

Experimental study on temperature characteristics and energy conversion in packed bed reactor with dielectric barrier discharge

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Abstract The temperature characteristics and energy conversion in packed bed reactor combined with a dielectric barrier discharge (DBD) plasma was investigated experimentally. The pellet temperatures of two types packed bed reactor, cylindrical reactor and parallel-plate reactor, was measured in conditions of various inlet voltage of DBD plasma. The relationship between pellet temperature of the packed bed and applied voltage of DBD plasma was discovered. The experimental result indicates a tendency that the pellet temperature of packed bed increases as the applied voltage of inlet plasma increases. When the voltage of inlet plasma is high enough, the pellet temperature increment decreases. Simultaneously, the packed bed temperature is sensitive to the inlet plasma energy and there is a potential application to heat exchanger. Moreover the proportion of energy consumption of plasma inputting into packed bed reactor was analyzed and calculated. The mechanisms that electrical energy of inlet plasma is transformed into heat energy in the two phases, gaseous and pellets of the packed bed reactor are different. The energy consumption in pellet phase is dielectric polarization loss and depends on packed bed geometry and DBD plasma etc. The energy consumption in gaseous phase is plasma sheath procedure. The important factors effecting on gas discharge are gaseous component and voltage, frequency of power.

1 Introduction

The so-called packed-bed reactor is constructed by placing dielectric pellets within the discharge region inside plasma reactor. The pellets could be either non-catalytic or catalytic. The latter is also termed as a plasma catalysis system. The plasma catalysis technique is widely used in Ozone generation [1], production of hydrogen from hydrocarbon reforming [2], gaseous pollutant removals [3] and gas heating. Packed-bed reactors have been proven to achieve higher energy efficiency than conventional plasma reactors either for ozone generation [4, 5] or for pollutant removal [6, 7].

The atmospheric plasma used in packed-bed plasma reactor can be divided into two types, i.e., thermal plasma and non-thermal plasma [2, 8], depending on the gas temperature. In thermal plasma, like plasma torch, all the charged species and neutral species are approximately in thermal equilibrium. The typical gas temperature ranges from 1,000 to 10,000 K. On the other hand, non-thermal plasma, such as corona discharge, DBD and surface discharge, are in thermally non-equilibrium state, in which the electron temperature (10,000–100,000 K or 1–10 eV) is much higher than the gas temperature (order of magnitude is about 100 K).

The plasma catalysis technique can be classified into two systems, i.e., single-stage and two-stage [1], depending on the position of catalyst. The single-stage type is constructed by packing catalyst pellets within the plasma zone or coating catalyst on the surface of electrode(s). The catalysts could completely or just partially overlap with the plasma zone. As for the two-stage type, the plasma zone is located either upstream or downstream the catalyst bed, which is termed as plasma pre-processing and plasma post-processing, respectively.

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The comparison between some intrinsic characteristics of these two different systems could provide useful information for determining which plasma is better. In general, two-stage combination is the only option for thermal plasma. On the other hand, for non-thermal plasma, the single-stage combination might be a better choice [1–3].

In general, heat transfer in packed bed can be divided into steady-state heat transfer and unsteady-state heat transfer. The analytical solution of temperature distribution and a variety of empirical formula of effective thermal conductivity and wall heat transfer coefficient have been researched thoroughly. Coberly and Marshall [9] studied coefficient of heat conductivity by experiments. And Ranz [10] proposed radial fluid hybrid model to investigate heat conductivity theoretically. The research of effective thermal conductivity of quiescent bed mainly includes two types of assumption. One assumption is that heat current is mono-directional [11–15] while the other assumption is that heat current is 2-D [16–19]. However, the heat transfer theories of packed bed are not enough for packed-bed plasma reactor because of the electrohydrodynamics (EHD) characteristic of plasma. Berges [20] summarized the studies of EHD enhancement of heat transfer and indicated that strong electric field will lead to fluid mixture near the heat transfer surface.

Previous research focused on the plasma chemical reaction. Whereas, it is worth noting that the temperature has a significant impact on the catalyst performance. The aim of this paper is to investigate the temperature characteristics of single-stage type packed bed plasma reactor by experiments and investigate the characteristics of energy conversion in the pellet phase.

2 Experiments and procedure

2.1 Packed bed plasma reactor

The geometry of a packed-bed reactor used in our experiments, as shown in Fig. 1, could be either cylindrical (or so-called ‘coaxial’) or parallel-plate.

Figure 1a illustrates the cylindrical packed-bed reactor. The $\gamma\text{-Al}_2\text{O}_3$ pellets (mean diameter d_p 3 mm) were set in quartz glass tubes (inner diameter d 54 mm and 116 mm) with a stainless steel rod (diameter 1.5 mm) at the center as a discharge electrode. The outer surface of the quartz glass tubes are covered with aluminum sheets of 100 mm and 1000 mm length as a ground electrode, respectively. Some asbestos was used to keep pellets’ position.

Figure 1b illustrates the parallel-plate packed-bed reactor. The $\gamma\text{-Al}_2\text{O}_3$ pellets (mean diameter d_p 3 mm) were set between two parallel plates. One plate was stainless steel meshes with different area ratio from 13, 24

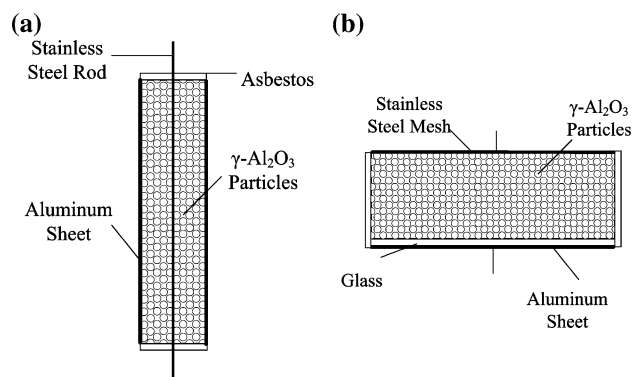


Fig. 1 Illustration of two types of packed-bed reactors with DBD. **a** Cylindrical. **b** Parallel-plate

to 32% and the other plate is glass plate covered with aluminum sheet. The stainless steel mesh and aluminum sheet are used as two electrodes. The distance between two plates l is 30 and 50 mm, respectively. And the plate area is $19\text{ cm} \times 11\text{ cm}$.

2.2 Experimental setup

Figure 2 shows the sketch of the experimental setup used in this study. The experimental measurements were carried out at room temperature under normal pressures. The power supply consists of an isolating transformer, a voltage regulator, and an impulsing power source. The isolating transformer is used to keep the isolation of the experimental setup from the other equipments. The output voltage of the voltage regulator ranges from 0 to 250 V.

It should be pointed out that two types of reactor were drawn up in Fig. 2 at the same time, while in the actual process the experiments were done for each reactor respectively.

2.3 Measurement of pellets temperature

A thermometer was set in the center of packed bed as shown in Fig. 2. The temperature changing with time is

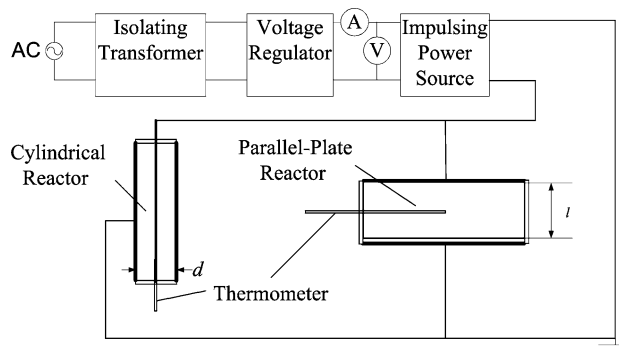


Fig. 2 Schematic diagram of the experimental setup

measured by the thermometer in conditions of various inlet voltage of DBD plasma in two types packed bed reactor. And the energy conversion from electric energy into heat energy in catalyst pellets can be calculated from the increase of temperature. The power input can be measured by the ammeter and voltmeter as shown in Fig. 2.

It is worth noting that the ambient temperature has a great impact on the measurement of temperature. Thus, the slope of temperature rise curve can be used to calculate the energy consumption instead of the absolute temperature.

3 Results and discussion

3.1 The temperature change in cylindrical reactor

Figure 3 shows the temperature change in cylindrical reactor under different voltage with different diameter. In Fig. 3a, the diameter d is 58 mm. Generally, the pellet temperature increases as the applied voltage of inlet plasma increases. When the voltage below 50 V, the slope of temperature rise curve increases with increasing of voltage. It is indicated that the greater the power input is, the more the heat production will be in pellet phase. However, the slope decreases with increasing of voltage when the voltage beyond 50 V. From Fig. 3 we can conclude an interesting phenomenon. When applied voltage is smaller, the proportion of energy consumed in pellets to the power input increases with increasing of applied voltage. However, when applied voltage is high enough, as ionizability increases, the proportion of energy consumed in pellets to the power input will decrease with increasing of applied voltage.

In Fig. 3b, the diameter is 120 mm which lead to lower electric field strength relative to that of Fig. 3a under the same applied voltage. And the slopes of curves increase as the voltage increases. But when the voltage is 130 V, the increase of slope relative to that of 110 V is not noticeable.

These characteristics indicate that the energy consumed in pellets will not always increase with applied voltage. The details of variation tendency of energy consumed on pellets should be calculated and in Sect. 4 some results will be shown.

3.2 The temperature change in parallel-plate reactor

Figures 4, 5 and 6 show the temperature change in parallel-plate reactors with different steel mesh electrode area ratio and different distance between two plates. Generally, the pellet temperature of packed bed increases with increasing of applied voltage of inlet plasma. Moreover, the slopes of temperature curves increase as the voltage increases.

The distance between two plates determines the electric field strength which has a significant impact on the creation of plasma. The shorter the distance between two plates is, the higher the electric field strength is in the plasma sheath, leading to higher ionizability. Another factor is that the area ratio of steel mesh which determines the secondary electron emission. The higher the area ratio of steel mesh is, the higher the secondary electron emission is, leading to higher current in the whole system.

Careful comparison of these three figures shows up some relationship between these two factors and the slopes of pellet temperature rise curves. Figure 4 indicates that the slopes of temperature rise curves with 30 mm distance between two plates are higher than that in the reactor with 50 mm distance when the area ratio of steel mesh is 13%. Conversely, when the area ratio is 24%, the slopes of temperature rise curves with 50 mm distance are higher than that with 30 mm distance as Fig. 5 indicates. However, when the area ratio is 32%, the slopes of curves with two distances between plates are approximately equal as Fig. 6 indicates. When the distance between plates is fixed as 30 mm, the slopes of temperature rise curves decreases in the order of the area ratio of steel mesh 13, 32 and 24%. However, when the distance is fixed as 50 mm, the slopes

Fig. 3 The temperature change in cylindrical reactor under different voltage with different inner diameter: **a** $d = 54$ mm, **b** $d = 116$ mm

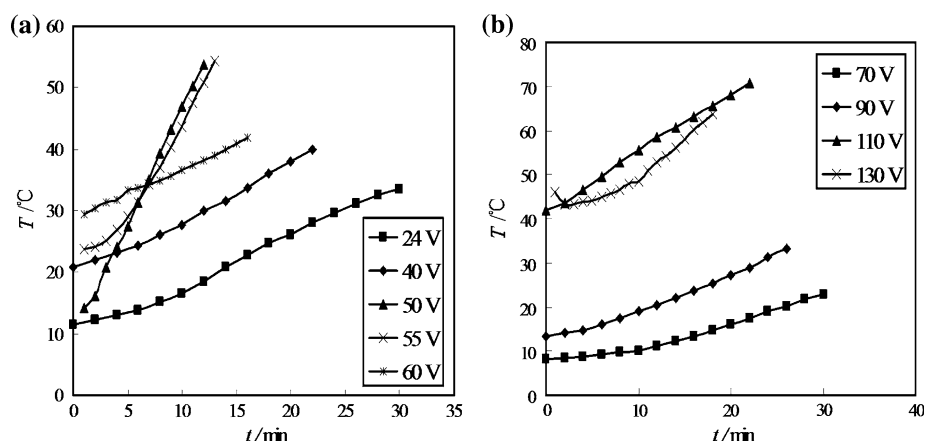


Fig. 4 The temperature change in parallel-plate reactor with the steel mesh electrode area ratio is 13% and the distance between two plates d is a 30 mm and b 50 mm

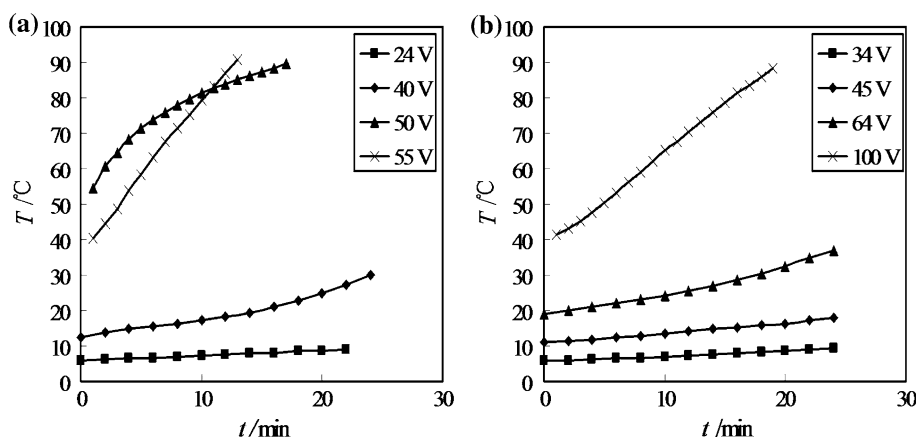


Fig. 5 The temperature change in parallel-plate reactor with the steel mesh electrode area ratio is 24% and the distance between two plates d is a 30 mm and b 50 mm

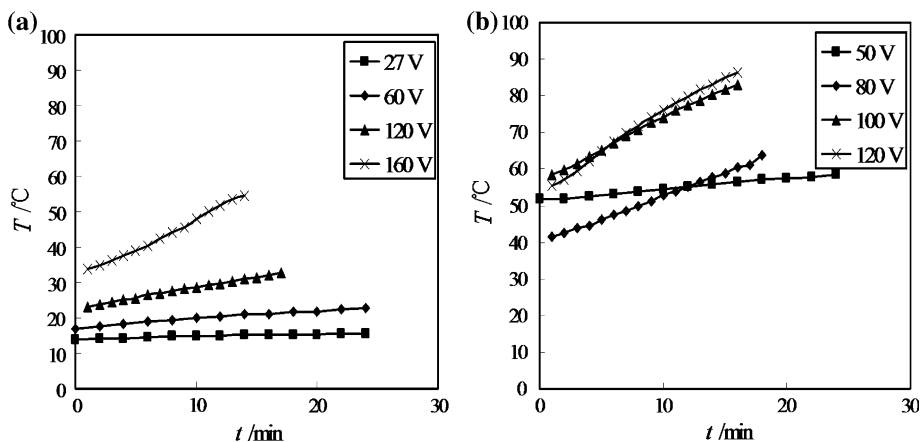
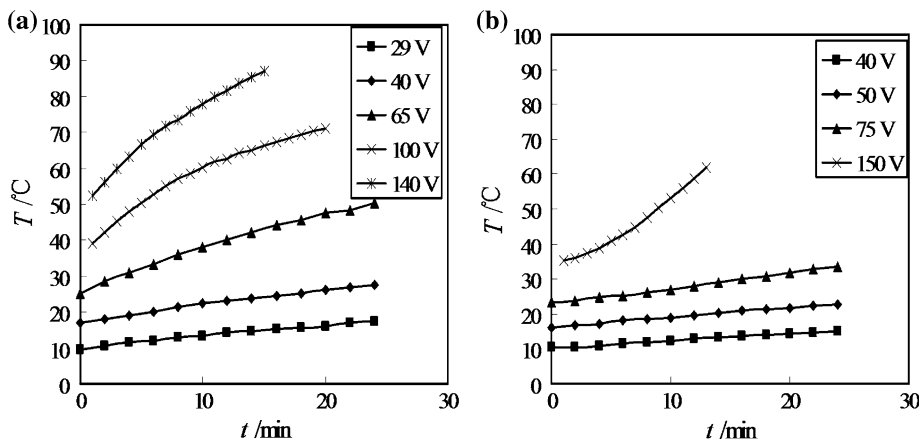


Fig. 6 The temperature change in parallel-plate reactor with the steel mesh electrode area ratio is 32% and the distance between two plates d is a 30 mm and b 50 mm



of temperature rise curves decreases in the order of the area ratio of steel mesh 13, 24 and 32%.

4 Analysis of energy conversion

The mechanisms that electrical energy of inlet plasma is transformed into heat energy in the two phases, gaseous and pellets of the packed bed reactor are different. The

energy consumption in pellets phase is dielectric polarization loss. And the energy consumption in gaseous phase is plasma sheath procedure.

4.1 The characteristics of energy consumption in the pellets

The energy consumption on the pellets is dielectric polarization loss which will increase the temperature of pellets

finally. The polarization in dielectric includes displacement polarization and relaxation polarization. Thus the energy loss should include two parts [21]:

$$W_p = V_d I_a = \left[\frac{\gamma S}{d} + \frac{\varepsilon_0(\varepsilon_s - \varepsilon_\infty) S}{d} \frac{\omega^2 \tau}{1 + (\omega\tau)^2} \right] V_d^2 \quad (1)$$

where the W_p is the polarization loss in pellets, γ is the electrical conductivity of pellets in AC electric field, S and d is the equivalent area and the equivalent thickness respectively, ε_0 , ε_s and ε_∞ is permittivity of vacuum, DC field dielectric constant, and optical dielectric constant respectively, τ is relaxation polarization time constant, ω is the pulsance of alternative electric field, V_d is the effective voltage dropped on the pellets, and I_a is the effective current.

In Eq. 1, V_d and I_a can give the power when there is no phase shift. However, it is very difficult to determine the electrical conductivity of every kind of dielectric pellets. And the voltage drop on the pellet and the equivalent thickness is difficult to be determined in packed bed. Thus, it is difficult to calculate the energy consumption on the pellets directly based on the voltage and the characteristics of pellets. In the following calculation, the energy consumption in pellets is calculated from the temperature rise of pellets.

The proportion of energy consumed in pellets to energy input in parallel-plate reactor can be calculated from experimental data as:

$$\beta = \frac{W_{pe}}{W_0} = \frac{m_p c_p \Delta T / t}{I_0 \cdot U_0} \quad (2)$$

where, β is the proportion of energy consumed in pellets to energy input, W_{pe} is the calculated value from experimental data of energy consumed in pellets, m_p and c_p is the mass and specific heat of pellets respectively, ΔT is the change of temperature in a duration of t . Figure 7 shows the characteristics of this proportion, W_0 and I_0 and U_0 is power input in reactor and the current and the voltage measured by the ammeter and voltmeter respectively. As mentioned, I_0 and U_0 can give the power when there is no phase shift. In Fig. 7a the distance between two plates is 30 mm and in Fig. 7b the distance is 50 mm. And the steel mesh electrode area ratios changes from 13, 24, to 24% in each figure.

In these curves we can get a conclusion that the tendency of proportion of energy consumed on pellets is not monotonic with applied voltage.

In Fig. 7a, the relationship between this proportion of energy and applied voltage is similar to a parabola with the area ratio of 13 or 24%. This proportion reduces firstly and increases afterward with the increase of applied voltage. Furthermore, the proportion with the ratio of 13% is bigger

than that with the ratio of 24%. However, when the area ratio is 32%, the proportion is bigger than that in two former cases. And it decreases with the increasing of applied voltage.

In Fig. 7b, the relationship between this proportion of energy and applied voltage is similar to a parabola with the area ratio of 24 or 32%. This proportion increases firstly and reduces afterward with the increasing of applied voltage when the ratio is 24% while it reduces firstly and increases afterward when the ratio is 32%. Furthermore, when the ratio is 24%, the proportion is bigger than that with the ratio of 32%. However, when the area ratio is 13%, the proportion is bigger than that in two former cases. And it increases with increasing of applied voltage.

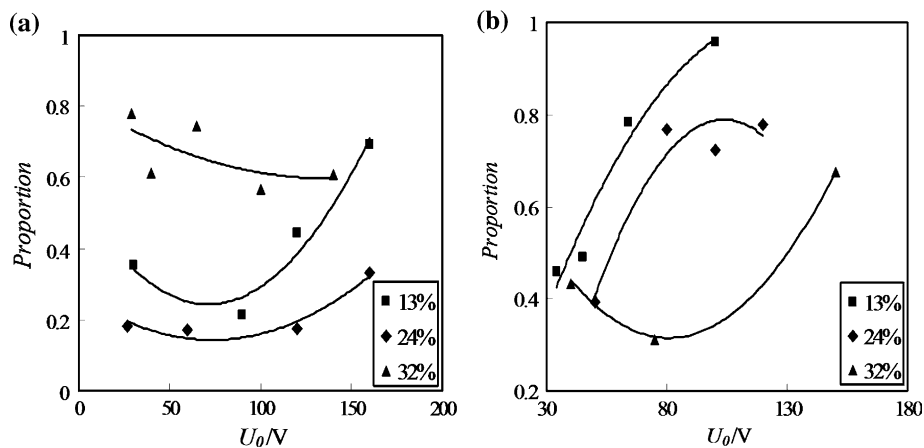
In packed bed plasma reactor, the power input is consumed in gaseous phase and pellet phase. The energy consumption in gaseous phase will change since the ionizability changes. In addition, the ionization reaction and exciting reaction have impact on the energy consumption. As a result, the proportion of energy consumption in pellet phase to electric energy of inlet plasma is complex.

4.2 Energy conversion in the gaseous phase

The energy conversion mechanism in gaseous phase is different from that in pellet phase. The electrons and ions gain energy from the alternating electric field. The energy absorption of electrons raises the electron temperature. However, this energy does not raise the macro temperature of whole gas. This is due to that the energy transfer from electrons to heavy particles in collision is very small since electron mass is far less than that of heavy particles. If the energy absorption of electrons is high enough, the ionization and excitation will be promoted in collision between electrons and heavy particles. However, generally, the mean energy gained by ions between two collisions is not enough to lead to ionization or excitation. This part of energy transfers to other gas molecules through frequent collisions and raises the macro gas temperature. In plasma energy calculation, the important parameters are electric field strength and electron density.

Because of the difference between the dielectric constants, the electric field strength is not uniformly distributed in packed bed plasma reactor. Takaki et al. [22] developed a one-dimensional parallel-plate model to evaluate the important plasma parameters for the reactor packed with spherical pellets in N_2 plasma. The time and cross-sectional averaged electric field (E_x) inside an approximately

Fig. 7 Proportion of energy consumed on pellets to the energy input with different distance between two plates: **a** 30 mm, and **b** 50 mm, and the steel mesh electrode area ratio changes from 13, 24, to 32%



spherical void and corresponding electron density (n_e) can be expressed by Eqs. 3 and 4,

$$E_x \approx \frac{V}{d} \frac{3\epsilon_p}{2\epsilon_p + \epsilon_g} \quad (3)$$

$$n_e \approx \frac{P}{V\alpha A e (\mu_0 E_0^\psi) E_x^{1-\psi}} \quad (4)$$

where, V is the applied voltage, d is the separation distance between the electrodes, ϵ_g and ϵ_p are the dielectric constants (or relative permittivities) of discharge gas and packing pellets, respectively, α and A are the void fraction and cross-sectional area of a packed-bed reactor, respectively, e is the electric charge of electrons, μ_0 is the electron mobility at reference electric field E_0 , and ψ is an empirical coefficient. P is the power consumed in the reactor. In general, the dielectric constant of pellets is bigger than that of discharge gas. Thus the electric field in void will be enhanced. However, the void between spherical packing pellets is usually not spherical and the electric field inside is more complicated. In fact, the maximum electric field near the contact point between pellets can be $10\text{--}10^4$ times higher than that in a spherical void, depending on the contact angle, curvature, and dielectric constant of the packing pellets [23–25]. Generally, the electric field and electron density in plasma is not uniform, and some empirical coefficients are difficult to be determined. Thus, the refined calculation is difficult. In some industry field, the plasma and dielectric pellets can be considered as two equivalent capacitances to compute the energy consumed in plasma.

The power consumed in gaseous phase discharge can be expressed as Eq. 5 [26]:

$$P = \frac{8\pi}{\omega} C_d \left(\frac{V_{op}}{V_g} - \frac{C_d + C_g}{C_d} \right) V_g^2 \quad (5)$$

where the P is the power consumed in gas discharge, C_d and C_g is the capacitance of pellets and discharge gas respectively, V_g and V_{op} is the biggest voltage on discharge gas and the applied voltage on the reactor respectively.

And here V_g is an average value, depending on gas component, pellet concentration and the gap width.

These two types of energy consumption, in gaseous phase and pellet phase, can be thought as connected in series. Some parameters such as electric field strength, the dielectric constant, the biggest voltage in plasma and empirical coefficient are impacted by the geometry and the characteristics of plasma. Thus the tendency of the proportion of these two types of consumption is complex as the experimental data has shown.

It should be pointed out, however, that the important factors effecting on gas discharge are gaseous component and voltage, frequency of power. In addition, the polarization loss in pellet phase depends on packed bed geometry and DBD plasma etc. These factors are not considered in this simple analysis. Furthermore, the heat transfer between two phases is not considered. Therefore, this model is only the qualitative analysis on the energy conversion.

5 Conclusions

The temperature characteristics and energy conversion in packed bed reactor combined with DBD plasma was investigated experimentally. The pellet temperatures of two types packed bed reactor, cylindrical reactor and parallel-plate reactor, was measured in conditions of various inlet voltage of DBD plasma. The relationship between pellet temperature of the packed bed and applied voltage of DBD plasma was discovered. Moreover the proportion of energy consumption of plasma inputting into packed bed reactor was analyzed and calculated. From these studies some conclusions can be drawn.

1. The experimental result indicates a tendency that the pellet temperature of packed bed increases with the applied voltage increases. When the voltage of inlet plasma is high enough, the pellet temperature increment decreases. Simultaneously, the packed bed

temperature is sensitive to the inlet plasma energy and there is a potential application to heat exchanger.

2. In a plasma catalysis system, the temperature has a significant impact on the catalyst performance. The experimental data indicates the electric energy input will increase the catalyst temperature. However, it is worth noting that the plasma has activation function. Therefore, when the gas in the discharge gap is prone to chemical reaction, the voltage choosing should be careful.
3. The proportion of energy consumed in pellets has a complex tendency because the complexity of the factors effecting plasma discharge and pellet loss. These factors include gaseous component, voltage and frequency of power, and packed bed geometry etc. A simple model was taken to analyze the energy conversion in two phases. And then some qualitative conclusions were drawn out to explain the complex tendency.

A complete theoretical analysis of the packed bed plasma need a variety of subject knowledge such as plasma chemistry, plasma physics, mass transfer and heat transfer etc. And a complete analysis will be taken in the future.

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