EXCHANGEABILITY OF SYNTHETIC GASES FROM SOLID FUELS WITH PIPELINE NATURAL GAS

By Joseph Grumer and Margaret E. Harris
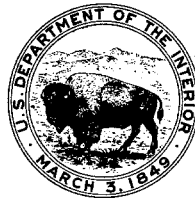
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by

Joseph Grumer[1] and Margaret E. Harris[2]

ABSTRACT

The range of composition of gases synthesized from solid fuels which are exchangeable with natural gas on current gas appliances was evaluated by theoretical principles. These principles consider primarily the flashback, blowoff, and yellow-tipping characteristics of air-entraining burner flames. It was found that complete identity between the synthetic and natural product is not necessary. Gases such as may be obtained by hydrogasification of coal and other solid fuels without the removal of unreacted hydrogen offer advantages in flame characteristics. Between 25 to 37 percent hydrogen can be tolerated in the sendout gas.

INTRODUCTION

By evaluating the exchangeability of synthetic gases with pipeline natural gas, the present investigation seeks to contribute to the effort made by the Bureau of Mines to meet future needs of consumers of gaseous fuels. The gas industry is the sixth largest in this country, supplying about 35 million homes, businesses, and industries (1, 31).[3] Forecasts have been made (20) that the United States does not have within its borders natural gas supplies adequate for this century. It has been estimated variously (6-7, 20) that the need for supplemental gas from coal, shale, or lignite may arise between 1975 and 1980; other estimates (26, 33) predict that the need will not arise so soon. In 1962, for the first time since 1945, the life index, or reserve-production ratio, of U.S. natural gas declined to less than 20 years (25); it was 19.9 years in 1962 and 18.7 years in 1963. While the index is

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[3] Underlined numbers in parentheses refer to items in the list of references at the end of this report.

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strongly influenced by many factors, it is still one of the most widely used measures of future famine or plenty (31).

Moreover, it is uncertain that other energy sources can readily and economically take up the load, should natural gas fall into short supply. In 1963, the life index of crude oil stood at 11.9 years, and that of natural gas liquids at 14.9 years. Both are also declining. Use of atomic energy in homes or even in small industry is not an immediate prospect. Existing distribution lines cannot carry large increases in electrical energy to consumers and they will be costly to replace. Currently is is cheaper to pipe energy to the consumer as gas than as electricity. For example, in 1960, the actual average cost to the consumer of a million Btu of energy in the form of electricity from coal was $7.20 (6-7). Had synthetic pipeline gas been available, the average cost to the consumer of a million Btu of energy in the form of gas from coal would have been $2. Contemplated advances in conveying coal by pipeline would not materially affect this comparison (32). It also appears unreasonable to discontinue use of the expensive pipeline transmission system and of the gas utility distribution systems now located throughout the country. Thus, though large natural gas reserves presently exist, consideration must be given to seeking future sources of synthetic gas suitable as a substitute or as a supplement for pipeline natural gas.

Two additional reasons may be given for this search. The cost to the consumer of natural gas is constantly rising, owing to the economics of supply and demand; if economically competitive, synthetic gas could stabilize costs. Synthetic gas can be made from the large deposits of coal, lignite, and oil shale available in different regions of the country; the choice of raw material for synthetic gas is likely to vary with locale. One requirement throughout the country is that the synthetic gas must operate satisfactorily (be exchangeable) on existing equipment burning natural gas; conversion of appliances to accommodate nonexchangeable gas is prohibitively costly and troublesome. Therefore the present plans of the gas industry generally call for supplementing pipeline natural gas with a synthetic gas from coal that is practically identical with the current natural product. However, such a gas may not be the cheapest and most abundant synthetic gas available. Before the final choice of processes to be developed is made, consideration should be given to the possibility that the national interest may better be served by a synthetic gas from coal, shale, or lignite which differs somewhat from the natural product.

A second incentive for the manufacture of synthetic substitute and supplemental pipeline gas is the economically depressed status of the coal industry. The United States has enormous reserves of coal, and the use of coal to generate electrical power will undoubtedly increase in the years to come. Use of coal to generate synthetic gas is another avenue whereby this resource can be utilized to the fullest benefit of the country. If pipeline gas from coal were to replace only 10 percent of the natural gas consumed in 1961, over 90 million tons of coal would be gasified (2-3).

The Bureau of Mines has long been engaged in process development and economic studies of the gasification of coal to give gaseous or liquid fuels.

It now appears timely to evaluate the composition of gases which can be synthesized from coal and other solid fuels in large supply in the United States and which are usable (exchangeable) without rebuilding or replacing existing gas burner appliances. The present evaluation is based on theoretical concepts previously published by the Bureau; no effort has been made here to evaluate the engineering problems of gasification processes, or their economics.

Concern over the exchangeability (traditionally and sometimes inappropriately called interchangeability) of fuel gases on consumer and industrial burners is not new. To cope with the problem of exchangeability, the Bureau has developed fundamental concepts of flame stabilization (24), yellow tipping (15-16), and entrainment of air in gas burners (36). It has also developed necessary data to implement these concepts (17). This and related information (10-11, 13-14, 18, 23) has been combined into relationships predicting the flashback, blowoff, and yellow-tip performance of burners in a community when fuel gases are exchanged without requiring preliminary appliance testing (10, 16).[4] Application of these relations requires only certain fuel gas data that are readily available: Density, heating value, stoichiometry, gas line pressure, and flame stability diagrams for both fuels involved in the exchange.

STANDARD OF SATISFACTORY EXCHANGEABILITY

One preliminary point should be raised, the distinction between "substitute gases" and "supplemental gases" as the terms are used in this report. From the standpoint of burner performance, all gases are either the adjustment gas (fuel a), that is, the gas for which the burner is originally adjusted, or the substitute gas (fuel x). A supplemental gas is a mixture of the adjustment gas with another gas; as far as the burner is concerned this is also a substitute gas. Accordingly, the gases shown in all figures of this report are either adjustment or substitute gases. However, for the purpose of identifying the product from a synthetic gas plant, a distinction is made in the discussions between gases which can be used without mixing with natural gas (substitute gases) and those which can be used only after mixing (supplemental gases).

The experience of gas utilities has shown that a 1,400-Btu propane-air mixture may be substituted for natural gas in distribution systems designed for residential and industrial uses (9, 21, 38). A theoretical analysis of this exchange is shown in figure 1; the theory is presented in references (10) and (16). Trouble zones are marked "burners giving blowoff with x" and "burners giving flashback with x." Much larger trouble zones on the figure would indicate nonacceptability of the exchange. The gas appliance approval program of the American Gas Association tends to eliminate marginal appliances which may be troubled by this exchange of propane-air for natural gas. That program requires that both 1,400-Btu butane-air gas and natural gas be used in testing appliances intended for natural gas service. The butane-air gas has characteristics very similar to those of the propane-air gas.

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[4] Of the references cited, 16 and 17 contain enough information for the reader desiring only "working" material. Helpful comparisons of old and new units are given in references 11 and 17. Publications by other authors dealing with interchangeability are too many to list; several fairly recent articles are noted as references 5, 19, 27, 29-30, 34, and 40.
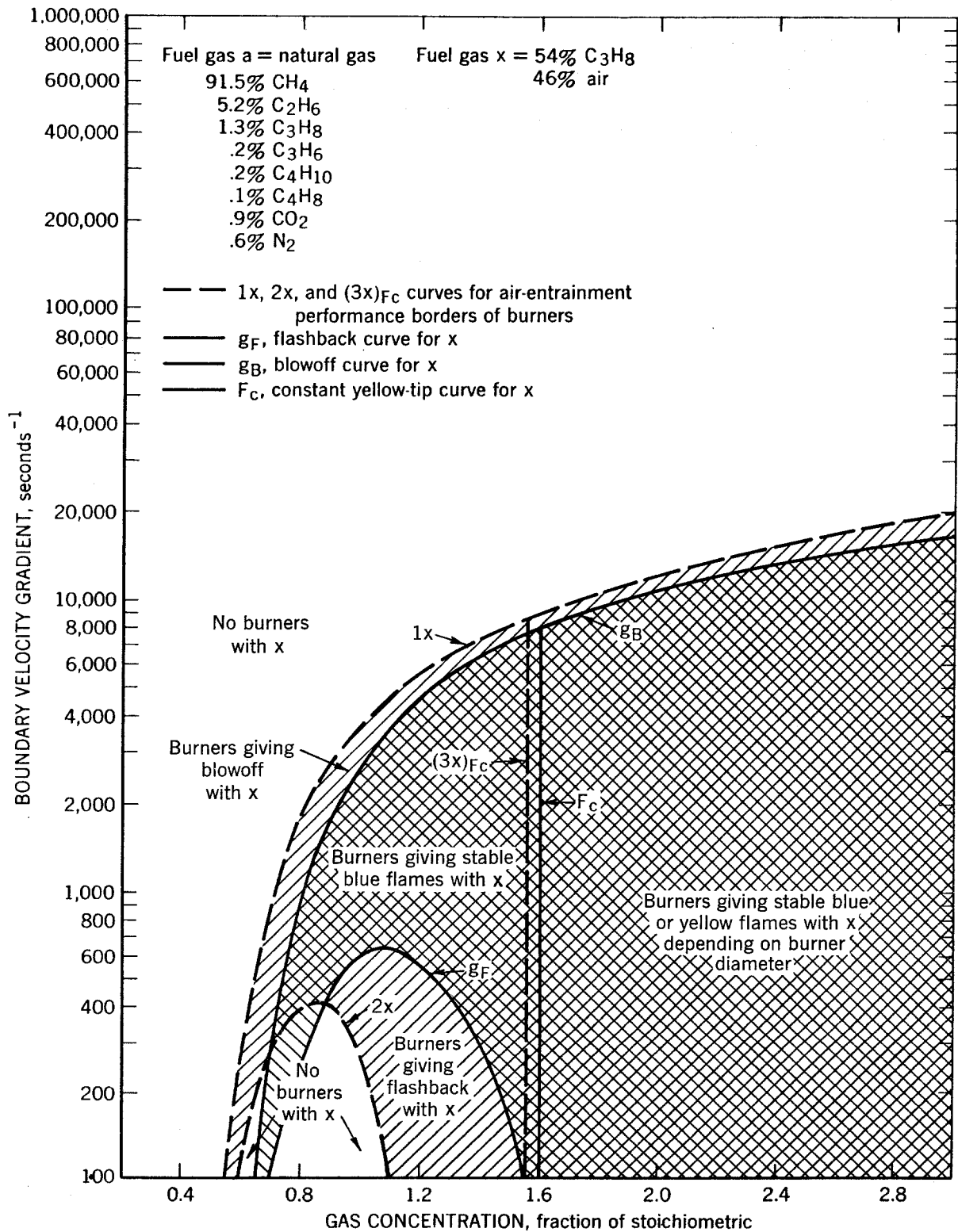
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FIGURE 1. - Predicted Burner Performance When Changing From Natural Gas to 54 Percent C_3H_8-46 Percent Air With Air-Entraining Burners.

For the exchanges presented in this report, the trouble zone limiting exchangeability is the flashback zone. Experience of gas utilities using natural gas (35, 39) has shown that flashback is a problem when the peak of the flashback trouble zone for a substitute gas exceeds a gradient somewhere between 600 and 1,000 reciprocal seconds (18).

FUELS FROM HYDROGASIFICATION OF COAL, OIL SHALE, OR LIGNITE

Substitute Pipeline Gas For Natural Gas

Analyses of the type made for the propane-air fuel were also made for product gases such as may be obtained from hydrogasification of coal, oil shale, or lignite. Depending on the process variables and methods of operation, the gases produced can range from 90 to 50 percent methane, the remainder being largely hydrogen. Representative gases that can be produced by this process have been taken to be mixtures of 75 percent methane and 25 percent hydrogen or 50 percent methane and 50 percent hydrogen. The exchangeability analyses are shown in figures 2 and 3. Compositions and other characteristics of various mixtures used in this study are given in table 1. Two natural gases are listed in these figures and in the table; for the purpose of this discussion, the differences are negligible. Figure 2 shows no blowoff trouble zone and a much smaller flashback trouble zone than that in figure 1, indicating that a mixture of 75 percent methane and 25 percent hydrogen could be substituted directly into gas pipelines and would be exchangeable with natural gas on domestic and industrial air-entraining burners. On the other hand, figure 3 shows no blowoff trouble zone but a more severe flashback trouble zone which would preclude the mixture of 50 percent methane and 50 percent hydrogen from being exchangeable for natural gas.

TABLE 1. - Characteristics of natural gas and candidate substitute gases

| | Fuel gases a | | | Fuel gases x | | | |
| | Natural gas | | Propane-air | Hydrogasification gases | | | |
	2,3,4	1	1	2	3	2	4
Methane................percent..	96.6	91.5	-	75.0	50.0	73.3	61.7
Ethane....................do....	1.6	5.2	-	-	-	0.8	0.4
Propane...................do....	-	1.3	54.0	-	-	-	-
Propylene.................do....	-	0.2	-	-	-	-	-
Butane....................do....	-	0.2	-	-	-	-	-
Butylene..................do....	-	0.1	-	-	-	-	-
Carbon dioxide............do....	-	0.9	-	-	-	-	-
Nitrogen..................do....	-	0.6	-	-	-	-	-
Hydrogen..................do....	-	-	-	25.0	50.0	25.0	37.5
Air.......................do....	-	-	46.0	-	-	-	-
Inerts....................do....	1.8	-	-	-	-	0.9	0.4
Stoichiometric fuel gas..pct of fuel.	9.5	9.04	7.43	11.3	14.3	11.4	12.7
Specific gravity..................	0.570	0.611	1.30	0.433	0.312	0.441	0.377
Heating value..........Btu/cu ft..	1,009	1,070	1,389	842	670	839	755
Lean limit of flammability ....pct of fuel[1]..	4.8	4.8	4.1	4.7	4.6	4.7	4.6
Maximum flame temperature.....° C..	1,950	1,950	1,925	~2,000	~2,000	~2,000	~2,000
Maximum burning velocity...ft/sec..	1.26	1.26	1.52	1.63	1.66	1.63	1.64
Change in stoichiometry, fuel x/a..	-	-	0.85	0.96	0.88	0.94	0.90
Heat input, fuel x/a..............	-	- ·	0.89	0.96	0.90	0.94	0.92
Heating value, fuel x/a...........	-	-	1.30	0.83	0.66	0.83	0.75
Specific flame intensity, fuel x/a.	-	-	1.37	1.25	1.28	1.25	1.27

[1] Upward flame propagation.

BOUNDARY VELOCITY GRADIENT, seconds^{-1}

1,000,000
800,000
600,000
400,000

200,000

100,000
80,000
60,000

40,000

20,000

10,000
8,000
6,000

4,000

2,000

1,000
800
600

400

200

100

Fuel gas a = natural gas
    96.6% CH_4
    1.6% C_2H_6
    1.8% inerts

Fuel gas x = 75% CH_4
    25% H_2

— — 1x, 2x, and (3x)$_{Fc}$ curves for air-entrainment
    performance borders of burners
——— g_F, flashback curve for x
——— g_B, blowoff curve for x
——— F_c, constant yellow-tip curve for x

g_B

No burners with x

No burners with x

1x

F_c

Burners giving stable blue flames with x

Burners giving stable blue or yellow flames with x depending on burner diameter

(3x)$_{Fc}$

Burners giving flashback with x

g_F

2x

No burners with x

0.4   0.8   1.2   1.6   2.0   2.4   2.8

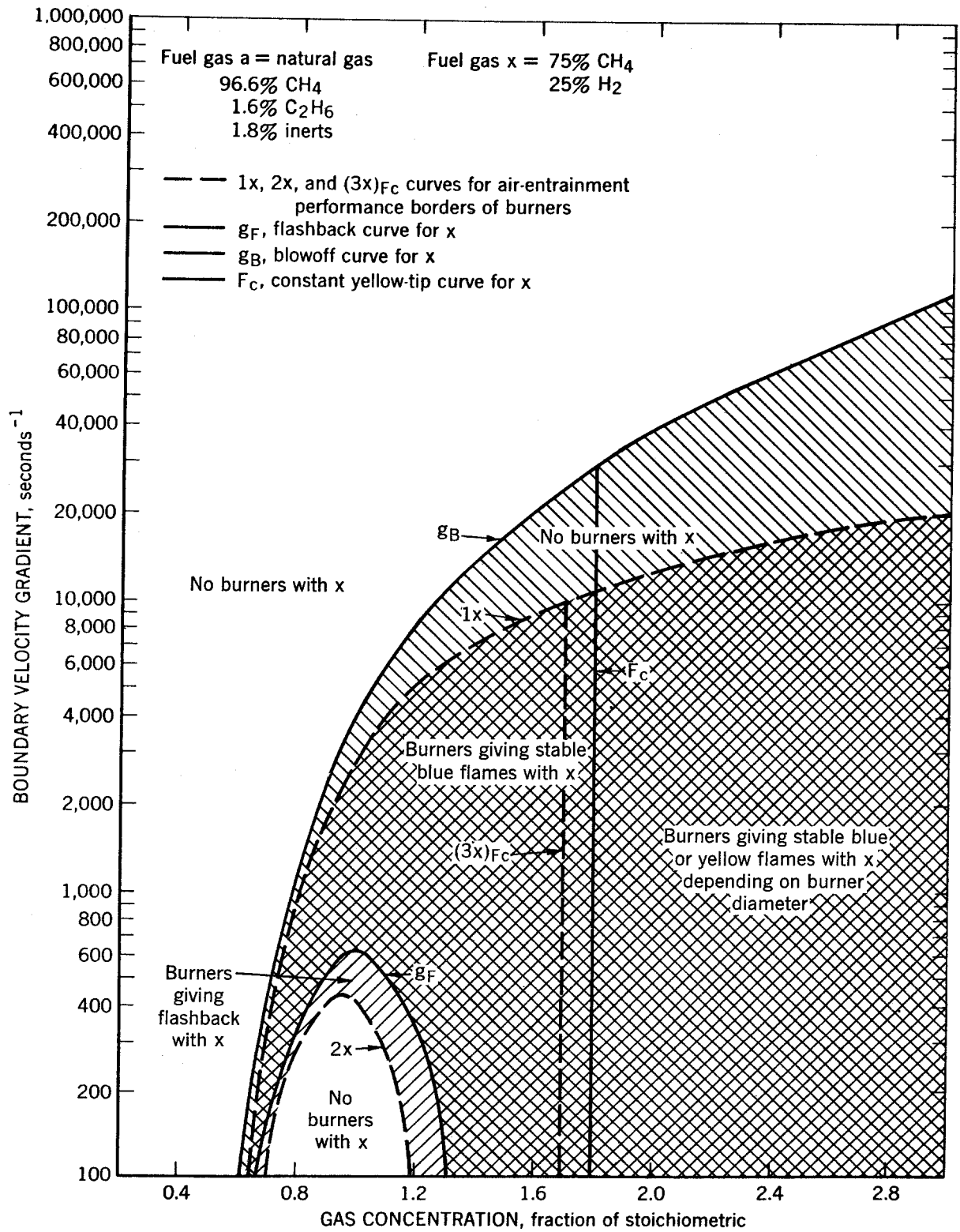GAS CONCENTRATION, fraction of stoichiometric

FIGURE 2. - Predicted Burner Performance When Changing From Natural Gas to 75 Percent CH_4-25 Percent H_2 With Air-Entraining Burners.
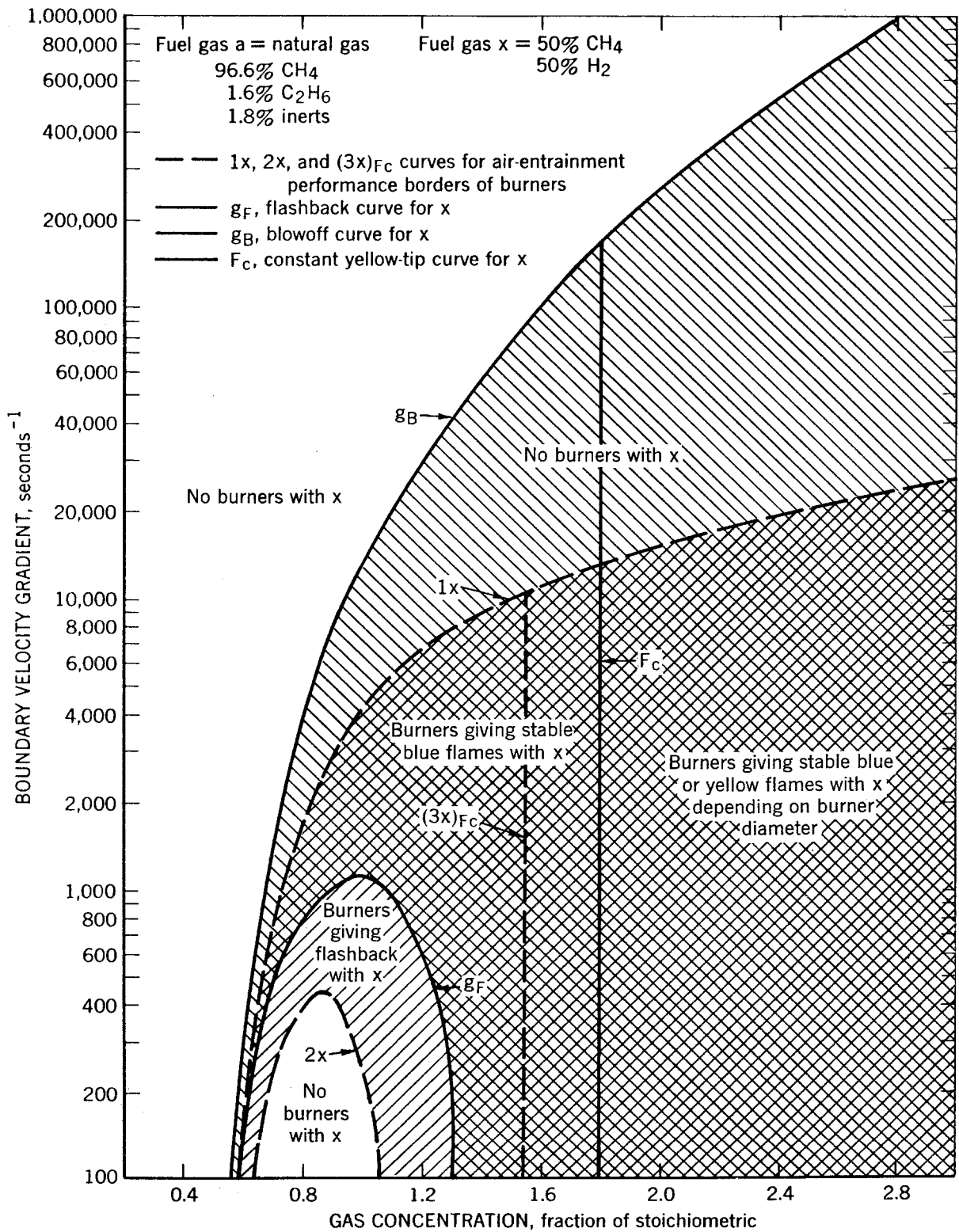
FIGURE 3. - Predicted Burner Performance When Changing From Natural Gas to 50 Percent CH₄-50 Percent H₂ With Air-Entraining Burners.

No difficulty with yellow tipping is anticipated in any of the three exchanges represented in figures 1 through 3, based on the fact that the dashed line, $3x(F_c)$, is to the left of the F_c line (16). Nor will flashback, blowoff, or yellow-tipping tendencies be much affected by minor constituents in the synthetic gas, such as nitrogen, carbon monoxide, or carbon dioxide.

Table 1 shows that substitution of acceptable hydrogen-methane fuels for natural gas results in a somewhat smaller shift in stoichiometry and heat input (Btu/hr) than does the use of the propane-air fuel. The heating values (Btu/cu ft) of the substitute gases are lower, but this is not of prime importance in burner performance. The heating value of the gas sent to the public has to conform with public utility regulatory requirements. These requirements are changeable; flame characteristics of a fuel gas are not. Another consideration is that the carrying capacity of a gas distribution system varies directly with heating value and inversely with the square root of the density of the gas being distributed. Variations in the carrying capacity of a gas utility system are similar to those given in the heat input line in table 1. There appears to be no problem in this respect, or in regard to the cost of cross-country transmission of hydrogen-bearing pipeline gas (8). Actually, there may be some advantage to the consumer if a synthetic pipeline gas contains a little hydrogen. As shown in table 1, the specific flame intensity (12), which is a measure of the concentration of the heat output of the flame, is a little greater with the hydrogen-bearing fuels than with natural gas. An increase in specific flame intensity makes it feasible to generate a given amount of heat with a smaller flame, so combustion chambers can be more compact. (For the distinction between heat input, heating value, and specific flame intensity, see appendix.) Flame temperatures and burning velocities (28) of these substitute gases vary little from those of natural gas. The presence of hydrogen will be helpful in any future fuel cell application; it will also increase stability limits of diffusion flames used in nonaerated burners (22). Yellow tipping and carbon formation are less likely with hydrogen-bearing fuels than with natural gas or with the 1,400-Btu propane-air substitute. The lean limits of flammability, which may be used as a measure of explosion hazard due to leakage, are about the same as with natural gas (4).

Supplemental Pipeline Gas With Natural Gas

Although 50 percent methane-50 percent hydrogen is not suitable as a substitute for natural gas in pipeline transmission systems, it is possible to use it as a supplemental gas by mixing it with natural gas at the gate of a distribution system of a utility. The predicted exchangeability for a 1-to-1 mixture of natural gas and the 50 percent methane-50 percent hydrogen fuel is the same as the exchange shown in figure 2. Thus, the resulting supplemental mixture is exchangeable.

A 1-to-3 supplement (prepared with 1 part natural gas and having a final hydrogen content of 37.5 percent) would result in marginal acceptability as judged by a comparison of figures 1 and 4 and by gas industry experience. Flames on burners would not blow off, but as stated previously, gas utility experience has shown that flashback can be a problem when the peak of the
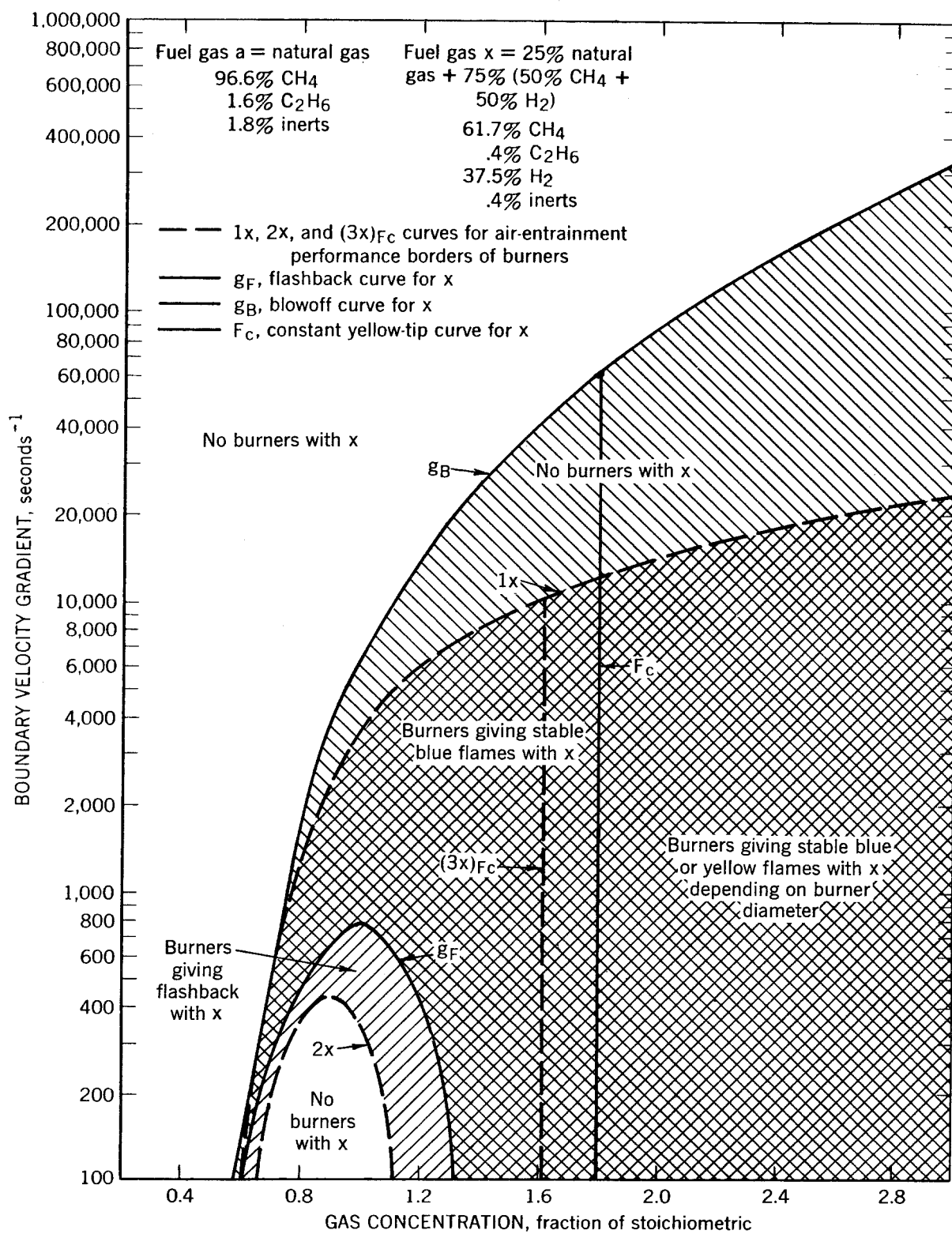
FIGURE 4. - Predicted Burner Performance When Changing From Natural Gas to 25 Percent Natural Gas + 75 Percent (50 Percent CH_4+50 Percent H_2) With Air-Entraining Burners.

flashback trouble zone is much over a gradient of 600 seconds^{-1}. Such is the case in this exchange (fig. 4), and therefore some flames will flash back. As shown in table 1, the stoichiometry of primary combustion for the exchange using a 1-to-3 supplement would become leaner, shifting to about 90 percent of the value with natural gas, and the heat input with fuel x would be 92 percent that of natural gas. This is less of a change than that accepted with 1,400-Btu propane-air.

NONACCEPTABLE SUBSTITUTE GASES

One nonacceptable substitute gas, 50 percent methane-50 percent hydrogen, has already been identified. In addition, the study considered the possibility of using producer gas, Lurgi gas, and other mixtures containing hydrogen plus carbon monoxide in excess of 30 percent, and mixtures containing large quantities of inert gases. These were not exchangeable.

CONCLUSIONS

The preceding considerations indicate that burner performance does not preclude the use in natural gas appliances of synthetic gases, other than a 1,000-Btu fuel consisting almost entirely of methane. A range of mixtures of methane and hydrogen may be used. Because of the approximations involved in this theoretical study, it is improper to pinpoint the maximum amount of hydrogen that may be acceptable in a synthetic pipeline gas. While 25 percent is acceptable, 37 percent is marginal. There are some advantages inherent in the use of hydrogen-bearing gases such as those obtainable from the hydrogasification of coal, oil shale, or lignite. In addition, it is possible that cost to customers of gas from solid fuels and the capacity of plants to make such gas would favor leaving the hydrogen in the hydrogasification product. The disadvantage of using a gas with a lower heating value is only apparent because heat input is a more appropriate criterion than heating value.

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APPENDIX.--Heating Value, Heat Input, and Specific Flame Intensity

The ratio of heating values of two fuels, x and a, is

$$\frac{(\text{Heating value})_x}{(\text{Heating value})_a} = \frac{(H_o)_x}{(H_o)_a} , \tag{1}$$

where H_o is the gross heating value (Btu/ft^3).

The heat input (Btu/hr) to an appliance is $V_o H$, where V_o is the flow of fuel (ft^3/hr). The flow is given by equation (2),

$$V_o \sim (2 \, p_o / \rho_o)^{\frac{1}{2}}, \tag{2}$$

where p_o is the gas line pressure (inches) and ρ_o is the density of the fuel gas (lb/ft^3). Accordingly the corresponding ratio of heat input is

$$\frac{(\text{Heat input})_x}{(\text{Heat input})_a} = \frac{(d_o)_a^{\frac{1}{2}}}{(d_o)_x^{\frac{1}{2}}} \frac{(H_o)_x}{(H_o)_a} , \tag{3}$$

where d_o is the specific gravity of the fuel gas.

The concept of specific flame intensity is useful in characterizing the concentration of heat available from a flame. Flame temperatures are inadequate because they only relate to the level of heat and not to the rate of its release.

The specific flame intensity is the rate of heat released per surface unit of primary flame (inner cone) and is given by equation (4),

$$I = \frac{VH}{A} , \tag{4}$$

where I is the specific flame intensity ($Btu/sec \; ft^2$ of area of the inner cone of flame), A is that area (ft^2), V is the volumetric flow of primary gas-air mixture, corrected to standard conditions (ft^3/sec), and H is the net heating value of the primary mixture at standard conditions (Btu/ft^3).

If the flame is laminar, equation (5) holds:

$$A = \frac{V}{v} . \tag{5}$$

Here v is the burning velocity (ft/sec) of the primary mixture. Substituting equation (5) into equation (4) results in equation (6),

$$I = vH. \tag{6}$$

To evaluate H, four types of flames must be considered:

1. <u>Lean flames</u> (>100 percent primary air),

$$H = H_o^* X,$$ (7)

where H_o^* is the net heating value of the pure fuel at standard conditions and X is the decimal fraction of combustible gas in the primary mixture. H_o is the gross heating value when defining heat input and heating value.

2. <u>Stoichiometric flames</u> (100 percent primary air),

$$H = H_o^* X_s,$$ (8)

where X_s is the decimal fraction of combustible gas in the primary mixture which has the proportion of gas and air balanced for complete combustion.

3. <u>Rich flames</u> (<100 percent primary air),

$$H = H_o^* X_s \frac{1 - X}{1 - X_s}.$$ (9)

Equation (9) is correct only if there is no appreciable diffusion of secondary air into the primary flame and each reacting fuel molecule burns to completely oxidized products. It is reasonable to expect that these conditions are met by a nearly stoichiometric rich flame such as one with maximum burning velocity.

4. <u>Very rich flames</u> (<<100 percent primary air)

Diffusion of secondary air into the primary flame is important, and some fuel molecules are partly oxidized and decomposed. The situation is too involved for a simple and general analysis.

The ratio of specific flame intensities is

$$\frac{I_x}{I_a} = \frac{(v\, H_o^* X)_x}{(v\, H_o^* X)_a} \frac{(1 - X_s)_a}{(1 - X_s)_x} \frac{(1 - X)_x}{(1 - X)_a}.$$ (10)

Equation (10) is for fuel rich mixtures; for other mixtures, only the first fraction applies. The ratios of specific flame intensities given in table 1 are for stoichiometric mixtures.