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APPENDIX

APPENDIX A

Assumptions and calculation procedures

To facilitate the calculations, some valid assumptions were made as follows:

1) All the gaseous behaviors obey the ideal gas law.
2) Pressure drop across the system is very small and can be negligible.
3) The pressure in the system equals atmospheric pressure (1 atm.)
4) The temperature change due to the reactions is very small and can be negligible. All experiments are assumed to be carried out at the ambient temperature.
5) The flow rate change across the reactor due to the variation in the gaseous compositions during the reaction time is very small and is assumed to be negligible.

The total molar flow rate of the gaseous stream can be calculated from the following equation:

$$N = \left(\frac{P}{RT}\right) \times q$$

where

P = Total pressure of the system (i.e., 1 atm)

q = Total volumetric flow rate (determined by using soap bubble meter)

R = Gas constant

T = Absolute ambient temperature (K)

With this, the molar flow rate of each component can also be determined by multiplying its percent volume derived from the GC analysis with the total molar flow rate.

The conversion is defined generally as:

$$\%Conversion = \left(\frac{\text{Mole reactant In} - \text{Mole reactant Out}}{\text{Mole reactant In}} \right) \times 100$$

The percent selectivity of each product is, however, defined strictly on the basis of the amount of carbon converted from the reactant into any specified products. In this case, the product selectivity was defined as follows,

When methane was the reactant,

$$\%C_2\ Selectivity = 2 \times \left(\frac{\text{Mole of } C_2 \text{ produced}}{\text{Mole of } CH_4 \text{ converted}} \right) \times 100$$

$$\%C_3\ Selectivity = 3 \times \left(\frac{\text{Mole of } C_3 \text{ produced}}{\text{Mole of } CH_4 \text{ converted}} \right) \times 100$$

and

$$\%C_4\ Selectivity = 4 \times \left(\frac{\text{Mole of } C_4 \text{ produced}}{\text{Mole of } CH_4 \text{ converted}} \right) \times 100$$

Here, the number "2", "3" or "4" in the above equations can be viewed as the ratio of the number of carbon atoms in the product divided by the number of the carbon atoms in the reactant. The product selectivity for the case of either ethane or propane was the reactant were also defined in the

similar ways except for the change in the factor of the carbon ratio. Thus, as a general terminology, the product selectivity is defined as:

$$\%C_P \; Selectivity = \left[\frac{P}{R}\right] \times \left(\frac{\text{Mole of } C_P \text{ produced}}{\text{Mole of } C_R \text{ converted}}\right) \times 100$$

where

P = number of carbon atom in product

R = number of carbon atom in reactant (i.e., R=1 for the case of methane)

APPENDIX B

Experimental Data

Table B.1 Methane conversion and product selectivities for pure methane inlet system at different voltages (total inlet flow rate = 20 ml/min)

Voltage (V)	Methane Conversion (%)	Product Selectivities (%)			
		Ethylene	Ethane	Propane	Butane
3750	0.470	0.000	21.332	0.000	0.000
4500	7.980	4.574	33.701	0.966	4.927
5000	8.557	4.254	36.168	11.741	5.938
6250	13.814	1.701	36.353	16.192	7.430
8750	22.185	1.762	33.691	15.707	9.821
10937	24.947	1.713	33.139	15.961	10.267

Table B.2 Methane conversion and product selectivities for pure methane inlet system at different voltages (total inlet flow rate = 40 ml/min)

Voltage (V)	Methane Conversion (%)	Product Selectivities (%)			
		Ethylene	Ethane	Propane	Butane
4500	2.330	10.792	34.660	0.000	0.000
6250	6.443	3.030	39.231	14.923	5.165
8750	9.398	2.655	39.424	16.388	6.759
10937	10.493	2.667	39.463	16.420	7.383

Table B.3 Methane conversion and product selectivities for pure methane inlet system at different flow rates (applied voltage = 6250 V)

Voltage (V)	Methane Conversion (%)	Product Selectivities (%)			
		Ethylene	Ethane	Propane	Butane
6250	23.052	1.907	34.566	15.670	10.136
6250	9.242	3.091	34.706	13.488	5.480
6250	4.057	3.642	25.355	8.423	0.000

Table B.4 Methane conversion and product selectivities for pure methane inlet system at different voltages (total flow rate = 20 ml/min; immediate voltage rising mode)

Voltage (V)	Methane Conversion (%)	Product Selectivities (%)			
		Ethylene	Ethane	Propane	Butane
4500	8.844	4.787	35.165	11.336	6.052
6250	23.052	1.907	34.566	15.670	10.136
7500	26.744	1.616	33.160	15.765	10.647

Table B.5 Methane conversion and product selectivities for the inlet system of methane and helium mixture at different voltages (total inlet flow rate = 20 ml/min; CH_4/He = 1:1)

Voltage (V)	Methane Conversion (%)	Product Selectivities (%)			
		Ethylene	Ethane	Propane	Butane
3750	13.137	0.000	27.641	12.123	4.673
5000	30.696	0.000	21.449	10.187	4.267
6250	29.230	0.000	30.365	15.258	8.425

Table B.6 Methane conversion and product selectivities for the inlet system of methane and helium mixture at different voltages (total inlet flow rate = 20 ml/min; CH_4/He = 3:1)

Voltage (V)	Methane Conversion (%)	Product Selectivities (%)			
		Ethylene	Ethane	Propane	Butane
3750	5.356	4.207	32.439	7.989	2.879
5000	9.757	2.282	39.275	16.434	7.649
6250	9.818	2.219	39.343	16.538	7.628

Table B.7 Methane conversion and product selectivities for the inlet system of methane and ethane mixture at different voltages (total inlet flow rate = 20 ml/min)

Ethane Content in Feed (%)	Voltage (V)	Methane Conv. (%)	Ethane Content in Outlet Stream (%)	Product Selectivities (%)		
				Ethylene	Propane	Butane
10.92	4500	8.46	10.51	10.38	24.52	12.46
10.92	6250	24.08	9.69	3.53	25.98	18.57
20.40	4500	7.77	17.72	17.40	43.42	30.03
20.40	6250	23.05	13.70	5.41	36.59	28.11

Table B.8 Ethane conversion and product selectivities for the inlet system of ethane and helium mixture at different voltages (total inlet flow rate = 20 ml/min; 20 % ethane in helium)

Voltage (V)	Ethane Conversion (%)	Product Selectivities (%)			
		Methane	Ethylene	Propane	Butane
5000	40.32	1.42	3.23	13.33	25.72
6250	49.66	2.90	3.35	13.79	24.19

Table B.9 Propane conversion and product selectivities for the inlet system of propane and helium mixture at different voltages (total inlet flow rate = 20 ml/min; 20 % propane in helium)

Voltage (V)	Propane Conversion (%)	Product Selectivities (%)				
		Methane	Ethylene	Ethane	Propylene	Butane
5000	36.72	2.44	5.55	11.81	5.22	11.86
6250	49.82	3.86	5.15	11.60	3.47	11.21

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