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The Heat Transfer Mechanism and Fire Insulation Properties of Some Intumescent Materials

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Heat transfer mechanisms and fire insulation properties of some intumescent materials are reviewed.

KEY WORDS Heat insulation, fire insulation, intumescence.

INTRODUCTION

At present the problems of heat and fire insulation of different surfaces and constructions from the effects of radiation and convectional heat flow $(1, \ldots, 2 \times 10^5 \text{ W/m}^2)$ occurring during fires, are very actual.^{1,2}

One of the methods of realizing the solution to the problem of increasing the thermal stability of objects in fire conditions is the creation and application of heat and fire insulation coatings.¹⁻³ They provide temperature decreases to safe levels near insulated surfaces, and thus protect them from destruction. Intumescent coatings are most optimal in this case.²⁻⁴ They provide the highest insulation effect with minimal initial thickness. In addition to being inexpensive, the technology of their application is simple.

Intumescent coatings have many components: substrate, polymer binder and additives, providing adhesion, gas generation, intumescence and hardening of carbonized residue. The formulation of such coatings varies greatly¹⁻⁵ and the emergence of new types makes the problem of studying heat transfer processes and their description very actual. Since the transfer mechanism and chimism of occurring processes are complex and not well understood for the warmup process, it is necessary to use parameters that characterize this process in an integral way and that are determined reliably from experiment at the first stage of investigation. Such an approach allows us to solve the practical problems of thermal projection of heat and fire insulation systems with different intensity and duration of external heat requirements.

EXPERIMENTAL

In Reference 6 it is considered, that intumescence proceeds within an infinite thin layer and process description is fraught to Stefan problem. A physical picture of the intumescence process is given in Figure 1, where (a) initial, (b) intermediate, and (c) finite stages are shown for typical heat and fire insulation coatings with initial thickness h_0 applied to the surface.

In this paper two types of coatings with various relative increased thicknesses h/h_0 are investigated. The mass loss $\Delta m/m_0$, effective sample density ρ , and value $\Delta h/h_0$ against thermostatting temperature T in gas flow (nitrogen, air)⁷ were measured experimentally. Experimental dependencies of these values for coating types are given in Figure 2. The curves $\rho(T)$ are shown to have an S-form character at T > 900 K tending to the asymptote corresponding to a final state (the carbonized layer in Figure 1). The increase in thickness $\Delta h/h_0(T)$ occurs in the range of 400 K < T < 700 K and, as is shown in Figure 2, it can proceed for several stages and



FIGURE 1 Physical picture.



FIGURE 2 (1) Mass losses $\Delta m/m_0$; (2) effective density ρ ; and (3) relative increased thickness $\Delta h/h_0$ against temperature of thermostatting T for two types of coatings: (a) $\Delta h/h_0 \approx 0, \ldots, 1.4$; (b) $\Delta h/h_0 \approx 0, \ldots, 1.4$; (b) $\Delta h/h_0 \approx 0, \ldots, 1.5$ (relatively marked as SGK - 1).

differs for various materials, which is due to the variety of their formulation: $a - \Delta h/h_0 = 0, \ldots, 1.4; b - \Delta h/h_0 = 0, \ldots, 1.5.$

The fire insulation ability of coatings under the exposure of a naked flame of an acetylene-oxygen burner was investigated on a rotatable table (1) of the test bed.⁸ (See Figure 3.) Heat insulation coating was applied to the surface of steel plates (2) with dimensions of $100 \times 100 \times 2$ mm and layer thickness $h_0 = 2.5$ mm in correspondence with Reference 5. The disposition scheme of the tested samples (3) is given in Figure 3. Plates with coatings were arranged with their faces along normal to the vector of velocity of running on flow of high temperature gas (flame). Lateral and reverse plate faces were insulated from the environment by an asbestos layer (4). Thermocouples (5) of CA (chromel-alumel) type with a thickness of ≈ 50 mkm were embedded on reverse plate faces (2) and on the surfaces of the tested samples. In addition, thermocouples were arranged at different distances from the samples surfaces (2, 10, 20, 30, 30, 40 mm) in order to record interfaces of media in the intumescence process of the samples during warmup, as was suggested in Reference 4. Four plates with coating applied to their surfaces and detectors for the recording of total q_s (6) and radiant q_L (7), thermal flows were simultaneously placed on the test bed.

An experiment was carried out as follows. One of the tested samples was placed against the source of convective warmup, the shutter was opened and the sample underwent the influence of high temperature gas flow during the given time interval. At the end of annealing the flame was intercepted by shutters and another sample was placed by a 60° turn of the table. After the annealing of all of the samples, radiant and convective thermal flows were measured with detectors. Signals from thermocouples and detectors were recorded by an oscillograph H-115 (9). According to detector records, the convective and radiant components of thermal flow were determined, and the convective heat exchange coefficient α and the gas flow temperature T_E were calculated according to the method in References 7 and 9.

A typical oscillogram of temperature recordings T(y, t) in coating SGK-1 made at different distances from surface of metal plate y_i is given in Figure 4.



FIGURE 3 Principle scheme of the test bed: (1) rotatable table; (2) plates; (3) samples; (4) heat insulator; (5) thermocouples; (6) detector of total heat flow; (7) detector of radiant heat flow; (8) shutter; (9) recording equipment.



FIGURE 4 Temperature T(y, t) and linear dimensions $\Delta y(t)$ changes of SGK sample in flow of hightemperature gas (temperature $T_E = 1440$ K, convective heat exchange coefficient $\alpha = 76$ W/m² K).

RESULTS AND DISCUSSION

During time t from the beginning of warmup, the sample temperature T_w increases and reaches the maximum of T = 610 K at t = 18 sec; subsequently a decrease in $T_w(t)$ is observed due to an increase in thickness and absorption of thermocoupling by the intumescent mass of the SGK-1 sample. At the moment of contact of the sample surface with thermocouples, on curves $T_{s,i}$ ($i = 0, 1, \ldots, 5$) there occur inflection points (A, B, C, D, E) characterizing the position of the media interface in space and time. The presence of characteristic points on curves $T_{s,i}(t)$ allows us to follow the dynamics of linear dimensions enlargement $\Delta y(t)$ with sample warmup in gas flow. In this case with $t \approx 0.4$ min a sharp decrease in temperature is observed as the end-peak on the temperature curve $T_{s,0}(t)$ (at the moment of t = 0 the thermocouple is placed on the sample surface).² Apparently this effect is connected with the simultaneous influence of the thermal destruction front of the polymer binder and the intumescence front on the thermocouple junction during the initial warmup period. With time they diverge in opposite directions (see Figure 1).

The behavior of the curve $\Delta y(t)$ speaks to the two-stage character of the thermal destruction process of the polymer binder of fire insulation coating SGK-1. The first stage proceeds in the range of 450 < T < 550 K, the second at T > 550 K. Superposition of two processes (intensive intumescence and absorption of heat due to thermal chemical transformations) leads to the appearance of inflection points on temperature curves T(y, t). Data in Figure 2 confirm the presence of these effects. The process became regular when t > 10 min. During a subsequent 60 min warmup of a carbonized sample the temperature $T_s(t)$ is slowly increased to T = 610 K on the reverse face of the metal substrate.

Analysis of curves $T_s(t)$, given in Figure 5, allows us to conclude that the reaction zone of thermal decomposition occupies total sample volume up to $t \approx 4.5$ min. At this moment its thickness is ≈ 25 mm. When $t \approx 5$ min, the 9-mm thickness layer, burning against the surface, has been completely carbonized. An inflection point is observed on curves T(y, t) (curve $3 - y \approx 9$ mm; curve $- y \approx 12$ mm). A temperature of $\approx 930-950$ K is the temperature of completeness of the materials thermal decomposition process (see also the dependencies $\Delta h/h_0$ and $\Delta m/m_0$ in Figure 2b).

Attention should be paid to the sharp decrease in velocity of isotherm dy_T/dt displacement when t > 5.1 min (see Table I) which also explain the infumescent process.

When T(y) > 700 K, velocity dy_T/dt decreases more than two times, due to the intensification of heat exchange in the subsurface carbonized layer. As was shown



FIGURE 5 Profiles of temperatures in the SGK-1 sample at different moments: (1) t = 3.5 min; (2) t = 4.3 min; (3) t = 5.1 min; (4) t = 6.6 min; (5) t = 10 min. ($T_E = 1440$ K, $h_0 = 2.5$ mm.)

TABLE I

Displacement velocity of isotherm dy_T/dt (mm/sec) according to sample thickness at different moments			
	<i>t</i> , min		
T(y), K	4.3	5.1	6.6
575	0.21	0.21	0.12
700	0.16	0.16	0.07
800	0.16	0.16	0.07
900	_	0.15	0.06
1000	-	-	0.03

by structural investigations the porosity of this layer reaches ≥ 0.8 when $t \ge 6.6$ min. During sample warmup, curves T(y) (4 and 5 in Figure 5) with two distinct temperature plateaus correspond to the character of the intumescence curve $\Delta h/h_0(T)$ in Figure 2b at low and high temperatures.

THEORETICAL

The description of nonstationary warmup of coating, applied to substrate, under exposure of external heat requirements was made on the basis of a simple model. The law of energy conservation and alternative relations of integral type, determined from the experiment, are the basis for this model:

$$(\rho c)_{ef} \frac{\partial T}{\partial t} = \frac{\partial}{\partial y} \left(\lambda_{ef} \frac{\partial T}{\partial y} \right) - (\rho c)_{ef} \upsilon \frac{\partial T}{\partial y} Q \dot{R}(T)$$
(1)

$$T(O, y) = T_h, \quad y(y_o, y_w(t))$$
 (2)

Boundary conditions:

$$\lambda_{ef} \frac{\partial T}{\partial y} y - y_w(t) = \alpha (T_E - T_w) + \varepsilon_w \sigma (\varepsilon_E T_E^4 - T_w^4)$$
(3)

$$\lambda_{ef} \frac{\partial T}{\partial y} y = y_0^+ = \lambda_M \frac{\partial T_M}{\partial y} y = y_0^-$$
(4)

Thermal state of substrate:

$$(\rho c)_{M} \frac{\partial T_{M}}{\partial t} = \frac{\partial}{\partial y} \left(\lambda_{M} \frac{\partial T_{M}}{\partial y} \right)$$
(5)

$$T_{\mathcal{M}}(O, y) = T_{\mathcal{M}} \tag{6}$$

$$\lambda_M \frac{\partial T_M}{\partial y} y = 0 = 0 \tag{7}$$

Here T is the temperature; v the current velocity of sample points; ρ , c, λ the density, heat capacity, and heat conduction of the material; Q, R(T) the thermal effect and velocity of material thermal decomposition process; α , ε the convective heat transfer and blackness coefficient; σ the Stefan-Boltzmann constant; y_o , $y_M(t)$ the coordinates of internal and external coating surfaces: indices H, W, E, M, ef correspond to the intial state, external coating surface, flame, substrate and effective value, respectively.

In Equation (1) the term comprising velocity v is responsible for deformation of the temperature field, which is due to the increase in thickness during the intumescence process. When writing (1), the enthalpy of escaping gaseous products was neglected. The solution of this equation requires determination of the value V through warmup characteristics. Since intumescence is the result of complex and less understandable physical-chemical processes occurring in material, then experimental dependence of relative increased thickness $\Delta h/h_0$ (see Figure 2) of the elementary sample, obtained under isothermic warmup conditions was used to complete the model. The given dependence was approximated by temperature function, by means of which the current velocity of the material and the coordinate of the warmup surface were calculated:

$$v(y, t) = \frac{d}{dt} \int_{y_0}^{y} \frac{\Delta h}{h_0} (T) \, dy, \qquad y_w(t) = \int_{y_0}^{y_M} \frac{\Delta h}{h_0} (T) \, dy \tag{8}$$

In boundary condition (3) convective and radiant components of the total heat flux were underlined, and the reradiation of the surface was taken into consideration. On the external face of the coating the condition of ideal thermal contact was supposed (4), and the reverse face of the substrate was considered to be thermally insulated (6).

For substrate material with high heat conduction λ_M , e.g. metal, equation (5) is not necessary due to the infinitesimal size of the temperature gradient according to thickness. As a result (4)–(6) are generated into the condition of thermal balance:

$$(\rho c \delta)_M \frac{\partial T}{\partial T} w = \lambda_{ef} \frac{\partial T}{\partial y} y = y_0$$

where δ_M is the substrate thickness.

The thermal-physical properties c_{ef} and λ_{ef} are determined by the structure of the porous layer. They were found from the solution of the inverse problem of heat conduction for thermally stabilized sample conditions by the use of analytical solutions.⁷

Numerical solution of equations (1) and (5) with boundary conditions (3), (4), and (8) was made by a unique method on the basis of an implicit difference scheme, having first-order approximation according to time t and coordinate y. The latter was connected with the irregularity interval of the difference network according to the thickness of the intumescent material. The presence of the convective term in (1) with completed relations (8) gives an iterative character to obtaining the solution to Equation (1).

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FIGURE 6 Temperature fields in intumescent coating.

The calculations of unsteady warmup of intumescent coating with pure convective warmup with parameters $T_E = 780$ K, $\alpha = 117$ W/m² K were carried out. The initial thickness of coating was $h_0 = 4.5$ mm and $T_H = 300$ K. Material (Figure 2a) with small value $\Delta h/h_0$ was considered (the last term in Equation (1) was neglected).

Experimental (solid) and calculated (dashed) curves of temperature $T_w(t)$ changes of sample surfaces according to time and in its interval points with coordinates y_i = 4.15 mm and y_i = 3.65 mm from the reverse face of the material are given in Figure 6. Here the coordinate $y_w(t)$ of a heated surface (current thickness) is given. It is seen that calculated and experimental data are in satisfactory agreement not only according to the integral value (current thickness $y_w(t)$), but according to local characteristics of warmup (temperatures in internal points).

CONCLUSIONS

- 1. Integral characteristics (sample density ρ and relative increased thickness $\Delta h/h_0$) have been determined for two types of intumescent coatings under conditions of isothermic warmup. It is shown that the intumescent process has a two-stage character.
- 2. The unsteady temperature field in intumescent coating of high multipleness has been investigated by means of thermocouple measurements and the phenomenology of the intumescence process is given.
- 3. A simple method is suggested for calculation of the temperature field in intumescent material, applied to substrate. It is in satisfactory agreement with measurement data, which allows us to provide designs for fire insulation constructions by means of choice of type and thickness of coatings for various heat requirements.

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