controlled by circulating a heating - cooling agent in interior heat exchangers 9-9'. Methanol and regenerating air are fed through connection 5-5' and sieves 4-4' with 100 l/hours, respectively 1000 NI/hour.

The circulation of the catalyst between reactor and regenerator is realised by nitrogen as
 20 lift gas through transfer lines 12-12'. Automatic control level of catalyst bed in R1 and R2 is achieved by keeping constant the pressure drops with regulators 11-11', which act the catalyst flow rate regulating valves 10-10'. Reaction products with catalyst entrained are evacuated at the top of R1 and R2 and separated in cyclone systems 6-7 and 6'-7'.
 25 Through the conduits 8-8' the entrained catalyst is recycled in the reactor-regenerator reaction zone. About 2 kg catalyst is withdrawn from the bottom of separation cyclone 7 or 7' in each 48 hours and is replaced with the same quantity of fresh catalyst through the charge device 15 or 15'. So the irreversible deactivation of the catalyst is compensated.

The coked catalyst in the conduit 13 has 4.3 wt.% coke and the regenerated catalyst in
 30 conduit 13' has a coke retention level of 1.7 wt.%. Reaction products after the exit from cyclone 7 are cooled in heat exchanger 14 and separated in the separation vessel 16 into a non-condensed hydrocarbon fraction and a liquid fraction which contains the process water, dimethylether and unconverted methanol. The gaseous hydrocarbon fraction is sent to a conventional olefins separation unit. From the liquid fraction is separated by
 35 distillation dimethylether and methanol which are recycled to the reactors R₁.

Regeneration gases after cooling in heat exchanger 16' and washing in the column 14' are evacuated into the atmosphere.

The composition of the fluxes is shown in Table 2.

Table 2. Effluent composition obtained in Example 1.

5

Component	Non-condensed organic phase vessel 16 wt.%	Liquid phase vessel 16 wt.%	Regeneration gases, exit cyclone 7' wt.%
Oxygen	-	-	1,5
Nitrogen	-	-	82.7
Carbon monoxide	-	-	4.3
Carbon dioxide	-	-	11
Hydrogen	0.2	-	0.5
Methane	1.6	-	-
Ethane	0.3	-	-

Component	Uncondensed organic phase vessel 16 wt. %	Liquid phase vessel 16 wt. %	Regeneration gases, exit cyclone 7' wt. %
Ethylene	46.8	-	-
Propane	2.5	-	-
Propylene	40	-	-
Butanes	0.53	-	-
1-Butene	1.74	-	-
iso-Butene	0.71	-	-
2-Butenes	4.04	-	-
C ₅ + hydrocarbons	1.58	-	-
Methanol	-	0.5	-
Dimethylether	-	-	-
Water	-	99.5	-

Example 2

Using the catalyst and installation of Example 1 by temperature and space velocity modification the ethylene/propylene ratio is changed in relatively large limits of 0.69-1.29.

Some illustrating results are shown in Table 3.

Table 3. Reaction products compositions, in different reaction conditions.

Experience number	1	2	3	4	5	6
a. Reaction conditions						
Temperature, °C	400	405	410	435	470	490
LHSV, h ⁻¹	1.1	0.6	1	1.9	1.5	2.7
b. Uncondensed organic phase analysis, wt. %						
Hydrogen	0.08	0.23	0.13	0.12	0.71	0.1
Carbon oxydes	-	-	-	0.14	0.21	-
Methane	0.62	1.78	0.81	1.12	2.42	0.91
Ethane	0.26	0.9	0.42	0.64	-	-
Ethylene	28.3	48.5	34.1	43	42.45	36.8
Propane	2.3	5.3	2.4	2.43	5.9	2.27
Propylene	41.2	35.7	42.9	42	32.95	45.4
Butanes	0.9	0.88	0.8	0.68	0.8	0.72
1-Butene	1.94	1.19	1.75	1.63	1.94	1.7
iso-Butene	0.77	1.23	0.75	0.44	1.04	0.2
2-Butenes	7.22	2.56	6.68	4.87	7.13	5.2

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1. Brent M.Lok, Celeste M. Messina, Robert L. Pation, Richard T. Gajek, Thomas R. Cannan, Edith M. Flanigen (Union Carbide Corporation), U.S. Patent No. 4440871 (April 3, 1984), Int. Cl. B01 j 27/14; U.S. Cl 502/241.
2. Ajit V. Sapre (Mobil Oil Corporation), U.S. Patent No.4590320 (May 20, 1986), Int. Cl. Co7C1/20; U.S. Cl.585/324; 585/315
3. Nicholas Davidiuk, James Haddad (Mobil Oil Corporation) U.S. Patent No. 4328384 (May 4, 1982), Int. Cl. C 97c1/20, U.S. Cl. 585/469; 585/639; 585/733.

Experience number	1	2	3	4	5	6
C5 + Hydrocarbons	2.41	0.95	2.84	1.58	4.45	1.75
Dimethylether	14	0.78	6.42	1.35	-	4.95
c. Liquid phase analysis wt.%						
Dimethylether	1.5	-	0.5	-	-	-
Methanol	12	3	9	1.5	0.1	1.5
Water	86.5	97	90.5	98.5	99.9	98.5
d. Coke deposits on the catalyst, wt.%						
Reactor R1 exit	4.9	5	4.8	4.5	4.6	4.7
Regenerator R2 exit	1.2	2.8	0.6	0.9	1.2	1.9
e. Ethylene/propylene ratio	0.69	1.36	0.79	1.02	1.29	0.81
f. Methanol conversion	93.3	98.3	95	99.2	100	99.2

The above description of the prior art is not intended to be, nor should it be interpreted as, an indication of the common general knowledge pertaining to the invention, but rather to assist the person skilled in the art in understanding the developmental process which lead to the invention.

Throughout the specification and claims the word “comprise” and its derivatives are intended to have an inclusive rather than exclusive meaning unless the context requires otherwise.

Claims

1. A method for the preparation of light olefins from methanol conversion using a microspherical catalyst including SAPO-34 zeolite, the method including the step of contacting methanol with a fluidised or moving bed of said catalyst wherein said
5 SAPO-34 zeolite is prepared from alumina, silica and a preformed template tetraethylammonium phosphate.
2. A method according to claim 1 including the further step of:

continuously regenerating the catalyst of said bed with air, and

substituting a portion of used catalyst with fresh catalyst in order to keep
10 constant the activity and selectivity of the catalyst.
3. A method according to claim 2, in which the regeneration with air is performed at 480-610°C.
4. A method according to claim 2 or claim 3, in which the contacting and the
15 regeneration steps are performed in a reactor and regenerator respectively with continuous circulation of the catalyst between said reactor and said regenerator.
5. A method according to any one of the preceding claims, in which methanol is contacted with said catalyst at a temperature of 400-490°C and liquid space velocities of 0.6 to 2.7 h⁻¹.
6. A method according to claim 1, in which the preformed template
20 tetraethylammonium phosphate is prepared from triethylamine, ethylbromide and phosphoric acid.
7. A method according to claim 1, in which the microspherical catalyst is obtained by the steps of:

25 atomising a mixture of said SAPO-34 zeolite and silica under pressure and at 400-450°C for form microspheres, and

calcinating said microspheres at 350-580°C to remove the template to obtain said microspherical catalyst.
8. A method for preparation of light olefins from methanol conversion substantially as herein described with reference to Example 1 or Example 2 and/or Figure 4.

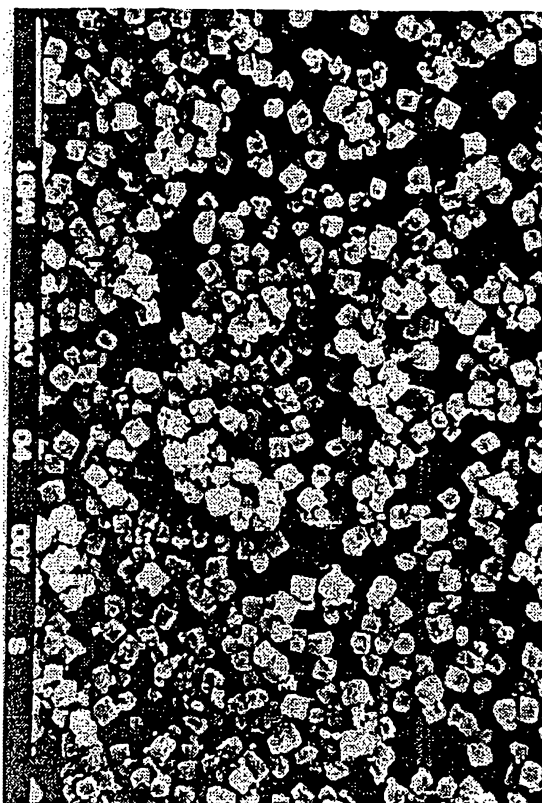


Figure 1. SEM microphotography of Exemple 1 zeolite

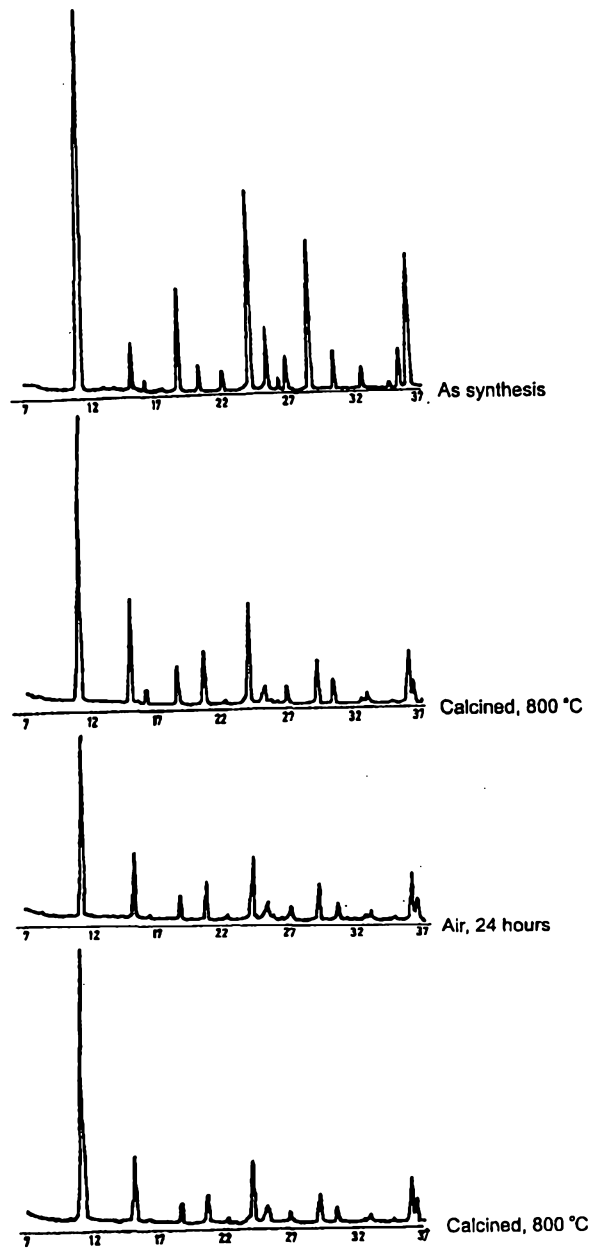


Figure 2. XRD-spectra of Exemple 1 sample

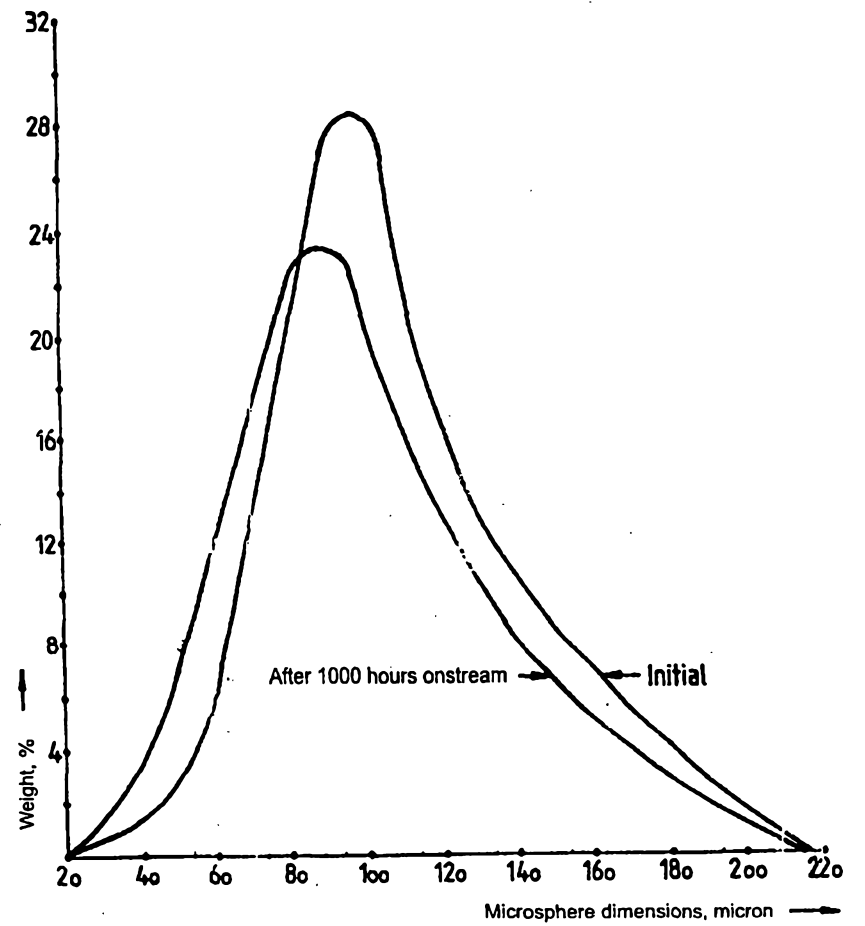


Figure 3. Dimension distribution curves of the catalyst, Exemple 1

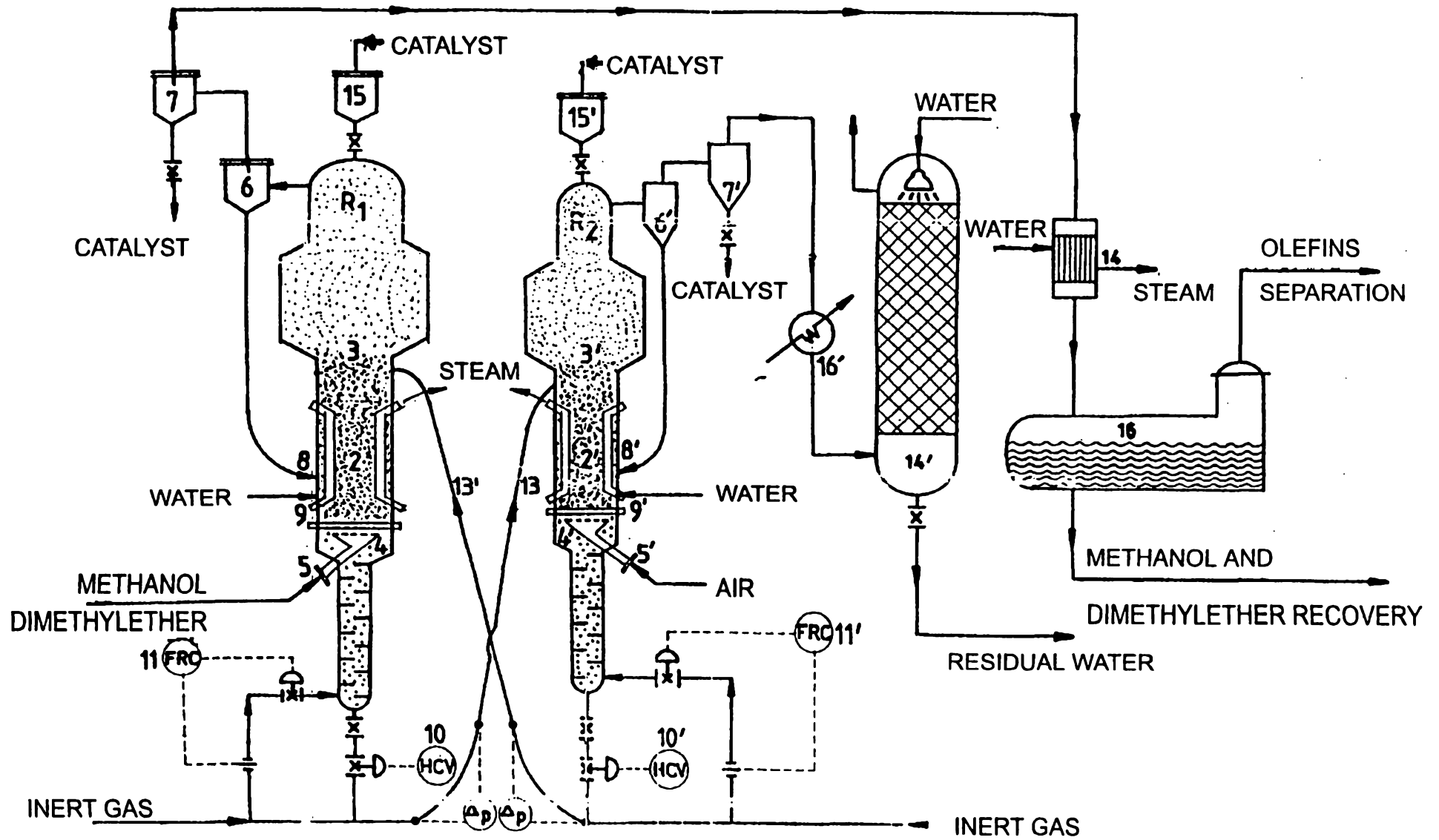


Figure 4. Installation for methanol conversion to olefins