Catalyst Nature and Frequency Effects on the Oligomerization of Methane via Microwave Heating

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Microwave-induced oligomerization of methane has been extensively investigated for many years. Very important factors that control the oligomerization of methane were studied, including factors such as microwave frequency, time the materials are in the reaction zone, and nature of the catalysts. Activated carbon, iron, and nickel were tested for the activation of methane. Experiments were conducted at fixed frequencies of 2.4 and 4.6 GHz and at variable frequency over the range of 2−7 GHz with a sweep rate of 0.5 s. The power used for all experiments was 500 W. Changes in product distribution, because of changes in frequency, have been observed, most probably because of the different transient heating patterns that occur at different values of frequency. Oligomers ranging from C_{2s} to C_{8s} compounds were obtained, in which C_{2s} compounds were the most abundant. Benzene was produced, with a selectivity of 28%.

I. Introduction

Natural gas fields, which are prospected and developed under conditions somewhat similar to those of oil, could have a dominant role in the next decade. The proven world reserves of oil seem approximately constant, whereas those of natural gas are regularly increasing and could meet the world consumption for 300 years at the present level of usage.[1]

Natural gas is essentially methane (83−97 vol %, depending on the origin) and, therefore, difficult to liquefy and quite chemically unreactive. Methane is thermodynamically stable, with respect to its elemental components. The reactions to make other hydrocarbons, all of which are less stable than methane at ∼1000 °C, have unfavorable free energies of reaction and are strongly limited by equilibrium. They need considerable energy input; therefore, temperatures of >1000 °C are required to transform methane to benzene, acetylene, ethylene, and ethane. At 550 °C, hydrogen and carbon are more stable.[2] Therefore, it appears that natural gas (or, more precisely, methane) is an interesting source of petrochemicals, but its use and transformation are difficult and expensive. Methane is a very valuable fuel, from the environmental point of view, and, so far, the production of energy is its main use. However, a more rational application of this resource is using methane to make either petrochemical or gasoline components.

Microwave energy interacts with materials at the molecular level. During the microwave heating of a dielectric, internal electric fields are generated within the material. Such fields can produce translation of electrons and ions and cause rotation of charged species. This movement can be opposed by friction, inertia, and other forces that can lead to attenuation of the electric fields and to volumetric heating of the material.[3] The ability of the microwave electric field to polarize molecules, and the ability of these molecules to follow the rapid reversal of the electric field, result in the conversion of electromagnetic energy to heat within the irradiated material.

Microwave heating has been investigated for several years over the past decade to activate methane catalytically, to produce more-valuable products (such as higher hydrocarbons and hydrogen).[4−6] No other heterogeneous catalytic studies using microwave radiation and involving either direct or indirect frequency effects, the nature of the catalyst, or the irradiation time have been published. In this work, we studied the systematic effects of the frequency, the nature of the catalyst, and the irradiation time on the conversion and product distribution of methane oligomerization via microwave heating.

II. Experimental Section

A. Microwave Apparatus and Gas System. The oligomerization of methane was performed in a flow reactor system. A Lambda Technologies variable-frequency unit (Vari-Wave model LT-502Xb) was used for the experiments at different frequencies. Microwave radiation was transmitted from the magnetron to the cavity by means of a high-power coaxial cable. This coaxial line is connected to a high-power coax-to-waveguide (WRD-250) and finally to a broadband microwave cavity launcher. The operational power range for this unit is 0−500 W. The incident center frequency can be varied over a range of 2.4−7.0 GHz, with a bandwidth sweep rate of 100 to 0.1 s.

The processing cavity is a 12 in. × 15 in. × 12 in. cavity manufactured from stainless steel. The cavity is accessed from the front by way of a swing door that provides a positive locking mechanism and enables the unit to have multiple microwave seals. Unless the door is properly sealed, microwave energy cannot be introduced into the processing cavity. The reactor, a straight, $^3/_8$-in. quartz tube, was mounted horizontally inside the cavity. CH_4 gas, as well as helium diluent gas, was mixed in a panel when necessary and flowed into the quartz reactor. Details of the experimental setup can be seen in Figure 1. High purity grade methane (obtained from Matheson) and ultrahigh-purity helium (purchased from Airgas) were used.

Various frequencies and modes were produced in the microwave cavity. This cavity allows TM_{01n} modes ($n = 2−9$)

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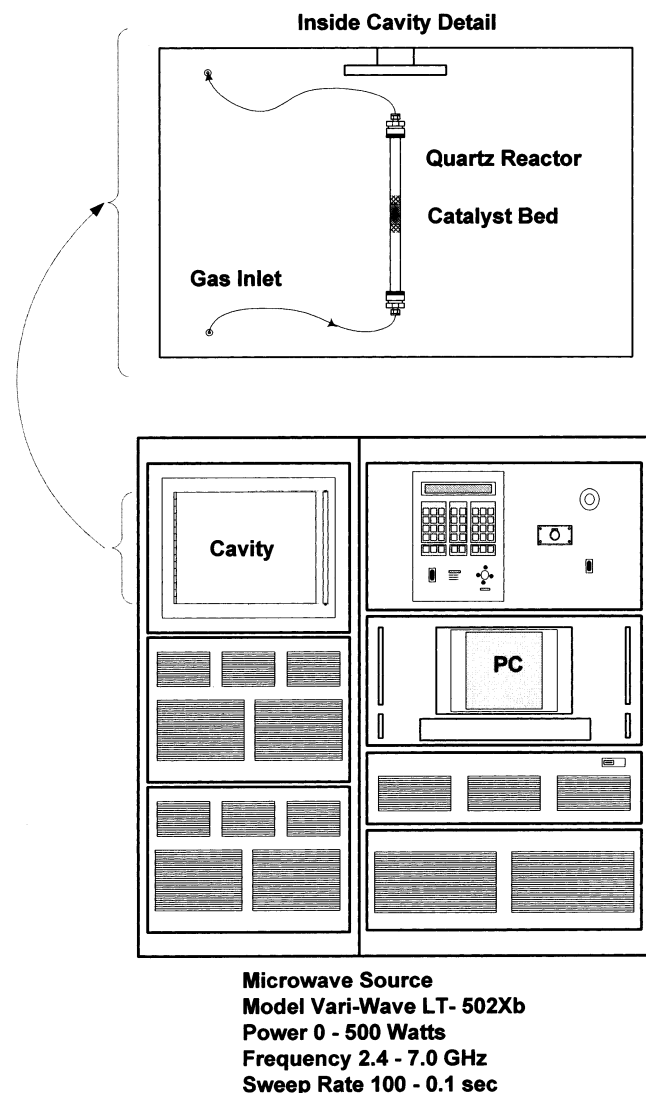
Figure 1. Microwave applicator and reactor.

over a frequency range of 2.4−7.0 GHz. The cavity was designed by Lambda Technologies to have different modes for each frequency. Experiments were performed at 2.4 GHz (TM$_{012}$), 4.6 GHz (TM$_{018}$), and a variable frequency within a range of 2.4−7.0 GHz with a sweep rate of 0.5 s. Thermal paper was inserted into the microwave cavity and used to map the heating patterns of the various modes.

B. Catalyst. Three different catalysts were tested in the oligomerization of methane. Most of the materials were purchased from different companies, and they were used as received. Nickel powder with a morphology resembling that of branched filaments was used for these experiments. The diameter of the filaments was 2.5 μm, as measured by scanning electron microscopy (SEM).[7] Clusters of nickel metal particles were aggregated to form the filaments. Activated carbon (MAXSORB, produced by Kansai Coke and Chemicals Co.) was also used. MAXSORB is a very high surface area (2200 m^2/g) activated carbon, produced from petroleum coke and coconut shell char, which results in a low ash content material (<2%).[8] X-ray diffraction analysis suggested that the activated carbon was entirely amorphous. The other catalyst tested was iron powder. As reported by Marun et al.,[9] the dielectric properties[10] of these materials do not seem to be an important factor in the oligomerization of methane via microwave heating. Approximately 100 mg of catalyst was placed inside the quartz

reactor. The catalyst bed was plugged at both ends with high-purity quartz wool. The catalytic quartz reactor was placed inside of the applicator, so that the material was positioned in the center of the cavity. To purge the atmosphere, helium gas was flowed through the reactor for 30 min prior to starting the microwave absorption.

The experiments were conducted in a 3:1 mixture of helium and pure methane, respectively, at atmospheric pressure, at a flow rate of 12 mL/min. Samples for analyses were taken after 3 and 20 min of irradiation at 500 W. Each experiment at different frequencies was performed with a fresh catalyst load.

C. Product Analysis. Products were trapped using a four-way gas sampling valve and analyzed using a Hewlett Packard model HP5890 Series II chromatograph that was equipped with a mass detector. Details about the gas chromatography/mass spectrometry (GC/MS) procedure have been published elsewhere.[11]

A carbon balance was used to estimate total conversion. All the compounds, multiplied by the number of C atoms present in that molecule, were summed. We report selectivities (C_{is}) as the selectivities toward the sum of compounds with a specific "i" number of C atoms.

The compounds analyzed using GC were as follows: methane, carbon dioxide, and carbon monoxide; ethylene, acetylene, and ethane (C_{2s} compounds); propene, propane, 1,2-propadiene, and propyne (C_{3s} compounds); 2-methylpropane, 2-butene, 1-buten-3-yne, 1,2-butadiene, butadiyne, 1-butyne, and 2-butyne (C_{4s} compounds); 3-pentene-1-yne (C_{5s} compounds); benzene (C_{6s} compound); toluene (C_{7s} compound); and ethylbenzene, ethynylbenzene, and ethenylbenzene (C_{8s} compounds).

The conversion in percentage is calculated as follows:

$$X(\%)_{CH_4} = (C_T - C_{CH_4 exit})/C_T \times 100 \qquad (1)$$

where C_T is the sum of the concentration of each compound (C_i) multiplied by the number of C atoms present in the molecule i (n_i), i.e.,

$$C_T = \sum n_i C_i \qquad (2)$$

The selectivities in percentage toward compounds with an "i" number of C atoms (S_i) were calculated using eq 3:

$$S_i (\%) = (\sum n_i C_i)/(C_T - C_{CH_4 exit}) \times 100 \qquad (3)$$

III. Results

Microwave-induced oligomerization of methane was performed at two different frequency levels (2.40 and 4.60 GHz). Variable-frequency reactions were also conducted by varying the frequency from 2.4 to 7.0 GHz with a sweep rate of 0.5 s. The effects of microwave frequency, irradiation time, and nature of the catalyst on the oligomerization of methane were studied. A summary of the results can be seen in Table 1.

No visible arc formation was detected during these runs, and the catalyst bed remained the same after reaction (checked using SEM photographs). In general, C_{2s} compounds were the most abundant products for the majority of the cases. For iron powder, the amount of carbon monoxide and carbon dioxide was also important. Among C_{2s} compounds, the product distribution regarding abundance was ethane, ethylene, and acetylene, where acetylene was the least abundant. Benzene was also produced, with a maximum selectivity of 28%. As we reported previously,[12] there are some absorbed species on the surface of the

TABLE 1: Summary of Results

frequency (GHz)	time (min)	conversion (%)	selectivity (%)						
			C2's	C6's	CO_2	CO	ethylene	acetylene	ethane
Nickel Catalyst, CH_4:He Feed, 3:9 mL/min Flow Rate, 500 W of Power									
2.4	3	4.1	81	5	3	7	0	0	81
	20	5.3	55	1	6	0	0	0	55
4.6	3	6.1	83	1	0	8	49	0	34
	20	8.5	72	10	0	12	26	16	30
V.F.[a]	3	17.4	83	2	6	0	0	0	83
	20	14.2	78	2	10	0	0	0	78
Iron Catalyst, CH_4:He Feed, 3:9 mL/min Flow Rate, 500 W of Power[b]									
2.4	3	10.8	21	0	35	42	9	0	12
	20	2.9	12	0	18	70	0	0	12
4.6	3	15.0	35	5	15	21	8	0	27
	20	9.0	23	5	10	35	2	0	21
V.F.[a]	3	10.0	68	12	11	5	3	0	65
	20	8.0	50	10	8	28	2	0	48
Activated Carbon Catalyst, CH_4:He Feed, 3:9 mL/min Flow Rate, 500 W of Power									
2.4	3	24.3	60	28	0	4	50	9	1
	20	30.5	58	25	0	8	43	8	7
4.6	3	31.2	72	25	0	2	58	12	2
	20	45.1	56	19	0	23	41	9	6
V.F.[a]	3	28.6	70	26	0	2	51	10	9
	20	38.9	62	21	0	15	47	9	6

[a] Variable frequency over a range of 2.4–7.0 GHz, with a sweep of 0.5 s. [b] For a frequency of 2.4 GHz, the power was 1130 W. Conversion below 700 W was zero.

catalyst, such as water, carbon monoxide, carbon dioxide, and molecular oxygen.

A. Frequency Effect on Nickel Catalyst. When irradiating nickel powder with low-frequency microwave energy at 500 W, the conversion increased slightly from 4% to 5% as time increased from 3 min to 20 min. Ethane is the major and only C_2 product, with a maximum selectivity of 81%. After 20 min at 500 W, the amount of produced ethane decreased to 55%, whereas some C_{3s} and C_{4s} compounds started to form. Figure 2a shows the detailed product distribution for these conditions.

When the microwave frequency was increased to 4.6 GHz, the amount of methane that was converted to products was higher than that of the low-frequency experiment, with an 8% conversion after 20 min of irradiation. Figure 2b shows the product distribution for the high-frequency experiments. Ethylene is formed at high microwave frequency, with a selectivity of 49%. Acetylene and benzene also became important after 20 min of irradiation, with a selectivity of 16% and 10%, respectively.

As seen in Figure 2c, the conversion of methane doubles when the reaction is performed at variable frequency. The frequency was set to vary over a range of 2.4–7.0 GHz within a range of 0.5 s at 500 W. The reaction was very selective to ethane, with a maximum selectivity of 83%. Carbon dioxide was the second-major product, with a selectivity of 15% after 20 min of irradiation.

B. Frequency Effect on Iron Catalyst. Panels a, b, and c of Figure 3 show the product distribution of the oligomerization of methane using iron powder for low-frequency, high-frequency, and variable-frequency experiments, respectively. At low frequency and 500 W, iron catalysts were not active for the investigated reaction. However, after 700 W, some carbon monoxide and carbon dioxide started to form, with a conversion of <1%. To obtain a reasonable conversion of ~10%, it was necessary to increase the incident power to 1130 W. Although carbon monoxide and carbon dioxide were still the major products under these conditions, C_{2s} compounds were also produced. The maximum selectivity reached for C_{2s} compounds was 21% after 3 min of irradiation, with ethane as the major C_2 product.

Interestingly, when the microwave frequency was increased to 4.6 GHz, the iron catalyst became active for the reaction at 500 W, with a conversion of 15%. When the time increased to 20 min, the conversion decreased to 9%. After 3 min of irradiation, the selectivity toward C_{2s} compounds reached 35%, with ethane as the major product. A considerable amount of C_{8s} compounds were formed, with a maximum selectivity of 18%. At 20 min, the selectivity toward C_{2s} compounds decreased to 23%, which is almost double that of the low-frequency experiments.
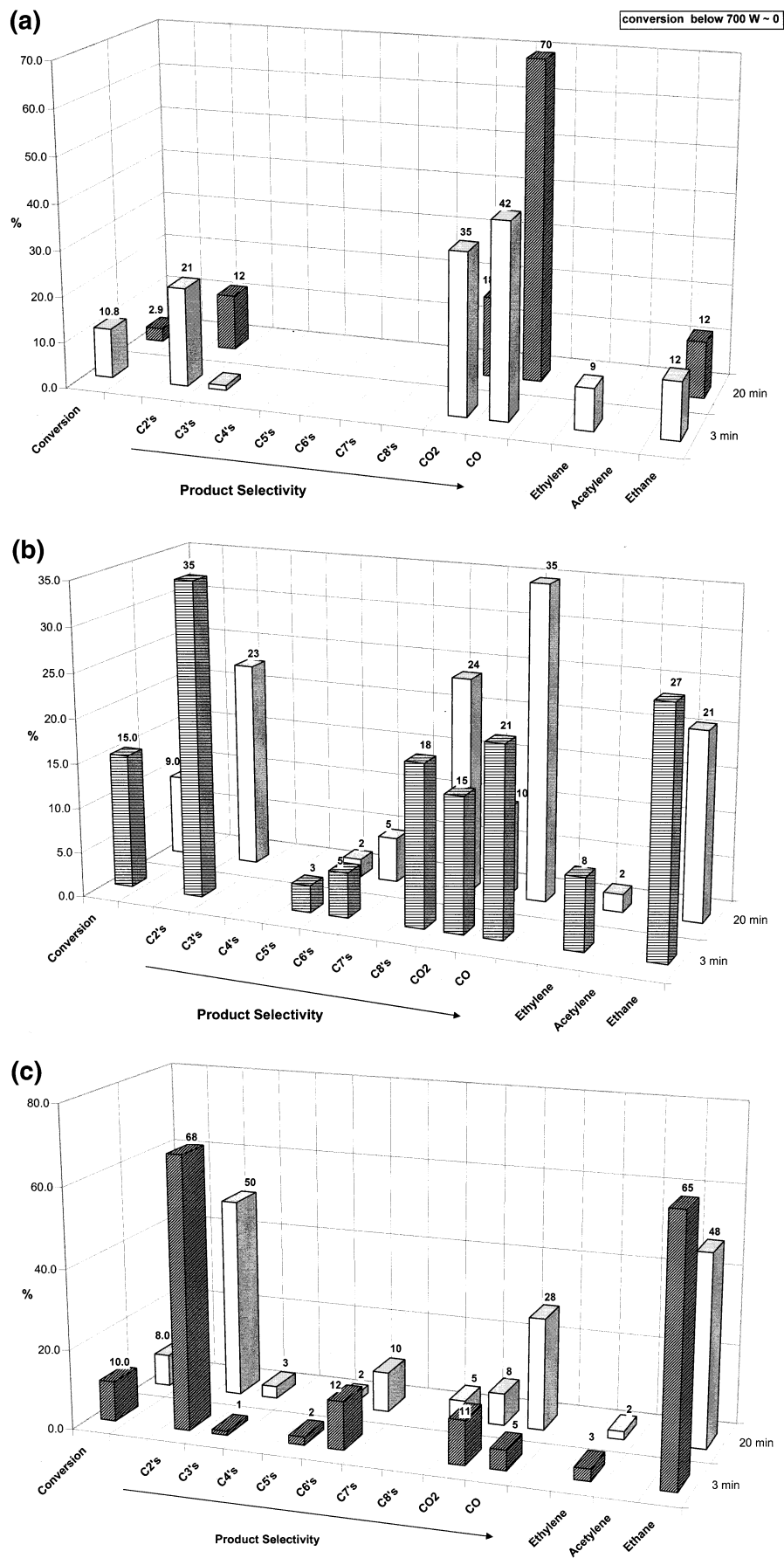
When experiments were performed at variable frequency and 500 W, the conversion was comparable to that at low frequency (10%). However, the selectivity toward C_{2s} compounds more than triples that of the those C_{2s} compounds at low frequency (68%). The major product was ethane, with a maximum selectivity of 65%. Benzene was also formed, with a selectivity of 12% after 3 min of irradiation. Selectivity toward carbon monoxide and carbon dioxide decreased considerably, to 11% and 6%, respectively.

C. Frequency Effect on Activated Carbon Catalyst. Unlike the other catalyst, activated carbon was very active for the oligomerization of methane at low power (500 W). Figure 4a shows the results for the reaction at low frequency. The conversion of methane to products increased from 24% to 30% as the irradiation time increased from 3 min to 20 min. The major products were C_{2s} compounds, with 60% selectivity. Distinctive from the other catalyst, the product distribution among C_{2s} compounds was ethylene, acetylene, and ethane, where ethylene was the most abundant and ethane the least abundant. The second-major product was benzene, with a maximum selectivity of 28% after 3 min of irradiation.

When the frequency increased to 4.6 GHz, conversion expectedly increased to 31%. In contrast to the other experiments, conversion increased while the irradiation time increased, as seen in Figure 4b. Although the selectivity toward C_{2s}
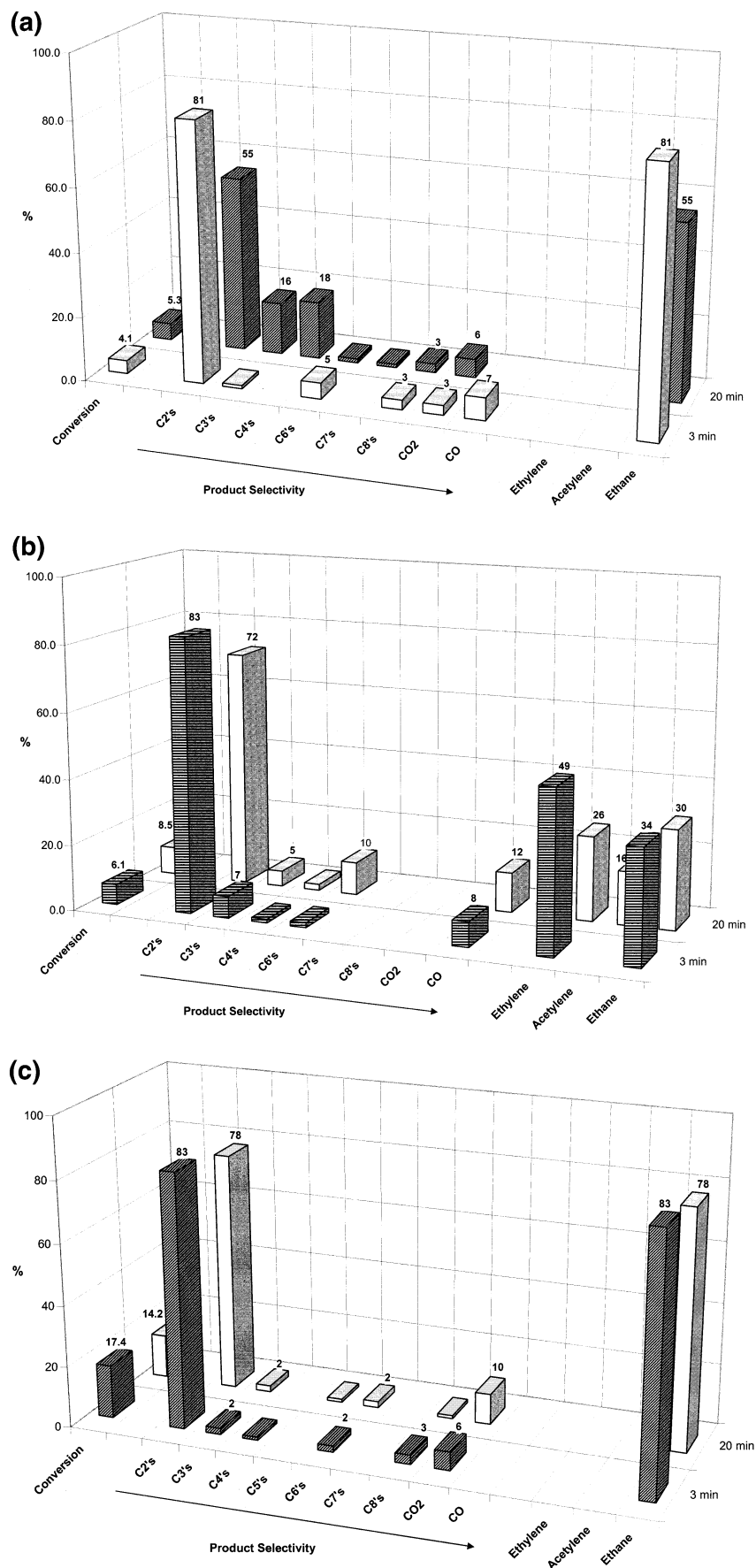
Figure 2. Effect of irradiation time on conversion and selectivity with nickel powder catalyst at 500 W using fixed frequencies ((a) 2.4 and (b) 4.6 GHz) and a variable frequency ((c) 2.4−7.0 GHz, with a sweep rate of 0.5 s).

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Figure 3. Effect of irradiation time on conversion and selectivity with iron powder catalyst at (a) 1130 W, using a fixed frequency of 2.4 GHz; (b) 500 W, using a fixed frequency of 4.6 GHz; and (c) 500 W, using a variable frequency of 2.4–7.0 GHz with a sweep rate of 0.5 s.
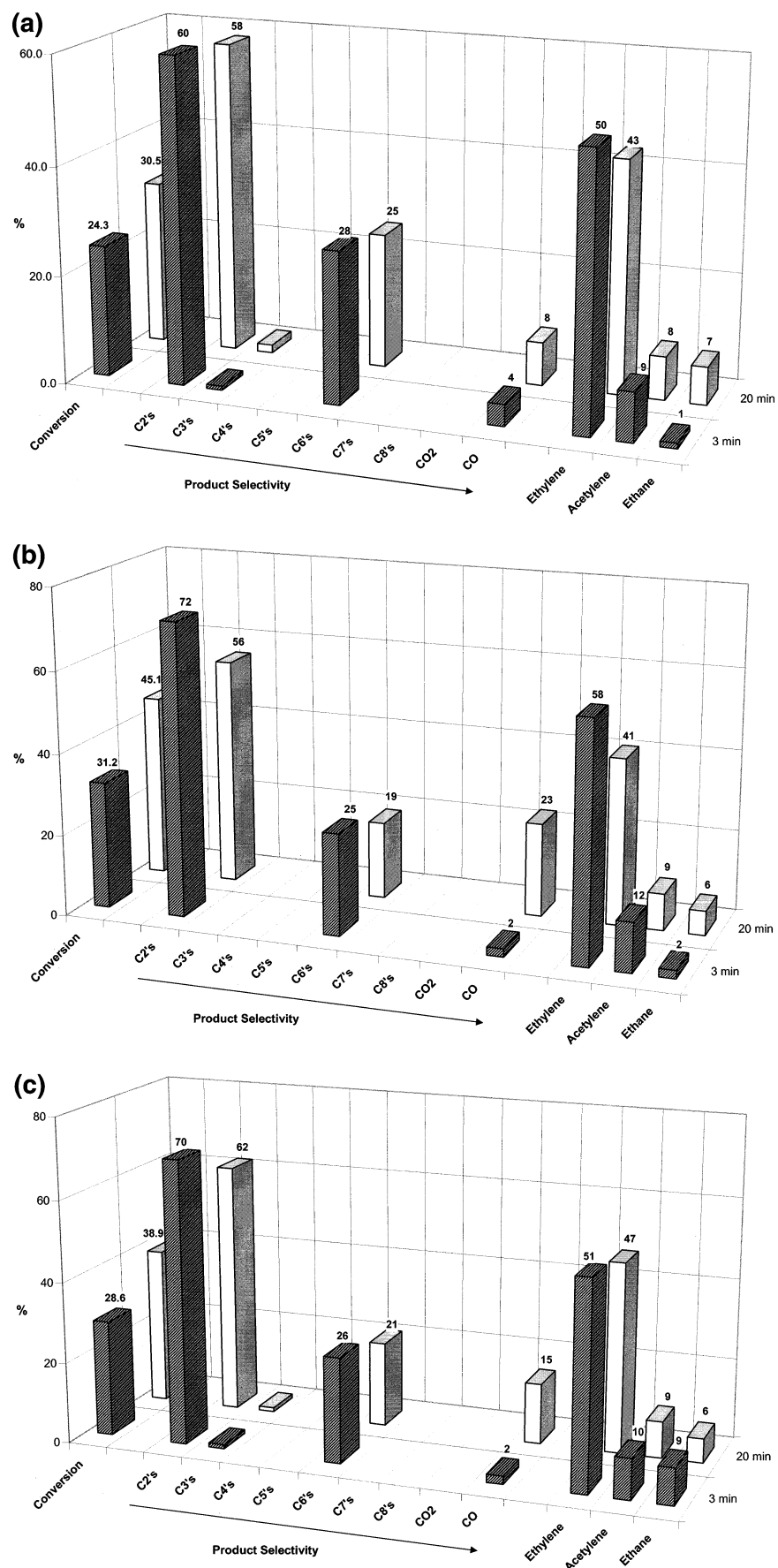
Figure 4. Effect of irradiation time on conversion and selectivity with activated carbon catalyst at 500 W using fixed frequencies ((a) 2.4 and (b) 4.6 GHz) and a variable frequency ((c) 2.4−7.0 GHz, with a sweep rate of 0.5 s).

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compounds increased to 72%, the product distribution remained fairly similar to that at low frequency. The selectivity toward benzene slightly decreased, from 25% to 19%, as the irradiation time increased from 3 min to 20 min.

Figure 4c shows the product distribution for the reaction at variable frequency. The conversion of methane under this condition was between that observed for the low- and high-frequency experiments, with a maximum of 39%. Again, the product distribution remains rather similar to those observed in previous experiments. The major product was ethylene, with a selectivity of 51%, followed by benzene, with a selectivity of 26%.
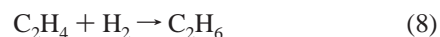
IV. Discussion

Three different catalysts were tested. Nickel powder, iron powder, and activated carbon were used for the activation of methane via microwave heating at 500 W. Interestingly, activated carbon was proven to be the most active catalyst under our system conditions. The maximum conversion was reached using activated carbon (45%) at a fixed microwave frequency of 4.6 GHz after 20 min of irradiation. Also, the amount of benzene was comparatively high (28%) when activated carbon was used as a catalyst. Although ethane was the major C_2 product for both the nickel and iron catalysts, ethylene was the major product for activated carbon. Acetylene was only produced when activated carbon was used as a catalyst. The formation of benzene may be explained by the well-known cyclization reaction. Bamwenda observed similar reactions, as reported in a previous publication.[13] This is in agreement with the fact that, for our system, the selectivity to benzene increases as the selectivity to acetylene also increases.

Activated carbon absorbs microwave radiation more efficiently than nickel powder.[14] Consequently, the temperature reached by activated carbon is higher than that reached by nickel and iron.[11] This condition of higher temperature on the active sites would favor the conversion of methane on activated carbon.

On the other hand, the surface area of activated carbon is 2 orders of magnitude larger than the surface area of nickel and iron powder. The conversion and activity of these materials are not proportionally related to the surface area of the catalysts we used. One possible explanation for the lack of a direct relationship between surface area and conversion might be the effect of the interaction of microwaves with surface functional groups such as OH groups and other catalytic intermediates (such as CH_3, CH_2, CH, and other species). Such functional groups may also influence the adsorption of the reactant species.

The reaction pathway for the catalytic oligomerization of methane using microwave heating most likely proceeds via the formation of free radical intermediates, as shown by Wan et al.[15] The primary decomposition fragments from methane are :CH2 and :CH radicals, which recombine on very hot surfaces to yield acetylene, whereas, on cooler surfaces, methane leads to the formation of ethylene. Ethane is favored for even cooler surfaces than those of ethylene. This finding is in agreement with our results, as seen in Figures 2−4. The figures show the formation of acetylene and ethylene for reactions with activated carbon, the most active catalyst. Ethylene was also formed using the other catalysts when working at the most energetic frequency (4.6 GHz). Reactions 4−11 summarize some of the proposed reactions.

$$CH_4 \rightarrow \cdot CH \tag{4}$$

$$CH_4 \rightarrow :CH_2 + H_2 \tag{5}$$

$$2CH_4 \rightarrow C_2H_4 + 2H_2 \tag{6}$$

$$2CH_4 \rightarrow C_2H_2 + 3H_2 \tag{7}$$

$$C_2H_4 + H_2 \rightarrow C_2H_6 \tag{8}$$

$$3C_2H_2 \rightarrow C_6H_6 \tag{9}$$

The presence of H_2 from reaction 5 leads to the hydrogenation of olefins and the production of ethane, as shown in reaction 8. The highly reactive methylene (:CH_2) intermediate can be used to elucidate the formation of ethylene and acetylene, as indicated in reactions 6 and 7. The formation of benzene may be explained by the well-known cyclization reaction given in reaction 9. Another viable mechanism for ethylene and acetylene formation could involve the dehydrogenation of secondary hydrocarbons:

$$C_2H_6 \rightarrow C_2H_4 + H_2 \tag{10}$$

$$C_2H_4 \rightarrow C_2H_2 + H_2 \tag{11}$$
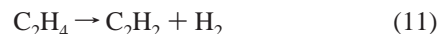
As discussed in a previous publication,[12] the effects of frequency on product distribution for the oligomerization of methane via microwave heating might be related to the diverse transient heating patterns that are generated by transverse magnetic modes at different frequencies. The quenching of intermediates has an important role in ethylene formation; thus, different transient heating patterns would affect the environment in which the reaction is taking place, consequently affecting the selectivities of the final products.

The general trend found was that activity increases as frequency increases. Previous studies[12] correlate the microwave radiation frequency with the activity of the nickel catalyst. As the radiation frequency increases, the particles trying to align with the electromagnetic field are going to move more rapidly, generating more heat than movement at lower frequency. Therefore, at the same power and irradiation time, the energy density in the material is higher for high-frequency radiation. The higher energy density in the material produces different and more-energetic transient heating patterns. When the temperature reached by the active sites is too high, the reaction leads to undesirable products, such as carbon monoxide and carbon dioxide.

The two parameters that define the dielectric properties of materials and concomitant heating patterns are dielectric constant (ϵ') and dielectric loss (ϵ''). The dielectric constant ϵ' describes the ability of the molecules to be polarized by the electric field. At low frequencies, this value will reach a maximum at the maximum amount of energy that can be stored in the material. As the frequency increases, the value of ϵ' decreases, as shown by Mingos et al.[16] The dielectric loss ϵ''' measures the efficiency at which microwave energy can be converted to heat and goes through a maximum as the frequency increases.[16] The relationship between these two parameters (ϵ''/ϵ') defines the dielectric loss tangent, tan δ, which measures the ability of a material to convert electromagnetic energy to heat at a given frequency and temperature. The tan δ value also goes through a maximum as the frequency increases. Therefore, higher formation of benzene−acetylene at higher frequency in our experiments can be explained by the increase of the dielectric loss as the frequency increases before going through the maximum. Greater

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tan δ values are directly related to greater temperatures, which are, consequently, related to ethylene and acetylene formation.[15]

When the frequency of the microwave radiation varies over a period of time, the transient heating patterns vary constantly, not allowing the formation of hot spots. Therefore, heating is completely uniform throughout the material. Although the variable-frequency microwave radiation can be highly energetic for activating methane, this does not lead to a large amount of undesirable products. From Figures 2−4 (panels b and c), it can be seen that, even though conversions are comparable for high- and variable-frequency experiments, the selectivities toward carbon dioxide and carbon monoxide are somewhat higher for the experiments at 4.6 GHz.

There are different sources of oxygen that account for the formation of carbon monoxide and carbon dioxide detected in the products. Activated carbon, iron, and nickel catalysts were used as received for the experiments. Temperature-programmed desorption analysis performed in previous studies[12] revealed the presence of oxygenated species absorbed on the surface of the catalysts, such as CO, CO_2, O_2, and H_2O. These absorbed species on the surface supply the oxygen for the formation of undesirable products.

V. Conclusion

The microwave heating oligomerization experiments have shown that nickel, iron powder, and activated carbon can act as selective catalysts for oligomerized products of methane. Oligomers ranging from C_2 to C_6 hydrocarbons have been prepared in good selectivity, depending on the nature of the catalyst, microwave frequency, irradiation time, and power levels used in the microwave reactor. Selectivities as high as 28% toward benzene were achieved when activated carbon was used as a catalyst. The changes in product distribution when using different microwave frequencies are most likely due to different transverse modes. Different transverse modes generate different transient heating patterns, consequently changing the dielectric properties of the catalyst (i.e., the dielectric loss and the dielectric constant).

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