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Received on 7.12.2006

UDC 536.46

ON ASCERTAINING NATURE OF THE CONCENTRATION BURNING LIMIT

K.O. Sabdenov, O.Yu. Dolmatov

Tomsk Polytechnic University E-mail: sabdenovko@mail.ru

The elemental theory of concentration limits of slow burning and detonation existence has been offered on the basis of development of Lewis' and Elbe's ideas, theoretical works of A.M. Klimov on convective flame extinguishing and works of Ya.B. Zeldovich on flame disappearance at decrease in burning temperature below the critical value.

Introduction

In atomic power engineering, as in any industry, assurance of fire- and explosion safety at enterprises is not the least of the factors. In this case, the incidents of unauthorized formation of gas mixtures on the basis of air as oxidizer are the most dangerous. Acetylene, hydrogen, methane, propane, oil vapors etc. are more often used as fuel [1]. One of the principle characteristics of combustible mixture, according to which its hazard is estimated, is relative percentage of combustible and oxidizer. It is also known that at excess of their concentration over a certain value (too depleted or concentrated mixture), burning is not possible at the given external conditions, i.e. concentration limit of flame spread occurs. To be more precise, concentration limit of flame existence appears as abrupt burning termination at combustible structure deviation from stoichiometric value [1-3]. Inspite of the important applied value, the satisfactory theory of this phenomenon has not been developed yet.

Yu.U. Linnet and Yu.S. Simpson (1956) suggested that experimentally determined limits are not mixture fundamental property [2]. Concentrating or depleting excessively the mixture, the convective streams which depend on experiment conditions start influencing greatly the combustion. Facts of dependence of concentration limits on combustion front moving direction relative to gravity vector indicate as well the important part of convective streams. Later B. Lewis and G. Elbe reached the conclusion about flame suppression near concentration limits by convective streams [2]. But they did not developed the detailed theory allowing calculating the dependence of combustion rate on mixture structure by deviating from stoichiometric composition and explaining a number of laws of concentration limits. The aim of this work is to suggest a simple explanation of concentration limit existence on the basis of development of ideas of Linnet, Simpson, Lewis and Elbe.

The main positions of the theory and its content

Two facts

- upper and lower concentration limits of deflagration and detonation propagation are placed just symmetrically relative to stoichiometric composition with respect to rearrangement «fuel ↔ oxidizer»;
- 2) at combustion limit with considerable decrease of flame velocity its temperature differs insignificantly from the value corresponding to stoichiometric composition. serve as the experimental base of the theory given below [2].

Let us disclose more detailed the content of the given facts. The effect of termination of combustion wave propagation is observed in very rich or in very weak mixtures which are supposed to consist only of fuel A and oxidizer B and having close molecular masses. But weak mixture may be considered as a rich one relative to oxidizer. In both cases the excess part of oxidizer or fu-

el may be considered as chemically «inert» addition, but, strictly speaking, it is not the case. Thus, presence of symmetry is observed. Its essence consists in indistinguishability of rich and weak mixtures at assigning of concentration limit. It should be noted here that symmetry given in general terms has an approximation character. The mechanism of chemical reactions behavior is not examined in detail.

The second fact implies burning termination at rather high temperature which exceeds considerably a new mixture temperature (in 5...7 times and more). Flame temperature at combustion limit decreases in 1,5...2 times at most relative to stoichiometric composition.

The theory suggested below should be considered as a rough conception on factors determining existence of concentration burning limits; but, at the same time, taking into account only the principle physical aspects.

Symmetric consequence given as the first experimental fact should be induced by such physical magnitude by nature being one of parameters determining combustion wave distribution. Only thermal effect of chemical reaction may be selected from a full list which is not given here. Only it can possess the kind of symmetry given above and only it can respond for existence of concentration limit.

Near the concentration burning limit the flame front (in rather wide tube where its walls do not influence the process) decays into separate sites in form of spherical calottes between which gas remains cold: chemical reaction does not occur there [2]. One of such sites with direction of flow of cold fuel round it is given in Fig. 1. One can suppose as well that combustion starts with such single site at supply of initial initiation energy to the mixture. Then the task consists in determining such combustible gas mixture composition at which burning is still possible.

Burning zone cooling with unburnt gas slip flow may result in combustion impossibility. Small quantity of B. Karlovits K which takes various forms depending on concrete situation is the condition of combustion existence. If it is matter of spherically symmetric flame with diameter d, then [2]

$$\mathbf{K} = \frac{\kappa}{u_n d} \frac{\rho_0}{\rho_b},$$

where u_n is the normal velocity of the flame; κ is the coefficient of thermal diffusivity; ρ_0 , ρ_b is the densities of a new mixture and combustion products.

Parameter K in such form characterizes heat transfer from the reaction zone with combustion temperature T_b into a new mixture having temperature T_0 . It may be expressed by the ratio of enthalpy of heated layer H''and flame zone H'[2]:

$$\mathbf{K} = g_u \frac{\kappa}{u_u^2}$$

where λ_T is the heat conductivity coefficient; c_p is the heat capacity at constant pressure.

Burn termination may also occur in the case of flame propagation in gas with typical flow rate gradient g_{u} . Then

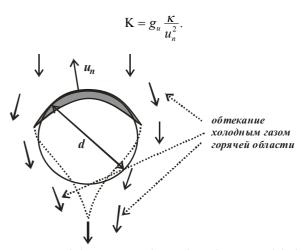


Fig. 1. Single burning site with typical size d. Region with high temperature is indicated with dotted line

Обтекание холодным газом горячей области – Cold gas flow round hot region

In such form number K reflects the degree of increasing burning surface or its «extension» which flame experiences in hydrodynamic distorted gas flow. Surface increase results in bend or turn of flame front. In this case flame becomes divergent. Heat flow directed from the reaction zone to unburnt mixture becomes divergent as well.

Equality of the given two forms of the criterion K is seen from presentation of typical gradient value for hydrodynamic flow shown in Fig. 1 in the form $g_u = u_n \rho_0 / (\rho_b d)$. But it should be noted that if the matter is flame propagation in the cases when gravity influence is considerable then characteristic of convective stream induced both by combustion front moving and gravity presence should be understood as parameter.

Dimensionless criterion K was for the first time introduced by B. Karlovits in 1953 as a measure of burning front extension by hydrodynamic flow. He also showed the key role of this parameter for estimation of constancy and existence of combustion.

Such important role of this parameter appears in the fact that new mixture particle volume, which pass through a unit of flame front area at its shift to this distance and which should be prepared by heating to chemical transformation, is higher in such flame than in the case of burning wave not subjected to extension. This feature results in increase of heat transfer velocity from the reaction zone to unburnt gas. Temperature in the reaction zone decreases. At excess of critical value by flame extension the equilibrium between heat release and heat loss in reaction zone may be completely disturbed and flame diminishes. Flame diminishes without any external heat absorbers (for example tube cold walls). A new mixture acts as quenching agent [2].

This physical picture stated for deriving ignition condition (spherical site) remains valid also for a single burning site shown in Fig. 1. In other words, a new mixture warming up with heat flow from chemical reaction zone is required for flame independent existence. At the same time this flow should not be too large, otherwise chemical reaction slows down so that it becomes impossible owing to the burning zone overcooling. To prevent this it is necessary to have rather high thermal effect of chemical reaction which depends, in its turn, on stoichiometric composition of combustible mixture, its dilution rate with inert additives as well as on conditions of carrying out the experiment (initial temperature and pressure).

According to the above said the burn termination in a single site may be spontaneous. If it is so then it is appropriate to say not about extinguishing but about *selfquenching* that reflects more precisely the nature of internal mechanisms resulting in existence of concentration limits of combustible mixtures. Then these limits get the quality of mixture fundamental property even if extinction occurs as a result of spontaneously appearing convective flows owing to diffuse-thermal or hydrodynamic flame instability.

Let us pay attention again to the fact that gravity influence is not considered here although in some cases its presence may influence greatly the limits of burning propagation [2]. These limits occur in conditions when the flame propagates horizontally and gravity may only shift numerical values of ultimate concentration.

A single burning site may exist if Karlovits's number is less than a critical value K_{cr} . K<K_{cr}. Replacing here inequality sign to equality we find minimum flame velocity u_n^{\min} :

$$u_n^{\min} = \frac{\kappa \rho_0}{d \mathbf{K}_{\rm cr} \rho_b}$$

Presence of K_{cr} follows from the experimental fact of existence of minimum ignition energy [2]. Strict theoretical calculation of K_{cr} by flame extinguishing in distorted hydrodynamic flow was carried out by A.M. Klimov [4].

Later on u_n^{\min} is supposed to be known. The value of this velocity for each mixture may be found from the solution of the problem on burning site propagation shown in Fig. 1.

It follows form the given reasoning that physical mechanisms determining combustible mixture ignition are the same as for flame propagation condition. In both processes thermal effect of chemical reaction is in the center.

Let us find the dependence of reaction thermal effect Q on mixture composition. Let the reaction between the fuel A and oxidizer B occur in burning wave:

$$v_a A + v_b B = C$$

where v_a , v_b are the stoichiometric coefficients; C are the reaction product. A new mixture consists of N_A molecules of sort A and N_B molecules of sort B. Appropriately, their molar portions are equal

$$\mu_a = \frac{N_A}{N_A + N_B}, \quad \mu_b = \frac{N_B}{N_A + N_B}$$

It is obvious that $\mu_b = 1 - \mu_a$. If substance A is in excess then only

$$N_A' = \frac{v_a}{v_b} N_b$$

from all N_A molecules takes part in reaction. Another part in quantity

$$N' = N_A - N'_A = N_A - \frac{v_a}{v_b} N_B$$

remains chemically «inert». Molar portion μ ' of this excess part is given by obvious equality

$$\mu' = \mu_a - \frac{v_a}{v_b} \mu_b$$

If Q_{st} is the thermal effect of the reaction at stoichiometric mixture composition then the reduction value of general thermal effect should be, in any event, proportional to molar portion μ' of «inert» part of mixture. Then we can write down

$$Q = Q_{st} - \mu' C_1 Q_{st} = Q_{st} \left(1 - C_1 \frac{v_b \mu_a - v_a \mu_b}{v_b} \right), \quad (1)$$

for thermal effect Q, where the proportionality coefficient $C_1=C_1(\mu_a,\mu_b)$. Similar reasoning for the case of substance B excess results in expression

$$Q = Q_{st} \left(1 - C_2 \frac{v_a \mu_b - v_b \mu_a}{v_a} \right), \tag{2}$$

where the second proportionality coefficient $C_2=C_2(\mu_a,\mu_b)$ is introduced and assumption of equality of fuel and oxidizer molecular masses accepted above is used.

If the coefficients C_1 and C_2 are not taken into account then formulas (1), (2) are applicable respectively only to positive differences $v_b\mu_a - v_a\mu_b$ and $v_a\mu_b - v_b\mu_a$, the second of which represents the opposite value from the first one.

It is clear that for the thermal coefficient Q there should be the unique expression, valid regardless of the fact what substance – A or B is in excess. But change of sign at transfer from formula (1) to formula (2) does not result in changing the sign before the proportionality coefficient. It means only one thing: the coefficients C_1 and C_2 themselves depend on the given differences and $C_1 \sim v_b \mu_a - v_a \mu_b$, $C_2 \sim v_a \mu_b - v_b \mu_a$, that remains numerators symmetrical in fractions from formulas (1), (2) relative to place rearrangement of $v_b \mu_a$ and $v_a \mu_b$. And right parts from proportionality sign may have any other uneven degree. Here the smallest of them equal unit is taken as the one resulting in the simplest sought-for formulas. Coefficients C_1 and C_2 may be certainly given in form of uneven degree series from $v_b \mu_a - v_a \mu_b$.

Thus, we come to conclusion about quadratic dependence of thermal effect Q on difference $v_b\mu_a - v_a\mu_b$ as the simplest possible form.

The next stage of refining the type of coefficients Следующий этап уточнения вида коэффициентов C_1, C_2 consists in requirement of converting the reaction thermal effect to zero in two ranges: at unlimited concentration of mixture with fuel $\mu_a \rightarrow 1$ and oxidizer $\mu_b \rightarrow 1$. This requirement fulfillment results in the following expression

$$Q = Q_{st} \left[1 - \left(\frac{v_a \mu_b - v_b \mu_a}{v_a \mu_b + v_b \mu_a} \right)^2 \right], \qquad (3)$$

If deflection factor from stoichiometric composition $\sigma = v_b \mu_a / v_a \mu_b$ is introduced here then formula (3) may be written down in more compact form:

$$Q = Q_{st} [1 - (\frac{\sigma - 1}{\sigma + 1})^2],$$
 (4)

It was shown above that the nature of existence of concentration limit of burning wave propagation (deflagration or detonation) is thermal. In order to determine the limiting ratio for concentration the same process of reasoning as in the theory of flame existence thermal limit of Ya.B. Zeldovich may be applied [5]; here the reason of extinction is heat extraction from burning zone to the wall of tube with small diameter. It is acceptable as in the theory of Ya.B. Zeldovich critical temperature drop and burning rates do not depend on conditions of heat loss from burning zone. Therefore, they are valid also in the case of convective extinguishing.

The thermal effect value affects directly on flame temperature T_b at adiabatic conditions. They connect to each other by a simple equality

$$T_{b} = T_{0} + \frac{Q}{c_{p}} = T_{0} + \frac{Q_{st}}{c_{p}} [1 - (\frac{\sigma - 1}{\sigma + 1})^{2}].$$
 (5)

Flame temperature T_b^* , below which the wave chemical transformation becomes impossible without external heat supply to burning zone, meets the critical value σ_* corresponding to occurrence of concentration limit. Total flame temperature T_b^m is implemented at stoichiometric composition:

$$T_b^m = T_0 + \frac{Q_{st}}{c_p}.$$
 (6)

Writing down this expression the weak dependence of heat capacity c_p of mixture composition is neglected. Burning rate at temperature T_b^m is maximal and equal

$$u_n^{\max} \sim \exp\left(-\frac{E}{2RT_b^m}\right).$$

Appropriately, burning rate at lower temperature limit has minimal value:

$$u_n^{\min} \sim \exp\left(-\frac{E}{2RT_b^*}\right).$$

Maximal drop of burning rate equals

$$\frac{u_n^{\max}}{u_n^{\min}} = \exp\left[\frac{E}{2R}\left(\frac{1}{T_b^*} - \frac{1}{T_b^m}\right)\right]$$

This ratio usually amounts to 2...6. At the same time, activation energy value is rather high to suppose small difference between T_b^m and T_b^* that coincides well with the second experimental fact given below. Let us present that

$$T_b^* = T_b^m - \phi \, \frac{R(T_b^m)^2}{E},$$

where ϕ is the numerical literal which may be found from the analysis of stability and existence of single site burning. Using here the expression (5) at value $\sigma = \sigma_*$ and (6), after simple transformations we obtain:

$$\frac{\sigma_{\pm} - 1}{\sigma_{\pm} + 1} = \pm \sqrt{\phi \frac{R(T_b^m)^2}{E(T_b^m - T_0)^2}}$$

Two roots $\sigma_{+}>1$, $\sigma_{*}<1$ for deflection coefficient from stoichiometric composition correspond here to excess and deficiency of combustible component A.

In more usual notations $\beta = RT_b^m/E$, $\mu_0^b = T_0/T_b^m$ we have:

$$\sigma_{+} = \frac{1 + \sqrt{\phi \beta / (1 - \mu_{0}^{b})}}{1 - \sqrt{\phi \beta / (1 - \mu_{0}^{b})}}, \ \sigma_{-} = \frac{1 - \sqrt{\phi \beta / (1 - \mu_{0}^{b})}}{1 + \sqrt{\phi \beta / (1 - \mu_{0}^{b})}} = \frac{1}{\sigma_{+}}.$$
(7)

Ratio of maximum burning rate to its value at burning extension equals

$$\frac{u_n^{\max}}{u_n^{\min}} \approx \exp(0, 5\phi). \tag{8}$$

For maximum flame velocity drop in six times we find $\phi \approx 3,6$. Value β for the majority of gas mixtures amounts to about 0,1...0,01. Appropriately, the temperature at flame extension decreases in $T_b^m/T_b^* \approx 1,56...1,04$ times from maximum possible value for stoichiometric value.

Maximal change of deflection coefficient from stoichiometry is found respectively in the ranges ($\mu_0^{b}=0,14$) $\sigma_{+}=4,7...1,5$ at fuel excess and $\sigma_{-}=0,21...0,67$ – at its deficiency.

Formula (4) does not cover all possible reasons of decreasing thermal effect at deflection from stoichiometric composition. It should be considered as one of the simplest expressions for the reaction thermal effect.

Comparison of theoretical conclusions with the experimental data

Let us present the ratio of the effective value of flame velocity u_n to the peak value (at stoichiometric composition)

$$\frac{u_n}{u_n^{\max}} = \exp\left[-\frac{E}{2RT_b^m} \frac{T_b - T_0}{T_b} (1 - \varpi^2)\right], \qquad (9)$$
$$\varpi = \frac{\sigma - 1}{\sigma + 1}.$$

Comparison of this formula with the experimental data in flame velocity dependence on part from deflection coefficient σ shows good qualitative and quantitative coincidence for combustible gases diluted with air (Fig. 2, 3).

Worth agreement of theoretical conclusions with the experimental data is observed if pure oxygen acts as oxidizer. Relative error for the mixture of methane with oxygen at σ =2,0 amounts to 40 %, but not more.

Burning temperature calculations are not given here. But it was ascertained that accuracy in calculation of burning temperature T_b influences considerably the success of theoretical predictions of flame velocity. Owing to the roughness in determining T_b the taken values of dimensionless activation energy for air mixtures turned out to be overrated. Relative error in calculations of burning temperature achieves 20 %; it is rather high as the temperature is in exhibitor with a large factor. Therefore, the theory should be further improved by correcting thermal effect dependence on mixture composition.

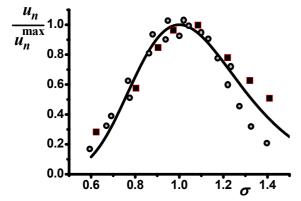


Fig. 2. Calculated and experimental [2] data of Kh.F. Kauard, F.Yu. Hartwell (●), G. Jan, A.R. Denewes, U.Yu. Haff (■) for the ratio of flame velocity to its maximum value of methane-air mixture as function from stoichiometry coefficient. Value T_b^m=2250 K

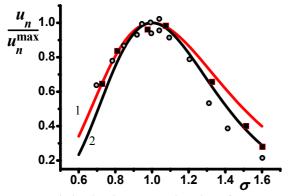


Fig. 3. Calculated and experimental [2] data of Yu.M. Zinger, Yu. Grumer, E.B. Kuk (●), B. Lewis, G. Elbe (■) for the ratio of flame velocity to its maximum value of propaneair mixture as a function from stoichiometry coefficient. Value T_b^m=2240 K

In this context, another important circumstance consists in a slight shift of burning velocity maximum and maximum burning temperature relative to stoichiometric composition observing in experiments. In formulas (5) and (9) such shift is not contained.

As for the concentration limits of detonation existence then here the case is somewhat easier: detonation velocity U_D is directly and mainly determined by thermal effect of chemical reaction [6]: $U_D \sim Q^{1/2}$. This ratio is valid up to disappearance of detonation combustion.

Physical mechanism defining detonation concentration limit is, obviously, the same as fro slow combustion. It may be argued by the fact that detonation starts at burning site. And if heat release is not high enough in it then expanding combustion products take heat away fast from chemical reaction zone resulting in its overcooling.

Thus, concentration limit of burning propagation whether it is laminar flame or detonation is explained from the uniform point of view. It is explained by the fact that concentration limits for both combustion modes either coincide or differ insignificantly [2, 7].

The above said results in conception of critical thermal effect of chemical reaction below which the independent combustion (laminar flame or detonation) is impossible at fixed environment.

The equality $\sigma_+\sigma_-=1$ follows from the second ratio (7) $\sigma_+\sigma_-=1$, that means the presence of strict symmetry of values σ_+ and σ_- relative to the unit on axis σ . For the borders of concentration limits in percentage of combustible component Y_{A^+} , Y_{A^-} the last equality may be formed as follows

$$\frac{Y_{\rm A}^+ Y_{\rm A}^-}{\left(Y_{\rm A}^{\rm st}\right)^2} = 1.$$
 (10)

In order to check up the accuracy of this value ratio all data at lower and upper concentration limits of slow burning are taken from the book [2], and for detonation – from monographs [7]. As calculations show the equality (10) is not well fulfilled for combustible mixtures with pure oxygen. So, ethane mixture with oxygen has the following characteristics: $Y_{\rm A}^{-}=4,1$ %, $Y_{\rm A}^{+}=50,5$ %, $Y_{\rm A}^{\rm sl}=22,0$ %. We obtain $Y_{\rm A}^{+}Y_{\rm A}^{-}/(Y_{\rm A}^{\rm sl})^2=0,43$ for it.

The ratio of symmetry (10) is better implemented for mixtures with air (Table) as well as for detonation concentration limits. For example, for propane-air mixture $Y_A^+Y_A^-/(Y_A^{st})^2=1,2$, and for mixture of ethylene with air $-Y_A^+Y_A^-/(Y_A^{st})^2=1,14$.

 Table.
 Check up of the ratio (10) for the mixture of a number of combustibles with air

| Combustible | Y _A ⁻, % | Y _A ⁺ , % | Y _A st , % | $Y_{\rm A}^{+}Y_{\rm A}^{-}/(Y_{\rm A}^{\rm st})^{2}$ |
|-------------|---------------------|---------------------------------|----------------------------------|---|
| Methane | 5,0 | 15,0 | 10,0 | 0,75 |
| Ethane | 3,0 | 12,5 | 6,0 | 1,04 |
| Propane | 2,2 | 9,5 | 4,02 | 1,23 |
| Propylene | 2,0 | 11,1 | 4,67 | 1,02 |
| Butane | 1,86 | 8,41 | 3,23 | 1,5 |
| Pentane | 1,4 | 7,8 | 2,63 | 1,58 |
| Hexane | 1,18 | 7,4 | 2,33 | 1,6 |
| CO | 12,5 | 74,2 | 42,0 | 0,53 |

Conclusion

Thus, the explanation of gas mixture concentration limits is suggested. Let us briefly state physical picture of the processes occurring in this case. Its matter is in the following: burning temperature decreases at withdrawal from stoichiometric composition ($\sigma \rightarrow 0$ or $\sigma \rightarrow \infty$) that involves rapid drop of flame velocity. As a result, heat quantity in a warmed up layer increases, mainly owing to its growing size equal approximately κ/u_n . It involves heat loss increase from flame zone. Losses are also conditioned by decrease of burning site size d at reaching the concentration limit, as $1/d \sim S/V$, where S is the site surface; V is its volume. Slight decrease of burning temperature results in drop of heat flow from burning site but it is not capable of com-

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pensating completely the increasing losses owing to drop of velocity u_n and size d. This tendency, finally, results in the lowest possible temperature $T_b^*(\sigma_{\pm})$ and burning rate $u_n^{\min}(T_b^*)=u_n^{\min}(\sigma_{\pm})$.

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Received on 7.12.2006

UDC 504.3.054:629

NUMERICAL MODELING OF THE EVAPORATION PROCESS OF UNSYMMETRIC DIMETHYLHYDRAZINE DROPS IN THE EARTH'S ATMOSPHERE

A.E. Dolotov, G.V. Kuznetsov, T.N. Nemova*

Tomsk Polytechnic University *Tomsk state architectural-construction university E-mail: elf@tpu.ru

The evaporation process of unsymmetric dimethylhydrazine drops at their movement to the Earth's surface after seal failure of fuel tanks of launch rockets at heights up to 50 km has been simulated.

Introduction

The problem of air pollution with unsymmetric dimethylhydrazine (NDMG) being in tanks of liquid engines of launch rockets [1, 2] after first steps work out, is still urgent. It is conditioned to a large scale, by the fact that a concrete mechanism of appearing this pollutant on the Earth's surface is not determined so far. On the one side there are no direct evidences of appearing this substance in soil or phytocoenosis structure. On the other side, in the regions of separation of launch rockets first stage (for example, Gorny Altai [3]) there are examples of abnormal development of some biological systems while there are no other negative influencing factors on these systems. The known results of modeling the processes of «evolution» of liquid NDMG cloud [4, 5] after seal failure of fuel tanks at heights do not allow forming the definite answer to the question of NDGM phase state in atmosphere.

The aim of this work is the numerical simulation of evaporation process of NDMG drop at its motion to the Earth's surface subject to inhomogeneous temperature field of a drop, the conditions of heat exchange with the environment changing in time and resistance force.

Statement of a problem

At problem set the assumption that NDMG drop is not deformed at its motion and remains of spherical form during the whole flying till the evaporation process stop was accepted. The convective heat exchange of NDMG with the environment the parameters of which (temperature, pressure and density) were considered to be variable in height according to distributions introduced in [6] was taken into account. Radiation heat exchange with the environment was not taken into account. Calculating the drop rate of motion υ it was accepted that gravity and resistance forces of gaseous medium influence it. The convective heat exchange α NDMG with air was computed subject to the dependence α on density ρ and rate of motion of NDMG drop υ .

The special feature of the problem is a shift of the drop external boundary because of NDMG evaporation with time variable velocity *w*. Magnitude *w* depends on drop surface temperature which changes nonlinearly by its motion to the Earth's surface. The problem in such statement is reduced to solution of the following differential equation system with proper boundary and initial conditions.