THE VAPOR-PHASE CATALYTIC HYDROGENATION OF OLEFINS

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This paper presents a review of the literature on the hydrogenation of the olefin hydrocarbons over solid catalysts. Although primarily aimed at flow systems, references to certain basic work in static systems are included.

The survey includes theoretical and experimental reports on adsorption and transfer of heat and mass as well as kinetic studies of the reaction mechanism. An appendix contains tables of references to the physical and thermodynamic properties of hydrogen, the olefin hydrocarbons, the respective alkanes, and mixtures of these components.

This review includes only that information which is scattered throughout the literature; data easily available in standard handbooks have been omitted.

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I. INTRODUCTION

The hydrogenation of the olefin hydrocarbons has been extensively studied during the past fifty years of research. Few, if any, studies have been directed at the problem of the reaction in tubular reactors. In such reactors the flow patterns and fluid dynamical behavior under any flow rate are more thoroughly understood than in the packed bed, and for this reason it should be possible to separate the diffusional steps from the chemical reaction steps, both experimentally and theoretically. This would make it possible to establish the effect of each of the operating variables on the overall rate in the reactor and, in particular, upon the diffusion and reaction steps independently. Each of these separate phenomena may then be treated as an individual occurrence and a correlation of these steps into an overall rate equation may then be studied.

This paper, which is a review of the literature on the hydrogenation of the olefin hydrocarbons over solid catalysts, is the result of a literature study prepared prior to the initiation of the experimental work under a contract from the Office of Ordnance Research of the Department of the Army on the kinetics of these reactions in tubular reactors. Tables of the available data on the properties of the olefin hydrocarbons, hydrogen, and mixtures of these are included to facilitate further experimental work in this field. The data which are easily available in standard handbooks have been omitted and only those data scattered throughout the chemical literature and thought to be of value in such a research project have been included.

II. THE CATALYTIC HYDROGENATION OF OLEFINS

A. Mechanism

Many theories have been advanced purporting to explain the mechanism and kinetics of the hydrogenation of the olefin hydrocarbons. Bennewitz and Neuman (9), Bodenstein (11), and Rideal (56) were among the first to offer mechanistic explanations of the phenomena observed. Since these early attempts a great many ideas and theories have been proposed. The most recent reviews of this field are those of Eley (19, 20, 21, 23), Eyring and his coworkers (25), Farkas (26), Taylor and Dibeler (70), and Laidler (40).

Hypotheses to explain the action of the catalyst in the reaction have been listed by several authors (2, 24, 25, 61, 79). The controversy between the proposed "associative" mechanism for the adsorption of the olefin on the metal catalyst and the "dissociative" mechanism seems to be still unsettled. Eley (22) supports the associative mechanism based upon calculations of the heat of adsorption. Beeck and his coworkers (6) support the second, through evidence on the observed self-hydrogenation of ethylene. It is possible that the actual mechanism lies somewhere between the two extremes and that the molecule in the adsorbed state possesses some of the properties of both; in short, that the proton is not fixed in position but possesses a certain amount of mobility.

However, the overall picture of the reaction seems to be clear. Below 200°C. and above −80°C. the reaction is of the first order with respect to the hydrogen concentration and independent of the olefin concentration for all partial pressures of olefin above a few millimeters of mercury. This situation reverses itself at higher temperatures and lower partial pressures of olefin.

The situation seems best explained by either of two approaches: viz., that gas-phase hydrogen attacks the surface covered with adsorbed olefin, or that there remains a small percentage of the surface upon which the hydrogen is adsorbed in competing amounts and that it is upon this surface that reaction proceeds. Farkas, Farkas, and Rideal (28) present experimental evidence for the latter, indicating that at least 5 per cent of the surface is still free to adsorb hydrogen in amounts permitting the reaction to proceed upon these areas. There is still no general agreement, however, as evidenced by two papers in a Discussion of the Faraday Society (No. 8, 1950; see reference 25), one of which, by Eyring, presents evidence based on theory in support of the reaction between adsorbed ethylene and molecular hydrogen in the gas phase, while in the other Laidler presents theoretical reasoning in support of a mechanism involving both molecules in the adsorbed state.

Considerable attention has been devoted to the exchange reactions between olefin hydrocarbons and deuterium and deuterated olefins. Such reactions are thought to be of possible significance in explaining the mechanism of the olefin hydrogenation. In general, the experimental evidence has led to the conclusion that the exchange reaction within the olefin systems is either extremely slow under all conditions studied to date or, possibly, does not occur at all (12, 15). Infrared and mass spectrometry were used as analytical tools. From such data it would seem that the "dissociative" adsorption is most unlikely in olefin systems. Taylor and his coworkers (67) have shown that exchange reactions within alkane systems do occur with measurable speed and from this it seems reasonable to propose a dissociative adsorption mechanism for these compounds. It is likewise well established that the exchange reactions between olefins and HD and D_2 occur simultaneously with the hydrogenation of the olefin (12, 28, 77, 80). These papers are reviewed by Taylor and Dibeler (70) and an excellent evaluation of their implication insofar as the hydrogenation mechanisms possible is presented.

Beeck and his coworkers have been extremely active in the study of the geometric factor in catalysis and its influence on the mechanism and rate of the hydrogenation of olefin hydrocarbons. Their work is summarized in three papers of considerable importance (6, 7, 8). Briefly, the importance of the lattice parameter as a guide in predicting catalytic activity is indicated. Through the technique of obtaining films of catalytically active metals in oriented and unoriented crystal states and by comparing the hydrogenation rates over each, conclusions may be drawn concerning the optimum geometry of the catalyst surface.

As a result of this work, it is proposed that the reaction of adsorbed hydrogen with ethylene from the gas phase is the fast reaction, whereas the slow or rate-determining step is the removal of adsorbed ethylene from the surface by hydrogenation with adsorbed hydrogen.

B. Experimental

The literature of experimental studies of this reaction is voluminous. The kinetics of the hydrogenation of the olefins (mainly ethylene) has been extensively determined in both homogeneous and heterogeneous systems for varying temperature and pressure ranges. Static and flow systems have been utilized in these experiments. Pease (47) found that the hydrogenation of ethylene in a static homogeneous system is a second-order reaction at 475–500°C. The same author carried out intensive investigations of the catalytic hydrogenation of ethylene over copper catalysts in the range of temperatures between 0°C. and 220°C. in static systems (45, 46, 48, 49).

Nickel has been used by many investigators as a catalyst for the determination of the kinetics of the hydrogenation of ethylene. Melville (42) studied the reaction on a nickel wire catalyst at temperatures from 100°C. to 300°C. and pressures from 1 mm. to 760 mm. Zur Strassen (65) did similar work with a nickel ribbon as a catalyst. Schwab and Zorn (63) and Aoyama, Matsuzawa, and Takahashi (1) determined the hydrogenation kinetics in flow systems using a 75 per cent nickel–25 per cent copper alloy as catalyst. Toyama (74, 75) studied the hydrogenation of ethylene (at 99–165°C.) and of propylene (at 25–178°C.) in a static system with nickel filaments as catalysts and found the initial rates of the hydrogenation of ethylene to be somewhat larger than those for propylene.

The kinetics of the hydrogenation of ethylene over platinum catalysts has been studied by Farkas and Farkas (27) and Postovskaya (50). A palladium wire was used by Fischer and Knorr (29).

Schuster (62) studied the kinetics of the hydrogenation of a series of olefins—ethylene, propylene, 1-butene, 2-butene, pentene—over several contact surfaces and determined the half-time period and energy of activation for these reactions. Several other authors also list activation energies for the hydrogenation of some of the olefins (58, 64, 78).

The adsorption effects in the low-temperature hydrogenation of ethylene over pure and promoted iron catalysts were studied by Hansford and Emmett (33). They found that only hydrogen is actively adsorbed between $-40°$ and $-89°$C. and, therefore, plays the dominant part in the reaction. This is contrary to most other findings at higher temperatures and with different catalysts (see next section). A general study of the catalytic effects of a group of metals in the hydrogenation process was performed by Schmidt (60). Klar (38) determined the relationship of the adsorption ratios of the reactants and products in the ethylene hydrogenation reaction to the observed rate constants.

III. BASIC PHYSICAL AND CHEMICAL MECHANISMS OCCURRING IN FLOW REACTORS

A. Adsorption

Many studies have been made of the adsorption of hydrogen and ethylene on palladium and other metals. These are reviewed from the point of view of the adsorption mechanism involved in the first section of this paper. The following is a summary of a portion of the experimental work done to date. Taylor and Burns (68) measured the adsorption of hydrogen and ethylene by finely divided nickel, cobalt, iron, copper, palladium, and platinum. The effect of repeated heating and different carriers on the adsorption capacity of palladium for hydrogen was determined by Sabalitschka (59). The adsorption of hydrogen by the platinum metals was measured by Muller and Schwabe (43) and Gutbier and Schieferdecker (32). The effect of the presence of water on the rate and amount of hydrogen adsorption by palladium black and palladium metal has been studied by Temkin and Bakh (71). Tarama, Myakawa, and Morishima (66) determined the static adsorption of hydrogen on palladium black at constant pressure within the ranges 0–180°C. and 0.125–1 atm.

Ubbelohde and Egerton (81) reported the removal isotherms of hydrogen from palladium at 306°C., 218°C., 150°C., and 100°C. A summary of the data on the adsorption of hydrogen and ethylene on various metals was given by Nyrop (44). Reyerson and Cines (53) measured the adsorption of propylene and propane on palladized silica gel at $-25°$C., 0°C., and 35°C., and concluded that the propylene covered the surface at all partial pressures above a few millimeters of mercury. The results were found to conform reasonably well to the B.E.T. theory.

A theoretical mechanism for the adsorption of hydrogen and ethylene, based on the activated complex theory, was postulated by Eyring, Colburn, and Zwolinski (25).

B. Mass and heat transfer in experimental studies

The importance of insuring mass and temperature equilibrium in reactors in which these hydrogenation reactions are being studied cannot be overemphasized. Some of the earlier experimental results are today subject to question because of failure on the part of the experimenter to provide for these factors.

The theoretical aspects of chemical flow reactors with regard to basic transfer mechanisms have been treated intensively only during the last few years. Trautz (76) was the first to relate viscosity, heat conductivity, and diffusion in gas mixtures with chemical reactivity by employing kinetic and quantum theory. Eddy diffusion of carbon dioxide and hydrogen into air was measured at different Reynolds numbers by Towle and Sherwood (73). Kirkwood (37) has sketched the outline for a general statistical mechanical theory of the transport properties and response to time dependent external force fields.

McCarter, Stutzman, and Koch (41) derived the equations for eddy diffusivities as a function of the parameters of the system and described two experimental methods for studies in turbulent gas flow. The characteristics of reaction- and diffusion-controlled heterogeneous reactions have been reviewed by Zimmerman (86), and criteria for the recognition of a heterogeneous reaction under joint activation and diffusion control have been set up. From the kinetic theory of gases, Curtiss and Hirschfelder (16) developed equations for the rate of change of entropy in a flow system, explicitly taking into account heat conductivity, viscosity, diffusion, and chemical reactions.

Wilke (83) derived an expression from the Stefan–Maxwell diffusion equations for the effective diffusion coefficient of a gas with respect to a multicomponent mixture of stagnant gases. Approximate methods for the calculation of rates of diffusion in complex mixtures for stagnant films, and also with simultaneous diffusion of all components, are presented.

Most of the theoretical and experimental work on flow reactors has been performed on reactors in which the catalyst was used in the form of coatings on various shapes of carrier packings. While this does not correspond to the apparatus to be used in the present study, some of the more basic treatments of the flow, diffusion, and heat-transfer problems which are encountered in packed-bed operation and which are discussed in the literature will be presented here.

Thiele (72) formulated a mathematical analysis of the activity of a catalyst particle as related to its structural dimensions and to the diffusivities and concentrations of the gases flowing through the bed. Pshezhetskiĭ (51) presented a theory for the rate of diffusion of a gas from the interior of a catalyst and from the surface of the catalyst particle to the gas current. He claimed that the difference between the apparent and the true reaction constants is proportional to the velocity of the gas flow and to the square of the grain diameter. Pshezhetskiĭ and Rubinshtein (52) derived a general quantitative theory for the steady state of heterogeneous reactions taking place when a gas streams through a bed of catalyst, taking into account the chemical and diffusion steps. A general relation between contact time and space velocity in packed-bed reactors was obtained by Brinkley

(13) by comparing the Euler and Lagrange forms of hydrodynamic steady-state equations for the composition of a flowing system which undergoes chemical reaction with accompanying volume change. He, however, assumed that diffusion effects could be neglected.

Bernard and Wilhelm (10) solved the differential equation for turbulent diffusion in packed beds through which a fluid is flowing for the special case when the tube wall provides a boundary condition. Wynkoop and Wilhelm (85) studied the hydrogenation of ethylene over a copper–magnesia catalyst and developed kinetic equations for the flow system. They discussed the flow equations and the precautions that must be used in their development.

A mathematical treatment of the temperature distribution in catalyst masses situated in reactors of various shapes has been given by Gel'perin (31). Wilson (84) combined the fundamental rate equations with a heat balance for flow reactors. With these he developed procedures for calculating longitudinal temperature gradients and overall heat-transfer coefficients.

Kayser (36), in connection with a study of the hydrogenation of propylene, developed a method of deriving the basic heat- and mass-transfer equations in terms of the analogy between heat, mass, and momentum transfer. He also presented a method of determining the rate expression defining a process over a range of compositions, temperatures, and flow rates in which the productive area of the catalyst is a function of the velocity of the gas flowing through the bed.

The fundamental heat-balance equations for reactors have been presented by Wilhelm *et al.* (82). A method for rapidly analyzing thermal problems by means of an electrical network has also been described. For use in temperature studies of flow reactors, Balandin (4) has described a differential thermocouple with automatic continuous recording of temperatures.

Although many empirical and semiempirical studies have been made of kinetics in packed-bed reactors, theoretical studies of the various phenomena which take place in catalytic reactors have been relatively few and recent. Since the packed bed offers an extremely complex picture insofar as mass and heat transfer and reaction surface are concerned, the most promising apparatus for investigation of the basic mechanisms occurring in catalytic converters is the tubular reactor.

The first attempt to formulate a theory for a flow reactor where both mass transport and chemical steps have to be taken into account was made by Forster and Geib (30) in 1934. They formulated equations for a first-order homogeneous flow reaction where the gas velocity is of such a magnitude that molecular diffusion (in the axial direction) becomes of importance. Their derivation was limited to laminar flow, and, what is a more serious restriction, plug flow was assumed.

Damköhler (17), with a more general approach to the problem, formulated the differential equations for the course of a homogeneous chemical reaction in a flow system by taking into account requirements for the conservation of mass, momentum, and energy. From these equations he obtained dimensionless parameters relating flow, temperatures, and concentrations. In a later paper (18), Damköhler presented calculations for the influence on the yield of an isothermal, homogeneous, constant-volume, first-order reaction of flow velocity and diffusion. He also derived the theory of Forster and Geib in a more simple manner and with more general assumptions (e.g., taking into account a parabolic velocity distribution). The influence of diffusion is estimated and found to be appreciable only at reduced pressures.

Baron, Manning, and Johnstone (5) have extended Damköhler's analysis to reversible reactions. They have applied their derivation to the analysis of experimental data on the flow oxidation of sulfur dioxide in order to determine the relative importance of the diffusion step in the reaction. This work was performed in a tubular reactor which had the catalyst supported on its wall. The same article includes a very elegant theoretical study of the influence of molecular collisions on the desorption reaction.

The problem of obtaining true chemical rate constants from experimental work with tubular reactors has been treated in several recent papers. Hoelscher (34) and Klinkenberg, Krajenbrink, and Lauwerier (39) have solved the differential equation governing diffusion in flow systems to obtain expressions for the concentration profiles in such systems for different flow patterns. While these solutions were derived specifically for laminar flow, the inclusion of the proper eddy diffusivity terms should permit their extension to the turbulent region. With expressions for the concentrations in flow reactors as a function of the radius available, it will now be possible to calculate the reaction rate constants of a reaction taking place at the tube wall by taking samples of the outlet gas composition at a properly specified radial position, e.g., the center.

Chaitt (14) has applied Hoelscher's work to the development of equations for a first-order reaction taking place in a tubular reactor at different velocities but with constant contact time. This leaves the reaction rate and the flow velocity as the only variables to be considered and permits a clearer understanding of the effects of these two quantities on the conversion. From Chaitt's results it is apparent that at low flow rates the reactions in a tubular reactor are completely controlled by the diffusion process toward the tube wall, while at higher flow rates the processes come under control of the depletion rate at the tube wall.

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V. APPENDIX

The literature abounds in physical and chemical data relative to the 1-olefin hydrocarbons, the corresponding saturated hydrocarbons, hydrogen, and mixtures of these. For the purposes of this report, however, table 1 and its bibliography will serve as sufficient reference.

TABLE 1

Properties of alkane, olefin, and alkane–olefin–hydrogen systems

A. Physical properties

Property	Gases*	References
Thermal conductivity	Hydrogen	Aberdeen, J., and Laby, T. H.: Proc. Roy. Soc. (London) **A113,** 459 (1926). Hercus, E. O., and Laby, T. H.: Phil. Mag. [7] **3,** 1061 (1927). Ibbs, T. L., and Hirst, A. A.: Proc. Roy. Soc. (London) **A123,** 134 (1929). Kannuluik, W. G., and Martin, L. H.: Proc. Roy. Soc. (London) **A144,** 496 (1934). Ulsamer, J.: Z. Ver. deut. Ing. **80,** 537 (1936). Weber, S.: Ann. Physik **82,** 479 (1927). Weizsacker, W.: Physik. Z. **37,** 641 (1936).
	Hydrogen, butane, ethane, propane	Delaplace, R.: Compt. rend. **203,** 1505 (1936).
	Hydrogen, propane	Trautz, M., and Zundel, A.: Ann. Physik **17,** 345 (1933).
Critical data	Butane	Beattie, J. A., Simard, G. L., and Su, G. J.: J. Am. Chem. Soc. **61,** 924 (1939).
	Butane, ethane, ethylene, pentane, propane, propylene	Brescia, F.: J. Chem. Phys. **14,** 501 (1946).
	Butane, ethane, ethylene, propane, propylene	Pickering, S. F.: Natl. Bur. Standards (U.S.), Sci. Papers, **No. 541,** 597 (1926).
	Butane, ethane, hexane, pentane, propane	Anon.: Oil Gas J. **23,** 115 (1945).
	Ethane	Beattie, J. A., Su, G. J., and Simard, G. L.: J. Am. Chem. Soc. **61,** 924 (1939).
	Ethane, ethylene, pentane, propane, propylene	Maass, O., and Wright, C. H.: J. Am. Chem. Soc. **43,** 1098 (1921).
	Propane	Sage, B. H., Schaafsma, J. G., and Lacey, W. N.: Ind. Eng. Chem. **26,** 1218 (1934).
Density	Butane	van Bogaert, L.: Bull. soc. chim. Belg. **36,** 384 (1927). Beckers, M.: Bull. soc. chim. Belg. **36,** 559 (1927).
	Butane, ethane, hexane, pentane, propane	Anon.: Oil Gas J. **23,** 115 (1946).
	Ethane, ethylene	Staharfoss, K.: J. chim. phys. **16,** 175 (1918).
	Ethane, ethylene, pentene, propane, propylene	Maass, O., and Wright, C. H.: J. Am. Chem. Soc. **43,** 1098 (1921).

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* Where more than one gas is listed in a heading, the articles under that heading will often give the property of mixtures of all or of some of the gases listed.

TABLE 1—*Continued*

Property	Gases	References
Density—*continued*	Pentane	Beall, I. N.: Refiner Natural Gasoline Mfr. **14,** 588 (1935).
	Pentane, hexane	Sugden, S.: J. Chem. Soc. **1927,** 1780.
	Propane	Sage, B. H., Schaafsma, J. G., and Lacey, W. N.: Ind. Eng. Chem. **26,** 1218 (1934).
Dimensions	Butane, 1-butene, ethane, hydrogen	Livingston, H. K.: J. Colloid Sci. **4,** 447 (1949).
	Hydrogen	Lunn, E. G., and Bichowsky, F. R.: Phys. Rev. **35,** 563 (1931).
Equations of state, *CPVT* relationships	Butane	Thornton, D. P., Jr.: Natl. Petroleum News **37,** No. 1, R31 (1945).
	Butane, ethane, ethylene, propane	Su, G. J., and Chang, C. H.: J. Am. Chem. Soc. **68,** 1080 (1946).
	Ethylene, hydrogen	Trautz, M., and Gursching, M.: Z. anorg. allgem. Chem. **179,** 1 (1929).
	Hexane	Herz, W.: Z. Elektrochem. **30,** 604 (1924).
	Hydrogen	Wohl, K.: Z. physik. Chem. **B2,** 77 (1929).
	Pentane	Beattie, J. A., Levine, S. W., and Douslin, D. R.: J. Am. Chem. Soc. **74,** 4778 (1952).
	Propane	Sage, B. H., Schaafsma, J. G., and Lacey, W. N.: Ind. Eng. Chem. **26,** 1218 (1934).
Moments of inertia	Ethylene	Badger, R. M.: Phys. Rev. **45,** 648 (1934).
	Hydrogen	Hyman, H. H., and Jeppesen, R.: Nature **125,** 462 (1930).
		Birge, T. R., and Jeppesen, R.: Nature **125,** 463 (1930).
Structure	Butane, ethane, propane	Bruche, E.: Ann. Physik [5] **4,** 387 (1930).
	Ethane, ethylene	Penney, W. G.: Proc. Roy. Soc. (London) **A144,** 166 (1934).
Twisting frequency	Ethylene	Penney, W. G.: Proc. Roy. Soc. (London) **46,** 333 (1934).
Viscosity	Butane	Sage, B. H., Yale, W. D., and Lacey, W. N.: Ind. Eng. Chem. **31,** 223 (1939).
	Butane, 1-butene, ethane, ethylene, 1-pentene, propane, propylene	Titani, T.: Bull. Inst. Phys. Chem. Research (Japan) **8,** 433 (1929).
	Butane, hydrogen	Keyes, F. G.: Trans. Am. Soc. Mech. Engrs. **73,** 589 (1951).
	Butane, hydrogen, propane	Wobser, R., and Muller, F.: Kolloid-Beih. **52,** 165 (1941).
	Butane, propane	Lipkin, M. R., Davison, J. A., and Kurtz, S. S.: Ind. Eng. Chem. **34,** 976 (1942).
	Butane, propylene	Trautz, M., and Husseini, T. I.: Ann. Physik **20,** 121 (1934).

TABLE 1—*Continued*

Property	Gases	References
Viscosity—*continued*	Ethane, hydrogen, propane	Trautz, M., and Kurtz, F.: Ann. Physik **9,** 981 (1930).
		Trautz, M., and Kurtz, F.: Ann. Physik **10,** 81 (1931).
	Ethane, hydrogen, propane, propylene	Adzumi, H.: Bull. Chem. Soc. Japan **12,** 199 (1937).
	Ethylene	Zimmer, O.: Ber. physik. Ges. **1912,** 471.
	Ethylene, hydrogen	Trautz, M., and Melster, A.: Ann. Physik [5] **7,** 409 (1930).
		Trautz, M., and Stauf, F. W.: Ann. Physik [5] **2,** 737 (1929).
	Hexane, pentane	Geist, J. M., and Cannon, M. R.: Ind. Eng. Chem., Anal. Ed. **18,** 611 (1946).
	Hydrogen	Clark, R.: Trans. Roy. Soc. Can. **13,** sect. iii, 177 (1919).
		van Itterbeck, A., and Claes, A.: Physica **5,** 938 (1938).
		Rammler, E., and Breitling, K.: Wärme **60,** 620, 636 (1937).
		Timiriazev, A.: Ann. Physik **40,** 971 (1913).
	Hydrogen, propane	Klemenc, A., and Remi, W.: Monatsh. **44,** 307 (1924).
	Pentane	Bleakney, W. M.: Physics **3,** 123 (1932).
		Day, R. K.: Phys. Rev. **40,** 281 (1932).
	Propane	Sage, B. H., and Lacey, W. N.: Ind. Eng. Chem. **30,** 829 (1938).
General	Butane, 1-butene, ethane, ethylene, propane, propylene	Smith, M. L., and Hanson, G. H.: Oil Gas J. **44,** No. 10, 119 (1945).
	Butane, propane	Finley, G. H.: *The Handbook of Butane-Propane Gases*, 2nd edition. Western Gas Co., Los Angeles, California (1935).
		Denny, L. C., and Wickstrom, H. W.: *The Handbook of Butane-Propane Gases*, 3rd edition. Jenkins Pubs., Los Angeles, California (1947).
	Ethylene, propylene	Coffin, C. C., and Maass, O.: Trans. Roy. Soc. Can. [3] **21,** No. 3, 33 (1928).
	1-Pentene	Sherrill, M. L., and Walter, G. F.: J. Am. Chem. Soc. **58,** 742 (1936).

B. Thermodynamic properties

Property	Gases	References
Bond energies	Butane, ethane, hexane, pentane, propane	Glockler, G.: J. Chem. Phys. **17,** 747 (1949).
	Ethane, ethylene	Glockler, G.: J. Chem. Phys. **16,** 842 (1948).
	Ethylene	Huckel, W.: Ber. **53B,** 1277 (1920).

TABLE 1—*Continued*

Property	Gases	References
Enthalpy	Butane, ethane, hexane, pentane, propane	Scheibel, E. G., and Jenny, F. J.: Ind. Eng. Chem. **37,** 990 (1945).
	Ethane, ethylene, hydrogen	Justi, E.: Feuerungstech. **26,** 313 (1938).
	Ethylene	Keesom, W. H., Bijl, A., and Monte, L. A.: Appl. Sci. Research **A3,** 261 (1952). Michels, A., de Groot, S. R., and Geldermans, M.: Appl. Sci. Research **A1,** 55 (1947) York, R., Jr., and White, E. F., Jr.: Trans. Am. Inst. Chem. Engrs. **40,** 227 (1944).
	Hydrogen	Kobe, K. A., and Long, E. G.: Petroleum Refiner **28,** No. 11, 127 (1949).
Entropy	Butane	Parks, G. S., Shomate, C. H., Kennedy, W. D., and Crawford, B. L., Jr.: J. Chem. Phys. **5,** 359 (1937). Vol'kenshteĭn, M. V.: Compt. rend. acad. sci. U.R.S.S. **49,** 111 (1945).
	Butane, 1-butene, ethane, ethylene, hexane, 1-hexene, pentane, 1-pentene, propane, propylene	Thacker, C. M., Folkins, H. O., and Miller, E. L.: Ind. Eng. Chem. **33,** 584 (1941).
	Butane, ethane, hexane, pentane, propane	Reis, T.: Rev. inst. franç. pétrole et Ann. combustibles liquides **1,** 33 (1946). Teck, P.: Verhandel. Koninkl. Vlaam. Acad. Wetenschap. Belg. **11,** 5 (1949).
	Butane, hexane, pentane	Bonino, G. B.: Chem. Zentr. **1943, II,** 712.
	1-Butene	Kassel, L. S.: J. Chem. Phys. **4,** 144 (1936). Todd, S. S., and Parks, G. S.: J. Am. Chem. Soc. **58,** 134 (1936).
	Ethane	Kemp, J. D., and Pitzer, K. S.: J. Am. Chem. Soc. **59,** 276 (1937). Mayer, J. E., Brunauer, S., and Mayer, M. G.: J. Am. Chem. Soc. **55,** 37 (1933).
	Ethane, ethylene, hydrogen	Justi, E.: Feuerungstech. **26,** 313 (1938).
	Ethylene	Kassel, L. S.: J. Am. Chem. Soc. **55,** 1351 (1933). Michels, A., de Groot, S. R., and Geldermans, M.: Appl. Sci. Research **A1,** 55 (1947). Vvendenskiĭ, A. A., and Vinnikova, S. G.: J. Gen. Chem. (U.S.S.R.) **4,** 120 (1934). York, R., Jr., and White, E. F., Jr.: Trans. Am. Inst. Chem. Engrs. **40,** 227 (1944).

TABLE 1—*Continued*

Property	Gases	References
Entropy—*continued*	Ethylene, hydrogen	Justi, E., and Luder, H.: Forsch. Gebiete Ingenieurw. **B6**, 209; Science Abstr. **38A**, 1110 (1935).
	Hydrogen	Keesom, W. H., and Houthoff, P. J.: Rapports et comm. 5th. Cong. intern. du froid, Apr.; Communs. Phys. Lab. Univ. Leiden, Suppl. **No. 65**, 31 (1928).
		Rodebush, W. H.: Proc. Natl. Acad. Sci. U.S. **15**, 678 (1929).
		Vrey, H. C.: J. Am. Chem. Soc. **45**, 1445 (1923).
	Pentane	Beall, I. N.: Refiner Natural Gasoline Mfr. **14**, 588 (1935).
	1-Pentene	Scott, D. W., Waddington, G., Smith, J. C., and Huffman, H. M.: J. Am. Chem. Soc. **71**, 2767 (1949).
	Propane	Sage, B. H., Schaafsma, J. G., and Lacey, W. N.: Ind. Eng. Chem. **26**, 1218 (1934).
Equilibrium constants (for hydrogenation of)	1-Butene	Kassel, L. S.: J. Chem. Phys. **4**, 144 (1936).
	1-Butene, ethylene, 1-hexene, 1-pentene, propylene	Kilpatrick, J. E., Prosen, E. J., Pitzer K. S., and Rossini, F. D.: J. Research Natl. Bur. Standards **36**, Research Paper No. 1722, 559 (1946).
	Ethylene	Frost, A. V.: Compt. rend. acad. sci. U.R.S.S. (N.S.) 161 (1933).
		Frost, A. V.: Khim. Tverdogo Topliva **4**, 171 (1933).
		Smith, H. A., and Vaughan, W. E.: J. Chem. Phys. **3**, 341 (1935).
		Teller, E., and Topley, B.: J. Chem. Soc. **1935**, 876.
		Vvendenskiĭ, A. A., and Vinnikova, S. G.: J. Gen. Chem. (U.S.S.R.) **4**, 120 (1934).
		Zeisie, H.: Z. Elektrochem. **48**, 30 (1942).
Free energy	Butane, ethane, hexane, pentane, propane,	Reis, T.: Rev. inst. franç. pétrole et Ann. combustibles liquides **1**, 33 (1946).
	1-Butene	Todd, S. S., and Parks, G. S.: J. Am. Chem. Soc. **58**, 134 (1936).
	Ethylene	Kassel, L. S.: J. Am. Chem. Soc. **55**, 1351 (1933).
		Keesom, W. H., Bijl, A., and Monte, L. A.: Appl. Sci. Research **A3**, 261 (1952).
		Michels, A., de Groot, S. R., and Geldermans, M.: Appl. Sci. Research **A1**, 55 (1947).
		Vvendenskiĭ, A. A., and Vinnikova, S. G.: J. Gen. Chem. (U.S.S.R.) **4**, 120 (1934).

TABLE 1—*Continued*

Property	Gases	References
Free energy—*continued*	Hydrogen	Giauque, W. F.: J. Am. Chem. Soc. **52,** 4816 (1930).
Free energy of formation	Butane, 1-butene, ethane, ethylene, hexane, 1-hexene, pentane, 1-pentene, propane, propylene	Thacker, C. M., Folkins, H. O., and Miller, E. L.: Ind. Eng. Chem. **33,** 584 (1941).
	Butane, ethane, hexane, pentane, propane	Prosen, E. J., Pitzer, K. S., and Rossini, F. D.: J. Research Natl. Bur. Standards **34,** Research Paper No. 1650, 403 (1945).
	1-Butene, ethylene, 1-hexene, 1-pentene, propylene	Kilpatrick, J. E., Prosen, E. J., Pitzer, K. S., and Rossini, F. D.: J. Research Natl. Bur. Standards **36,** Research Paper No. 1722, 559 (1946).
	Ethane	Thayer, V. R., and Stegeman, G.: J. Phys. Chem. **35,** 1505 (1931).
	Ethylene	Haas, M. E., and Stegeman, G.: J. Phys. Chem. **36,** 2127 (1932).
		Haas, M. E.: Univ. Pittsburgh Bull. **29,** 116; Science Abstr. **36A,** 479 (1933).
	Hydrogen	Kobe, K. A., and Long, E. G.: Petroleum Refiner **28,** No. 11, 127 (1949).
Heat capacity	Butane	Templeton, D. H., Davies, D. D., Felsing, W. A.: J. Am. Chem. Soc. **66,** 2033 (1944).
	Butane, 1-butene, ethane, ethylene, hexane, 1-hexene, pentane, 1-pentene, propane, propylene	Stull, D. R.: Ind. Eng. Chem. **35,** 1303 (1943).
		Thacker, C. M., Folkins, H. O., and Miller, E. L.: Ind. Eng. Chem. **33,** 584 (1941).
		Vvendenskiĭ, A. A.: Neftyanoe Khoz. **25,** No. 2, 47 (1947).
	Butane, ethane, ethylene	Spencer, H. M., and Flannagan, G. M.: J. Am. Chem. Soc. **64,** 2511 (1942).
	Butane, ethane, hexane, pentane, propane	Anon.: Oil Gas J. **23,** 115 (1945).
	Butane, hexane, pentane, propane	Pitzer, K. S.: J. Am. Chem. Soc. **63,** 2413 (1941).
	Butane, pentane, propane	Sage, B. H., Webster, D. C., and Lacey, W. N.: Ind. Eng. Chem. **29,** 1309 (1937).
	Butane, propane	Sage, B. H., and Lacey, W. N.: Ind. Eng. Chem. **27,** 1484 (1935).
	1-Butene	Todd, S. S., and Parks, G. S.: J. Am. Chem. Soc. **58,** 134 (1936).
		Wacker, P. F., Chenny, R. K., and Scott, R. B.: J. Research Natl. Bur. Standards **38,** Research Paper No. 1804, 651 (1947).

TABLE 1—*Continued*

Property	Gases	References
Heat capacity—*continued*	Ethane	Dixon, H. B., Campbell, C., and Parker, A.: Proc. Roy. Soc. (London) **100A,** 1 (1921).
		Kistiakowsky, G. B., and Rice, W. W.: J. Chem. Phys. **7,** 281 (1939).
		Thayer, V. R., and Stegeman, G.: J. Phys. Chem. **35,** 1505 (1931).
	Ethane, ethylene	Eucken, A., and Parts, A.: Z. physik. Chem. **B20,** 184 (1933).
		Heuse, W.: Ann. Physik. **59,** 86 (1919).
	Ethane, ethylene, hydrogen	Frost, A. V.: Trans. Exptl. Research Lab. "Khemgas" (Leningrad), Materials on Cracking and Chemical Treatment of Cracking Products (U.S.S.R.) **3,** 27 (1936).
		Justi, E.: Feuerungstech. **26,** 313 (1938).
	Ethane, hexane, pentane, propylene	Spencer, H. M.: J. Am. Chem. Soc. **67,** 1859 (1945).
	Ethylene	Haas, M. E., and Stegeman, G.: J. Phys. Chem. **36,** 2127 (1932).
		Haas, M. E.: Univ. Pittsburgh Bull. **29,** 116; Science Abstr. **36A,** 479 (1933).
		Michels, A., de Groot, S. R., and Geldermans, M.: Appl. Sci. Research **A1,** 55 (1947).
		Sweigert, R. L., and Beardsley, M. W.: Georgia School Technol., State Eng. Expt. Sta. Bulletin **No. 2** (1938).
	Ethylene, hydrogen	Justi, E., and Luder, H.: Forsch. Gebiete Ingenieurw. **B6,** 209; Science Abstr. **38A,** 1110 (1935).
	Hexane	Jatkar, S. K. K.: J. Indian Inst. Sci. **22A,** Pt. II, 19 (1939).
		Waddington, G., and Douslin, D. R.: J. Am. Chem. Soc. **69,** 2275 (1947).
	Hexane, pentane	Bennewitz, K., and Rossner, W.: Z. physik. Chem. **B39,** 126 (1938).
	Hydrogen	Fenning, R. W., and Whiffin, A. C.: Trans. Roy. Soc. (London) **A238,** 149 (1939).
		Giauque, W. F.: J. Am. Chem. Soc. **52,** 4816 (1930).
		Heck, R. C. H.: Mech. Eng. **62,** 9 (1940).
		Kobe, K. A., and Long, E. G.: Petroleum Refiner **28,** No. 11, 127 (1949).
	Pentane	Beall, I. N.: Refiner Natural Gasoline Mfr. **14,** 588 (1935).
	1-Pentene	Scott, D. W., Waddington, G., Smith, J. C., and Huffman, H. M.: J. Am. Chem. Soc. **71,** 2767 (1949).
	Propane	Kistiakowsky, G. B., and Rice, W. W.: J. Chem. Phys. **8,** 610 (1940).

TABLE 1—*Continued*

Property	Gases	References
Heat of adsorption	Hydrogen	Maxted, E. B., and Hassid, N.: J. Chem. Soc. **1931,** 3313. Tryling, C. F.: J. Phys. Chem. **30,** 818 (1926).
Heat of dissociation	Ethylene, hydrogen	Justi, E., and Luder, H.: Forsch. Gebiete Ingenieurw. **B6,** 209; Science Abstr. **38A,** 1110 (1935).
Heat of formation	Butane, 1-butene	Prosen, E. J., Maron, F. W., and Rossini, F. D.: J. Research Natl. Bur. Standards **46,** 106, Research Paper No. 2181 (1951).
	Butane, 1-butene, ethane, ethylene, hexane, 1-hexene, pentane, 1-pentene, propane, propylene	Thacker, C. M., Folkins, H. O., and Miller, E. L.: Ind. Eng. Chem. **33,** 584 (1941).
	Butane, ethane, hexane, pentane, propane	Prosen, E. J., Pitzer, K. S., and Rossini, F. D.: J. Research Natl. Bur. Standards **34,** 403, Research Paper No. 1650 (1945).
	1-Butene, ethylene, 1-hexene, 1-pentene, propylene	Prosen, E. J., and Rossini, F. D.: J. Research Natl. Bur. Standards **36,** 269, Research Paper No. 1702 (1946).
Heat of hydrogenation	Butane, 1-butene	Prosen, E. J., Maron, F. W., and Rossini, F. D.: J. Research Natl. Bur. Standards **46,** 106, Research Paper No. 2181 (1951).
	1-Butene, ethylene, 1-hexene, 1-pentene, propylene	Prosen, E. J., and Rossini, F. D.: J. Research Natl. Bur. Standards **36,** 269, Research Paper No. 1702 (1946).
	1-Butene, propylene	Kistiakowsky, G. B., Ruhoff, J. R., Smith, H. A., and Vaughan, W. E.: J. Am. Chem. Soc. **57,** 876 (1935).
	Ethylene	Teller, E., and Topley, B.: J. Chem. Soc. **1935,** 876. v. Wortenberg, H., and Krause, G.: Z. physik. Chem. **A151,** 105 (1930). Kistiakowsky, G. B., Romeyn, H., Jr., Ruhoff, J. R., Smith, H. A., and Vaughan, W. E.: J. Am. Chem. Soc. **57,** 65 (1935).
	General	Melville, H. W.: Science Progr. **32,** 515 (1938).
Heat of vaporization	Butane, ethane, hexane, pentane, propane	Anon.: Oil Gas J. **23,** 115 (1945).
	Butane, propane	Dana, L. I., Jenkins, A. C., Burdick, J. N., and Timm, R. C.: Refrig. Eng. **12,** 387 (1926).

TABLE 1—*Concluded*

Property	Gases	References
Heat of vaporization—*continued*	Ethane	Satterly, J., and Patterson, J.: Trans. Roy. Soc. Can. **13**, Sect. iii, 123 (1919).
	Pentane	Aries, E.: Compt. rend. **168**, 444 (1919).
Spectrum	Butane, ethane, hexane, pentane, propane	DesLandres, H.: Compt. rend. **213**, 98 (1941).
	Butane, ethane, hexane, propane	Roberts, R. H., and Johnsen, S. E. J.: Anal. Chem. **20**, 690 (1948).
General	Butane, ethane, hexane, pentane, propane	Penson, W. B., and Pimentel, G. C.: J. Am. Chem. Soc. **75**, 532 (1953). Sage, B. H.: Ind. Eng. Chem. **42**, 631 (1950).
	Butane, propane	Finley, G. H.: *The Handbook of Butane-Propane Gases*, 2nd edition. Western Gas Co., Los Angeles, California (1935). Denny, L. C., and Wickstrom, H. W.: *The Handbook of Butane-Propane Gases*, 3rd edition. Jenkins Pubs., Los Angeles, California (1947).
	Ethylene, propylene	Coffin, C. C., and Maass, O.: Trans. Roy. Soc. Can. [3] **21**, No. 3, 33 (1928).