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PREPRINT

Lanthanum Cerium Manganese Hexaaluminate Combustion Catalysts for Compact Steam Reformers

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Abstract

Several substituted hexaaluminates were synthesized via a surfactant-mediated process and tested for their catalytic propane combustion activity. After calcination at 1100 °C, catalyst surface areas were as high as 78 m²/g. The best catalysts contained lanthanum, cerium, and manganese, with La_{0.6}Ce_{0.4}MnAl₁₁O₁₉ representing the optimum. A parallel plate catalytic combustion reactor with a catalyst supported on Fecralloy foam burned propane and JP-8 jet fuel with a low pressure drop, and demonstrated a sufficiently-uniform temperature profile, and stable self-sustained operation. This combustion reactor will be suitable for use in compact steam reformers producing hydrogen for mobile fuel cells.

Keywords

Hexaaluminate, manganese, lanthanum, cerium, catalytic combustion, propane, JP-8, surfactant, Fecralloy, foam

Introduction

Fuel cells promise to provide significant advantages over conventional diesel generator sets for mobile electric power applications. They can provide power more fuel-efficiently, more quietly, and with a smaller footprint and higher reliability than conventional diesel generators. The most practical fuel for fuel cells is hydrogen from the steam reforming of hydrocarbons.

The steam reforming reaction is fast and endothermic, so the rate of hydrogen production in the steam reforming reactor is generally limited by the rate of heat transfer from the heat source. When the heat source is an open-flame or radiant burner and the steam reformer is a packed bed, the rate of heat transfer is limited by wall film and bed resistances. Heat transfer can be improved by replacing the burner/ packed bed system with parallel channels containing Fecralloy foam inserts alternately coated with a combustion or reforming catalyst. In this way heat is transferred by conduction directly from the source to the sink, allowing for faster hydrogen production in a more compact reactor [1]. This approach, however, requires the development of active, stable combustion catalysts that can be coated directly onto Fecralloy supports. Also, the combustion reactor must be able to develop a relatively flat temperature profile along the axis of flow, to allow for adequate residence time of reactants in the heated section of the adjacent steam reforming channel.

Metal-substituted hexaaluminates are currently being developed for use as combustion catalysts in high temperature lean natural gas turbines in order to reduce NO_x emissions [2–22]. Unlike standard γ -alumina catalyst supports, they maintain substantial surface areas at temperatures exceeding 1000 °C and thus are well-suited for high temperature catalytic combustion. They have the general formula $\text{AM}_x\text{Al}_{12-x}\text{O}_{19}$ where A is a large cation from the alkaline earth metals or the lanthanides and M is a transition metal cation close in size to the Al^{3+}

cation. The A cation creates a layered structure that inhibits sintering while the M cation catalyzes combustion.

These experiments were designed to select a substituted hexaaluminate for use as a combustion catalyst in a compact steam reformer. Different catalyst formulations were synthesized as granules and tested in a packed bed reactor for catalytic activity and high-temperature stability in the combustion of propane/air mixtures. A catalyst formulation with superior activity was then tested as a washcoat on Fecralloy foam in a parallel plate catalytic combustion reactor. Both propane and JP-8 were used as fuels.

Experimental

The catalysts were synthesized using a modification of the surfactant-mediated procedure devised by Cho *et al.* [12]. Stoichiometric quantities of acetates of the A and M ions (Aldrich) and 10 g cetyltrimethylammonium chloride (25 wt. % in water, Aldrich) were mixed together; 25 g of alumina sol (10 % Al_2O_3 , 10-100 nm, Nissan) was gradually stirred in at a rate of 2.5 g every 2 min; 15 g urea (Fisher) was added, followed by stirring for 15 min and ultrasonic agitation for 1 h. The mixture was heated at 100 °C for 24 h to decompose the urea into carbon dioxide and ammonia. This increased the pH and caused the condensation of the sol. The mixture was calcined in air at 550 °C overnight and at 1100 °C for 6 h to form the hexaaluminate, drive off the ammonia, and burn off the surfactant and acetate. BET surface areas of catalyst granules were measured using a Micromeritics Flowsorb 2300.

A packed bed reactor was used to evaluate the catalysts. It was a 30.5 cm long, 0.64 cm OD, 0.48 cm ID alumina tube heated by a tube furnace. For the light-off curves, the reactor was fed 94 sccm air, 3 sccm propane, and 3 sccm neon. Gas samples were taken through a septum downstream of the furnace and analyzed by gas chromatography using a 30 meter Carboxen

1010 PLOT column (Supelco), argon carrier gas, and a thermal conductivity detector. The neon in the gas mixture provided an internal standard to correct for detector drift and variation in injection volume.

A good catalyst ($\text{LaMnAl}_{11}\text{O}_{19}$) was tested for stability by increasing the packed bed gas flows to 238 sccm air and 10 sccm propane. A Kanthal heating wire wrapped around the reactor initiated self-sustained catalytic combustion. Light-off curves with a feed of 94 sccm air, 3 sccm propane, and 3 sccm neon were acquired both before and after the test to detect catalyst deactivation.

A parallel plate catalytic combustion reactor was constructed from two stainless steel plates (7.62 cm x 3.56 cm x 0.16 cm), a stainless steel frame (7.62 cm x 3.56 cm x 1 cm with a 5 cm x 1 cm x 1 cm cavity), and two Fecralloy foam strip catalyst supports (Porvair, 40 pores/inch, 5% density, dimensions 5 cm x 1 cm x 0.5 cm) stacked inside the frame cavity. The assembly is pictured in Figure 1. The plates and frame were bolted together, with Resbond 907GF 2350 °F Adhesive and Sealing Putty being used as a sealant. The catalyst supports were washcoated with $\text{La}_{0.8}\text{Ce}_{0.2}\text{MnAl}_{11}\text{O}_{19}$ using the following procedure. The catalyst supports were sonicated in acetone for 1 h, oxidized in air at 900 °C for 24 h to grow an alumina scale for improved catalyst adhesion, dipped in a 12% slurry of $\text{La}_{0.8}\text{Ce}_{0.2}\text{MnAl}_{11}\text{O}_{19}$ in ethanol, and dried with a heat gun. Three dipping/drying cycles were performed for each foam support. After assembly, the reactor was fed 3000 sccm air and found to be free of leaks detectable by Snoop Liquid Leak Detector (Swagelok).

The parallel plate reactor's performance was characterized for the catalytic combustion of propane. Fifty sccm propane and 1190 sccm air (providing a stoichiometric fuel/air ratio) were fed to the reactor without preheating. Combustion was initiated by resistively heating the reactor

with coiled heat rope. Upon light-off, resistive heating was discontinued and the reactor was allowed to reach steady state. The temperature profile of the reactor was measured at four external points (corresponding to the bolt positions) in a line along the axis of flow with a type E thermocouple probe. The probe had a 1.5 mm diameter stainless steel sheath. The experiment was repeated with a feed of 14 ml/h JP-8 jet fuel (dispensed by an Isco LC-5000 syringe pump) and 3630 sccm air. Using $C_{11}H_{21}$ as an approximate molecular formula for JP-8, this is calculated to be 72% excess air.

Results and Discussion

Table 1 lists surface area measurements for the substituted hexaaluminate catalysts. As transition metal substitution increases, surface area tends to decrease. For $x = 0.5$ transition metal cations per hexaaluminate unit, surface area ranges from 65 to 78 m^2/g ; for $x = 1$, surface area ranges from 46 to 70 m^2/g ; for $x = 2$, surface area ranges from 27 to 62 m^2/g ; and the catalyst with $x = 3$ has a surface area of 12 m^2/g . This is consistent with other results in the literature. Artizzu-Duart *et al.* found that surface area declined from 14–20 m^2/g for $x = 1$ to 6–10 m^2/g for $x = 4$ [7]. Likewise, Wang *et al.* reported that surface area declined from 49 m^2/g for $x = 0$ to 7 m^2/g for $x = 6$ [18]. Transition metal-catalyzed sintering has been proposed as the cause of this phenomenon [12, 18].

Table 1 also lists temperatures for 50% and 100% conversion and partial oxidation products for the light-off curves depicted in Figures 2–4. Figure 2 shows the relative performance of substituted hexaaluminate catalysts that have previously been reported in the literature. Figure 3 shows the effect of substituting cerium for manganese in $LaMnAl_{11}O_{19}$ and Figure 4 shows the effect of substituting cerium for lanthanum in $LaMnAl_{11}O_{19}$.

In general, the catalysts with 1–3 manganese cations per hexaaluminate unit are most active, have the lowest light-off temperatures, and completely oxidize propane to carbon dioxide and water. The catalyst rankings by T_{50} and T_{100} are identical save for the position of $\text{BaMn}_3\text{Al}_9\text{O}_{19}$. Its high manganese content provides it with a higher intrinsic activity than the other catalysts and thus the lowest T_{50} . Near full conversion, however, mass transfer limitations between the gas bulk and the catalyst surface become more important and the catalyst's low specific surface area causes its light-off curve to lag.

The catalysts with 0.5 manganese cations per hexaaluminate unit are less active. $\text{LaCo}_{0.5}\text{Mn}_{0.5}\text{Al}_{11}\text{O}_{19}$ and $\text{LaMg}_{0.5}\text{Mn}_{0.5}\text{Al}_{11}\text{O}_{19}$ reach full conversion at temperatures about 80 °C higher than the best catalysts. $\text{La}_{0.5}\text{Mn}_{0.5}\text{Al}_{11}\text{O}_{19}$ is competitive with catalysts with twice its manganese content because of its exceptionally high surface area. For all three catalysts, combustion is incomplete. Even when propane is completely consumed, hydrogen is present in the exhaust.

The catalysts without manganese are poor. They have high light-off temperatures and produce substantial amounts of hydrogen, carbon monoxide, and ethylene as partial oxidation products. Of the metals tested, iron is best, followed by cobalt and cerium.

Our results concur with Wang *et al.* [18] and Jang *et al.* [4], who also found that $\text{LaMnAl}_{11}\text{O}_{19}$ was the best hexaaluminate combustion catalyst — neglecting for the moment catalysts with cerium substitution for lanthanum, which have not yet been reported in the literature. Our $\text{LaMg}_{0.5}\text{Mn}_{0.5}\text{Al}_{11}\text{O}_{19}$ did not perform nearly as well as that of Groppi *et al.*, which marginally outperformed $\text{LaMnAl}_{11}\text{O}_{19}$ [15]. Our bimetallic manganese-cobalt catalyst was similarly lacking and did not exhibit the synergy reported by Yan and Thompson [3].

To improve upon $\text{LaMnAl}_{11}\text{O}_{19}$, we experimented with cerium substitution. Cerium substitution for manganese has been shown to augment manganese oxide catalysts. For the wet oxidation of ammonia, a manganese/cerium ratio of 7:3 was optimal [23]. For the wet oxidation of poly(ethylene glycol) or surrogate domestic wastewater, a manganese/cerium ratio of 1:1 was optimal [24, 25]. A manganese/cerium ratio of 6:4 was best for the wet oxidation of phenol [26]. However, our results show that partially replacing manganese with cerium in $\text{LaMn}_{1-x}\text{Ce}_x\text{Al}_{11}\text{O}_{19}$ actually reduces catalytic activity (Figure 2). It is interesting to note that the light-off curves for $\text{LaMn}_{0.5}\text{Ce}_{0.5}\text{Al}_{11}\text{O}_{19}$ and $\text{LaCo}_{0.5}\text{Mn}_{0.5}\text{Al}_{11}\text{O}_{19}$ are nearly identical and that in the absence of manganese, cobalt- and cerium-substituted hexaaluminates are equivalent catalysts. It thus appears that cerium and cobalt merely dilute manganese without providing any synergistic effects.

We therefore tried substituting cerium for lanthanum, while maintaining the manganese content at one cation per hexaaluminate unit. In perovskites of the composition $\text{La}_{1-x}\text{Ce}_x\text{MnO}_3$, cerium doping has produced mixed results. Marchetti and Forni found that $\text{La}_{0.9}\text{Ce}_{0.1}\text{MnO}_3$ was less active than LaMnO_3 for methane combustion [27]. Song *et al.* found that $\text{La}_{0.7}\text{Ce}_{0.3}\text{MnO}_3$ was less active than LaMnO_3 for methane combustion but more active for carbon monoxide combustion [28]. Zhang-Steenwinkel *et al.* found that for the range 0–0.3, the catalyst with $x = 0.2$ was most active for carbon monoxide oxidation [29]. Alifanti *et al.* found that for the range 0–0.5, $x = 0.1$ provided the highest activity for methane combustion [30, 31]. Our experiments found that for $\text{La}_{1-x}\text{Ce}_x\text{MnAl}_{11}\text{O}_{19}$, cerium substitution increased activity, with maximum activity occurring at a higher cerium content ($x = 0.4$) than is the case with perovskites (Figure 4). A substantial improvement of 30 °C in both T_{50} and T_{100} over $\text{LaMnAl}_{11}\text{O}_{19}$ was achieved.

When the high-temperature stability of $\text{LaMnAl}_{11}\text{O}_{19}$ was tested by running a self-sustained propane combustion reaction in a packed bed reactor, a red-hot spot formed at the entrance of the reactor and remained stationary over 14 cumulative hours of operation and four start-ups. Light-off curves from before and after the test are displayed in Figure 5 and are virtually identical. Since the reactor was not within a tube furnace for these light-off curves, the reactor was not isothermal and the measured exit temperatures are lower than the reactor temperatures in Figures 2–4. This experiment confirmed that manganese-substituted hexaaluminates are both active and stable enough to be used as combustion catalysts.

The parallel plate catalytic combustion reactor design overcame two drawbacks of packed bed reactors: high pressure drop and concentrated hot spots. Figure 6 shows that pressure drop was negligible for the operating flow rate range. Temperature profiles of the reactor exterior are shown in Figure 7. For 50 sccm propane (lower heating value of 75 W), the temperature was a rather uniform 480–490 °C over the first half of the reactor and then smoothly dropped to 417 °C near the exit. For 14 ml/h JP-8 (lower heating value of 133 W) the hot band was 100 °C hotter, but was located in the mid-section of the reactor, as the JP-8 was not preheated and thus required some residence time to vaporize. The reactor burned propane for 7 hours and JP-8 for 27 hours with no sign of catalyst deactivation.

Conclusions

Substituted hexaaluminates with high surface area (up to 78 m^2/g) can be easily and inexpensively synthesized via a surfactant-mediated process. The better formulations combine high-temperature stability with activity sufficient to sustain catalytic combustion at lower temperatures. $\text{La}_{0.6}\text{Ce}_{0.4}\text{MnAl}_{11}\text{O}_{19}$ was the most active catalyst tested. A parallel plate catalytic combustion reactor with substituted hexaaluminate catalyst supported on Fecralloy foam

successfully burned propane and JP-8. It exhibited a low pressure drop, a sufficiently-uniform temperature profile, and stable self-sustained operation. This unit will be suitable for use in compact steam reformers for mobile fuel cells.

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Table 1. Catalyst surface area and propane light-off performance.

Catalyst	Specific Surface Area (m ² /g)	T ₅₀ (°C)	T ₁₀₀ (°C)	Partial Oxidation Products
LaMnAl ₁₁ O ₁₉	53	350	400	-
BaMnAl ₁₁ O ₁₉	62	355	410	-
La _{0.5} Mn _{0.5} Al ₁₁ O ₁₉	78	365	410	H ₂
Sr _{0.8} La _{0.2} MnAl ₁₁ O ₁₉	70	375	410	-
BaFeMnAl ₁₀ O ₁₉	27	375	410	-
BaMn ₃ Al ₉ O ₁₉	12	345	420	-
LaCo _{0.5} Mn _{0.5} Al ₁₁ O ₁₉	67	390	450	H ₂
LaMg _{0.5} Mn _{0.5} Al ₁₁ O ₁₉	65	400	460	H ₂
BaFe ₂ Al ₁₀ O ₁₉	62	435	480	H ₂ , CO, C ₂ H ₄
LaCoAl ₁₁ O ₁₉	46	480	530	H ₂ , CO, C ₂ H ₄
BaCe ₃ Al ₉ O ₁₉	44	480	530	H ₂ , CO, C ₂ H ₄
LaMn _{0.95} Ce _{0.05} Al ₁₁ O ₁₉	57	365	410	-
LaMn _{0.7} Ce _{0.3} Al ₁₁ O ₁₉	63	365	430	H ₂
LaMn _{0.5} Ce _{0.5} Al ₁₁ O ₁₉	59	385	450	H ₂
LaMn _{0.05} Ce _{0.95} Al ₁₁ O ₁₉	59	470	520	H ₂ , CO, C ₂ H ₄
La _{0.9} Ce _{0.1} MnAl ₁₁ O ₁₉	61	355	410	-
La _{0.8} Ce _{0.2} MnAl ₁₁ O ₁₉	62	340	390	-
La _{0.7} Ce _{0.3} MnAl ₁₁ O ₁₉	57	340	390	-
La _{0.6} Ce _{0.4} MnAl ₁₁ O ₁₉	64	320	370	-
La _{0.5} Ce _{0.5} MnAl ₁₁ O ₁₉	67	330	380	-

Figure Captions

Figure 1. Drawing of parallel plate catalytic combustion reactor.

Figure 2. Light-off curves for propane combustion: (○) $\text{LaMnAl}_{11}\text{O}_{19}$, (▲) $\text{BaMnAl}_{11}\text{O}_{19}$, (*) $\text{La}_{0.5}\text{Mn}_{0.5}\text{Al}_{11}\text{O}_{19}$, (◆) $\text{Sr}_{0.8}\text{La}_{0.2}\text{MnAl}_{11}\text{O}_{19}$, (■) $\text{BaFeMnAl}_{10}\text{O}_{19}$, (●) $\text{BaMn}_3\text{Al}_9\text{O}_{19}$, (+) $\text{LaCo}_{0.5}\text{Mn}_{0.5}\text{Al}_{11}\text{O}_{19}$, (□) $\text{LaMg}_{0.5}\text{Mn}_{0.5}\text{Al}_{11}\text{O}_{19}$, (◇) $\text{BaFe}_2\text{Al}_{10}\text{O}_{19}$, (×) $\text{LaCoAl}_{11}\text{O}_{19}$, (△) $\text{BaCe}_3\text{Al}_9\text{O}_{19}$.

Figure 3. Light-off curves for propane combustion: (◆) $\text{LaMnAl}_{11}\text{O}_{19}$, (■) $\text{LaMn}_{0.95}\text{Ce}_{0.05}\text{Al}_{11}\text{O}_{19}$, (+) $\text{LaMn}_{0.7}\text{Ce}_{0.3}\text{Al}_{11}\text{O}_{19}$, (●) $\text{LaMn}_{0.5}\text{Ce}_{0.5}\text{Al}_{11}\text{O}_{19}$, (▲) $\text{LaMn}_{0.05}\text{Ce}_{0.95}\text{Al}_{11}\text{O}_{19}$.

Figure 4. Light-off curves for propane combustion: (*) $\text{La}_{0.6}\text{Ce}_{0.4}\text{MnAl}_{11}\text{O}_{19}$, (+) $\text{La}_{0.5}\text{Ce}_{0.5}\text{MnAl}_{11}\text{O}_{19}$, (▲) $\text{La}_{0.7}\text{Ce}_{0.3}\text{MnAl}_{11}\text{O}_{19}$, (●) $\text{La}_{0.8}\text{Ce}_{0.2}\text{MnAl}_{11}\text{O}_{19}$, (◆) $\text{LaMnAl}_{11}\text{O}_{19}$, (■) $\text{La}_{0.9}\text{Ce}_{0.1}\text{MnAl}_{11}\text{O}_{19}$.

Figure 5. Light-off curves for $\text{LaMnAl}_{11}\text{O}_{19}$ used in high-temperature stability test: (■) fresh, (●) used.

Figure 6. Parallel plate catalytic combustion reactor pressure drop curve.

Figure 7. Temperature profile of parallel plate catalytic combustion reactor.

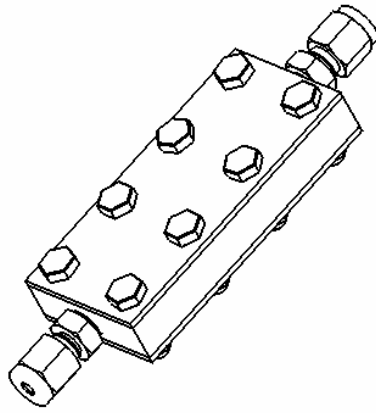


Figure 1.

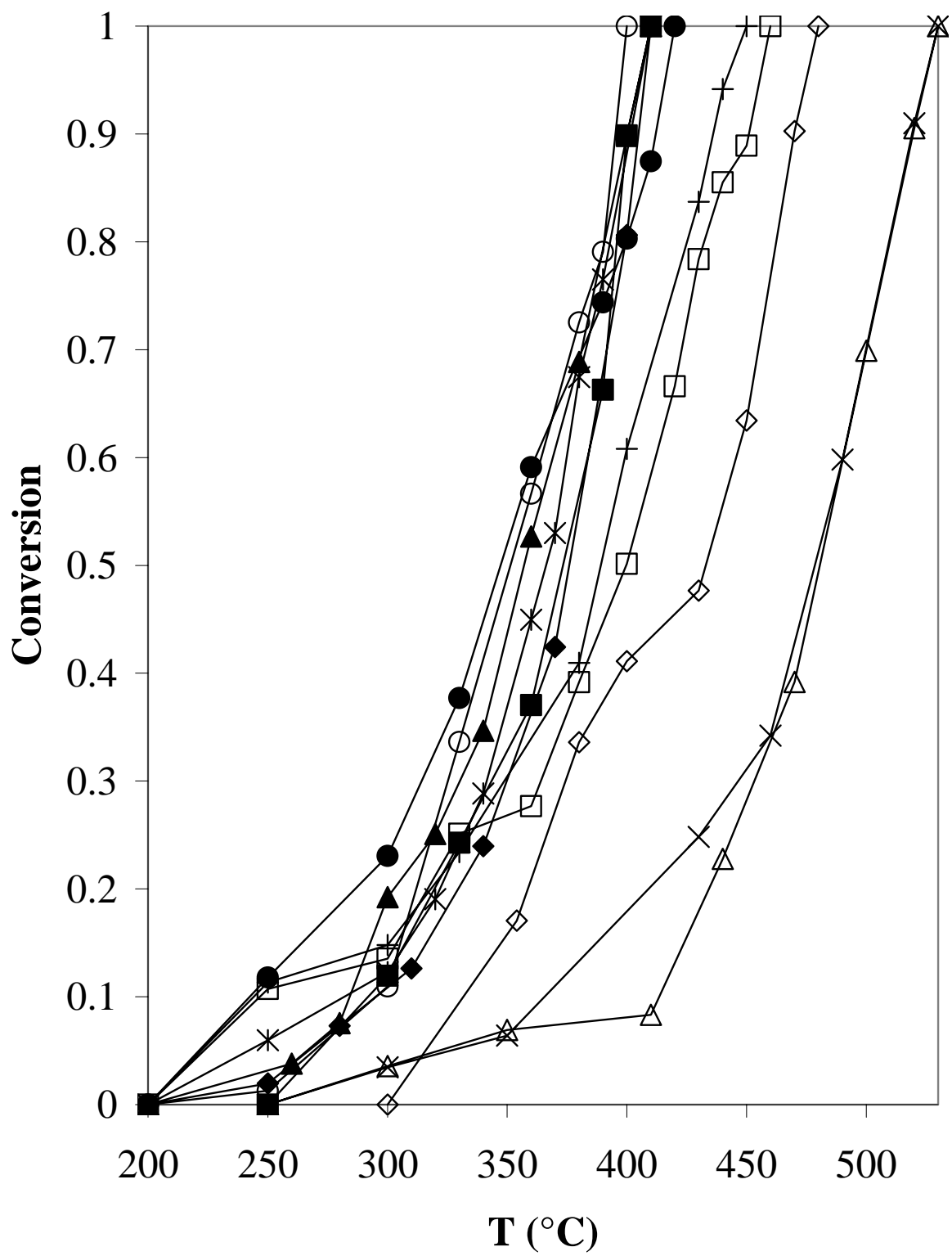


Figure 2.

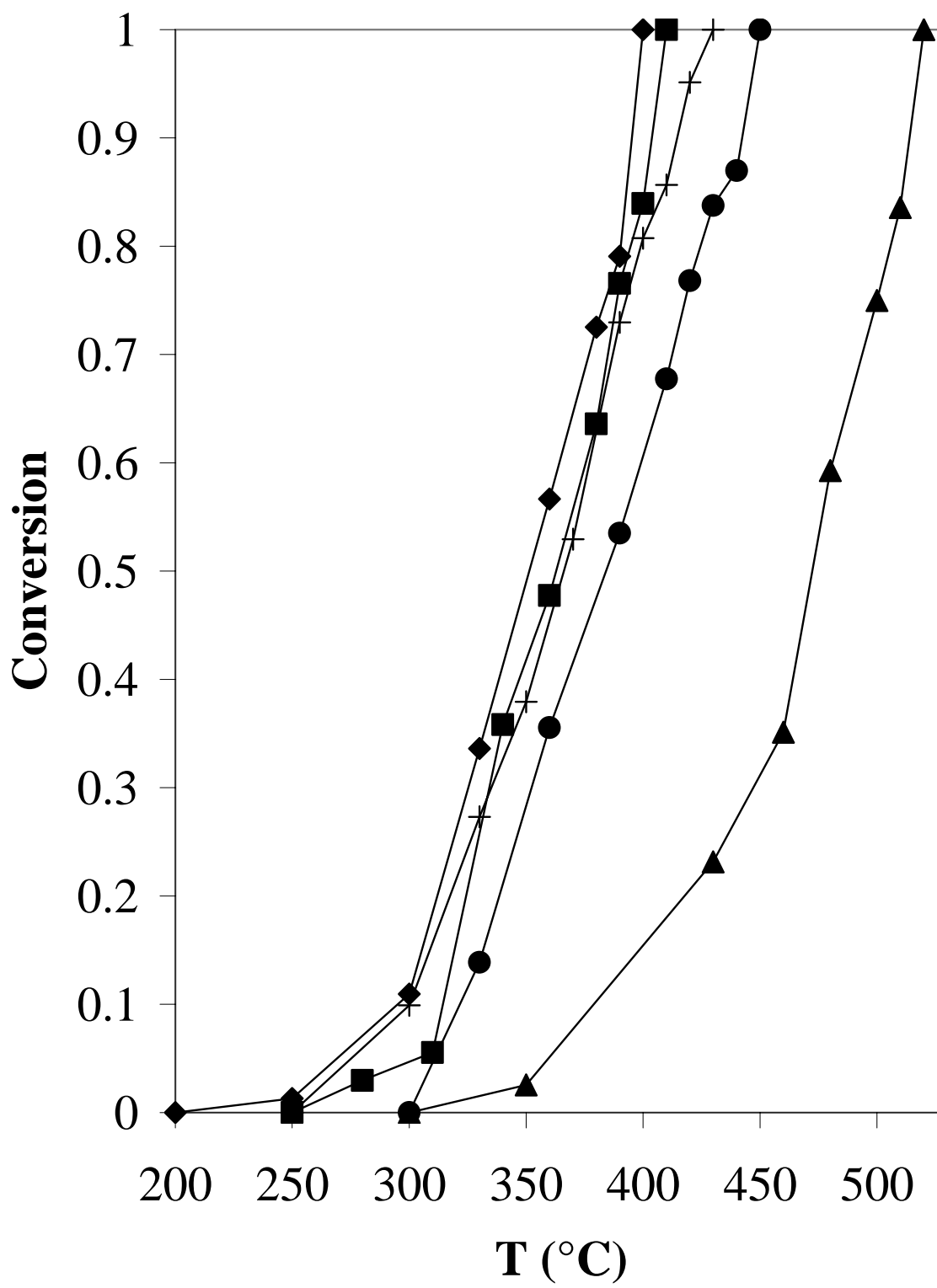


Figure 3.

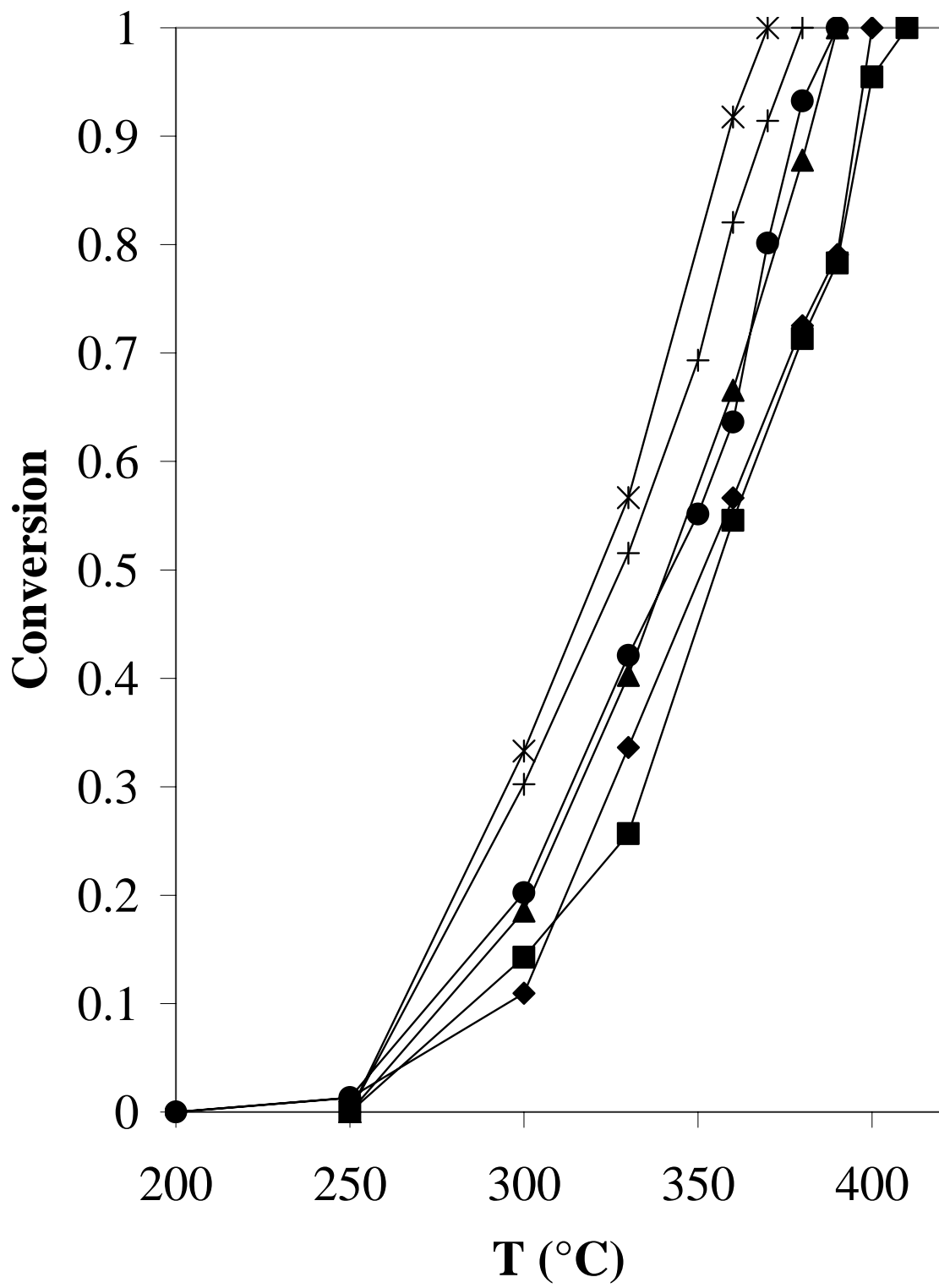


Figure 4.

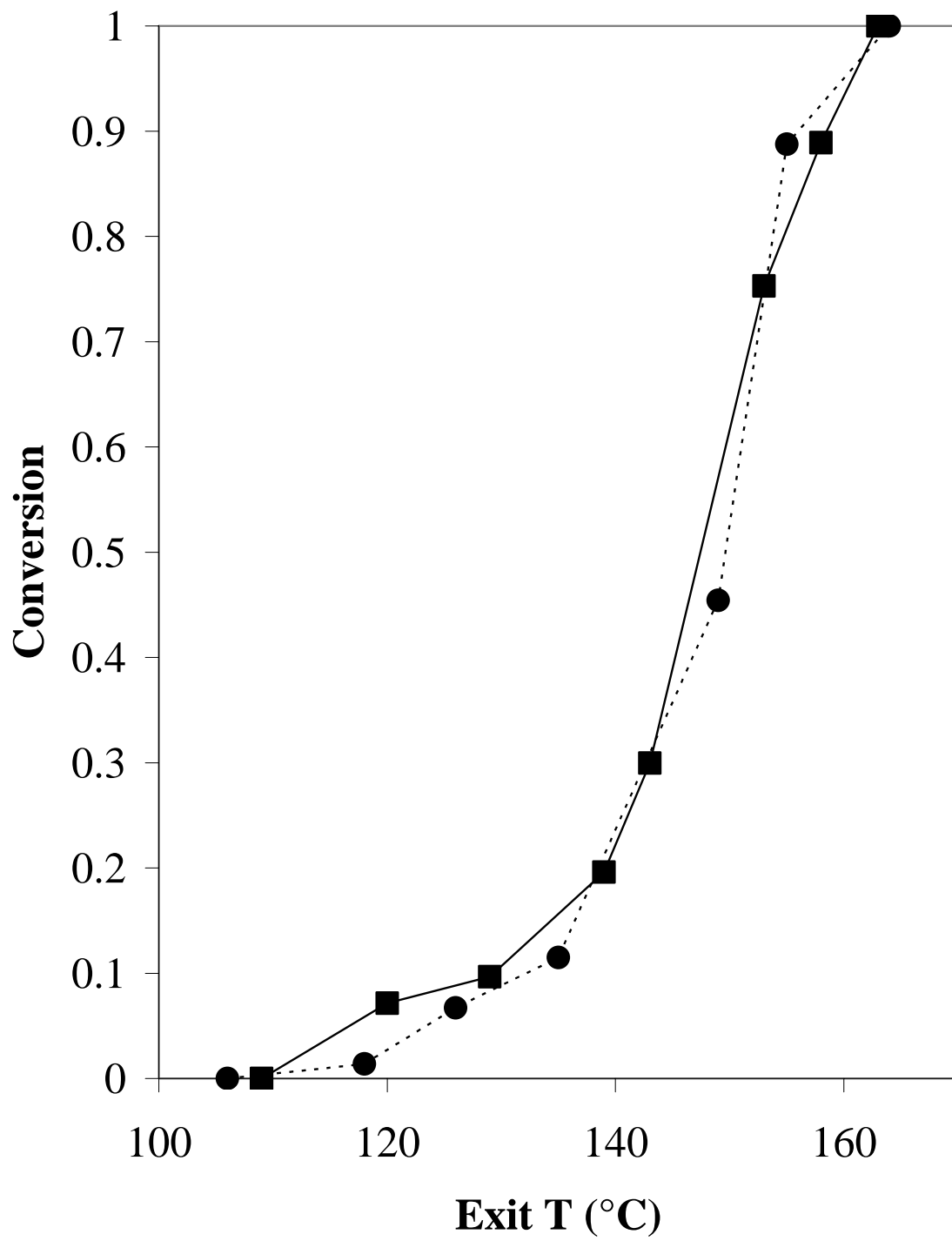


Figure 5.

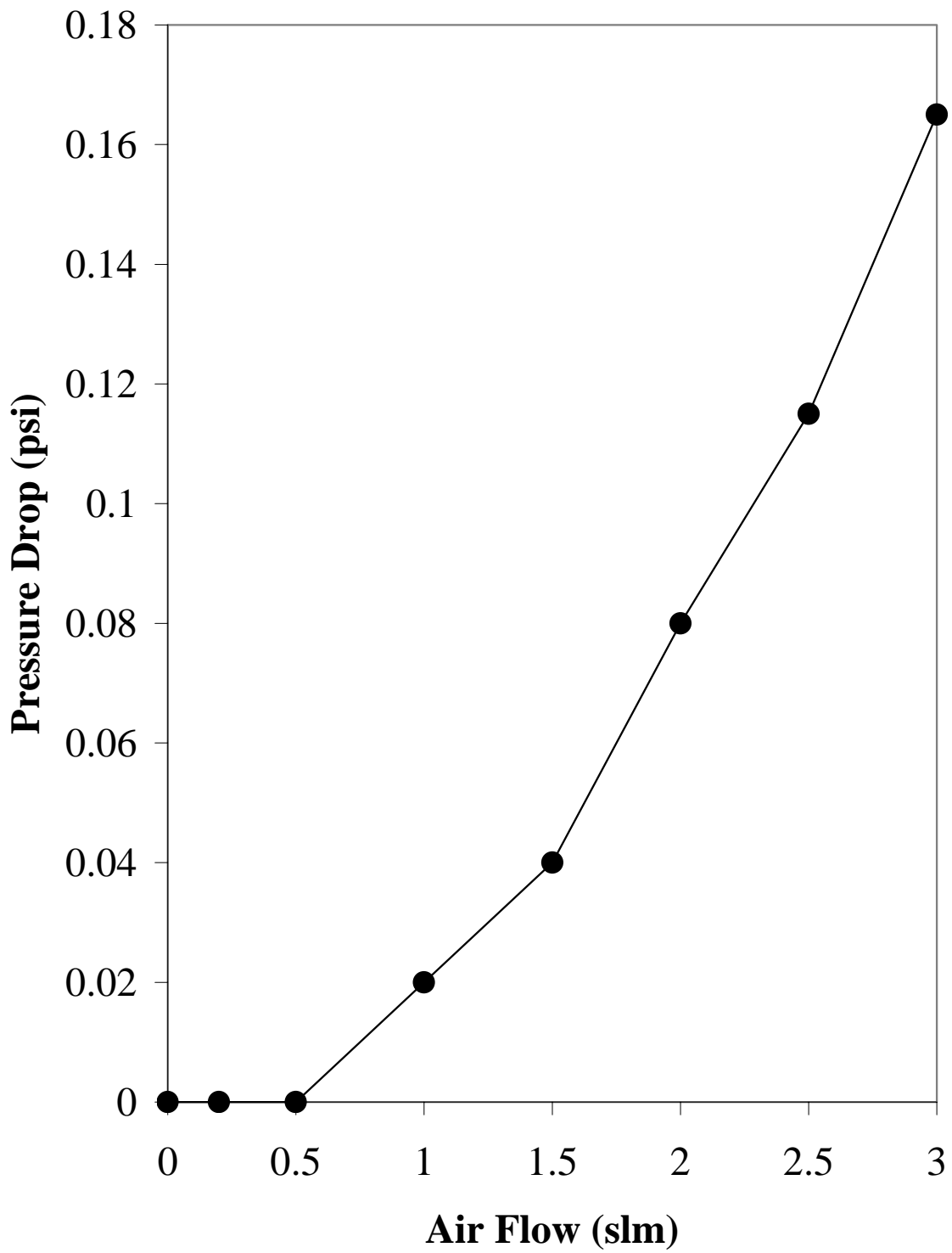


Figure 6.

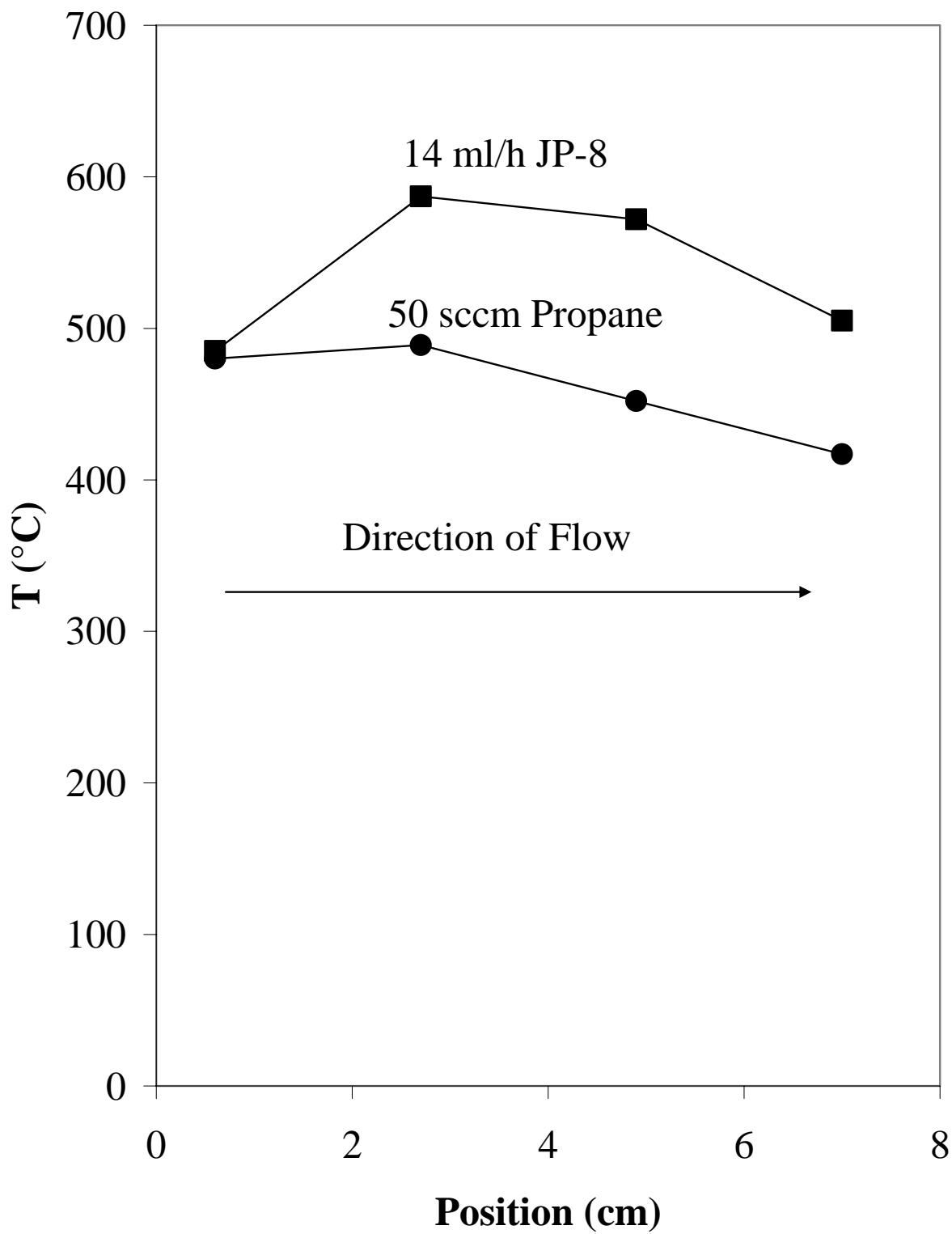


Figure 7.