Quenching the Plasma Reaction by Means of the Fluidized Bed

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BACKGROUND

The ability to maintain temperatures of up to 50,000°K. for indefinitely long periods has created a growing interest in ultrahigh-temperature chemical technology. The devices that can achieve and sustain these ultrahigh temperatures are the plasma generators, and their use as chemical reactors is being explored. Applied research in plasma chemistry is being directed toward chemical synthesis, extractive metallurgy, purification, and crystal growth (*1, 2, 5, 6, 7, 8, 9, 11, 13*).

It is significant to note that attainment of ultrahigh temperatures for favorable chemical equilibrium is far less of a problem than that of preventing undesirable back reactions as the products are cooled from the plasma temperatures. The problem of quenching the products in a manner that gives a desired selectivity and also in a way that utilizes their energy content is probably the greatest obstacle confronting the development of commercial processes which employ plasma reactions.

Many quenching methods have been proposed and are being investigated. Methods include expansion (*8, 10*), surface heat exchange, the injection of secondary components into the plasma, and the direct conversion of energy by magnetohydronamic effects.

Use of the fluidized bed to quench plasma reactions also deserves attention because of the high rates of heat transfer and the continuity of operation. The literature refers to the concept of fluidized bed cooling of high temperature gases (*3, 4, 14*), but there is little information about the behavior of the fluidized systems during quenching or about the quenching rates. And no information has been reported regarding the quenching of plasmas in fluidized beds. The purpose of this study was to examine the operation of a fluidized bed into which a plasma was being injected. Experiments were made to determine the heat transferred from the plasma and to estimate the rate of cooling of the plasma.

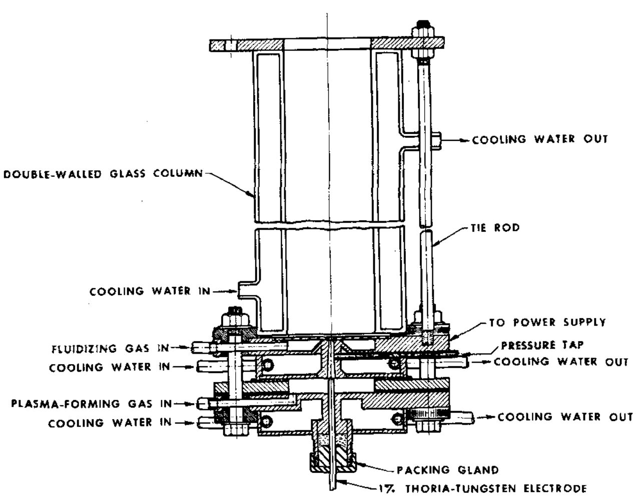

Fig. 1. Experimental apparatus.

EXPERIMENTAL

Apparatus

The transfer of heat from a plasma jet of argon to a bed of fluidized alumina was studied with the apparatus shown in Figure 1. The plasma generator was located beneath the fluidization vessel, and the argon plasma formed at the generator was injected vertically into the bed of fluidized solids. Additional fluidizing gas was provided to maintain a uniform degree of fluidization.

The plasma generator consisted of a ⅛-in. diameter thoriated tungsten electrode held in a vertical position by a packing gland located in a water-cooled copper base plate. The anode was also of copper construction and was water cooled. A slightly tapered hole of ¼-in. diameter was located in the center of the anode. The vertical axis of the electrode was centered within the hole in the anode. The width of the annular space between the electrode and anode decreased as the electrode was raised, and the vertical position of the electrode could be adjusted during operation.

The argon which formed the plasma entered the generator through a hole in the base plate. The gas moved upward through the annular opening between the electrode and anode where the plasma was formed. It then issued from the hole in the anode as a confined arc or jet. The cold wall of the anode served to stabilize and restrict the plasma, preventing it from contacting the copper wall. Argon was also used as the secondary fluidizing gas, and it entered beneath a distributor plate through a horizontal hole in the anode. A porous disk of sintered stainless steel was used to distribute the fluidizing gas evenly at the base of the fluidization vessel.

The fluidized bed was maintained in a double-walled glass column of about 2 in. I.D. Water was circulated between the walls to effect cooling of the bed and to provide an independent control of the bed temperature.

The electrical power was supplied by a d.c. motor-generator of 5-kw. output. A high frequency a.c. starter unit created an arc discharge between the electrode and anode to enable the d.c. discharge to be initiated.

Operation and Measurements

The usual method of operation was to first establish the plasma with the column free of solids. After a suitable flow of the auxiliary fluidizing gas was provided, the alumina was fed

TABLE 1. TYPICAL SIZE ANALYSIS OF ALUMINA USED

U.S. Mesh No.	Weight %
+ 80	2.0
−80 + 100	2.0
−100 + 150	6.0
−150 + 200	68.0
−200 + 270	19.5
−270 + 325	2.0
−325	0.5

slowly into the column from the top. Luminosity from the plasma decreased rapidly with the addition of solids, and almost no light emission was observed during the tests when bed depths exceeded 2 in. Except for the luminosity, the appearance of the fluidized bed was identical to the usual fluidized bed operating without plasma injection. A typical size analysis of the alumina used is given in Table 1.

Steady state operation was achieved within 10 min. for all tests. Constant temperatures within the fluidized bed and of the cooling water discharge were an indication of a steady state condition. After a run the alumina was easily removed through a vacuum line inserted from the top.

The position of the tungsten electrode with respect to the anode was found to be an important aspect of the operation. If it were too high, the arc discharge emerged from the jet opening and completed an electrical path to the outside edge of the opening. This caused hot spots and some melting and pitting of the copper wall in this location. Continued operation in this manner allowed particles from the bed to adhere to the wall. A gradual growth of fused bed material would result until it extended into the plasma jet causing the arc to become unstable. If the electrode tip was positioned at least ½ in. below the jet opening, arcing to the top edge did not occur. There was no tendency for the bed particles to fuse in this case, and the fluidized operation could be maintained indefinitely.

The electrical power input to the plasma was determined as the difference between the total electrical input to the plasma generator and the heat losses to the water used to cool the generator. The total electrical input was found by measuring the voltage and amperage of the motor-generator output. The energy loss to the cooling water, the heat transferred to the secondary, and the heat removed from the bed were determined from measured flow rates and differences in the temperature of the streams. Rotameters were used for flow-rate measurement. Example data showing the distribution of energy within the system are given in Table 2.

The temperature at various points in the fluidized bed was determined by measurement with bare thermocouples. A thermocouple head assembly was used which consisted of seven thermocouples spaced ¼ in. apart in a vertical plane. Chromel-alumel of 28 B and S gauge was used, except in the center-line position where platinum-platinum, 10% rhodium, 30 B and S gauge, was used for the higher power input runs. The thermocouples were connected through a thermocouple selector switch to a potentiometer. Insulated compensating thermocouple lead wire was used for all connections. It was found during operation at steady conditions that, except for the center-line temperatures, the measured temperatures at any point within the fluidized bed fluctuated less than 2°C. Along the center line below the 1½-in. level above the distributor, temperature swings of 100°C. were not uncommon, particularly when power input levels exceeded 1 kw. It was also noted that the measured exit gas temperature was the same as the temperatures within the bed at the upper bed levels.

A check of the axial symmetry of the fluidized bed behavior was made by comparing temperature profiles at various radial positions during operation with plasma injection. Measurements were made along three radial lines spaced 120 deg. apart, giving six temperatures spaced 60 deg, apart on concentric circles of ¼-, ½-, and ¾-in. radius. These measurements were repeated at three elevations above the distributor. Example data at the ¼-in. level above the distributor are given in Table 3. The data at higher bed levels also showed little temperature variation with angular position, and it was concluded that performance was essentially symmetric.

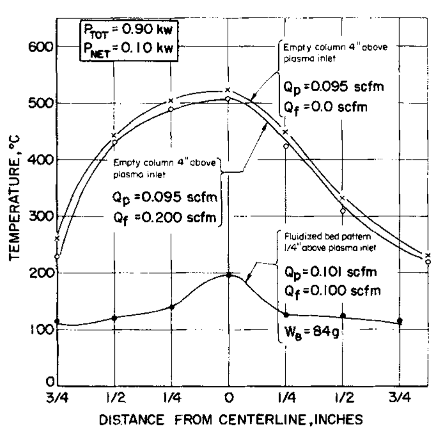

Fig. 2. The effect of the fluidized bed for quenching.

TABLE 2. TYPICAL DISTRIBUTION OF ENERGY

Plasma Generator Section

Run	E*	I†	Total power, kw.	Losses, kw. Anode	Losses, kw. Bottom	Net power, kw.
A-44	22	51	1.12	0.65	0.27	0.20
B-22	32	55	1.76	0.70	0.34	0.72

Fluidized Bed Section

Run	Q_P	Q_f	T_P, °K.**	W_B	Energy to jacket, kw.	Energy as sensible heat to gases, kw.	Sum energy out, kw.
A-44	0.08	0.12	6,320	50	0.16	0.01	0.17
B-22	0.33	0.13	5,660	100	0.69	0.03	0.72

* Voltage across electrodes, v.
† Current in plasma generator circuit, amp.
** Ionization of argon is negligible below 9,000°K. and average or equilibrium temperatures were calculated as

$$\frac{\text{net electrical energy input to plasma, cal./g.-mole}}{C_p \text{ ideal gas, cal./g.-mole-°K.}} + T°, °K.$$

RESULTS

Initial tests were made to determine the gross effect of the fluidized bed on the quenching of the plasma. Tem-

TABLE 3. TEMPERATURE ALONG THREE RADIALS AT $\frac{1}{4}$-IN. LEVEL ABOVE DISTRIBUTOR*

Run	Radial position, deg.	$-\frac{3}{4}$	$-\frac{1}{2}$	$-\frac{1}{4}$	0	$\frac{1}{4}$	$\frac{1}{2}$	$\frac{3}{4}$
		Temperature, °C. Distance from center line, in.						
A-9	0	93	97	102	132	97	95	94
A-10	120	92	94	98	138	98	96	92
A-11	240	92	96	99	136	98	94	92

* Power to plasma generator—1 kw., Q_P—0.068 std. cu. ft./min., Q_f—0.290 std. cu. ft./min., W_B—84 g.

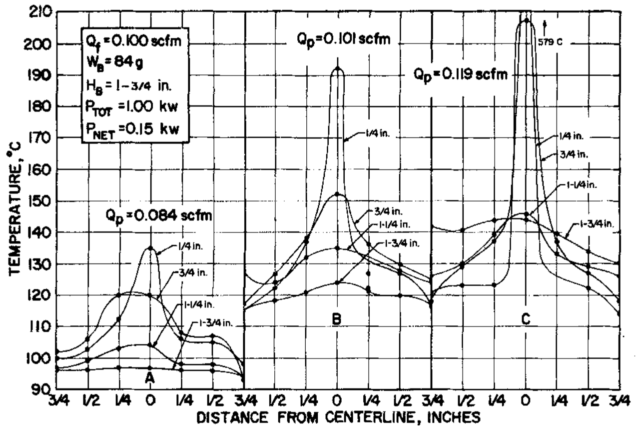

Fig. 3. The effect of plasma gas rate on temperature profiles in the fluidized bed.

perature profiles were taken during operation with the column free of solids and compared with profiles taken during fluidized bed operation at similar conditions. Figure 2 shows a typical comparison, and the increase in the quench rate due to the presence of the solids is apparent.

Experimental results which describe the temperature conditions in the fluidized bed during injection of the plasma are given. Figure 3 shows the effect of increasing the plasma gas rate on the temperature profiles within the bed. At a relatively low plasma rate (Figure 3), there was only a modest difference in center-line temperature. The temperature deviations within the bed became more pronounced as the plasma rate was increased (Figures 3*b* and 3*c*) with greater differences in center-line temperatures as expected. The remarkable ability of the fluidized bed to achieve a rapid quenching action within a short distance from the injection point is indicated by the fact that the average inlet temperatures of the plasma exceeded 4,000°K. during these experiments.

There appeared to be only limited lateral mixing, and heat transfer from the center to the wall was less pronounced at higher plasma rates. Note for example that although the peak temperature at any level increased with increased plasma rate, the plot of temperature vs. distance became narrower. Increases in the rate of auxiliary fluidizing gas appeared to increase the extent of both the lateral and longitudinal movement, causing a more uniform temperature within the bed (Figure 4).

Deeper beds were found to increase the rate of heat transfer from the plasma and thereby cause a more uniform temperature distribution within the fluidized bed. This effect is shown in Figure 5. The data given in Figure 5 also indicate that the mixing of the bed material with the plasma at the plasma inlet is better with deeper beds. The temperature-distance curves for the level ¼ in. above the distributor are shown separately in Figure 6 to illustrate the increased intermixing of the bed with the plasma at the injection point as the bed depth is increased.

ANALYSIS OF RESULTS

The pattern of the temperature distributions indicates that virtually no lateral spreading of the plasma jet occurs upon injection in the fluidized bed. Lateral temperature gradients in the vicinity of the injection point can exceed 4,000°C./in. as evident from the data shown in Figure 7. It appears that there is a net lateral movement of the solids toward the plasma at the low bed levels and some movement of the solids away from the center line at the higher bed levels. Measurement of pressures in the plasma below the injection point indicated that the high-velocity jet enters the bed at a lower pressure than the static pressure at the bottom of the fluidized column of solids. It therefore seems likely that solids become entrained in the boundary layer of the jet in a manner similar to the entrainment of surrounding air by an air jet as described by Taylor, et al. (*12*). This effect would explain the observations that at lower bed levels, temperatures ¼ in. from the center line decreased with increased plasma rate, while center-line temperatures increased sharply with increased plasma flow rate at corresponding bed levels.

The actual quenching of the plasma then involves the energy exchange between the plasma gas and the entrained gas and solids from the fluidized bed. Radiant energy transfer also occurs and probably accounts for the major part of the heat transfer in a lateral direction away from the plasma. The transfer of energy from the plasma is an exceedingly complex process. In addition to convective heat transfer and thermal radiation, the energy transfer involves the mechanisms of Bremsstrahlung radiation due to the interaction of atoms and ions with free electrons, radiative recombination to ions and electrons, and the reversion from excited metastable states back to ground states. The extent of energy transfer by any one of these mechanisms depends greatly on the energy level and degree of ionization of the plasma, and no attempt was made in this study to examine their relative importance.

It was found empirically that a semilogarithmic plot of the difference between the center-line temperature and that of the bulk-bed temperature vs. the distance from the plasma-jet opening approximated a straight line. The

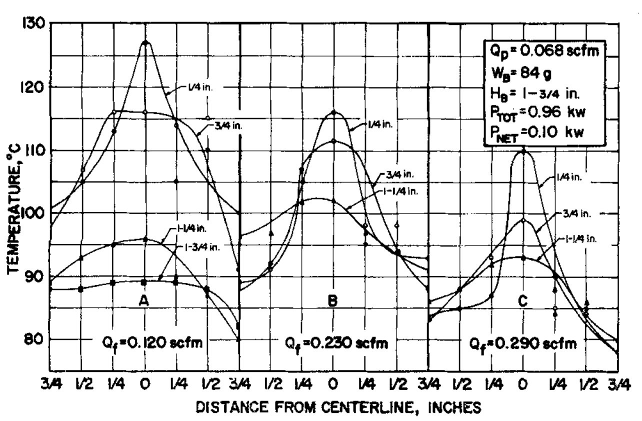

Fig. 4. The effect of fluidizing gas rate on temperature profiles in the fluidized bed.

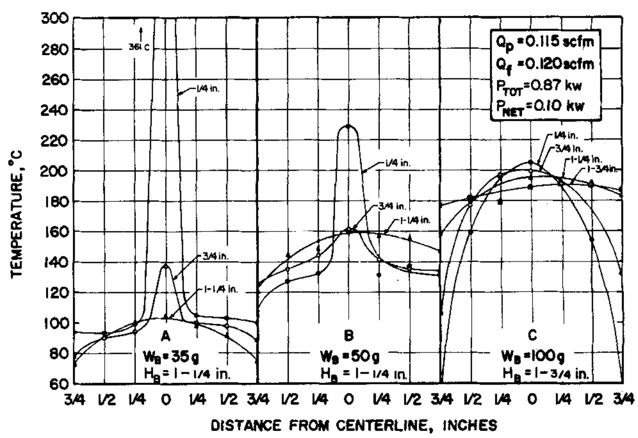

Fig. 5. The effect of bed depth on temperature profiles in the fluidized bed.

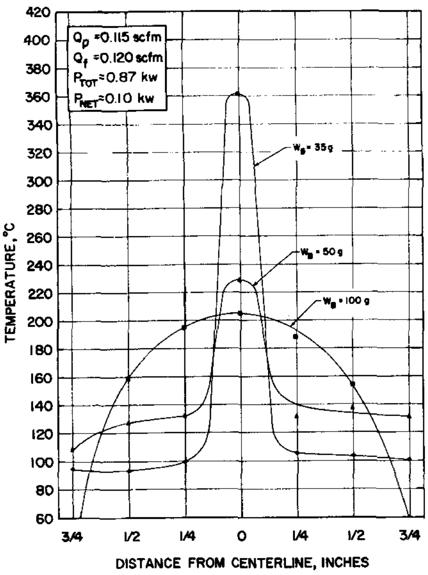

Fig. 6. Temperature profiles at the ¼-in. level above plasma injection point.

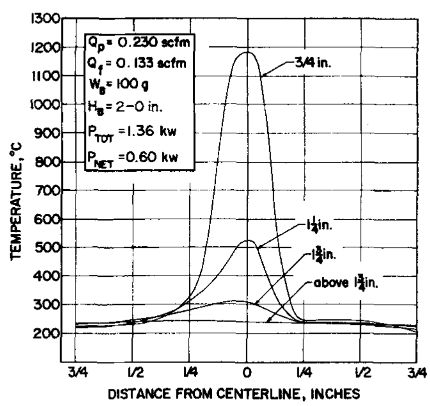

Fig. 7. Temperature profiles showing high lateral temperature gradient near plasma injection point.

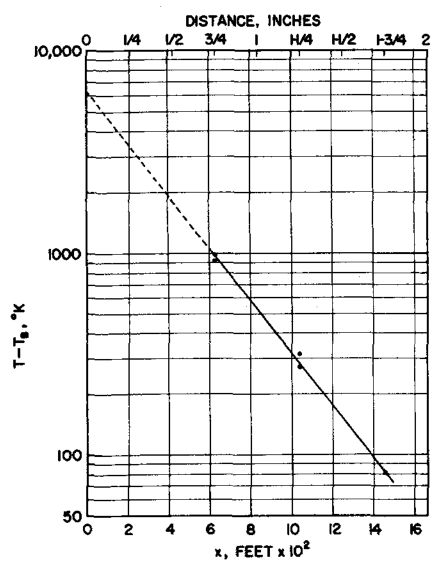

Fig. 8. Semilogarithmic plot showing relationship of center-line temperature in jet with distance above plasma injection point.

center-line temperatures given in Figure 7 are presented this way in Figure 8. Interestingly, the temperature found by extrapolation of this line to zero distance was found to be in close agreement with the average or equilibrium temperature of the plasma at the point of injection in the bed as calculated from an energy balance. A high degree of turbulence in the region of the center-line measurements and the integration of temperature by the thermocouple are believed to be the reasons that the extrapolation gives the average temperature rather than a hot core temperature as the plasma jet would be expected to have along the center line at the point of injection.

This correlation provides a means to estimate the temperature and rate of quenching of the plasma at various distances from the jet opening. The expression for this relationship is

$$\ln (T - T_B) = cx + \ln (T_P - T_B) \qquad (1)$$

Solving for the temperature in the jet one obtains

$$T = (T_P - T_B) \cdot e^{cx} + T_B \qquad (2)$$

Differentiating with respect to time one gets

$$\frac{dT}{d\theta} = (T_P - T_B) \cdot c \cdot e^{cx} \cdot \frac{dx}{d\theta} \qquad (3)$$

and substitution gives

$$\frac{dT}{d\theta} = (T_P - T_B) \cdot c \cdot e^{cx} \cdot v \qquad (4)$$

Assuming no dilution of the jet and a constant mass flow rate across the jet one obtains

$$v = Q_P \cdot \frac{1}{60} \cdot \frac{1}{A_o} \cdot \frac{T}{T^\circ}$$

where

A_o = 0.341 × 10^{-3} sq.ft. (jet opening ¼-in. diameter)
T° = 294.1°K. (70°F.)
v = 0.1662 $Q_P \cdot T$

For the data given in Figure 8, the coefficient c was determined to be -29.4 ft.$^{-1}$. The plasma gas rate for these runs was Q_P = 0.23 std.cu.ft./min., and the bulk bed temperature was about 230°C. (503°K.). The value $(T_P - T_B)$ was found to be 6,300°K.

Substitution of these values into Equation (4) yields

$$\frac{dT}{d\theta} = -\,7{,}080 \cdot T \cdot e^{-29.4\,x} \qquad (5)$$

TABLE 4. CALCULATED QUENCH RATES

Distance from inlet along center line, in.	x, ft.	T-T_B, °K.	T, °K.	v, ft./sec.	Quench rate, °K./sec. × 10^{-6}
0	0	6,300	6,803	260	48.2
¼	0.0208	3,400	3,903	149	14.8
½	0.0416	1,800	2,303	88	4.69
¾	0.0625	990	1,493	57	1.66
1	0.0833	540	1,043	40	0.64
1¼	0.1041	290	793	30	0.26
1½	0.1250	160	663	25	0.12

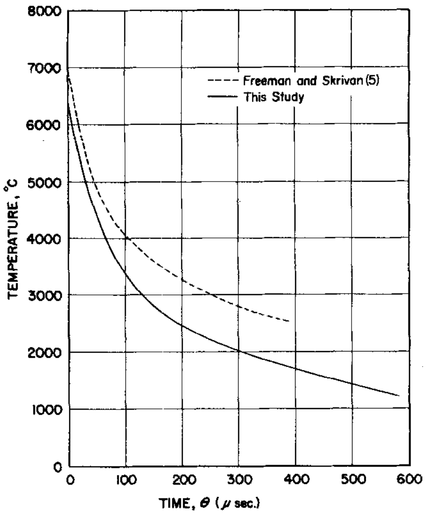

Fig. 9. Comparison of calculated temperature decay rates for fluidized-bed quench and cooling by surface heat transfer.

The quench rates calculated by Equation (5) for the example data are given in Table 4.

It is also interesting to compare the temperature decay rate calculated for fluidized bed quench with that calculated by Freeman and Skrivan based on data obtained by quenching an argon plasma in a ¼-in. I.D. water-cooled copper tube (5). The following development was used to make this comparison:

$$\theta = \int_{x=0}^{x=x} \frac{dx}{v} = \int_{x=0}^{x=x} \frac{dx}{0.1662\, Q_P \cdot T} \qquad (6)$$

Substituting for T in terms of x [Equation (2)] and for $Q_P = 0.23$ std.cu.ft./min. one gets

$$\theta = \frac{1}{(0.0382)} \int_{x=0}^{x=x} \frac{dx}{(T_P - T_B)\, e^{cx} + T_B} \qquad (7)$$

and integrating one obtains

$$\theta = \frac{1}{(0.0382)\; c \cdot T_B} \left(cx + \ln \frac{T_P}{T} \right) \qquad (8)$$

The calculated values with $c = -29.4$ ft.$^{-1}$ and $T_B = 503$°K. are shown in Figure 9 with those calculated by Freeman and Skrivan. Both sets of calculations were made for approximately the same flow rate, and they indicate an initial quench rate of about 50×10^6 °K./sec. for both cases. However the presence of the fluidized bed appears to increase the overall temperature decay rate over that obtained by heat transfer through a fixed surface in contact with the plasma. The degree of this change in quench rate from that obtained by surface heat transfer probably depends on those factors that affect the intermixing of the solids and gases of the bed with the plasma. Certainly the bed depth, bed temperature, and surrounding gas velocity, as well as the particle properties, will be important. These factors are being investigated.

CONCLUSIONS

There is little doubt that the fluidization method can be applied with advantage for rapid quenching of ultrahigh-temperature gases. The quench rate estimated from temperature measurements was found to be significantly higher than that determined for surface heat transfer or that reported for the DeLaval nozzle expansion. The average quench rates in cooling to below 1,000°C. were found to exceed 1×10^6 °K./sec., and the initial temperature decay rate was estimated to be about 50×10^6 °K./sec. during ordinary experimental conditions.

There are related uses of the fluidized solids system that should also be mentioned in connection with high-temperature technology. The economical recovery of energy may be possible with the fluidized quench system because a high level for heat rejection can be maintained in the fluidized bed. It may be possible to use this energy conveniently in a direct contact heat transfer system to either preheat the reactants or to conduct secondary reactions at temperatures to 1,500°C. Another possible application of the fluidized bed that appears unusually promising is the quenching of gaseous products and simultaneous agglomeration of any solid components that form during the cooling process. Ordinarily, the quenching of gases from which solids condense would be difficult by surface heat exchange or by expansion because of the effects of fouling and erosion.

In summary the fluidized solids method has many unique features. Their imaginative use will continue to serve the chemical industries as chemical processing is extended into the ultrahigh-temperature region.

NOTATION

- A_o = area of jet opening, sq.ft.
- c = coefficient, ft.$^{-1}$
- H_B = approximate height of fluidized, in.
- P_{NET} = net electrical power contained in plasma at point of injection into fluidized bed, kw.
- P_{TOT} = total electrical power supplied to plasma generator, kw.
- Q_f = flow rate of argon for auxiliary fluidizing gas, std. cu.ft./min.
- Q_P = flow rate of argon to plasma, std.cu.ft./min.
- T = center-line temperature of jet at $x = x$, °K.
- T_B = bulk temperature of fluidized bed, °K.
- T_P = average temperature of plasma at $x = 0$, °K.
- T° = reference temperature, 294.1°K. (70°F.)
- θ = elapsed time, sec.
- v = jet velocity at center line, ft./sec.
- W_B = bed weight, g.
- x = vertical distance above jet opening, ft.

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