MATHEMATICAL SIMULATION
OF THE GAS-PARTICLES REACTION FLOWS
IN INCINERATION OF METAL-CONTAINING WASTE

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ABSTRACT

A "quasi-equilibrium" approach for thermodynamic calculation of chemical composition and properties of metal-containing fuel combustion products has been developed and used as a part of the mathematical model of heterogeneous reacting flow which carry burning and/or evaporating particles. By using of this approach, the applicable mathematical model has been devised, which allows defining the change in chemical composition and thermal characteristics of combustion products along the incineration chamber.

As an example, the simulation results of the reacting flow of magnesium-sodium nitrate-organic mixture are presented. The simulation results on the gas phase temperature in the flow of combustion products are in good agreement with those obtained experimentally.

The proposed method of "quasi-equilibrium" thermodynamic calculation and mathematical model provide a real possibility for performing of numerical experiments on the basis of mathematical simulation of nonequilibrium flows of combustion products. Numerical experiments help correctly to estimate the work characteristics in the process of treatment devices design saving time and costs.

INTRODUCTION AND SOME PHYSICAL BACKGROUND

Recently, a complex project was initiated at SIA "Radon" with the final task to develop selective thermochemical processes in order to treat mixed and radioactive waste containing heavy metals, radioactive and organic contaminants (1-3). Thermodynamic simulations prior to experimental tests are of crucial importance. These simulations facilitate designing of selective to hazardous components powder metal fuels, which shall be used to minimize the releases of chemical and radioactive species.

Also in a design and development of the temperature control systems for the waste thermal treatment facilities, a composition and properties of combustion products generated should correctly be predicted in any points inside the treatment chamber. The combustion products of metal-containing waste are defined as a significantly nonequilibrium multiphase heterogeneous system with combustion and evaporation of metal and organic fuel particles and other heterogeneous processes. In particular, on combustion of the waste, containing metal powder and oxygen-bearing salts and organics, the combustion products flow in some chamber can be represented as follows (Fig.1).

On the burning surface ($x = 0$) and in the its immediate vicinity the salts and organics decomposition with the oxygen generation takes place. Some metal particles are ignited and have time to burn out on the burning mixture surface. The remaining particles are released from the burning surface, ignited and burn in the decomposition products.
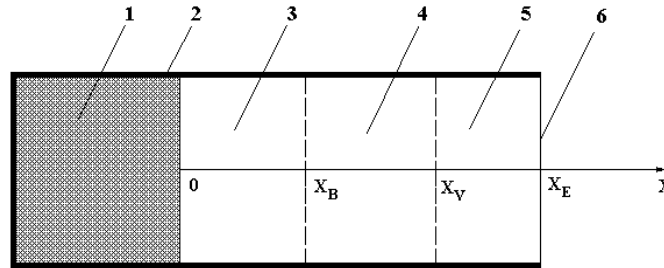
Fig. 1. The scheme of processes in chamber flow of metal-containing waste combustion products (at the deficit of oxidant).

1 – waste; 2 – chamber; 3 – area of oxygen "burnout"; 4 – area of metal drops evaporation;
5 – area of thermodynamic equilibrium; 6 – exit section of chamber.
$Y = Y_0$ at $x = 0$; $Y = Y_{max}$ and $Z = Z_0$ at $x = x_B$; $Z = Z_{max}$ at $x = x_V$.

The burning ends either when metal particles completely burn (at the stoichiometric or excess oxidant content in mixture) or then oxygen is completely used (at the deficit of oxidant in mixture). In the latter case, the metal particles, after the burning stops ($x = x_B$), begin to evaporate. The evaporation continues until a saturation of the metal vapor partial pressure in the combustion products shall be reached ($x = x_v$). As this takes place, a combustion products composition has become thermodynamically equilibrium. If the chamber length is too small then the equilibrium in combustion products is not attained.
The mixture of so-called "primary" combustion products generated in the chamber may reburn through an air feed into chamber.
The change in chemical composition and thermal characteristics of combustion products along the chamber can be defined on the basis of mathematical simulation.

GOVERNING EQUATIONS

In fact, the combustion products of waste being investigated are defined as a heterogeneous three-phase (gas-droplets-solid particles) system. The mathematical models for such three-phase system flows have recently evolved (4,5).
We are here deriving the equation set governing the outlined three-phase flow pattern along the incineration chamber under following simplifying assumptions:
- the flow is one-dimensional, steady, adiabatic, and subsonic;
- the droplets and solid particles are polydisperse and spherical;
- the condensed particles size distribution is assumed as discrete;
- the collisions and interactions of droplets and particles are not taken into account;
- the gravity effect is neglected.
It should also be noted that in the flow being considered all flow characteristics are strongly dependent on the degree of burnout and evaporation of disperse phase.
In view of these assumptions and comment, the equations, determining flow characteristics, can be written as:

$$\frac{1}{p}\frac{dp}{dx}+\frac{1}{\mu_g}\frac{d\mu_g}{dx}-\frac{1}{\rho_g}\frac{d\rho_g}{dx}-\frac{1}{T_g}\frac{dT_g}{dx}=0$$

(Eq.1)

$$\frac{d}{dx}(\rho_g w_g F)=M_g$$

(Eq.2)

$$\frac{d}{dx}(\delta_{q,i}w_{q,i}F)=M_{q,i}$$

(Eq.3)

$$w_g\frac{d}{dx}(\rho_g w_g)+\frac{dp}{dx}+\sum_q\sum_i\left(w_{q,i}\frac{d}{dx}(w_{q,i}\delta_{q,i})\right)=0$$

(Eq.4)

$$w_g\frac{d}{dx}\left[\rho_g\left(J_g+\frac{w_g^2}{2}\right)\right]+\sum_q\sum_i w_{q,i}\frac{d}{dx}\left[\delta_{q,i}\left(J_{q,i}+\frac{w_{q,i}^2}{2}\right)\right]=0$$

(Eq.5)

$$\frac{dw_{q,i}}{dx}=\frac{3}{8}\frac{c_{xq,i}\rho_g(w_g-w_{q,i})|w_g-w_{q,i}|}{\rho_q r_{q,i}w_{q,i}}$$

(Eq.6)

$$\frac{dr_{q,i}}{dx}=-K_q(\Omega)\big/\omega_q r_{q,i}^{\omega_q-1}w_{q,i}$$

(Eq.7)

$$\left(\frac{dJ_q}{dT}\right)\frac{dT_{q,i}}{dx}=\frac{3h_{q,i}(T_g-T_{q,i})}{\rho_q r_{q,i}w_{q,i}}$$

(Eq.8)

$$\Omega=\Omega(p,Y,Z),\text{ where }\Omega=T_b,T_v,T_g,\mu_g.\lambda_g,\eta_g,c_{pg}$$

(Eq.9)

$$Y=1-\sum_i\overline{g}(r_{b(i)0})(r_{b(i)}^3)\big/(r_{b(i)0}^3)$$

(Eq.10)

$$Z=1-\sum_i\overline{g}(r_{v(i)0})(r_{v(i)}^3)\big/(r_{v(i)0}^3),$$

(Eq.11)

where $c_{xq,i}=f(\mathrm{Re}_{q,i})$ - the drag coefficient; F - the area of flow section; $\overline{g}(r)$ - the fraction of radius r particle in normalized mass distribution function; J – the total enthalpy; K – the coefficient of burnout or vaporization law; M – the mass source function; $Nu_{q,i}=f(\mathrm{Re}_{q,i},\mathrm{Pr})$ - the Nusselt number; p – the pressure; $\mathrm{Pr}=\mu_g c_{pg}/\lambda_g$ - the Prandtl number; r – the particle radius; $\mathrm{Re}_{q,i}=2r_{q,i}|w_g-w_{q,i}|\rho_g/\eta_g$ - the Reynolds number; T – the temperature; w – the rate; x – the coordinate along flow; δ - the disperse phase mass content per volume unity; λ - the heat conductivity; μ - the molecular mass; η - the viscosity; ρ - the density; Ω - the sum total of

heterogeneous system properties; ω - the specific coefficient of burnout or vaporization law; *subscripts*: g – gas phase; q – disperse phase; i – size fraction of disperse phase; b – burnout particles; v – evaporating droplets.

For simplicity, the momentum and energy conservation Equations 4 and 5 are here written taking into consideration small absolute and relative rates of gas and disperse flow components.

The sum total of heterogeneous system properties Ω at each integration step is calculated by the special "quasi-equilibrium" thermodynamic approach.

"QUASI-EQUILIBRIUM" THERMODYNAMIC APPROACH

The distinguishing feature of a nonequilibrium heterogeneous system lies in different values of its individual component temperatures. The evaporating drop temperature depends on environmental conditions and is below environmental gas temperature. The burning drop temperature differs also from the ambient gas phase temperature. It can be stated generally, different temperature values occur in the multiphase nonequilibrium heterogeneous system not only between solid, liquid and gas phases but between particles of different sizes too.

The another feature of a heterogeneous reacting system relates to the high energetic of burning and evaporation processes. Therefore, the thermodynamic properties of combustion products of mixtures considered depend strongly on degrees of burnout and of evaporation of disperse individual components.

And finally, the feature of the nonequilibrium system being considered that is for us most important is that the burning and evaporation rates of disperse phases are significantly less than the rates of gas phase reactions. As a consequence, the gas phase composition may be thought of as equilibrium in a local section (point) of flow and the complete combustion products mixture may be considered as quasi-equilibrium.

From the above-mentioned characteristic properties of a heterogeneous reacting system, the "quasi-equilibrium" thermodynamic calculation method has been postulated and developed (6,7), but is briefly reviewed here.

Let us consider a heterogeneous system being of current interest concerning the problem of thermochemical treatment of spent ion-exchange resins (8,9). As a simplification example, it is considered a heterogeneous system, which consists of a metal fuel simulator and an oxygen-bearing salt simulator, that is in the system there is one fuel element M and one oxidant element L only.

Let us determine the burning degree Y and the evaporation degree Z of element M in the system being investigated by relations:

$$Y = 1 - (M_*)/(M_*^*)$$

(Eq.12)

$$Z = 1 - (M_{**})/[(M) - (M_*^*)]$$

(Eq.13)

where (M) is the total mole amount of element M; (M_*) is the mole amount of unburned element M; (M_{**}) is the mole amount of unevaporated element M; (M_*^*) is the mole amount of element M which may be oxidized by the (L) mole of element L.

It is clear that $\left(M_*^*\right) = (L)$ by $(M) > (L)$. Then for the given burnout degree Y the amount of unburned element M in the initial mixture may be determined by the Eq. 12:

$$(M_*) = (1-Y)\cdot(L) ,$$

(Eq.14)

where $0 \leq Y \leq Y_{max}$ and Y_{max} may reach 1.0.

The amount of unevaporated element M in the initial mixture at the given value of Z is according to the Eq. 13:

$$(M_{**}) = (1-Z)\cdot[(M)-(L)] ,$$

(Eq.15)

where $0 \leq Z \leq Z_{max}$ and Z_{max} depends on the element M saturated vapor pressure in the system being investigated.

In accord with the "quasi-equilibrium" principle, the combustion products resulting in the given system may be considered as the "three-temperature" system: burning droplets of metal M_* are of temperature T_*, evaporating droplets of metal M_{**} are of temperature T_{**}, all remaining products (gases, fine-disperse oxides) have the same temperature $T_{eq} = T_g$.

In this special case of the quasi-equilibrium system, the equation for the calculation of the combustion products total enthalpy $J_{c.p.}$ is

$$J_{c.p.} = \sum_i g_i J_i(T_{eq}) + g_* J_{Mg_*}(T_*) + g_{**} J_{Mg_{**}}(T_{**}) ,$$

(Eq.16)

where g_i are mass fractions of combustion products i (gas and condensed) having the same temperature T_{eq}; g_* and g_{**} are M_* and M_{**} mass fractions in combustion products; J is the total enthalpy.

The individual temperatures in the Eq.16 are defined by the successive approximation method. As an initial approximation, equilibrium thermodynamic calculation data obtained with anyone of known algorithms, for example, (10), are used. Then, we calculate the temperature T_* of burning M_* particles and the temperature T_{**} of evaporating M_{**} particles at the equilibrium temperature T of the gas phase of combustion products by the use of known physical models (11-13). Later, we calculate the total enthalpy of reaction products of the whole heterogeneous system according to Eq.16.

.The total enthalpy thus obtained is compared with the total enthalpy J_m of initial mixture. Furthermore, if it is required, we enter the correction ΔT to temperature T and recycle the calculation until equality $J_m = J_{c.p.}$ is reached within the given accuracy. As a result, we obtain values of temperatures T_{eq}, T_*, T_{**}, and the sum total of properties of the "quasi-equilibrium" mixture of the combustion products.

The developed method of "quasi-equilibrium" thermodynamic calculation was used as a part of the mathematical model of a combustion products flow of metal-containing mixtures

SOME CALCULATION RESULTS

The outlined mathematical model and "quasi-equilibrium" calculation algorithm have been realized into computer codes

To illustrate the devised code capability we present Figs. 2 - 4.

Fig. 2 shows a relationship between temperature T_{eq} and the degrees of metal burnout Y and evaporation Z that is obtained as one of calculated results for a mixture of magnesium-sodium nitrate with the mass percent composition 70/30.

 Fig. 3 shows, for example, the gas temperature variation along the chamber in the flow of combustion products of magnesium - sodium nitrate - organic mixture as a simulation result by using the mathematical model, Eqs. 1 - 11, coupled with "quasi-equilibrium" thermodynamic calculation method. In the presented simulation example, the initial diameter of magnesium particles was varied from 56 to 168 μm. It is clearly seen that the greater is the metal particle initial diameter, the longer is the particle burn-out path which correlates with the temperature maximum.

Really, the metal particles have a size distribution. For this case of the metal particles size distribution with a medium-diameter about 110 μm, the gas temperature variation along the chamber is shown in Fig. 4 as a solid curve.
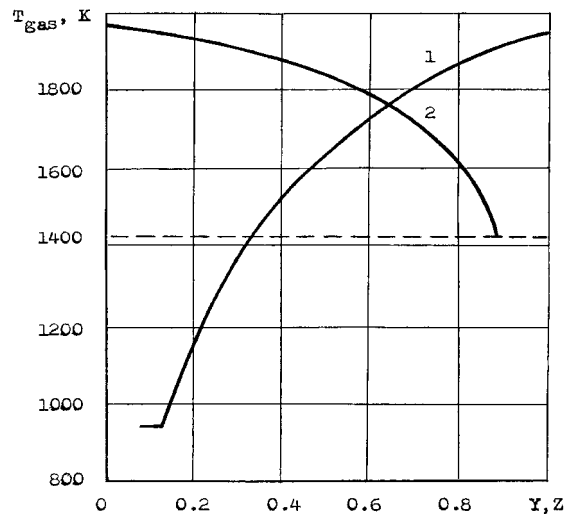

Fig. 2. The calculated gas temperature of combustion products as a function of
Y for Z=0 (curve 1) and of Z for Y=1.0 (curve 2).
- - - - -   *thermodynamic equilibrium*

The results of gas phase temperature measurements are presented at the same figure as solid circles. Considering experimental difficulties due to burning metal particles, the calculated and experimental data agree quite well.
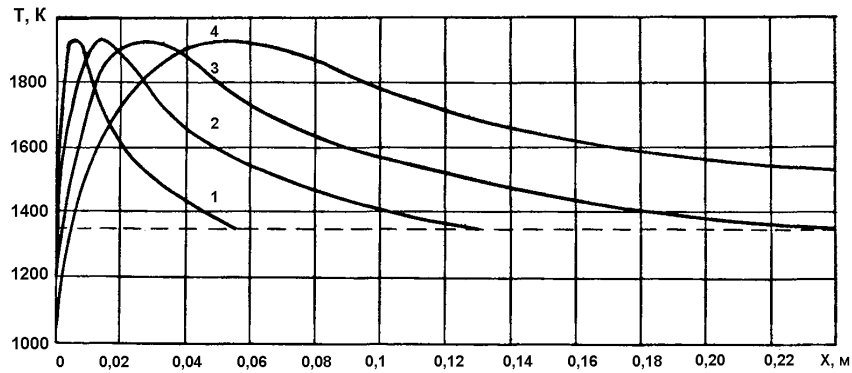
Fig. 3. The gas phase temperature variation along the chamber vs initial metal particles diameter: curve 1 – 56, 2 – 88, 3 – 120, 4 – 168 μm.
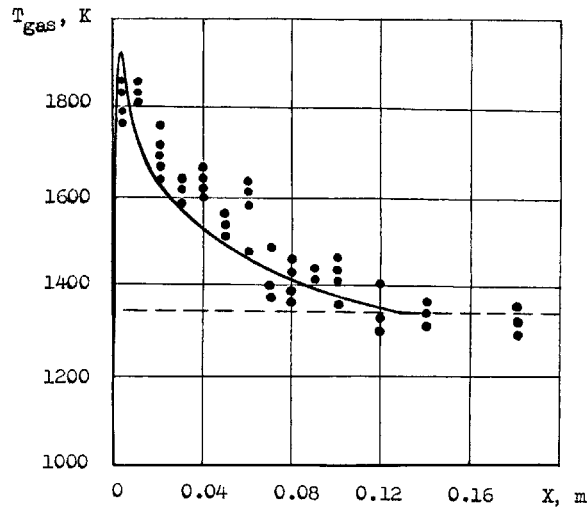
Fig. 4. The gas phase temperature variation in the flow along a chamber.
—————— *quasi-equilibrium calculation*
- - - - - *thermodynamic equilibrium*
• *experimental points*

CONCLUSIONS

In conclusion, it may be noted that the proposed mathematical model provides a real possibility for performing of numerical experiments on the basis of mathematical simulation of nonequilibrium flows of combustion products. Numerical experiments help correctly to estimate the work characteristics in the process of treatment devices design saving time and costs.

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