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G. R. Landolt[a]; G. T. Kerr[b]
[a] Process Research and Technical Service Division, Mobil Research and Development Corporation, Paulsboro, New Jersey [b] Central Research Division, Mobil Research and Development Corporation, Princeton, New Jersey

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METHODS OF SEPARATION AND PURIFICATION USING THE MOLECULAR SIEVING PROPERTIES OF ZEOLITES

G. R. Landolt
Process Research and Technical Service Division
Mobil Research and Development Corporation
Paulsboro, New Jersey 08066

and

G. T. Kerr
Central Research Division
Mobil Research and Development Corporation
Princeton, New Jersey 08540

I. Introduction

During the past ten years many natural zeolites have been produced synthetically in the laboratory, along with many new zeolites not found in nature. With the introduction of synthetic zeolite A[1] and zeolite X[2] by the Linde Company, the uses of molecular sieves has grown exponentially, particularly in the petroleum refining and petrochemical fields. While the main use of zeolites has been in the field of catalysis, much has been accomplished in separations and purifications. Many unique processes have been introduced in adsorption, separation, and treating which are definite advances in the field.

Several excellent reviews of the use of molecular sieves in purification[3], treating[4], and separation[5] have been presented. Here the authors will attempt to present a combined picture of the use of molecular sieves in separations and purifications along with an introduction to zeolites -- their preparation, structure, and molecular sieving properties.

A. Delineation of Coverage

The uses of molecular sieves which could possibly be included in this review are so manifold that a thorough coverage would require a very extensive treatise. Much of the fringe-area material will not be discussed in detail, but will be mentioned where appropriate. These include preparation techniques, structural considerations, molecular sieving properties and processes using molecular sieves. For persons interested in the field, the authors have included an extensive bibliography of literature and patent references (Appendix).

II. Zeolites - History and Definition

A. History

Naturally occurring zeolites have been known for well over 100 years, and the crystal structure analyses have been completed on about one-half of the presently known natural zeolites. Early findings by Weigel and Steinhoff[6] on the adsorption of water, methyl and ethyl alcohols, and formic acid in the natural zeolite, chabazite, and the exclusion of acetone, ether and benzene were recognized by McBain[7] who first used the term "molecular sieve." In 1938, Barrer[8] pointed out the molecular sieving properties of these zeolites and their uses in the separation of polar and non-polar gases. However, it was not until the disclosure of synthetic zeolites by the Linde Company that zeolites came into prominence. During the past ten years, a flurry of activity took place in the use of these synthetic zeolites, particularly for separations and purifications, resulting in the many unique processes used today. With the introduction of Durabead 5 molecular sieve cracking catalysts[9], further activity took place in the field of catalysis. Even today, investi-

TABLE I

Classification and Crystallographic Data of Zeolites

Species	Idealised unit cell Contents
Analcime group	
Analcime	$Na_{16}[Al_{16}Si_{32}O_{96}],16H_2O$
Natrolite group	
Natrolite	$Na_{16}[Al_{16}Si_{24}O_{80}],16H_2O$
Thomsonite	$Na_4Ca_8[Al_{20}Si_{20}O_{80}],24H_2O$
Edingtonite	$Ba_2[Al_4Si_6O_{20}],8H_2O$
Chabazite Group	
Gmelinite	$Na_8[Al_8Si_{16}O_{48}],24H_2O$
Chabazite	$Ca_2[Al_4Si_8O_{24}],13H_2O$
Erionite	$(Ca\ etc.)_{4.5}[Al_9Si_{27}O_{72}],27H_2O$
Levynite	$Ca_3[Al_6Si_{12}O_{36}],18H_2O$
Cancrinite hydrate	$Na_6[Al_6Si_6O_{24}],5H_2O$
Sodalite hydrate	$Na_6[Al_6Si_6O_{24}],4H_2O$
Phillipsite group	
Phillipsite	$(K,Na)_{10}[Al_{10}Si_{22}O_{64}],20H_2O$
Gismondite	$Ca_4[Al_8Si_8O_{32}],16H_2O$
Barrer's PI	$Na_6[Al_6Si_{10}O_{32}],15H_2O$
Heulandite group	
Brewsterite	$(Sr,Ba,Ca)_2[Al_4Si_{12}O_{32}],10H_2O$
Heulandite	$Ca_4[Al_8Si_{28}O_{72}],24H_2O$
Stilbite	$Na_2Ca_4[Al_{10}Si_{26}O_{72}],28H_2O$
Mordenite group	
Mordenite	$Na_8[Al_8Si_{40}O_{96}],24H_2O$
Dachiardite	$Na_5[Al_5Si_{19}O_{48}],12H_2O$
Epistilbite	$Ca_3[Al_6Si_{18}O_{48}],16H_2O$
Ferrierite	$Na_2Mg_2[Al_6Si_{30}O_{72}],18H_2O$
Bikitaite	$Li_2[Al_2Si_4O_{12}],2H_2O$
Faujasite group	
Faujasite	$(Na_2,Ca)_{32}[Al_{64}Si_{128}O_{384}],256H_2O$
Linde A	$Na_{96}[Al_{96}Si_{96}O_{384}],216H_2O$
ZK-5	$Na_{24}[Al_{24}Si_{72}O_{192}],90H_2O$
Paulingite	$(K_2,Ca,Na_2)_{152}[Al_{152}Si_{520}O_{672}],\sim 700H_2O$

Crystal data (with unit cell constants in Å)				Isostructural species
cubic Ia3d	$a = 13 \cdot 7$			Wairakite, leucite, pollucite, viseite, kehoite
orthorhomb, Fdd2	$a = 18 \cdot 30$	$b = 18 \cdot 63$	$c = 6 \cdot 60$	Mesolite, scolecite
orthorhomb. Pnn2	$a = 13 \cdot 07$	$b = 13 \cdot 08$	$c = 13 \cdot 18$	Gonnardite
orthorhomb. $P2_12_12$	$a = 9 \cdot 54$	$b = 9 \cdot 65$	$c = 6 \cdot 50$	
(pseudotetrag. $P42_1m$)				
hexagonal $P6_3/mmc$	$a = 13 \cdot 75$	$c = 10 \cdot 05$		
trigonal R3m	$a = 9 \cdot 44$	$\alpha = 94^\circ 28'$		
(hexagonal	$a = 13 \cdot 78$	$c = 15 \cdot 06$)		
hexagonal $P6_3/mmc$	$a = 13 \cdot 26$	$c = 15 \cdot 12$		Linde T
trigonal R3m	$a = 10 \cdot 75$	$\alpha = 76^\circ 25'$		
(hexagonal	$a = 13 \cdot 32$	$c = 22 \cdot 51$)		
hexagonal $P6_32$ (?)	$a = 12 \cdot 7$	$c = 5 \cdot 15$		
cubic $P\bar{4}3n$	$a = 8 \cdot 88$			Nosean, Zhdanov's G, danalite, tugtupite, (synthetic Ca-alumino-sulphate)
orthorhombic B2mb	$a = 9 \cdot 96$	$b = 14 \cdot 25$	$c = 14 \cdot 25$	Harmotome
monoclinic $P2_1/c$	$a = 10 \cdot 02$	$b = 10 \cdot 62$ $\beta = 92^\circ 24'$	$c = 9 \cdot 84$	
cubic Im3m	$a = 10 \cdot 0$			Linde B, garronite (?)
monoclinic $P2_1/m$	$a = 6 \cdot 77$	$b = 17 \cdot 51$ $\beta = 94^\circ 18'$	$c = 7 \cdot 74$	
monoclinic Cm	$a = 17 \cdot 71$	$b = 17 \cdot 84$ $\beta = 116^\circ 20'$	$c = 7 \cdot 46$	Clinoptilolite
monoclinc C2/m	$a = 13 \cdot 64$	$b = 18 \cdot 24$ $\beta = 128^\circ 0'$	$c = 11 \cdot 27$	
orthorhomb. Cmcm	$a = 18 \cdot 13$	$b = 20 \cdot 49$	$c = 7 \cdot 52$	
monoclinic C2/m	$a = 18 \cdot 73$	$b = 7 \cdot 54$ $\beta = 107^\circ 54'$	$c = 10 \cdot 30$	
monoclinic C2/m	$a = 8 \cdot 92$	$b = 17 \cdot 73$ $\beta = 124^\circ 20'$	$c = 10 \cdot 21$	
orthorhomb. Immm	$a = 19 \cdot 16$	$b = 14 \cdot 13$	$c = 7 \cdot 49$	
monoclinic $P2_1$	$a = 8 \cdot 61$	$b = 4 \cdot 96$ $\beta = 114^\circ 26'$	$c = 7 \cdot 61$	
cubic Fd3m	$a = 24 \cdot 7$			Linde X, Linde Y
cubic Fm3	$a = 24 \cdot 64$			ZK-4
(pseudo cell: cubic Pm3m	$a = 12 \cdot 32$)			
cubic Im3m	$a = 18 \cdot 7$			
cubic Im3m	$a = 35 \cdot 1$			

gations in the separation and catalysis fields are continuing at a rapid pace.

B. Definition

A zeolite, more commonly known as a molecular sieve, is a hydrous crystalline aluminosilicate having a tetrahedrally coordinated framework and capable of reversible hydration-dehydration, ion exchange, and the sorption of molecules having a smaller critical diameter than the uniform crystalline pores of the structure. From this definition, zeolites are readily distinguished from other crystalline aluminosilicates such as clays, feldspars, and mica.

III. Structural Considerations

Although some 57 varieties of naturally occurring and synthetic zeolites are known, detailed crystallographic structure of only about 25 have been identified. At present, seven groups of zeolites can be distinguished by structural considerations[10]. Meier designated these seven groups (Table 1) as the analcime, natrolite, chabazite, phillipsite, heulandite, mordenite and faujasite groups -- in which all members of a given group contain some common building unit. For example, the crystal structures of faujasite, Linde A, ZK-5 and paulingite all contain a truncated octrahedron (Sodalite cage) and/or a truncated cubooctrahedron[11] as their primary structural unit (Figure 1).

Some of the secondary building units of zeolite frameworks are shown in Table 2.

IV. General Properties

A. Formula

Zeolites can be represented by the general formula: $M_{\frac{1}{v}}^{}$ $(AlO_2.x\ SiO_2).y\ H_2O$, where x is the ratio of silicon to aluminum and y is the moles of water of hydration. The number of equivalents of M (of valence

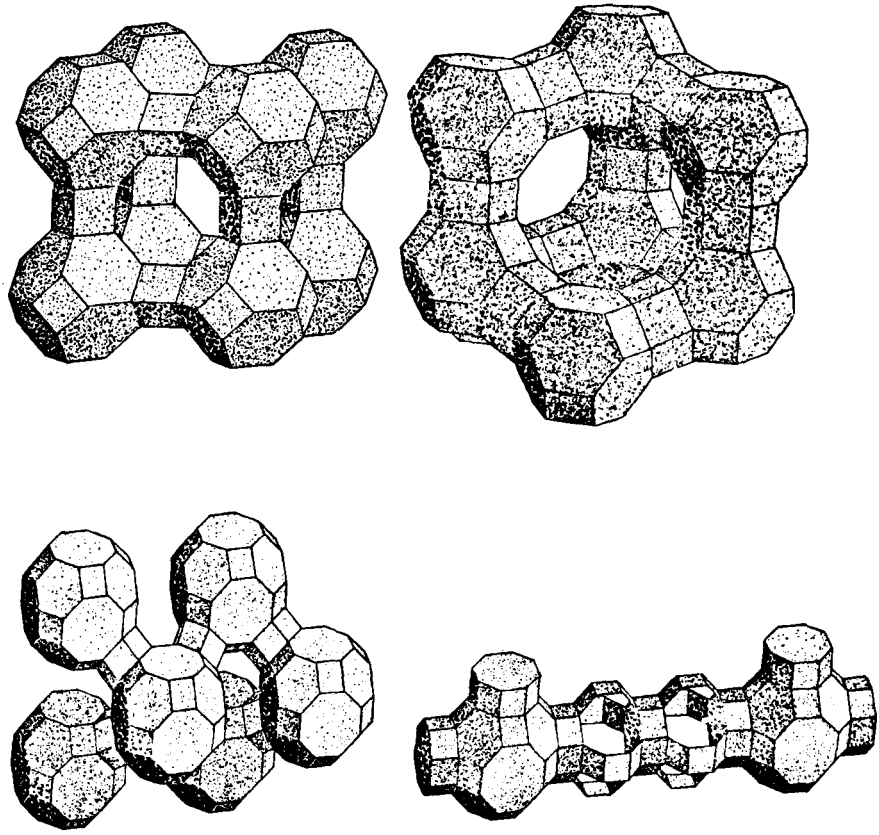

FIGURE 1

Arrangements of Truncated Octrahedra in the Framework Structure of Linde A (a) And Faujasite (b) And Of Truncated Cubo-Octrahedra In ZK-5 (c) And Paulingite (d)

v) cation is always equal to the number of moles of aluminum, and mixed cations can be present -- i.e., Na, K. Zeolites having the same crystal structure but varying silicon to aluminum ratios can be synthesized -- for example, zeolites X and Y have faujasite-type structures but different Si/Al values. Typical zeolite formulations are shown in Table 1.

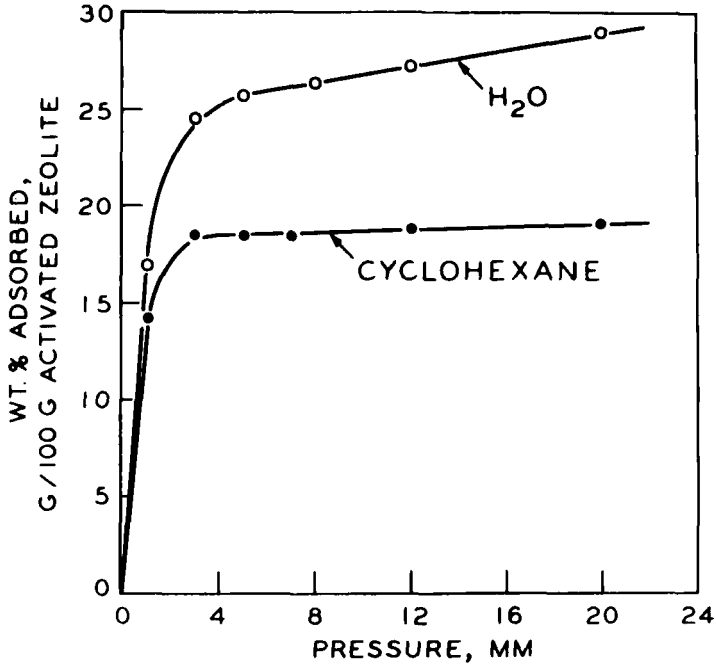

FIGURE 2

Adsorption Isotherms of Zeolite Y

B. Adsorptive Properties

Besides the elemental composition of the zeolites, also of interest are the physical properties of the zeolites in relation to their use in separation and purification processes. For example, one of the primary characteristics of a particular zeolite is its adsorptive properties. Complete adsorption isotherms are particularly helpful in determining equilibrium sorptive capacities, which can be useful in the evaluation of zeolites for specific separation and purification processes. Typical adsorption isotherms for Linde zeolite Y are shown in Figure 2.

Sorptive properties can be used to differentiate between large and small pore zeolites, as shown in

TABLE III

Effective Sorption Capacities of Molecular Sieves

	Amount Sorbed at 25 C (g/100 g Activated Sample)		
Zeolite	H_2O	n-C_6H_{14}	Cyclo-C_6
4A	24.5	-	-
5A	24.5	12.0	-
B	20.0	-	-
X	31.5	16.8	18.5
Y	28.0	16.5	19.0
Erionite	11.3	4.4	0.7
Offretite	16.6	8.6	5.3
Mordenite	13.3	6.1	7.3
Experimental Conditions		Temp. 25 C	
P (torr)	12	20	20
P/P_0	0.54	0.2	0.2
MCD (A)[a]	2.8	4.6	6.0

(a) Approx. molecular dimension measured with Courtauld space filling molecular models.

Table 3. Zeolite A, synthesized in the sodium form, is a relatively small pore zeolite with an effective pore diameter of about 4 Å. As expected, it sorbs water but excludes both normal hexane and cyclohexane, whose mean critical diameters (4.6 Å and 6.0 Å, respectively) are too large to allow entry into the planar octagonal ports. Zeolite X, however, is a relatively large port zeolite with an effective pore diameter between 7 and 10 Å, and therefore sorbs all three liquids.

C. Thermal Properties

Another physical property of interest is the thermal activation (loss of the water of hydration) or thermal stability of the zeolite crystal structures.

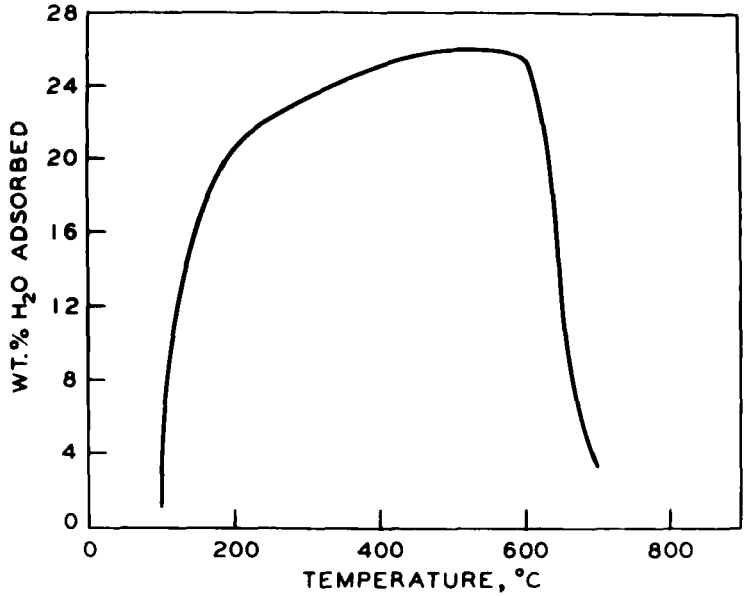

FIGURE 3

Thermal Stability of Zeolite A

For example, the sorptive capacity of zeolite A for water increases greatly after thermal activation from 100 to 300°C, then remains relatively constant, and finally decreases markedly above 600°C as the crystal structure collapses (Figure 3). Thermal stability would then be a prime prerequisite for high temperature adsorption-separation processes.

D. Effect of Ion Exchange

Ion exchange of the parent cations present in a zeolite with other cations can have a profound effect on the molecular sieving properties of the zeolite. For example, the effective pore diameter of zeolite A increases from about 4 to 5 Å as the parent sodium ions are replaced by divalent calcium ions. As expected, then, 4 A sieve sorbs water and low carbon number com-

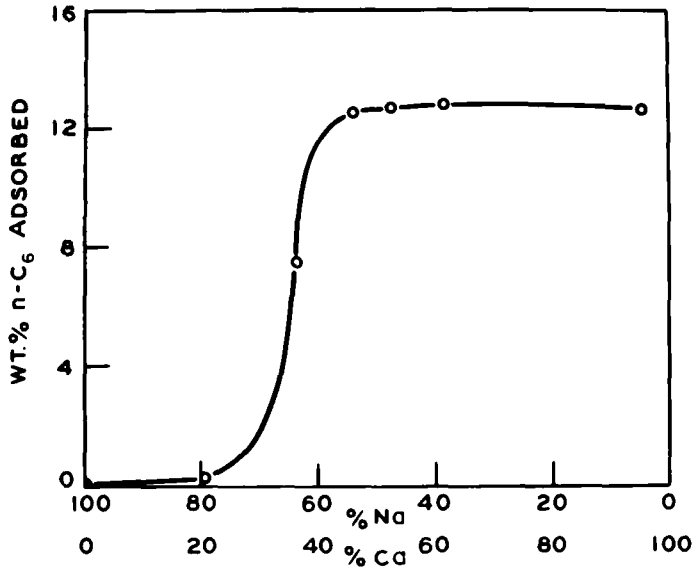

FIGURE 4

Effect of Ion Exchange on the Adsorption Properties of Linde Type A Zeolite

pounds such as methane and ethane while 5 A sieve sorbs higher molecular weight straight chain compounds such as n-hexane. Using n-hexane as a selected probe molecule, one can readily observe the change in the effective pore size in the zeolite as a function of ion content (Figure 4).

Molecular sieves can then be tailored to a certain degree by ion exchange to effect certain sorptive properties. The replacement of sodium ions in type X zeolite by the larger potassium ion has the effect of reducing the effective pore diameter (from 10 Å to about 7 Å). On the other hand, replacement of sodium ions in type Y zeolite by the smaller ammonium

ions which are subsequently reduced to hydrogen ions by calcination, increases the sorptive capacity of the zeolite.

V. Synthesis of Zeolites

A. Crystallization Techniques

The synthesis of zeolites is a field complete in itself, so only rudimentary and general procedures can be discussed herein. At least four components are necessary in the synthesis: a source of cations usually in the form of a strongly basic hydroxide, aluminate, silicate, and water. Mixed bases may also be present and the silicate and aluminate may be replaced with germanate and gallate. The tetrahedron required for the representation of the four-component systems can be examined by the use of ternary diagrams in which the fourth component - water - is held constant. Typical reaction compositions required for type A, X, Y, and sodalite in the Na_2O-SiO_2-Al_2O_3 system are shown in Figure 5. It is obvious that detailed exploration of the crystallization fields would require extensive and laborious experimentation. Indeed, some 1000-2000 experiments would be required in one four-component field to cover component variations as well as other variables, such as seeding, temperature, mixing variables, reaction time, etc. Typical crystallization fields used in the extensive study of hydrothermal synthesis by Barrer and co-workers[12] are shown in Figure 6. Even with the wealth of zeolite synthesis reported, large areas in the crystallization fields still remain to be explored.

These diagrams also include many metastable phases. For example, the metastable phase, after longer contact with the mother liquor, transforms into a more stable compound, i.e., type A will recrystallize

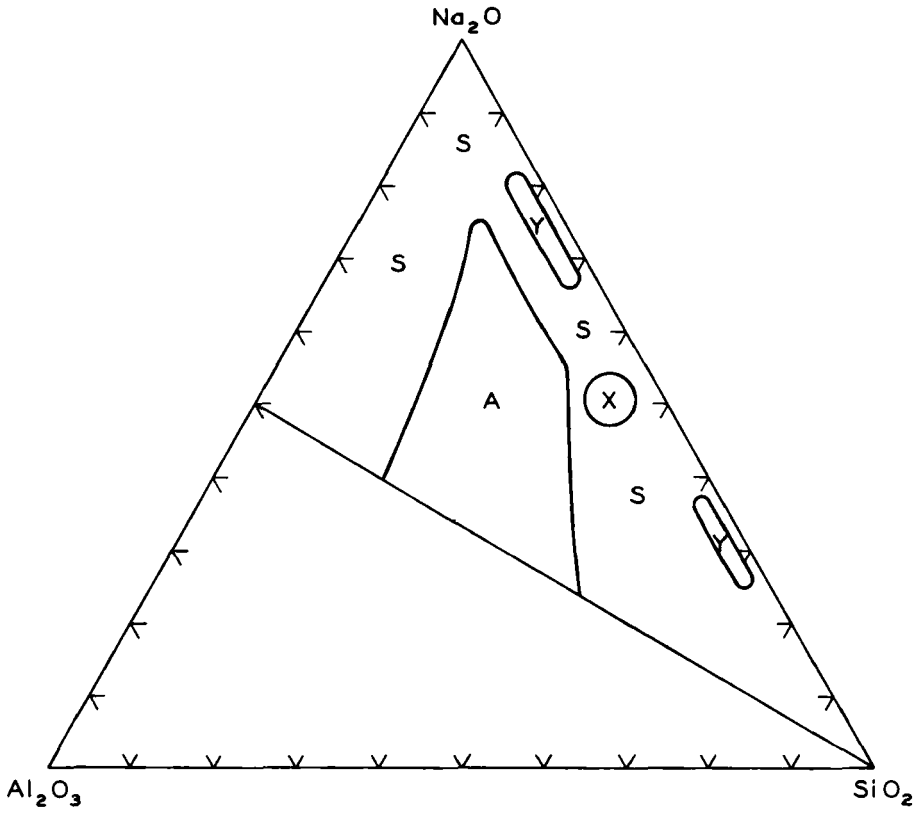

FIGURE 5

Reaction Compositions of Zeolite, A, X, Y, and Sodalite in the Na_2O - SiO_2-Al_2O_3 System

to sodalite and type X to phillipsite.

VI. Molecular Sieve Drying

A. Gas Drying

Molecular sieves provide an economical and efficient means of drying gases to dew points below -100°F and liquids to moisture contents less than 1 ppm. Two properties make molecular sieves among the most effective desiccants known: (1) they exert strong physical forces on molecules in their vicinity, and (2)

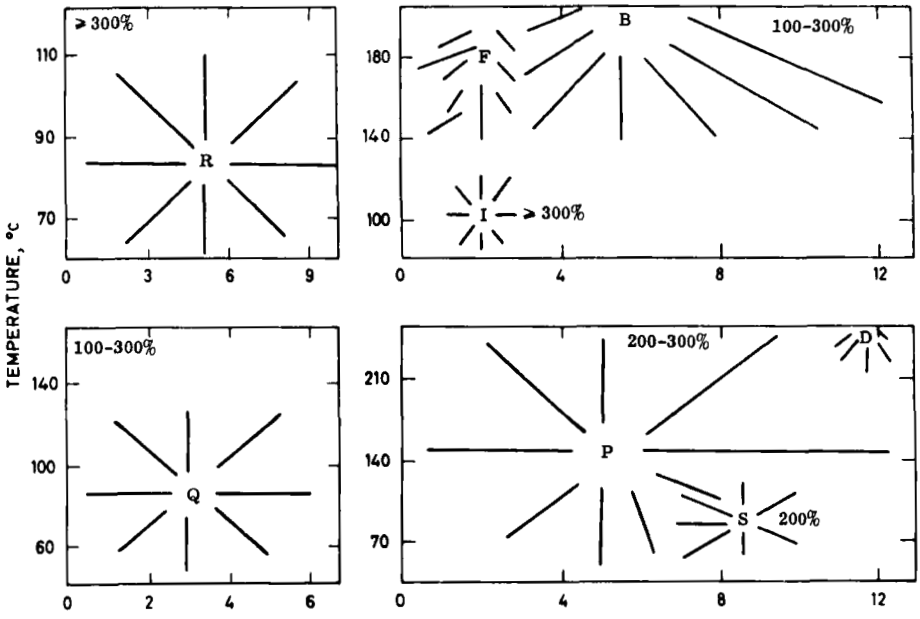

Fig. 6 Approximate areas of formation of some products of low-temperature crystallization of gels Na_2O, Al_2O_3, $nSiO_2$+ NaOH aq. The figures give the excess % of NaOH.

Na-Q	Synthetic Na-zeolite (Linde Sieve A)	$Na_2O,Al_2O_3,2SiO_2,4H_2O$
Na-B	Analcite	$Na_2O,Al_2O_3,4SiO_2,2H_2O$
Na-S	Near-chabazite or gmelinite	$Na_2O,Al_2O_3,4\cdot7SiO_2,6\cdot6H_2O$
Na-R	Near-faujasite (Linde Sieve X)	$Na_2O,Al_2O_3,2\cdot7SiO_2,6\cdot5H_2O$
Na-F	Basic nosean	$Na_2O,Al_2O_3,2SiO_2,xNaOH,yH_2O$
Na-I	Basic sodalite	$Na_2O,Al_2O_3,2\cdot1SiO_2,2NaOH,1\cdot5H_2O$
Na-P	Phases of phillipsite group	$Na_2O,Al_2O_3,3\cdot3SiO_2,4\cdot3H_2O$ to $Na_2O,Al_2O_3,5\cdot3SiO_2,5\cdot7H_2O$
Na-D	Mordenite	$Na_2O,Al_2O_3,10SiO_2,6\cdot7H_2O$

large molecules are excluded from the active sites. The cations in the crystal lattice act as strong positive charges, attracting the negative end of polar molecules. The more polar the molecule, the more strongly it will be adsorbed. This results in high capacity at low partial pressures (Figure 7) [13].

Although molecular sieves 4A, 5A and 13X have been used in gas drying processes, the preferred zeolite is 3A, a potassium-exchanged zeolite A, which restricts

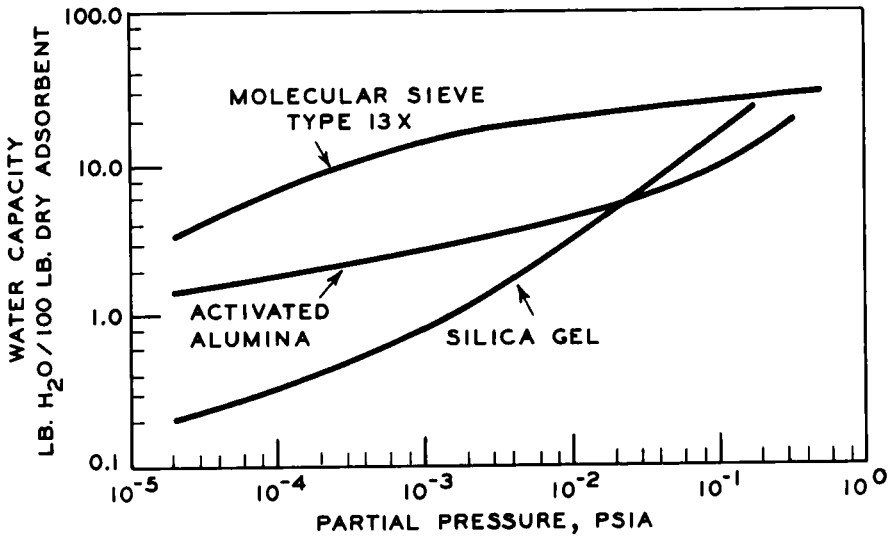

FIGURE 7

Gas Drying with Molecular Sieves Results in High Capacity at Low Partial Pressures

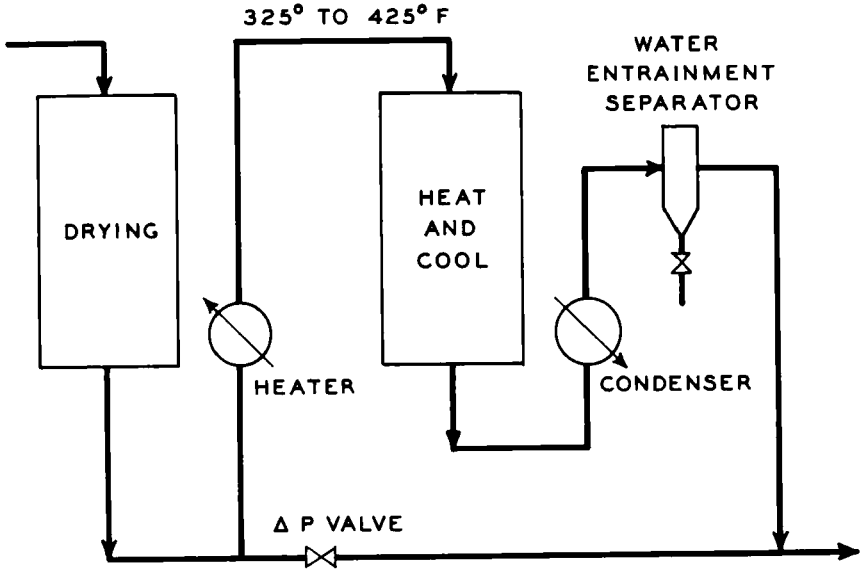

FIGURE 8

Modified Dehydrator

the effective pore size so that only water and inorganic gases are adsorbed. Water concentration as low as 35 ppb are obtainable over a wide range of operating conditions.

Numerous methods and processes are reported in the literature[14,15,16]. A modified dehydration process reported by Clark[17] is shown in Figure 8. Here the flow control valve is moved from the feed line as in conventional dehydration to the product line. The entire feed would pass through the dryer at full line pressure. The water content of the effluent would be approximately 0.1 lb/MMSCF. Approximately 10% of the total gas flow would be split off from the product gas, passed through a heater, raised to the bed temperature and passed down through the bed for regeneration. Using 4A sieve, design capacities in the range of 10-14 wt % are available.

For drying refinery off-gases, molecular sieves have been found superior to other desiccants, such as alumina. Cochrane[18] shows that effective drying is an essential process step where hydrogen-rich off gas from reformers is used as a raw material for the synthesis of anhydrous ammonia. Alumina containing varying amounts of molecular sieve has increased dryer capacity for conventional hydrogen dryers. Increase in capacity is almost directly proportional to the amount of molecular sieve used (Figure 9). Although sieves can increase the capacity of the dryers, they have no effect on the dew point of the effluent gas. Water content of the effluent gas is less than 10 ppm. Molecular sieves have proved superior to other dessicants where interference from aromatic and straight chain hydrocarbons in the gas stream is a problem.

The use of acid-resistant zeolites with approximately 4 Å pores (AW-500) has been used to dehydrate

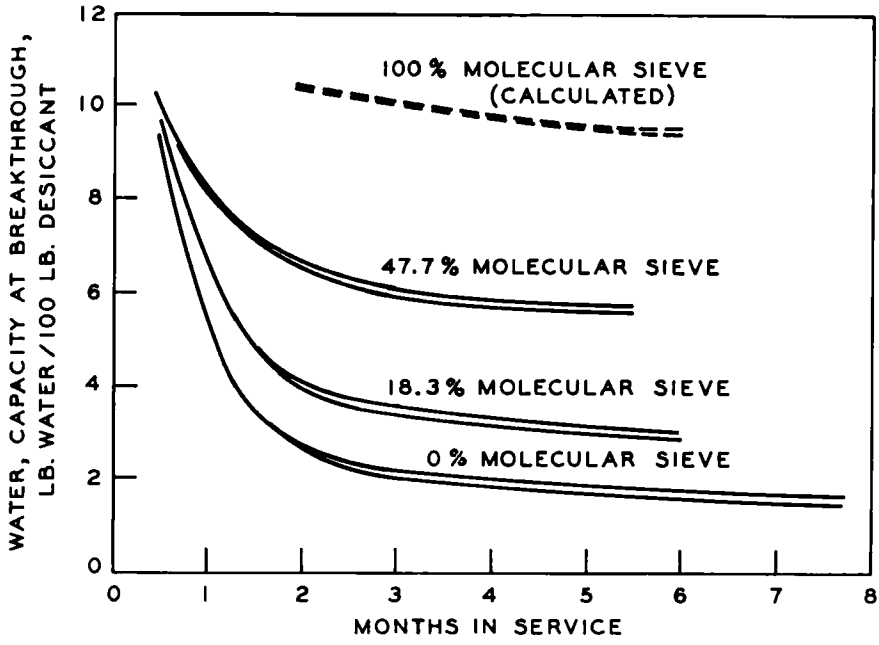

FIGURE 9

Molecular Sieve-Alumina Dryer Capacity Increases with Zeolite Content

high acid gas in field operations[19]. The process scheme in Figure 10 reduced the water content of the produced gas from 90 to 2.5 lb/MMCF or a dew point of 17°F. Field tests up to 3,150 regeneration cycles and the predicted capacity of the sieve desiccant are shown in Figure 11. A two-year service life has been achieved.

Pierce and Stieghan[20] have reported the use of 3 A sieve (potassium zeolite A) for dehydrating cracked gas streams and to prevent the polymerization of olefins in the feed stream on the desiccant. The desiccant activity of molecular sieve compared to silica gel and alumina is shown in Figure 12.

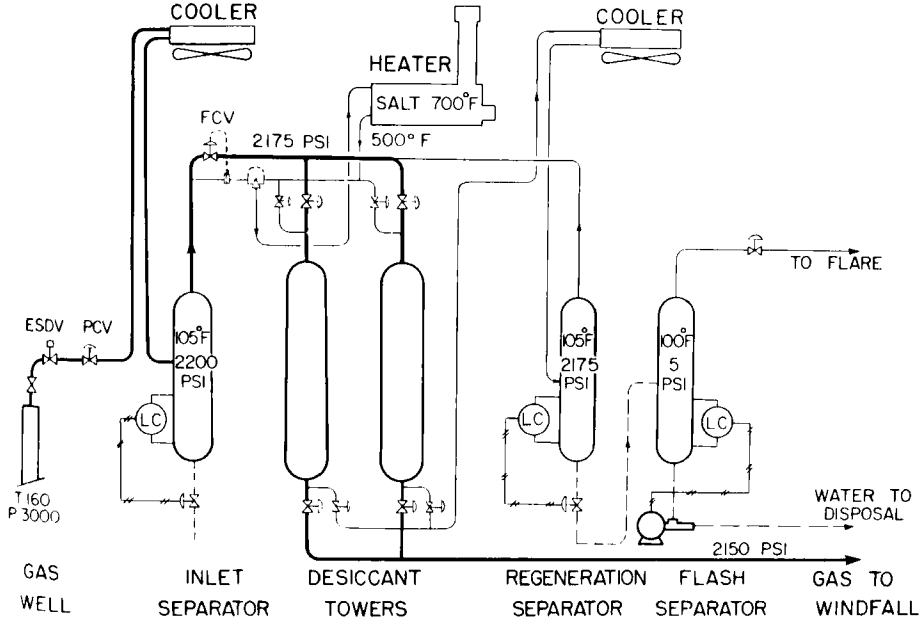

FIGURE 10

Dehydration of High Acid Gas

The small pore size of 3 A sieve precludes the adsorption of all olefins -- even ethylene.

The problem of CO_2 adsorption by the desiccant in propylene drying was solved by Sun Oil Company by using 3 A molecular sieve. McConnell[21] shows low CO_2 content in the propylene product even after the regeneration after 200-400 barrels of propylene had passed through the bed (Table 4).

The above examples exemplify the many uses of molecular sieves in gas drying. Today, virtually all gas chromatographic units, whether laboratory or commercial size, use molecular sieves to dry the carrier gas stream. Practically all the hydrogen produced from reforming operations is purified by molecular sieves be-

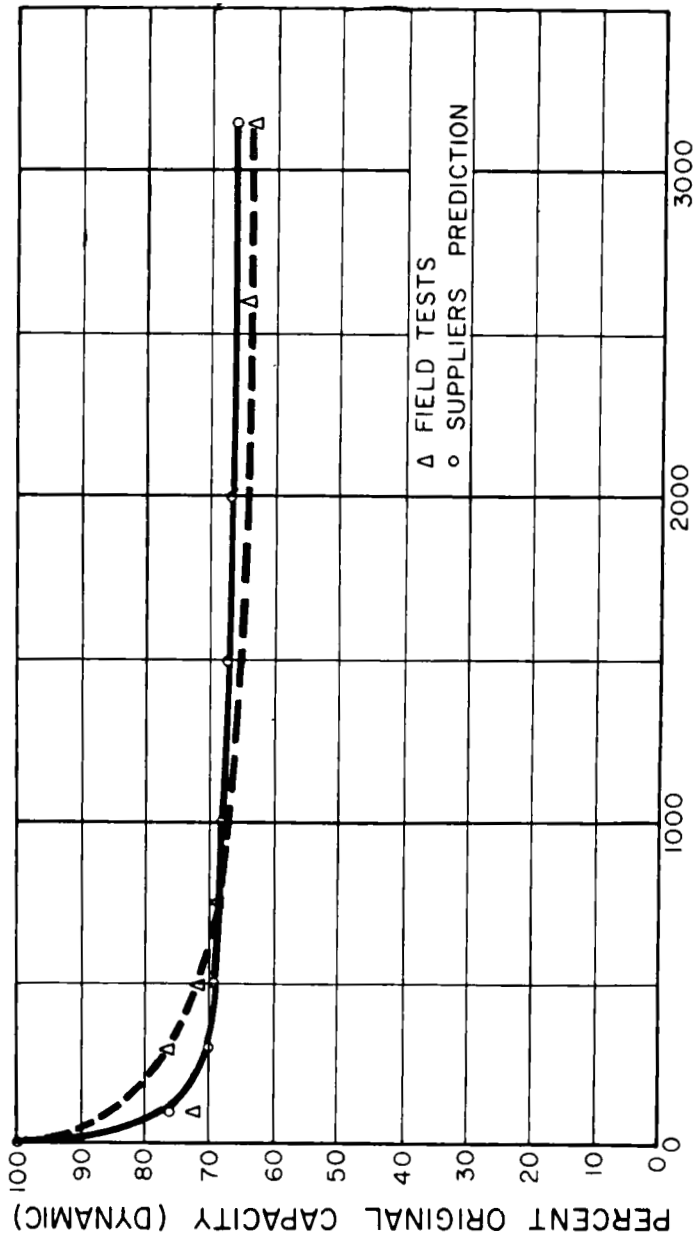

FIGURE 11

Performance of Acid-Resistant 4 Å Sieve

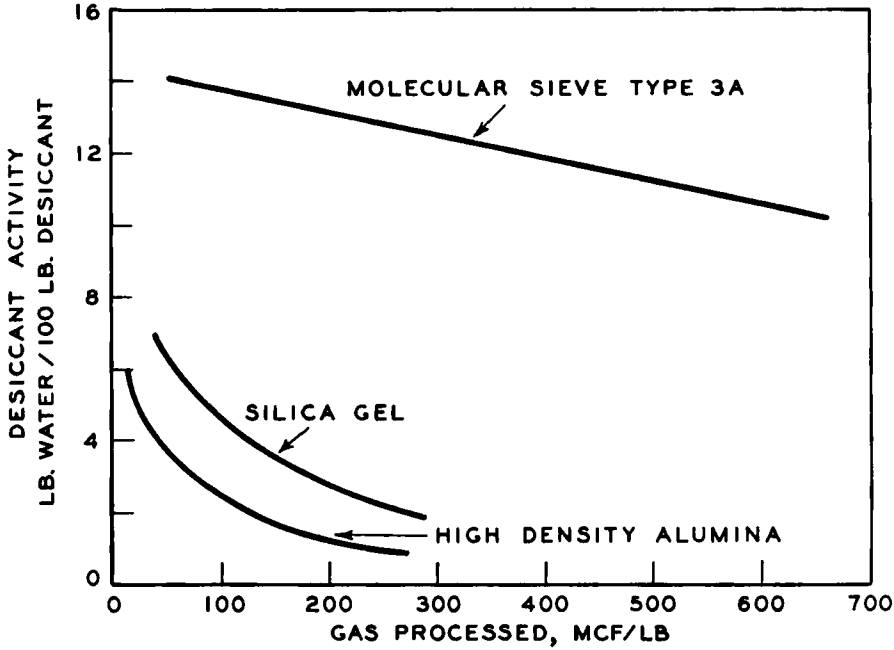

FIGURE 12

Desiccant Activity Versus Gas Processes

TABLE IV

A 3 ANGSTROM MOLECULAR SIEVE GAVE LOW CARBON DIOXIDE CONTENT IN THE PROPYLENE PRODUCT

Propylene Processed After Regeneration, Barrels	Carbon Dioxide in Propylene, ppm
1	3
30	2
45	1

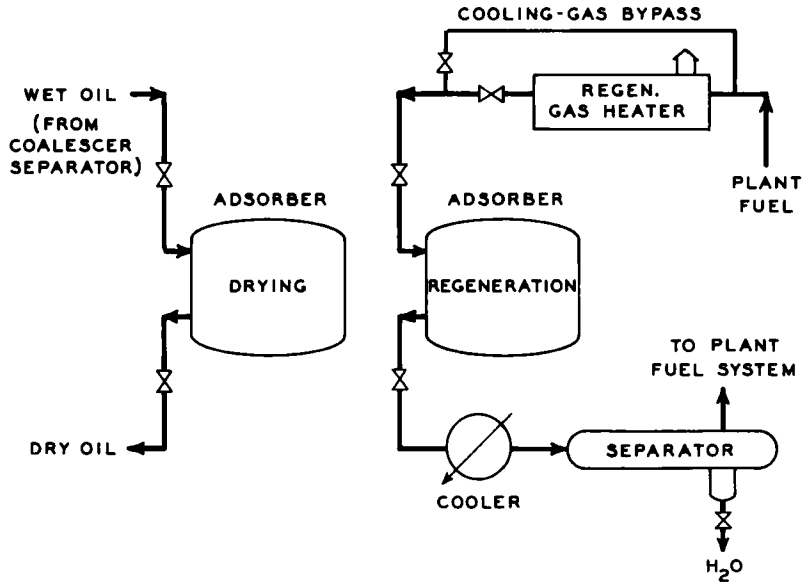

FIGURE 13

Processing Steps in Molecular Sieve Oil Dehydration

fore use as a process stream in other refinery operations, such as hydrocracking.

B. Liquid Drying

A molecular sieve dehydrator for drying adsorber oil normally used in low temperature oil-adsorption processes has been reported by Collins and Raidt[22]. A typical dual-bed dehydrator (Figure 13) provides lean adsorber oil with only 10-15 ppm water content. Dry lean oil produced by this method can be shown to give a greater hydrocarbon recovery from natural gas streams, i.e., more propane, butane, and ethane.

The superdrying of alkylation feeds for HF alkylation units has been reported by Collins[23]. Figure 14 shows a typical flow diagram for drying (<15 ppm) the olefin and isobutane feeds. Typical performance of the

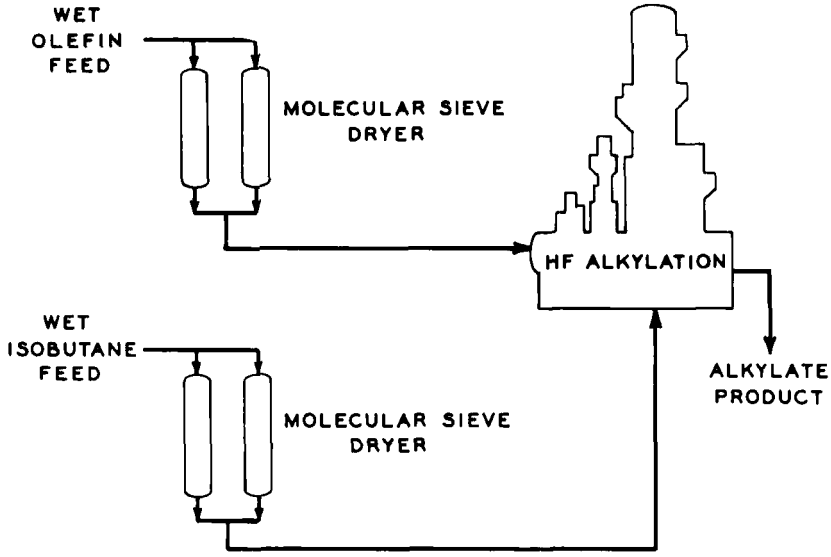

FIGURE 14

Flow Diagram Shows How Molecular Sieves
Dry Olefin Feed and Isobutane Makeup Stream

HF alkylation unit process are: less frequent catalyst regeneration, less corrosion and fouling, less acid consumption, and higher octane ratings of the product. Results of this molecular sieve drying (3 A sieve) are shown in Table 5.

In summary, molecular sieves can be used to dry many substances. For example, Gehrhart and Kyle[24] have reported the drying of benzyl alcohol by 5 A sieve. Some of the compounds that can be dried by molecular sieves are shown in Table 6[13].

VII. Liquid Phase Treating

The use of molecular sieves has become the standard approach to sweetening products from liquid propane and butane to natural gasolines. Conventional dehydration processes (Figure 15)[25] or variations

TABLE V

Commercial Performance of HF Alkylation Unit Using Molecular Sieve Feed Dryers

Water content of feed dryers, p.p.m. by wt.	100-450
Water content of dry effluents,, min. detectable, p.p.m.	<15 p.p.m.
Acid regeneration frequency, per month	1
Typical HF water content before acid regeneration, wt. %	1.2
Typical HF water content after acid refrigeration, wt. %	0.4

	Olefin feed	Isobutane makeup
Stream compositions:		
Propane, vol. %	7	8
Propylene, vol. %	15	...
i-Butane, vol. %	35	31
n-Butane, vol. %	12	60
i-Butylene, vol. %	12	...
Butene-2, vol. %	13	...
Pentanes, vol. %	5	1
Pentenes, vol. %	1	...
Sulfur compounds, p.p.m. by wt.	~100	~125

TABLE VI

Some Substances Which Can Be Dried by Molecular Sieves

Inorganic gases	Air, annealing gas, chlorine.
Hydrocarbons	Ethane, propane, propylene, *n*-butane, *iso*butane, butylene, butadiene, *iso*butylene, pentane, cyclohexane, *n*-heptane, isoprene, benzene, toluene, mixed xylenes.
Halogenated hydrocarbons	Methyl chloride, methylene chloride, carbon tetrachloride, ethylene dichloride, propylene dichloride, 2-ethylhexyl dichloride, *n*-butyl chloride, di- and mono-chlorodifluoromethanes.
Alcohols	Methanol, ethanol, *n*-propanol, ethylene glycol, 2-ethylhexanol.
Others	Dimethyl, diethyl and *iso*propyl ethers, amyl *n*-butylamine, 2-ethylhexylamine, dimethylformamide, acetone, acrylonitrile, pyridine.

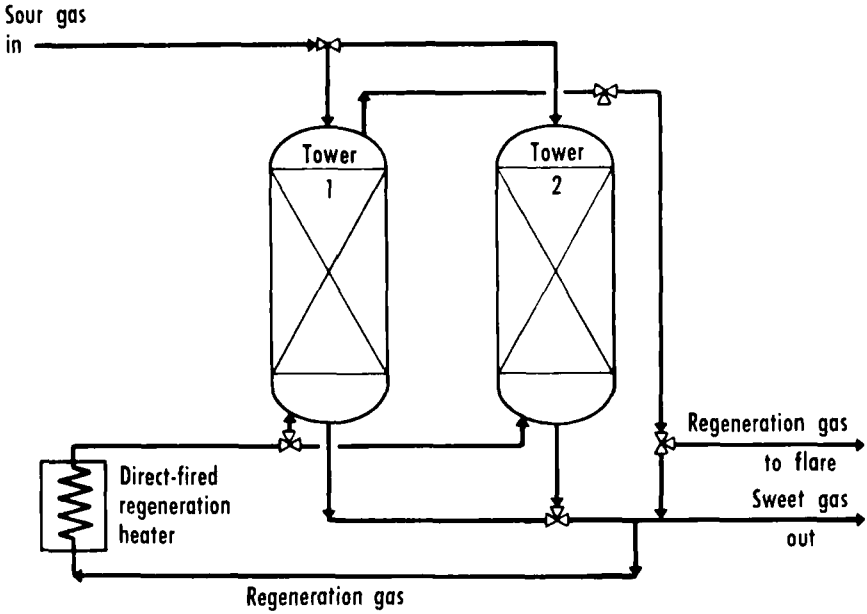

FIGURE 15

Typical Molecular-Sieve Sweetening

of the basic design, make a variety of sweetening processes available.

In the sweetening of liquid propane or butane, the products of such treatment are non-corrosive to copper strip, contain low total sulfur compounds, and are dry. Molecular sieve treatment provides all three advantages in a single step operation. Operation is simple and straight forward as in ordinary dehydration processes. In a typical operation the adsorption step is at ambient temperature with pressure high enough to prevent vaporization. The liquid flow is downward with the adsorbent bed full of liquid. On completion of the adsorption step, the liquid in the bed is drained, the pressure reduced, and the vaporized product sent to recovery or fuel. The adsorbent is regenerated with

TABLE VII

SULFUR SPECIFICATIONS

Natural-Gasoline Fraction	
C_3-C_4	3.0 ppm total sulfur (maximum or 0.3 grain/100 scf (maximum) and noncorrosive
iC_5	3.0 ppm mercaptan sulfur, doctor sweet and noncorrosive
nC_3 and Heavier	3.0 ppm mercaptan sulfur, doctor sweet and noncorrosive

a downward purge of hot, dry, sweet gas. The cycle is then repeated by the introduction of the liquid feed to the top of the bed.

In many cases molecular sieves can be substituted in the dehydrator of existing scrubbing-desiccant purification units. Operating costs are almost entirely the replacement cost of the sieve, which has at least a two-year life span. Today, over half of the propane produced is treated by molecular sieves. There are more than fifty light hydrocarbon sweeteners in operation today. Most of these units use 3 A, 4 A, or 5 A molecular sieves depending on the needs of the installation.

Sweetening of natural gasoline fractions by cyclic adsorption and regeneration can be accomplished using 13X molecular sieves[26]. Efficient desulfurization is necessary to enable the use of natural gasolines in motor gasolines of higher octane numbers and as charge stocks for catalytic conversion processes. Sulfur specifications for natural gasoline fractions are shown in Table 7.

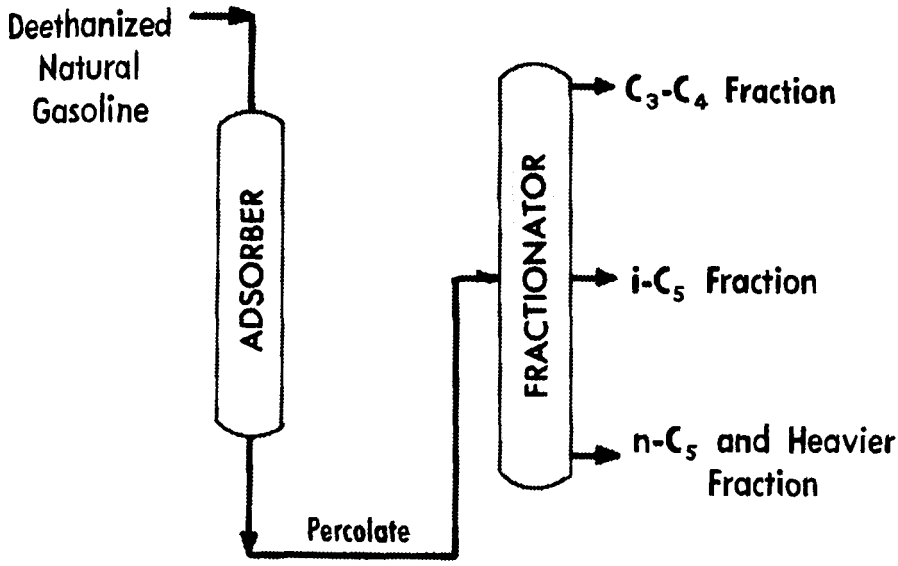

FIGURE 16

Flow Pattern for Sweetening Processes

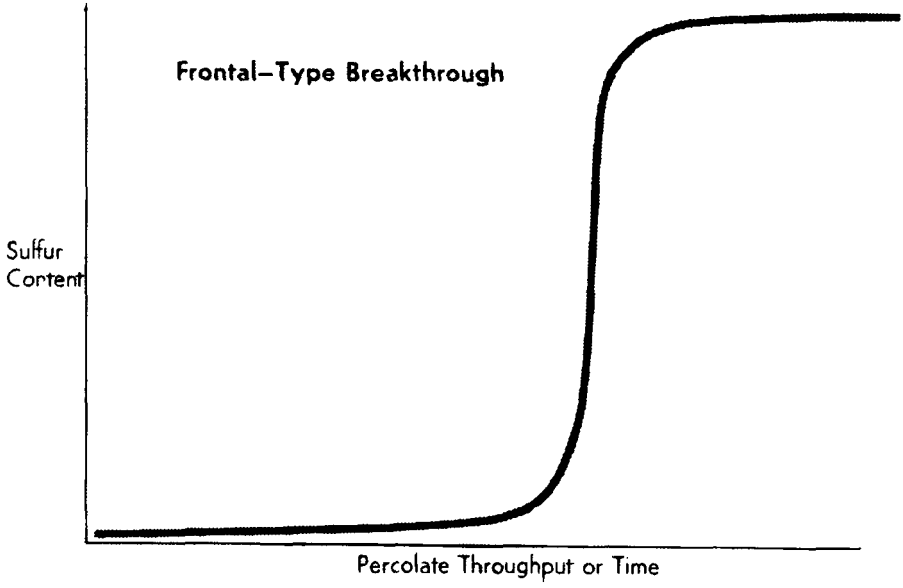

FIGURE 17

Frontal-Type Breakthrough is Must if Sulfur Compounds are to be Removed to Meet Stringent Sulfur Specifications

TABLE VIII

Composition of Natural Gasolines

Component—	% by Weight of Gasoline	Sulfur Content: p.p.m. Total	Sulfur Content: p.p.m. Mercaptan
Gasoline A		885	746
C_3–C_4	53.7	450	450
iC_5	23.2	1,062	918
nC_5 and heavier	23.0	1,717	1,219
Gasoline B		2,265	970
C_3–C_4	55.1	1,500	768
iC_5	23.3	1,800	1,500
nC_5 and heavier	21.6	4,700	900

TABLE IX

TYPICAL EFFLUENT SULFUR DATA

Adsorption: 100°F, 350 psig, Liquid Weight Hourly Space Velocity (LWHSV) – 1.68

Column: 2.1 Inches I.D. x 3.4 Feet Long – 1/8 Inch Pellets Molecular Sieves

Fraction	Instantaneous Throughput: Gallons of Gasoline/ Pound of Sieves	Sulfur Content: C_3-C_4 Total	Sulfur Content: C_3-C_4 Mercaptan	Sulfur Content: iC_5 Total	Sulfur Content: iC_5 Mercaptan	Sulfur Content: nC_5 Plus Total	Sulfur Content: nC_5 Plus Mercaptan
Feed B		1500	768	1800	1500	4700	900
Effluent	1.0	< 1	< 1	1.0	< 1	1.2	< 1
Effluent	2.0	< 1	< 1	1.3	< 1	1.5	< 1
Effluent	2.5	< 1	< 1	1.5	< 1	2.0	< 1
Effluent	3.0	< 1	< 1	3.5	< 1	10.0	< 1
Effluent	3.5	1.0	1.0	7.0	1.0	120	10.0
Effluent	4.0	3.0	3.0	20	5.8	350	43
Effluent	5.0	3.0	5.0	400	65	1800	190

Using a process scheme shown in Figure 16, deethanized gasolines are passed through a bed of 13X sieve pellets to remove the sulfur compounds. Since the sulfur specifications are low, the design and operation of the adsorption phase must give a frontal-type breakthrough for the sulfur compounds (Figure 17). This assures highest purity, as well as maximum adsorbent life. Typical gasoline compositions and effluent sulfur data are shown in Tables 8, 9.

VIII. Commercial Processes

A. The IsoSiv Process for Upgrading Gasoline

The Union Carbide Isosiv process presents certain advantages as a tool, either by itself or in combination with isomerization, for upgrading gasolines. Using Linde 5 A molecular sieve as the adsorbent, only normal paraffins are adsorbed from mixtures of normal paraffins, isoparaffins, aromatics and naphthenes. The process is a vapor-phase, isothermal, rapid-cycle, pressure-swing, fixed-bed adsorption process as shown in Figure 18[27]. Using a light naphtha feedstock, flow continues until the adsorption front reaches some predetermined point in the bed short of the breakthrough of the normal paraffins. The isomer gasoline is condensed, cooled and pumped to storage. The adsorber is then taken off stream, partially depressurized, and the bed desorbed by vacuum countercurrent to the feed step. As the pressure drops, the normal paraffins are desorbed, condensed and pumped to storage. The adsorber then is repressurized and put back on the feedstock stream. With two or more adsorbers, the continuous operation is achieved. Isosiv is extremely versatile and can handle feedstocks from natural gasoline through gas oil fractions (Figure 19)[28]. Depending on the boiling range of the feed and the purity of the normal paraffins ob-

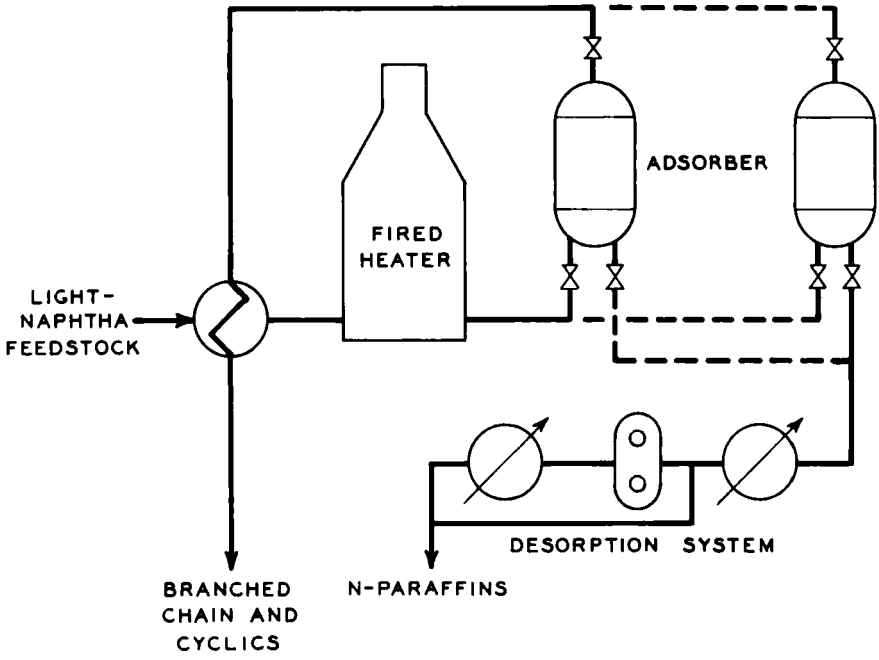

FIGURE 18

Isoviv Process for Separation of Normal Paraffins from Branched Chain and Cyclics

tained, the products have a variety of uses, such as specialty chemicals, pure solvents, biodegradable detergents, intermediate jet fuels, isomerization feedstocks, etc. Typical product distributions are shown in Table 10, 11, and 12[28].

A modified Isosiv process yielding high purity n-paraffins in the C_{11}-C_{20} range has been recently disclosed[29]. Operating conditions vary depending on feedstock composition, but the purge material, n-hexane, is always the same. A recent program shows the process can recover 95-97% of 98%-pure normals in the C_{15}-C_{20} range.

TABLE X

KEROSINE MATERIAL BALANCE, B D

	Feed	Non-Normals Product	Normals Product
Normal Paraffins	2,655	66	2,589
Isomers & Cyclics	5,832	5,832	20
Aromatics	1,400	1,395	5
Olefins	93	65	28
Total	10,000	7,358	2,642
Freezing Pt., °F (ASTM D-1477)	-24	-48	-
Smoke Pt., MM (ASTM D-1322-59T)	23	21	-
Existent Gum, mg/100 ml (ASTM D-381-61T)	3.6	0.4	-

TABLE XI

LIGHT NAPHTHA MATERIAL BALANCE, BPD

	Feed	Non-Normals Product	Normals Product
Non-Normal C_5	208	200	8
nC_5	196	32	164
Non-Normal C_6	2,759	2,715	44
nC_6	2,362	47	2,315
Non-Normal C_7	4,294	4,256	38
nC_7	158	1	157
Non-Normal C_8	23	23	-
Total	10,000	7,274	2,726
Octane No. Ft + 3 CC Tel	82.3	95.4	-

TABLE XII

HEAVY NAPHTHA MATERIAL BALANCE, BPD

	Feed	Non-Normals Product	Normals Product
Non-Normal C_5	80	78	2
nC_5	70	13	57
Non-Normal C_6	1,070	1,047	23
nC_6	760	15	745
Non-Normal C_7	3,100	3,031	69
nC_7	710	14	696
Non-Normal C_8	1,920	1,878	42
nC_8	430	4	426
Non-Normal C_9	1,610	1,568	42
nC_9	130	3	127
Non-Normal C_{10}	120	115	5
Total	10,000	7,766	2,234
Octane No. Ft + 3 CC Tel	79.2	88.4	-

In 1971, Shell and Union Carbide offered a combination process for upgrading gasoline[30]. The Total Isomerization Package (TIP) combines the Hysomer process and Isosiv. TIP provides a flexible design which can optimize operating efficiency over a wide range of feedstocks and product specifications. With narrow-cut feedstocks rich in n-pentane and n-hexane, the Isosiv process separates the product from the Hysomer unit and the unconverted normals are recycled back to the isomerization unit. For feedstocks high in benzene and C_7+'s, the Isosiv unit precedes the Hysomer unit, with the high octane components sent directly to blending, thereby reducing the size of the isomerization stage.

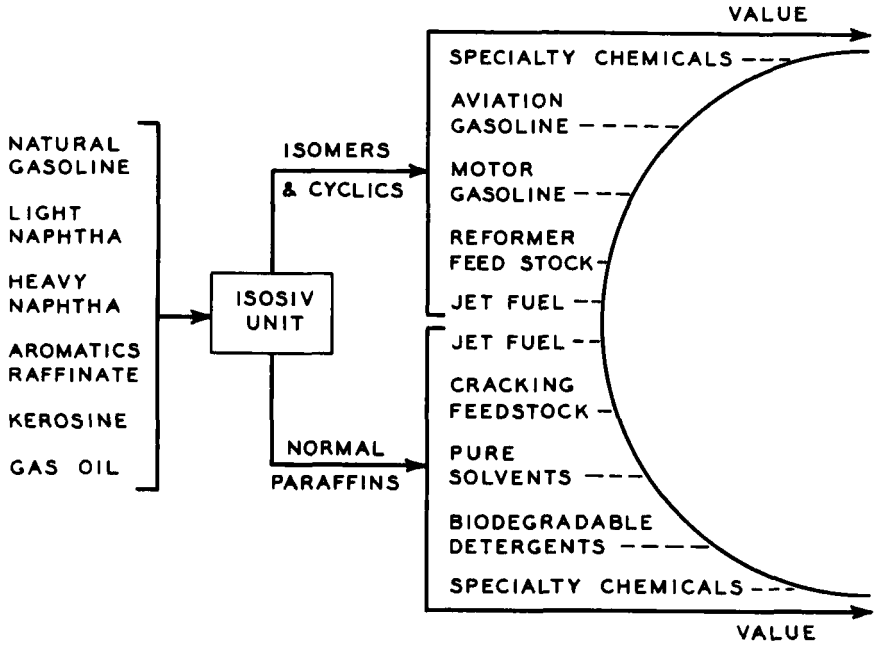

FIGURE 19

Isosiv Will Handle Any of These Feedstocks, But Economics Dictate End Use of Products

More recently, Union Carbide has announced a new development, Olefin Siv[29]. With identical equipment used in the Isosiv Process, high purity isobutene and n-butene are recovered from a mixed butenes feedstock. With a feed containing 50/50 iso-to-normal ratio, Olefin Siv yields 99.5% pure n-butenes.

B. Texaco Selective Finishing Process

The Texaco Selective Finishing Process (TSF) is a versatile process for upgrading gasoline by the removal of low octane normal paraffins by adsorption. The process is a typical adsorption-desorption fixed bed cyclic operation (Figure 20)[31] in which the bed of selective adsorbent (5 A sieve) becomes saturated

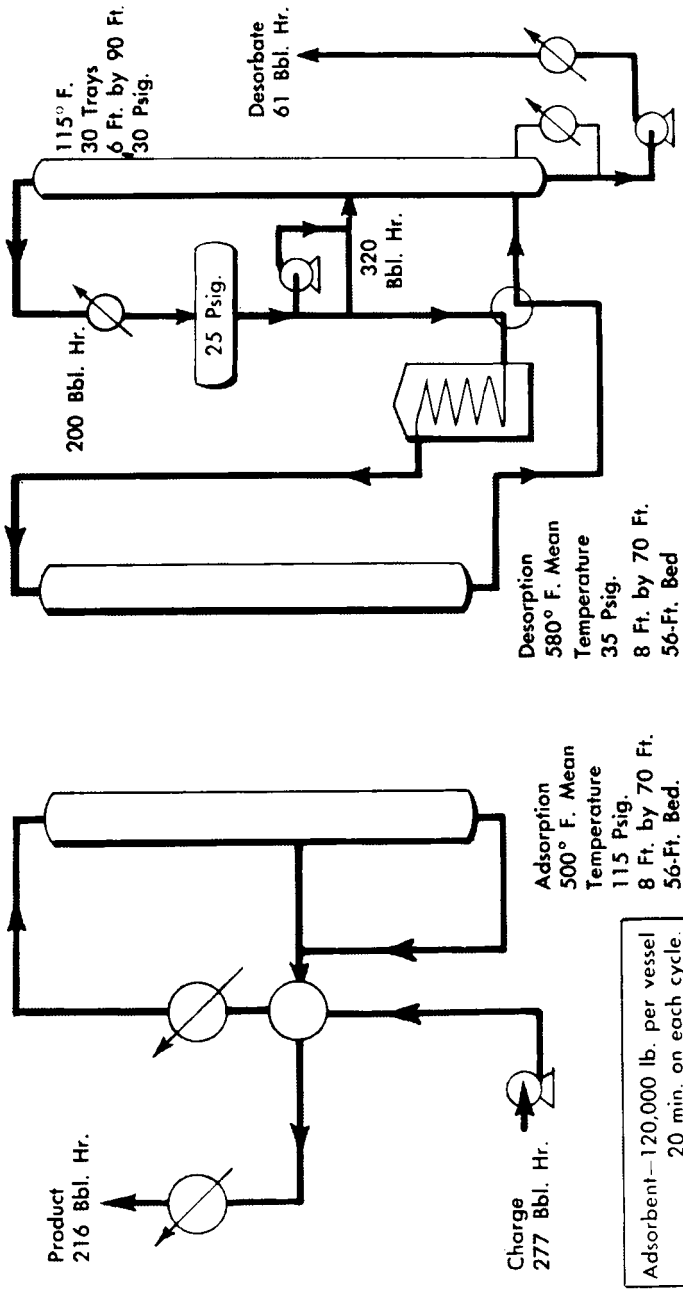

FIGURE 20

Texaco Selective Finishing Process

TABLE XIII

Response of Various Stocks to Texaco Selective Finishing

Stock	Research octane numbers: Charge to TSF, Clear	Charge to TSF, +3 cc TEL	Finished product, Clear	Finished product, +3 cc TEL	RON increase (clear)
Light straightrun naphtha	69.0	87.1	83.7	96.6	14.7
Heavy straightrun naphtha	44.1	66.4	70.6	...	26.5
Full-boiling-range platformate A	87.1	97.1	95.3	101.1	8.2
Full-boiling-range platformate B	94.0	100.8	98.1	104.0	4.1
Thermal cracked naphtha	76.0	88.5	89.1	96.1	13.1
Heavy FCC naphtha	88.0	94.0	94.4	98.8	6.4
Light FCC naphtha	93.8	99.2	95.7	100.6	1.9
Aviation alkylate	93.1	104.4	93.6	...	0.5
Propylene polymer gasoline	93.0	98.0	93.6	98.6	0.6

with straight chain paraffins until no further sorption takes place. The paraffins then pass through the bed without being sorbed and appear in the effluent product Charge to the bed is then stopped and desorption effected.

Using this method, TSF can increase the octane number (clear) of various naphthas from 4-27 octane numbers (Table 13) [32] with straight run naphthas responding best to TSF. Therefore, the TSF process can improve the octane number requirement without increasing the aromatic concentration.

C. The UOP Molex Separation Process

The Molex process is a fixed bed, isothermal, liquid-phase continuous separation operation (Figure 21) for the separation of normal paraffins from isoparaffins and cyclic compounds in a single processing step. In this process the feed and desorbent streams enter the sieve chamber simultaneously while the raffinate and extract streams are withdrawn. The four streams are routed through a distributing device which shifts the flow of these streams into another quartet of lines. Movement of the inlet and outlet points

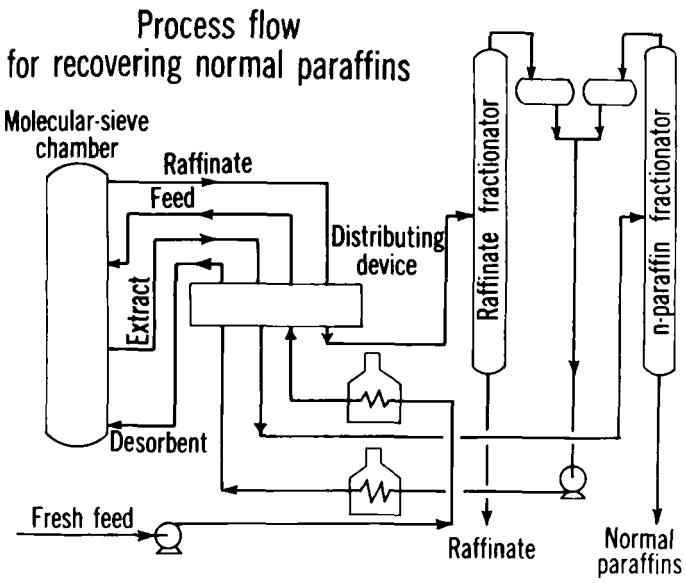

FIGURE 21

Process Flow for Recovering Normal Paraffins

from one location to the next adjacent location providing a flow situation where the molecular sieve (5 A) might be imagined as moving slowly through the chamber.

The process can be used for a wide range of charge stocks, including full range gasolines, light straight run gasolines, kerosines, jet fuels, and light gas oils up to 630°F EP. Molex has pronounced economics and technical advantages when complex separations are involved, such as a mixture of pentanes and hexanes[33].

A prime example of the versatility of this process is the separation of C_{11}-C_{14} normal paraffins from virgin crude fractions for use as a raw material in biodegradable detergent manufacture[34]. Results of processing a C_{11}-C_{15} kerosine containing 15.5% normal paraffins are shown in Table 14.

	Feed	Extract
Gravity, °API	41.8	53.6
ASTM Distillation, °F		
IBP	400	401
10 percent	414	416
50 percent	**436**	**432**
90 percent	470	466
EP	496	490
GLC Analysis, Wt-percent		
n-C_{10}	0.5	3.5
n-C_{11}	1.5	10.9
n-C_{12}	4.0	25.7
n-C_{13}	4.7	25.3
n-C_{14}	3.3	21.3
n-C_{15}	1.4	7.7
n-C_{16}	0.1	0.9
Total *n*-Paraffins	15.5	95.3
Aromatics by FIA Analysis, Vol-percent	12	....
Bromine Index	124	....

TABLE XIV

Commercial Molex Processing of A C_{11}-C_{15} Kerosine

TABLE II. Secondary Building Units of Zeolite Frameworks.

Species	Single rings			Double rings		Complex or multiple units		
	4	6	8	4-4	6-6	4-1	5-1	4-4-1
Analcime	+	+						
Natrolite Thomsonite Edingtonite						+ + +		
Gmelinite Chabazite Erionite Levynite Cancrinite Sodalite	+ + + (+) + +	+ + + + + +	+ (+)		+ +			
Phillipsite Gismondite Barrer P1	+ + +		+ + +	 +				
Brewsterite Heulandite Stilbite	+							+ +
Mordenite Dachiardite Epistilbite Ferrierite Bikitaite							+ + + + +	
Faujasite Linde A ZK-5 Paulingite	+ + + +	+ + +	+ + + +	 +	+ +			

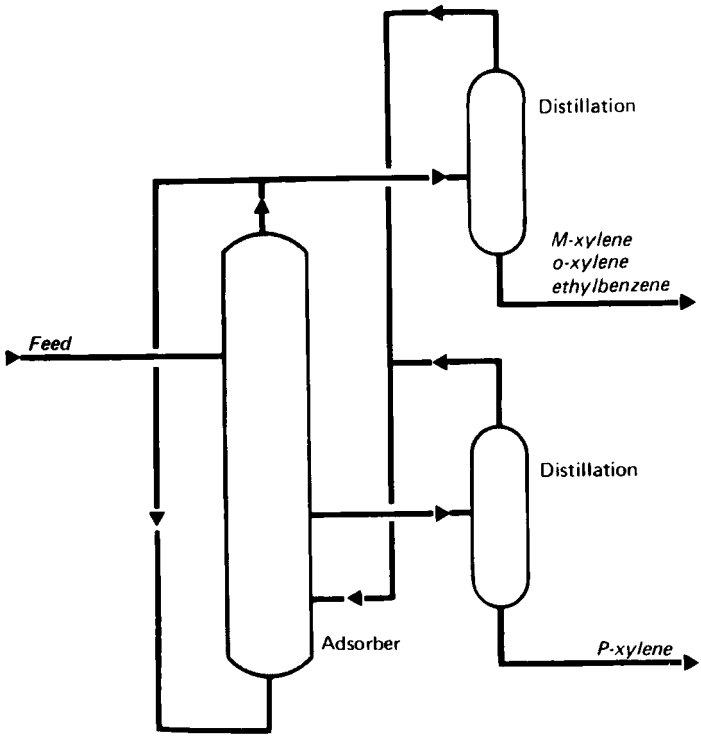

FIG. 22. U.O.P. Parex adsorption process for p-xylene.

The process has been shown to be economical not only when in competition with fractionation but also when called upon to perform separations impossible with fractionation.

D. The UOP Parex Adsorption Process for Paraxylenes

One of the major advances in separations is the separation of paraxylene from other C_8 isomers using molecular sieves instead of the usual crystallization processes. The Universal Oil Product Parex process is a prime example. This process is based on a unique adsorbent with a strong affinity for paraxylene and a weak affinity for the other three isomers normally associated with paraxylene. The adsorbent used is a potassium or barium exchanged type X zeolite.

The continuous separation process (Figure 22)[35] is carried out in the liquid phase at 120-175°C at moderate pressures using a countercurrent flow of adsorbent and liquid without actual movement of the solids. Pilot plant performance shows that this continuous system requires only 1/25 of the adsorbent inventory required in a batch process[36]. Paraxylene with a purity of 99.5% is obtained with extraction efficiencies of 98.4%. Typical pilot plant performance for treating a C_8-aromatic isomer mixture from a reformate extraction are shown in Table 15[36].

U.O.P. claims that this process offers both lower capital investment and operating costs than present crystallization methods (Table 16)[35]. Four Parex units have been licensed and are in the design and construction stage.

IX. Novel Uses in Separation and Purification

Many unique uses of molecular sieves have been developed over the years. Novel uses include the drying of pyrolysis gas[37], separation of hydrogen

TABLE XV

Pilot Plant Performance When Treating C_8-Aromatic Isomers Derived From a Reformate Extract

	Feed (Udex C_8-Arom.)	Extract	Raffinate
p-Xylene, wt.-%	14.3	99.5	<0.1
Ethylbenzene, wt.-%	32.5	0.3	37.9
m-Xylene, wt.-%	35.4	0.1	41.3
o-Xylene, wt.-%	17.8	0.1	20.7
Total	100.0	100.0	100.0

isotopes[38], vacuum drying of iodine[39], drying of low molecular weight fatty acids[40], separation of ammonia and methyl amine[41], separation of inorganic phosphates[42], separation of lithium isotopes[43], adsorption of heavy water[44], and the drying of dimethyl formamide and acrylonitrile[45]. Molecular sieves have been suggested as cigarette filters, packaging desiccants, desiccants for fast drying cements, and the encapsulation of volatile or poisonous gases and liquids for storage. The uses of molecular sieves appears to be unlimited.

X. Looking Ahead with Molecular Sieves

The many unique and commercial separation and purification methods reported here show that molecular sieves have an extended future. Adsorptive separations indicate many new and novel processes will appear as more new synthetic zeolites with unique molecular sieving properties are discovered. It is only a question of time until molecular sieve separation processes replace crystallization and fractionation as the major techniques for the production of the many products required in the petroleum and petrochemical fields. Combination processes in which molecular

Process	Phillips Petroleum Co.		Maruzen Petrochemical Ltd.		UOP "Parex"	Krupp
Feed Composition						
Ethylbenzene %	6.84	24.90	20	24.90		27.60
P-xylene %	21.21	24.70	20	24.70		21.20
M-xylene %	48.11	49.10	40	49.10		47.60
O-xylene %	20.06	1.30	20	1.30		3.60
Capacity tons/year	45,000 p-X	45,000 p-X	45,000 p-X	45,000 p-X	50,000 p-X	50,000 p-X
Cost of plant erected ($)	4,000,000	3,500,000	3,360,000	3,120,000	2,636,000	2,462,500
Total investment ($)	5,900,000	5,250,000	5,100,000	4,800,000	3,636,000	1,800,000
Processing costs ($/ton)						
Direct labor costs	9.27	7.60	8.20	7 53	12.35	7.02
Indirect costs	31.42	29.80	29.22	28.52	10.52	7.50
Overheads (8% of labor costs)	included	included	included	included	2.84	0.63
Taxes and Insurance (2% of fixed capital)	included	included	included	included	1.05	0.98
Depreciation (8% of investment)	included	included	included	included	15.87	15.87
Processing costs ($/ton) (not including feedstock)	40.70	37.40	37.67	36.06	42.64	10.2
Feedstock	51.60	51.60	51.60	51.60		
Processing costs ($/ton) (including feedstock)	92.30	89.00	89.02	87.66		
Utilities ($/ton)	4.05	2.77	3.75	3.47	2.86	5.21

TABLE XVI

Economic Comparison of Processing Costs
of P-Xylene Crystallization

sieves are used both as catalysts and adsorbents are rapidly being developed. These developments will continue to engage the interest of research workers and engineers alike, and will provide a tremendous stimulus to search for new uses.

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XII. Appendix

Bibliography of Literature and U.S. Patents on the Use of Molecular Sieves for Separation and Purification.

In view of the wide interest in molecular sieves, the following extensive bibliography is provided. Abstracted from Chemical Titles and API Literature References from 1953 to date, the listing will serve as a basic guide to most published papers. The authors humbly apologize to anyone not referred to in the listings. The references are arranged chronologically.

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823. Tolmachev, ZH. Fiz Khim, 46, 700 No. 3 (1972)
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B. U.S. Patents

Separation

2,813,139 (1957)	2,956,015 (1960)	3,068,627 (1962)
2,818,137 (")	2,956,089 (")	3,069,470 (")
2,818,449 (")	2,957,927 (")	3,070,542 (")
2,818,455 (")	2,958,708 (")	3,078,635 (1963)
2,850,549 (1958)	2,963,519 (")	3,078,636 (")
2,858,901 (")	2,966,451 (")	3,078,640 (")
2,859,256 (")	2,966,528 (")	3,078,641 (")
2,859,257 (")	2,966,531 (")	3,078,642 (")
2,866,835 (")	2,971,993 (1961)	3,078,643 (")
2,881,862 (1959)	2,972,643 (")	3,078,644 (")
2,886,522 (")	2,974,179 (")	3,078,645 (")
2,889,893 (")	2,977,346 (")	3,085,380 (")
2,899,379 (")	2,978,407 (")	3,086,339 (")
2,900,430 (")	2,982,715 (")	3,094,483 (")
2,901,519 (")	2,984,620 (")	3,094,569 (")
2,906,795 (")	2,985,589 (")	3,095,288 (")
2,912,473 (")	2,987,471 (")	3,096,380 (")
2,917,556 (")	2,988,502 (")	3,098,814 (")
2,920,037 (1960)	2,988,503 (")	3,103,425 (")
2,920,038 (")	2,988,577 (")	3,106,591 (")
2,921,026 (")	2,992,703 (")	3,106,593 (")
2,921,104 (")	2,996,558 (")	3,111,387 (")
2,921,970 (")	2,999,861 (")	3,114,782 (")
2,924,639 (")	3,002,911 (")	3,121,625 (1964)
2,925,380 (")	3,007,545 (")	3,121,757 (")
2,925,381 (")	3,010,894 (")	3,126,425 (")
2,930,447 (")	3,012,630 (")	3,132,079 (")
2,935,467 (")	3,014,078 (")	3,133,126 (")
2,935,539 (")	3,024,262 (1962)	3,140,931 (")
2,938,864 (")	3,029,242 (")	3,144,307 (")
2,940,926 (")	3,029,575 (")	3,146,277 (")
2,944,092 (")	3,037,338 (")	3,155,468 (")
2,944,627 (")	3,053,913 (")	3,160,581 (")
2,945,804 (")	3,064,003 (")	3,164,454 (1965)
2,950,336 (")	3,066,116 (")	3,176,444 (")
2,952,630 (")	3,067,271 (")	3,176,445 (")

Separation (Cont'd)

3,182,017 (1965)
3,183,182 (")
3,184,406 (")
3,184,518 (")
3,201,344 (")
3,201,490 (")
3,204,006 (")
3,205,166 (")
3,208,200 (")
3,211,644 (")
3,226,913 (1966)
3,226,914 (")
3,227,647 (")
3,231,631 (")
3,239,455 (")
3,242,070 (")
3,243,470 (")
3,244,619 (")
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3,251,766 (")
3,260,667 (")
3,265,750 (")
3,266,221 (")
3,268,440 (")
3,271,303 (")
3,274,100 (")
3,285,701 (")
3,291,725 (")
3,291,726 (")
3,294,858 (")
3,303,231 (1967)
3,306,006 (")
3,306,847 (")
3,306,848 (")
3,308,058 (")
3,309,311 (")
3,309,415 (")
3,310,486 (")
3,321,396 (")
3,331,882 (")
3,338,030 (")
3,342,726 (")
3,346,485 (")
3,347,783 (")
3,350,471 (")
3,350,472 (")
3,355,509 (")
3,360,582 (")
3,362,904 (1968)
3,365,394 (1968)
3,370,002 (")
3,373,103 (")
3,378,486 (")
3,392,113 (")
3,395,097 (")
3,405,057 (")
3,418,235 (")
3,420,772 (1969)
3,422,003 (")
3,422,004 (")
3,422,005 (")
3,437,590 (")
3,442,066 (")
3,450,629 (")
3,451,924 (")
3,455,815 (")
3,458,592 (")
3,461,177 (")
3,468,791 (")
3,485,748 (")
3,489,808 (1970)
3,501,545 (")
3,508,382 (")
3,510,423 (")
3,517,484 (")
3,520,113 (")
3,520,801 (")
3,524,895 (")
3,531,400 (")
3,531,539 (")
3,533,220 (")
3,537,237 (")
3,539,501 (")
3,539,502 (")
3,558,730 (")
3,558,732 (")
3,600,453 (")
3,636,180 (1972)
3,653,184 (")
3,654,145 (")
3,656,278 (")
3,662,074 (")
3,665,046 (")
3,668,266 (")
3,668,267 (")
3,686,342 (")
3,686,343 (")

Purification

2,810,454 (1957)
2,843,219 (1958)
2,893,512 (1959)
2,910,139 (")
2,934,167 (1960)
2,935,540 (")
2,950,319 (")
2,971,607 (1961)
2,971,824 (")
2,977,346 (")
2,979,548 (")
3,011,589 (")
3,015,369 (")
3,022,143 (1962)
3,024,867 (")
3,024,868 (")
3,033,642 (")
3,078,634 (1963)
3,078,637 (")
3,078,638 (")
3,978,639 (")
3,080,436 (")
3,085,379 (")
3,085,380 (")
3,087,291 (")
3,100,685 (")
3,102,012 (")
3,150,942 (1964)
3,151,178 (")
3,151,959 (")
3,164,453 (")
3,170,986 (")
3,185,540 (")
3,186,789 (")
3,197,942 (")
3,209,050 (")
3,221,073 (")
3,228,995 (1966)
3,242,641 (")
3,300,324 (")
3,306,945 (1967)
3,322,842 (")
3,330,778 (")
3,331,190 (")
3,345,135 (")
3,358,421 (")
3,363,401 (1968)

Purification (Cont'd)

3,398,208 (1968)
3,490,865 (1970)
3,538,168 (")
3,557,531 (1971)
3,566,611 (")
3,597,169 (")
3,634,533 (1972)
3,654,144 (")
3,660,967 (")
3,278,624 (1966)
3,346,328 (1967)
3,392,508 (1968)
3,454,488 (1969)
3,470,677 (")
3,494,854 (1970)
3,549,529 (")

Treating

2,834,429 (1958)
2,859,170 (")
2,859,173 (")
2,886,508 (1959)
2,886,509 (")
2,888,394 (")
2,891,902 (")
2,894,998 (")
2,899,474 (")
2,909,538 (")
2,914,591 (")
2,917,449 (")
2,924,567 (1960)
2,937,133 (")
2,943,037 (")
2,947,683 (")
2,958,644 (")
2,975,222 (1961)
2,981,675 (")
2,981,679 (")
3,005,768 (")
3,007,863 (")
3,025,133 (1962)
3,051,646 (")
3,061,992 (")
3,063,933 (")
3,063,934 (")
3,067,002 (")
3,080,436 (1963)
3,081,255 (")
3,082,166 (")
3,100,745 (")
3,163,594 (1964)
3,167,506 (1965)
3,236,903 (1966)
3,243,366 (")
3,258,417 (")
3,278,422 (")

INDEX TO LITERATURE BIBLIOGRAPHY

Adsorption

Water - 90,95,154,165,213,224,276,318,321,324, 343,380,428,463,481,534,562,614,628,635, 651,717,736,739,762,763,780,796,814,822

Theory - 273,275,314,423,489,612,731,748,759,764, 812

Misc. - 85,87,89,91,92,93,131,153,156,157,164, 166,201,202,203,205,214,216,219,222,228, 267,268,283,286,290,319,320,322,323,331, 337,384,391,399,401,406,421,472,476,493, 404,537,540,553,570,609,617,619,622,632, 638,641,654,722,723,724,730,744,745,746, 747,749,754,756,758,761,804,807,819

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Gases - 14,21,49,83,126,163,147,191,200,209,234, 277,311,316,325,330,372,373,422,447,471, 475,498,499,500,640

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Gases - 16,32,47,79,99,128,139,141,158,168,169, 185,196,246,251,255,274,291,292,294,298, 304,351,365,395,409,415,430,432,448,455, 468,483,511,512,517,546,576,578,582,584, 590,593,652,685,688

Liquids - 54,67,98,107,108,113,141,145,179,185,207, 225,236,244,249,283,288,302,338,340,346, 355,357,360,387,390,393,415,448,465,542, 576,593,610,634,653,675,782,811,818

Treating

Gases - 12,27,30,39,49,62,73,78,79,82,112,137, 140,149,150,179,180,186,211,231,233,245, 252,272,344,359,368,370,371,436,482,502, 507,563,580,603,691,692

Liquids - 60,74,81,97,101,102,105,111,116,135,143, 188,492,581,600,625,649,668,725,734

Commercial Processes

Texaco Selective Finishing (TSF) - 17,24,37,40

Molex - 22,23,25,26,38,63,76,240,241,307,441

Isosiv - 64,68,100,109,144,189,193,237,238,253, 352,429,446,449,496,508,575,576,577, 579,592,671,681,683,686,693

Parex - 364,503,519,590,599,673,680,689,690