# POLYTETRAFLUORO-ETHYLENE ABLATION IN HIGH-TEMPERATURE JET OF DIFFERENT **CHEMICAL COMPOSITIONS**

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Аннотапия-В статье приводится описание экспериментальной установки-электродугового подогревателя, позволяющего получить высокотемпературную струю различного химического состава. Получены характеристики подогревателя и основных параметров генерируемой им струи (распределение скорости, давление и др.). Измерены средние тепловые потоки. Показано, что безразмерная скорость уноса политетрафторэтилена увеличивается с увеличением концентрации кислорода в набегающем потоке, особенно при низких температурах, что хорошо согласуется с теоретической формулой М. Адамса.

Показано, что вдув продуктов деполимеризации в пограничный слой может снизить тепловой поток по 25%.

## **NOMENCLATURE**

- $U_{\rm S}$ source voltage;
- P. pressure:
- $T_{\star}$ temperature:
- heat capacity;  $c_p$
- thermal conductivity:  $\lambda$ ,
- $density:$ ρ,
- viscosity:  $\mu$ .
- diffusion coefficient; D,
- molecular weight;  $M_{\cdot}$
- anode diameter;  $d_{-}$
- G. gas flow rate;
- $F_{\star}$ surface area;
- ŵ. relative velocity of jet;
- relative radius of jet:  $\overline{r}$ .
- $\bar{H}$ . mean-mass jet enthalpy:
- standard conditions enthalpy:  $H_{\alpha}$
- power exponent of jet velocity profile;  $n_{\cdot}$
- heat flux:  $q,$
- velocity of mass entrainment: m.
- heat of depolymerization:  $h_{\rm den}$
- combustion heat of depolymerization  $h_{\alpha}$ products:
- $C_{0}$ oxygen concentration in a gas flow:
- blowing factor:  $\gamma$ ,

 $\Delta m$ . increase of mass entrainment velocity due to chemical reactions.

# **Subscripts**

- at stagnation point;  $S_{\star}$
- at nozzle outlet; e.
- on impermeable surface: 0.
- at surface temperature; w.
- gaseous depolymerization products;  $v,$
- radiative:  $r_{\rm s}$
- arc.  $q_{\star}$

# **INTRODUCTION**

AT PRESENT intense studies are being carried out in the field of thermal shielding by ablating coatings. The survey of works in this field is presented in  $\lceil 1, 2 \rceil$ . Detection of physical and chemical processes causing the destruction of the outer layer of the coating is particularly complicated and at the same time very important in the above method of thermal shielding. Heat protecting materials are as a rule composite. The flowing by medium can react with the entrainment products and with the solid phase of the coating. Chemical reactions may also take place between individual components of

the material during its heating. This variety of physico-chemical processes is also complicated by the fact that all the phenomena listed proceed at the same time and affect one another. Theoretical analysis of the destruction process of composite materials is carried out with a number of simplifying assumptions. In this connection the experimental study of destruction of shielding materials under laboratory conditions in gas dynamic electric are installations becomes of great importance. Experimental study allows detection of the destruction mechanism of heat protecting coating and makes it possible to obtain the relations describing the dependence of destruction characteristics (mass entrainment velocity, effective enthalpy, surface temperature) on the parameters of the incoming flow (enthalpy, stagnation pressure and chemical composition of the flow).

There are many works concerned with destruction of heat protecting materials in the air.

Of great interest is the study of destruction of sublimating materials of polytetrafluoro-ethylene type in various media. In the present paper we have studied the process of destruction of polytetrafluoro-ethylene in argon, nitrogen, air and oxygen.

### EXPERIMENTAL INSTALLATION

For the generation of a high-temperature gas flow, an electric-arc heater with two 400 kW copper electrodes under cooling is used. The heater is supplied with electric energy from a mercury rectifier  $(U = 500 \text{ V}, I = 1500 \text{ A})$ .

The design of the heater and its characteristics are presented in  $\lceil 3 \text{ and } 4 \rceil$ . To reduce temperature and pressure oscillations in the flow and to level the parameters over the jet cross-section, a plenum chamber was mounted with cylindrical nozzles 14,15 and 18 mm dia. The jet discharged into atmosphere.

Argon, nitrogen, air and oxygen were used as working fluids. The gas flow rate changed within the following ranges, respectively:  $G_{Ar} = 13.0 -$ 14.5 g/s;  $G_{N_2} = 20-12$  g/s;  $G_{air} = 60-13.3$  g/s;  $G_{\text{O}_2} = 6.35 - 13.75$  g/s. Construction of the heater

and the procedure of measurement are shown in Fig. 1.

To obtain the required jet parameters, the necessary power was supplied to the charge. This was realized by regulating the ballast resistor *R* in series with the electric-arc heater

$$
R = (U - U_g)/I. \tag{1}
$$

In this case the value of the ballast resistance should be chosen so that the stability condition of the equilibrium state for the given gas flow rate be satisfied

$$
\frac{d}{dI}(U_g + U_R) \Big|_{U_g = U_{y_0}} > 0. \tag{2}
$$

Burning of the arc in the heater (and its voltage-current characteristic) is affected by a number of factors determining space-time distribution of heat sources in the arc and conditions of energy dissipation to the surrounding (heat entrainment by gas). The total effect of all the factors affects the behaviour of the voltagecurrent arc characteristics. The latter therefore depend on the value of the gas flow rate, properties of gas, its pressure and the way of supply, on geometric dimensions, the form of the electrodes, the electrode material, polarity of electrodes and finally on the resistance. The arc resistance in its turn depends on the temperature and the physical properties of the gas.

According to the molecular-kinetic theory of ideal gases, at  $p$ ,  $T = const$ , the physical properties of the gas are functions of a single physical value, i.e. of the molecular weight :

$$
c_P \simeq M^{-1}; \quad \lambda \simeq M^{-0.5}; \quad \rho \simeq M;
$$
  

$$
D \simeq M^{0.5}; \quad \mu \simeq M^{0.5}.
$$

For plasma generators with a non-constricted arc (as in the present case) and fixed geometric dimensions, it is reasonable to suppose that the voltage-current characteristic mainly depends on the gas properties with respect to some characteristic temperature and gas flow rate. The influence of some physical properties of gas upon the voltage-current characteristics



**FIG.** 1. Schematic drawing and measurement procedure. 1: electric arc heater; 2: sample; 3 : calorimeter ; 4 : model for measurement of pressure distribution ; 5 : turning device ; 6 : camera ; 7 : high-speed photo-recorder ; 8 : capacity with working gas ; 9 : reductor ; 10 : manometer ; 11: flow-rate regulator; 12: pump; 13: capacity with water; 14: tank for water; 15: mirror; 16: thermistor; 17 : thermocouple; 18 : connecting pipes; 19 : hot gas flow.

is therefore taken into account by a correction factor depending on the molecular weight.

The statistical voltage-current characteristics of the installation are given in criteria1 form in Fig. 2 [S]. The influence of the gas properties on the arc characteristics is quite obvious from Fig. 2. This is shown by the fact that the experimental points lie down on the straight lines typical for the given gas. It may be seen that the lower the molecular weight of the working gas is  $(N_z - 28.02, \text{ air} - 28.97, \text{ and } O_z - 32.0)$ , then the higher are the straight lines placed.

As a result of the studies it appeared possible to account for the influence of the physical properties via molecular weight. Voltage-current characteristics of the heater for gases with molecular weight  $28 \le M \le 32$  and for current from 200 to 1200 A can be calculated with  $\mp 15$  per cent error from the following formula

$$
U = \frac{5600}{10^{0.03M}} \left(\frac{G}{Id}\right)^{\frac{4}{3}} \left(\frac{d}{G}\right)^{0.05}.
$$
 (3)



**FIG. 2.** Generalized voltage-current characteristics of an arc heater operating with different gases  $d = 15$  mm;  $d_{\text{cath}} = 20 \text{ mm.}$  1: Oxygen: 13,  $G_{\text{O}_2} = 8.5 \text{ g/s}$ ; 14, 11.3; 15, 12.3; 16, 13.8. 2: Air: 17, 13.25 g/s; 18, 1.01 [19]; 19, 205 ( $d = 10$  mm) [19]; 20, 5.7–6.5; 21, 11.5. 3: Nitrogen: 4, 20 g/s; 5, 30; 6, 4.0; 7, 5.0; 8, 6.0; 9, 7.0; 10, 8.0; 11, 9.0; 12, 10.0.

## **PLASMA JET**

**The** study of thermal destruction processes of heat-protecting materials is closely connected with the study of a jet. Stagnation enthalpy

(temperature) and the velocity (stagnation pressure) are the main jet parameters. The knowledge of voltage-current properties of the heater, its performance and gas flow rate allows the determination of the mean-mass enthalpy of outflowing jet. Mean-mass velocity may be predicted by the formula

$$
\overline{w}_e = \frac{G}{\rho F}.
$$
 (4)

The installation allows the generation of a plasma jet with the following mean-mass parameters: (a) temperature  $3000-7000$ °K and (b) velocity  $300 - 1500$  m/sec.

The method of enthalpy and jet velocity determination from the energy balance suffers from an essential defect since it characterizes only mean parameters of flow and does not give an idea of distribution of the above parameters over the jet cross-section. Some results of jet temperature measuring by spectroscopic methods are given in  $\lceil 6 \rceil$  and 7.

The problem of determination of the local velocity of plasma jet is complicated by the fact that use of the probe methods known in hydrodynamics is hampered because of high temperature. Besides, the probes distort aerodynamic processes and virtually measure the disturbances which they themselves introduce.

A number of methods  $[8-11, 13]$  exists for measuring local velocity of a jet. The most popular one is the method of high speed photoscanning  $[8, 10, 11, 13]$ . It is also called the method ofbrightness fluctuations. In this method the velocity of brightness fluctuations movement in hypersonic flow is identified with the jet velocity. The reason of brightness fluctuations is in the burning mechanism of the arc in the plasma generator. The essence of the phenomenon lies in shunting the arc upon electrode wall  $\lceil 5, 14 \rceil$  which produces regions with various gas heating. Gas luminiscence changes sharply with temperature change.

The process of velocity measurement proceeds as follows. The flow picture is scanned by a rotating mirror in the direction normal to

the flow. This procedure was carried out with the help of a high-speed photo-registrating unit (Fig. 1) operating under the conditions of a photoregister. Velocity distribution was determined over the nitrogen plasma jet crosssection. Measurements were carried out for various operating conditions of a plasma generator with a plenum chamber and without it, with additional gas flow and without it. The velocities measured on the axis under operating conditions with a plenum chamber differ by  $1.5-12$  per cent from the mean-mass velocity (Table 1). In operations without a plenum chamber the difference is 58 per cent. This shows that the plenum chamber smoothes pulsations and levels the flow parameters over the jet cross-section.

The velocity distribution along the jet radius at the nozzle outlet for various combinations of parameters is given in Fig. 3. The relations obtained are described by the following empiric equation

$$
\widetilde{w} = 1 - 0.4 \widetilde{r}^2 \bigg( \frac{H_e}{H_0} \bigg)^n. \tag{5}
$$

It is found that the power exponent  $n$  depends mainly on the useful power and gas flow rate (Fig. 4).

The ablation process in heat-protecting materials (melting, mainly) is greatly affected by the pressure distribution on the ablating surface. The pressure distribution was measured on a model (Fig. 1) with a flat face end with the diameter equal to that of the sample (20 mm) under the following conditions: with nitrogen,  $G = 7.0$  g/s,  $d_c = 14$  mm,  $\overline{T_e} = 6000$ °K,  $\overline{w_e} = 900$  $m/s$ ,  $w_{axis} = 1000$  m/s, test holes were 1 mm dia. the distance between the model axis and the nozzle outlet was 10 and 20 mm.

The pressures were measured with standard manometers of the Bourdon type  $(1 \text{ kg/cm}^2)$ . For simultaneous records of the readings of all the manometers the whole instrument panel was photographed.

The profiles of total pressure measured at the above distances practically coincide and Table 1. Some data on the electric heater and the plasma jet parameters





FIG. 3. Radial velocity distribution. Experimental conditions are presented in Table 1. 1: regime 4; 2:6; 3:7; 4:3.

are correlated by the approximate formula within  $+8$  per cent.

$$
p = p_s \left( 1 - \frac{x^4}{R^4} \right). \tag{6}
$$



FIG. 4. Velocity profile exponent n vs. parameter  $\eta$ Ng/G.

The heat flux from the hot gas to the body is the parameter which essentially determines the behaviour the heat protecting material. The heat flux was measured with a water calorimeter  $\lceil 2, 15 \rceil$  the diagrammatic picture of which is given in Fig. 1.

With the water flow rate given  $(G_{\text{wat}})$  and calorimeter dimensions known, the only thing that is wanted for the calculation of the heat flux are the measurements of the temperature rise of cooling water  $(\Delta t)$ . If the heat leakage from the non-operating calorimeter surfaces is neglected (in the present case they are negligible), then the mean specific heat flux to the calorimeter surface is determined by

$$
q = c_p G_{\text{wat}} \Delta t / F. \tag{7}
$$

The great advantage of the calorimeter measurement is that the calorimeter may be exposed to a plasma jet for a long time. The essential disadvantage of the calorimeter is the low temperature of its working surface  $(T_w \simeq 700$ °K) which does not allow simulation of the temperature conditions in materials with 'higher surface temperature  $(T_w \ge 2000)$ °K). Therefore when determining heat flux to an impermiable surface of heat-protecting coating  $(q_0)$ , it is necessary to introduce the correction factor for the difference between the calorimeter surface temperature and the sample surface temperature

$$
q_0 = q \frac{\overline{H}_e - \overline{H}_{w_0}}{\overline{H}_e - H_w}.
$$
 (8)

In Fig. 5 the measured heat fluxes for various gases are compared with the values predicted by Fay and Riddell for a laminar equilibrium



**FIG. 5.** Plot of Nusselt number vs. Reynolds number. 1, argon; 2, nitrogen; 3, air; 4, oxygen; 5, Fay and Riddell's theory [16] for nitrogen,  $d_c = 14$  and 15 mm. Black dots  $d_{\text{rev}}$  $= 10$  mm, white  $d_{\text{rev}} = 20$  mm.

boundary layer. The comparison is presented in dimensionless coordinates  $Nu_w$  and  $Re_w$ . The diameter of the heat flux meter is taken as the characteristic dimension. The comparison shows good agreement between the measured and predicted [16] values. As it is seen from the diagram, the value of the heat flux in the studied region depends on the enthalpy and the flow velocity and does not depend on the kind of gas used within the experimental error range. Hence the conclusion follows that the value of  $Nu$  for the gases under study will mainly be determined by the enthalpy drop in a boundary layer and velocity gradient which is in agreement with the theoretical premises of work [16].

## **ABLATION OF POLYTETRAFLUORO-ETHYLENE**

Polytetrafluoro-ethylene (Teflon) is a product of tetrafluoro-ethylene polymerization. It possesses the chain  $(C_2F_4)$ , with a high degree of symmetry. Mean molecular weight of the polymer ranges from 389000 to 890000.

In intensive heating of polymer materials, depolymerization reaction takes place which is believed to result in production of a gaseous monomer. This is accompanied by absorption of about 1740 kJ/kg for the case of polytetrafluoro-ethylene. It may be supposed that if oxygen components are present in the main stream, the monomer can be involved into the chemical reactions

$$
(C_2F_4)_{gas} + O_2 \rightarrow 2COF_2
$$
  

$$
(C_2F_4)_{gas} + O_2 \rightarrow 2CO + 2F_2.
$$

These reactions are exothermie. Combustion heat per I kg of oxygen for polytetrafluoroethylene is about 23000 kJ/kg.

To compare the efliciency of a heat-insulating material, experimental values of the dimensionless entrainment velocity  $(\bar{G})$  and effective enthalpy  $(I_{\text{eff}})$  are usually used.

The dimensionless entrainment velocity is determined from the formula

$$
\overline{G} = \frac{\dot{m}_v}{(\alpha/c_p)_o}.
$$
 (9)

With no mass entrainment the heat-transfer coefficient is defined by

$$
(\alpha/c_{\rm P})_o = \frac{q_0}{\overline{H}_e - H_w}.
$$
 (10)

The effective enthalpy in the inert atmosphere may be determined from the following expression

$$
I_{\rm eff} = \frac{q_o - q_r}{\dot{m}_v}.
$$
 (11)

ment velocity was determined with the help of through the boundary layer to the surface. a camera. Depending on the operating conditions The experimental dimensionless entrainment and jet composition, the linear velocity of velocities are compared with those predicted polytetrafluoro-ethylene entrainment changed by the formula within the range  $0.17$  to  $0.62$  mm/sec for an argon jet,  $0.95-2.0$  mm/sec for a nitrogen jet and  $0.44-1.8$  mm/sec for an air jet and  $0.6-$ 1.3 mm/sec pure oxygen.

It follows from Fig. 6 that mass entrainment increases with the increase of enthalpy and



FIG. 6. Dimensionless mass entrainment velocity plotted FIG. 0. Dimensionless mass entrainment velocity plotted<br>against the main stream enthalpy for various gases: 1, argon; comes from the fact that within the runs the 2, nitrogen; 3, air; 4, oxygen; 5, air [20]. For all the points stagnation pressure was not maintained equal the stagnation pressure  $p_s \simeq 1.02$  bar; 6, air [12], stagnation pressure  $p_s \simeq 6.20$  bar. Solid lines (7, 8 and 9) are the values for inert atmosphere, air and pure oxygen as predicted by above it. In the present work the excessive (12),  $M<sub>n</sub> = 100$ .

in the incoming flow. The value of mass entrain-<br> $\frac{1}{20}$  increasing stagnation pressure. ment obtained in air exceeds by 30 per cent the increasing stagnation pressure.<br>If the value of the heat flux due to burning is corresponding value in an inert atmosphere supposed to be spent only on the increase of (argon, nitrogen) due to the increase of heat supposed to be spent only on the increase of flux at chemical reactions. In the atmosphere of pure oxygen this difference may be up to 90 per cent. The increased mass entrainment in pure oxygen, as compared with air, indicates that homogeneous chemical reactions near the ablating wall in the air proceed in the diffusion or<br>transition region and the process of oxidation is the inert and aggressive media is known.

In the experiments the linear mass entrain- controlled by the amount of oxygen diffused

$$
\overline{G} = \frac{1}{\frac{h_{\text{dep}} + c_P(T_w - T_\infty)}{\overline{H}_e - H_w + C_{o_2} \cdot h_q}}.
$$
(12)

This formula is obtained from Adams' formula [l, 121 including expressions (10) and (11) and allowing for the blowing effect.

The value ofthe blowing factor for the products ofdepolymerization was calculated by Mugalev's formula  $\lceil 17 \rceil$ 

$$
\gamma = 0.6 \left(\frac{H_w}{\overline{H}_e}\right)^{-0.03} \left(\frac{M_e}{M_v}\right)^{0.24} \tag{13}
$$

The surface temperature was assumed equal to 750°K [l].

For inert atmosphere and oxygen experimental data agree well with the predicted values. For air at atmospheric pressure experimental data fall somewhat above the predicted ones. This may be attributed to the fact that the polymer does not completely decompose into a monomer [12]. An additional explanation to the atmospheric pressure, but was usually pressure was up to  $0.6$  bar. The comparison of curve (10) with curve (8) and experimental points depends on the presence of oxygen components (3) and (5) has shown that the dimensionless<br>entrainment velocity increases rapidly with

enthalpy is calculated by the formula

$$
I_{\text{eff}}^* = \frac{q_o - q_p}{m_v + \Delta m_v}.
$$
 (14)

the inert and aggressive media is known,

the increase of heat flux due to chemical reactions can be evaluated

$$
q_g = \Delta \dot{m}_v I_{\rm eff} = q_o \bigg( \frac{I_{\rm eff}}{I_{\rm eff}^*} - 1 \bigg). \qquad (15)
$$

With the help of relations  $(9-11)$  this equation can be reduced to the form

$$
q_g = q_o \left(\frac{\bar{G}^*}{\bar{G}} - 1\right).
$$
 (16)

Estimation of the heat flux due to burning by relation (16) has shown that the mean value of the heat flux to the surface of polytetrafluoroethylene with enthalpy flux  $>5000$  kJ/kg increases up to 40 per cent in the air medium and 70 per cent in the oxygen medium.

Study of the decreasing effect of blowing into the laminar boundary layer on heat flux from a high-temperature gas flow to a solid body is of great practical importance. With depolymerization heat known, it is easy to determine the heat flux at the ablation surface

$$
q_w = \dot{m}_v h_{\text{dep}}.\tag{17}
$$

This allows to demonstrate the blowing effect of



FIG. 7. Effect of blowing depolymerization products into a boundary layer on heat-transfer close to the stagnation point. 1, argon ; 2, nitrogen.

tetrafluoro-ethylene depolymerization products on the heat transfer at the stagnation point. This effect may be described as the ratio of the heat fluxes at the impermeable surface  $(q<sub>a</sub>)$  to the heat fluxes with mass injection  $(q_w)$  v. the dimensionless blowing velocity.

In Fig. 7 this dependence is shown for an inert atmosphere (argon, nitrogen). The curve is linear which is in agreement with the theoretical predictions  $\lceil 17 \rceil$ . The heat flux may decrease due to the blowing effect by 75 per cent and more.

In aggressive atmospheres heat fluxes to the ablation surface are somewhat higher due to burning of the depolymerization products in a boundary layer.

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Abstract-In the paper an experimental unit of an electric heater is described which produces hightemperature jets of various chemical compositions. The characteristics of the heater and basic parameters of the jet produced (velocity distribution, pressure, etc.) are obtained. The mean heat fluxes are measured. The dimensionless entrainment velocity of polytetrafluoro-ethylene is shown to increase with oxygen concentration in the main stream, especially at low temperatures, which fact agrees well with the theoretical formula of Mac C. Adams.

Blowing of depolarization products is shown to decrease the heat flux by up to 25 per cent.

Résumé—Dans cet article, on décrit une unité expérimentale de réchauffeur électrique qui produit un jet à température élevée et de différentes compositions chimiques. Les caractéristiques du réchauffeur et les parambtres de base du jet produit (distribution de vitesse, pression, etc.) sont obtenues Lea flux de chaleur moyens sont mesurés. La vitesse d'entraînement non-dimensionalisée du polytetrafluoroéthylène augmente avec la concentration d'oxygène dans l'écoulement principal particulièrement aux faibles températures qui en fait sont en bon accord avec la formule théorique de Mac C. Adams.

On montre que le soufflage des produits de dépolarisation diminue le flux de chaleur jusqu'à 20 pour cent.

Zusammenfassung-In dieser Arbeit wird die experimentelle Anlage einer elektrischen Heizung beschrieben, die Hochtemperatur-Freistrahlen verschiedener chemischer Zusammensetzungen erzeugt. Die Charakteristiken der Heizung und der Grundparameter des erzeugten Freistrahls (Geschwindigkeitsverteilung, Druck usw.) werden erhalten. Die mittleren Wärmeströme werden gemessen. Es zeigt sich, dass die dimensionslose Mitreissgeschwindigkeit von Polytetrafluor-Äthylen mit der Sauerstoffkonzentration in der Kernströmung insbesondere bei niedrigen Temperaturen zunimmt. Diese Tatsache stimmt gut mit der theoretischen Beziehung von Mac C. Adams iiberein.

Weiterhin zeigt sich, dass das Einblasen von Depolarisationsprodukten den Wärmestrom bis zu 25% herabsetzt.