The Distribution of Energy in Overdriven Detonation Waves and its Influence on their Stability.

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Introduction.

It is very good known that detonation waves (DW) are instable. This lead to formation of the transversal waves and the cellular structure, they are the constant concomitants of the detonation in the Chapman-Jouguet (CJ) regime (see for example [1]). Qualitatively evident, that with wave speed increase (under overdrive) the temperature behind the shock wave and in the overall reaction zone will increase too. This leads to intensification of the dissociation of detonation product, and together them to decrease of the potential energy of the chemical bond ΔH_x^0, released during chemical reaction. The computation of the overdriven detonation carried out in the works [2,3] has showed that for the wide range of gas mixtures the heat release ΔH_x^0 passes zero and becomes negative at the ratio of overdriven wave velocity to Chapman-Jouguet velocity $D/D_{CJ} \leq 1,95$. With the overdrive the detonation degenerates in the shock wave (SW), which must be stable [4]. The experimental confirmation of the computations [2, 3] was received by the works in which transition phenomena had been investigated. For example, in [5] the transition of detonation to stationary regime from the fort overdrive at initiation by the spark discharge of the strong enough power was studied. With weakening of uniform wave at $D \approx 1,5 \div 1,6 D_{CJ}$ the characteristic cellular structure has arisen on the soot layer imprints.

The systematical experimental investigation of the influence of overdrive on the cellular DW structure has been carried out in [6]. For mixtures $2H_2 + O_2$ and $2H_2 + O_2 + 7Ar$ the transition from smooth DW to the cellular structure was fixed on the soot layer imprints and the sweep Schlieren photographs at the velocity of wave consistent (in computation [2]) with $\Delta H_x^0 = 0$.

For the mixture methane with oxygen also diluted by seven moles of argon ($D_{CJ} = 1640$ m/s) the computed velocity at which $\Delta H_x^0 = 0$ was ≈ 2360 m/s [3]. Nevertheless, in the experiments [6] it could observe the cellular structure at the wave velocity approximately up to 2700 m/s. The instability was conserved in the range of the velocities from 2300 to 2700 m/s in spite of $\Delta H_x^0 < 0$. We shall attempt to discuss these facts on more detail and to give explanations to these and a certain others adjacent phenomena.

The shocks and detonations Hugoniot curves computation data.

In the table the results of computation a number of parameters for the Chapman-Jouguet regimes («$_{CJ}$»), overdriven chemical equilibrium detonation and («$_e$») and the shock waves («$_S$») for a certain mixtures are shown. This data had been received (but not yet published) in [2, 3] with use of [7]. For wave overdriven the indexes «$_3$» and «$_4$» corresponds to the computed points neighboring with the point of $\Delta H_x = 0$: «$_3$» – the heat release is yet positive, «$_4$» the heat release already is negative. The ovrall values of the heat capacity ratios ($\gamma = C_p / C_v$) are listed corresponding to the "frozen" chemical composition, π – pressures relative to the initial ones, D – velocities of wave [m/s], molecular mass μ – [g/mole], temperature T [K] and the heat release ΔH_{xCJ}^0 in the CJ-point [cal/mole].

As we can see from the table, the mixtures may be separated in two types following to molecular mass change. In the mixtures of the type "B" containing in the initial composition molecules of hydrocarbon (CH_4, C_2H_2 and so on) with the great number of vibration freedom degrees and the triatomic molecules (N_2O), the molecular mass of the chemical reaction products in the CJ point is *less* than initial one ($\mu_0 > \mu_{CJ}$). For these mixtures "in average" the molecular structure of products becomes simpler against the initial state, and the number of internal freedom degrees decreases in the course of the chemical

Table.

The molecular masses some mixtures, parameters in Neumann spike, in CJ-point and for overdriven waves (from [2,3]). Initial temperature T_0=298,15 K.

Mixts→ Params.↓	Type "A"				Type "B"					
	$2H_2+O_2$	$2H_2+O_2$ $+7Ar$	$2CO+O_2$	$C_2H_2+24O_2$	CH_4+2O_2	CH_4+2O_2 $+2Ar$	CH_4+2O_2 $+7Ar$	$C_2H_2+5O_2$	$2C_2H_2+5O_2$ $+21Ar$	N_2O+CO
P_0(atm)	0,1	1,0	1,0	0,1	0,1	0,2	1,0	1,0	1,0	1,0
μ_0	12,01	31,56	29,34	31,76	**26,68**	**31,99**	**35,96**	**30,30**	**37,53**	**36,01**
μ_{CJ}	14,08	34,18	34,53	32,30	**20,44**	**27,44**	**33,72**	**23.31**	**35,97**	**31,50**
μ_3	12,37	32,24	30,77	30,59	17,99	25,52	31,55	20,12	33,57	28,57
μ_4	11,23	30,91	28,13	28,99	16,34	24,29	30,15	18,17	32,36	26,73
γ_0	1,397	1,554	1,396	1,385	1,360	1,462	1,554	1,328	1,528	1,325
γ_s	1,320	1,492	1,298	1,283	**1,176**	**1,245**	**1,357**	**1,194**	**1,414**	**1,209**
γ_{feCJ}	1,225	1,419	1,212	1,257	**1,227**	**1,288**	**1,384**	**1,266**	**1,442**	**1,242**
γ_{s3}	1,270	1,462	1,272	1,255	1,155	1,227	1,337	1,188	1,409	1,196
γ_{fe3}	1,254	1,443	1,245	1,258	1,260	1,310	1,416	1,317	1,485	1,274
γ_{s4}	1,251	1,448	1,264	1,241	1,151*	1,221	1,331	1,140*	1,406	1,190
γ_{fe4}	1,283	1,466	1,273	1,272	1,290	1,328	1,440	1,356	1,514	1,297
π_{sCJ}	30,12	28,74	32,66	24,22	50,74	43,10	34,04	64,59	38,96	45,97
π_{CJ}	17,32	16,85	18,48	13,20	27,10	25,26	19,48	33,84	22,14	24.58
π_{s3}	79,72	55,19	86,07	63,89	133,8	82,90	65,51	169,0	74,46	120,6
π_{e3}	78,81	53,66	83,97	60,99	124,2	75,10	61,71	155,0	59,83	111,9
π_{s4}	122,5	75,89	131,8	**97,97**	205,0*	113,9	90,70	257,6*	102,1	184,3
π_{e4}	125,2	77,54	133,1	**97,53**	197,4	108,3	89,05	245,1	100,2	176,9
D_{CJ}	2713	1693	1799	1484	2290	1961	1685	2425	1776	1884
D_3	4368	2331	2896	2390	3686	2699	2319	3904	2446	3033
D_4	5390	2726	3573	2949	4549	3156	2711	4817	2860	3744
T_{sCJ}	1646	2054	2384	1281	1791	1941	2002	2210	2384	1752
T_{eCJ}	3278	3077	3525	2272	3327	3313	3000	4215	3525	3479
T_{s3}	3522	3568	3661	2674	3656	3190	3388	4933	4195	3784
T_{e3}	3823	3906	4302	3165	3931	3789	3583	5331	4470	4505
T_{s4}	4991	4713	5275	3793	5156*	4122	4434	6650*	5590	5459
T_{e4}	4115	4319	4827	3551	4274	4048	3889	6101	5085	5320
ΔH_x^0	16050	8327	20346	11500	26028	17390	10771	29512	11221	23765

*)The data for shock waves given from [8].

reactions. The decrease of the capacities ratio in comparison with the value behind the shock wave ($\gamma_{fCJ} > \gamma_s$) says the same. In the limits of the velocities range under the computation for these mixtures, always, γ_e appears to be more than γ_s.

In the type "A" mixtures on the base of the H_2, CO containing in their initial composition the simple molecules with the single freedom degree (O_2, CO, N_2, H_2), or highly diluted by oxygen mixture of $C_2H_2 + 24O_2$ transition to the CJ-state occurs with an **increase** of the molecular mass ($\mu_0 < \mu_{CJ}$). The reaction leads to the complication of the molecules in comparison with the initial state, and, accordingly, to increase of the number of the internal freedom degrees ($\gamma_s > \gamma_{fCJ}$). For these mixtures as an overdrive is intensified, the molecular mass decreases also (as for type "B"), and between the points «3» and «4» (see fig.1) becomes less than initial one. Somewhere between the same points γ_s becomes less than γ_s. It means that with an intensification of overdrive as a result of shift of the equilibrium composition to dissociation the molecules also become simpler than in the initial state.
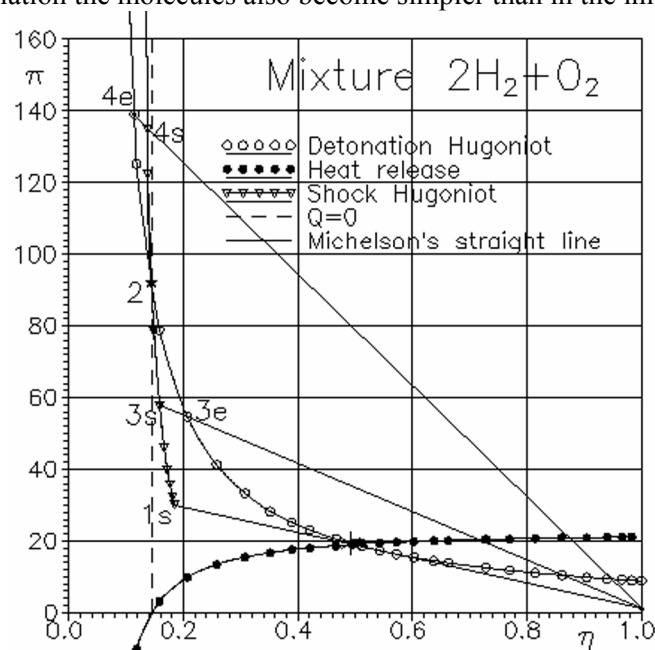

Fig.1 The shock and equilibrium Hugoniot and heat release curve at $D > D_{CJ}$ for the mixtures of the type "A".

In the figures 1÷3 the shock and detonation Hugoniot curves and the dependencies of the energy of the chemical bond per mass unit released in the course of reaction (in term of square of the initial sound velocity $Q = \Delta H_x^0 / c_0^2$) are shown. In the case of the normal detonation, the shock wave transfers a gas from initial state to a point 1s (fig 1, 2) corresponding to the state behind SW moving with the CJ velocity. If $D > D_{CJ}$ the points of type "3s" or "4s" satisfy to the shock pressed state.

When $Q > 0$, in the stationary part of the overdriven wave flow ("pressed by a piston") the pressure decreases just along Mikhelson-Raleigh's straight line from the point 3s to the point 3e up to the intersection with the equilibrium Hugoniot curve defined by the chemical and relaxation processes behind the shock or overdriven detonation.

When overdrive increases, the heat release Q decreases and finally changes its sign. The intersection of a straight line raised from the point $Q=0$ with the Hugoniot curves (the point 2 in fig.1) defines the parameters of wave under which the heat release of chemical reaction becomes equal zero. For the mixture $2H_2 + O_2$ and other mixture of type "A" just close the same point the detonation wave degenerates in shock wave. With the change of the sign of Q the pressure behind the shock wave (the point 4s in fig.1) becomes less than in the equilibrium point (4e), that is the transition to the

equilibrium followed by augmentation of the pressure, this case corresponds to heat absorption by the dissociation processes.
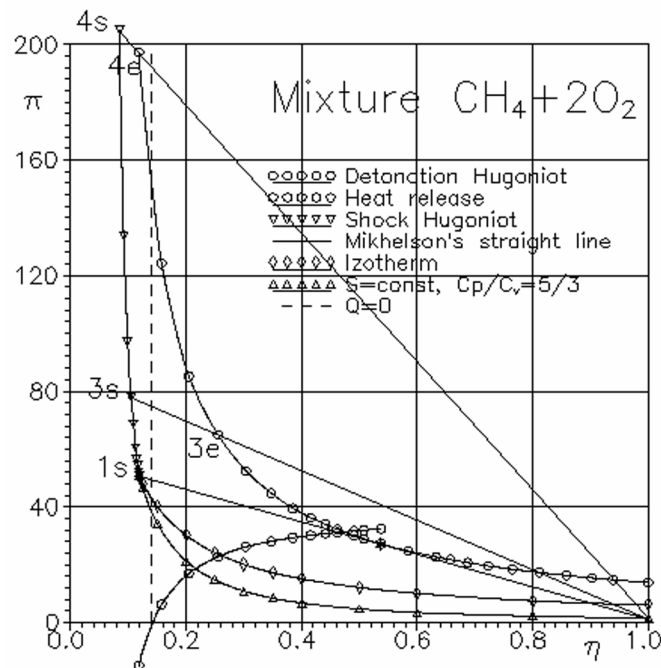

Fig.2 The shock and equilibrium Hugoniot and heat release curve at $D>D_{CJ}$ for the mixtures of the type "B".

Another we can see for the mixtures of type "B" (fig.2). There is no any intersection of the equilibrium and shock Hugoniots at $Q=0$ here. The point *1s* can lay more left than line $Q=0$. There is the range of overdriven wave velocities in which the transition to equilibrium is realized with pressure ***decrease*** in spite of $Q<0$, which corresponds to addition of some heat. The pressure in the shock wave (point 4s) remains yet higher than in the equilibrium point (4e). So anomalous pressure behavior and the observation in the experiments of the detonation front instability within this range of velocities are the evidence that some addition source of energy exists at the cost of which the heat brings to the flow
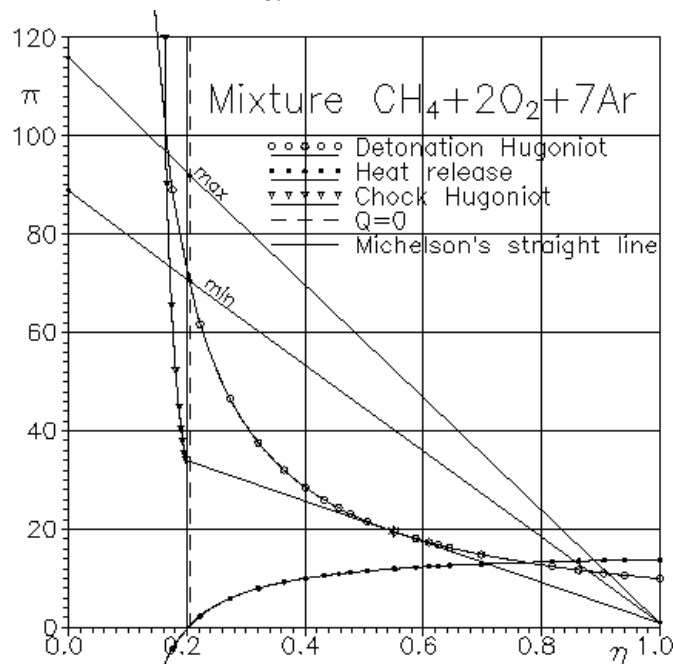

Fig.3 The shock and equilibrium Hugoniots and heat release curve and the area of instable regimes from [6] at $\Delta H_x^0 \leq 0$.

behind the shock wave. And only under the further very fort overdrive, then the dissociation absorbs this overall addition energy, the tendency to intersection of the shock and detonation Hugoniots curves are observed.

In the figure 3 Hugoniots curves are shown for the CH_4+2O_2+7Ar mixture used in the experiments [6]. Here Michelson's straight line (SM) «max» was drawn for the maximum velocity of the overdriven wave, higher which the cellular structure was not observed (D=2700 m/s) in the experiments. It passes near the point of the equilibrium and shock Hugoniots curves intersection. For minimal velocity (D=2360 m/s), lover which the smooth wave was not observed the SM «min» passes near the point of intersection the equilibrium Hugoniot curve with the line of Q=0. One can consider this fact as some accidental coincidence, but the boundaries of the area of the velocity values at which for this mixture it is possible to observe both the smooth waves and the cellular structure correlate in the limits of the measurement errors with area for which behavior of the pressure behind overdriven wave is anomalous.

In both types of mixtures «A» and «B», at Q>0 the gas temperature increases in rear of the shock when passing to equilibrium ($T_{s3}<T_{e3}$), but at Q<0 temperature decreases ($T_{s4}>T_{e4}$), that is the temperature behaves "in natural manner" (see the table) independently of the "anomalous" pressure behavior!

The instability detonation wave in gases. Theoretical findings.
The problem of instability was discussed in the works of K.I.Schelkin, R.M.Zaidel, V.V.Pukhnachev [9-12] in frame of model of the irreversible chemical reaction in the perfect gas in course of which the temp of heat release obeys Arrhenius law. The principal result of these works consists in that the self-sustained detonation wave is instable to infinitesimal disturbances if the chemical reaction activation energy (E_a) is more than some value. The influences on the stability of the chemical energy release and the overdrive was not investigated.

J.J.Erpenbeck [11,12] undertook the proper study for the same model of the medium. The results of computation showed that the boundaries of the stability area on the plane "overdrive – the transversal disturbance wave number" depend on the activation energy as well as the energy release and the overdrive degree. In the calculations a decrease of the activation energy caused the some stabilizing influence. But even at E_a=0 the detonation near the CJ-regime remains instable provided the non-dimensional quantity of heat brought to the gas mass unit at the cost of the chemical energy $q_x = \Delta H_x^0/\mu_0 R T_0 \approx >10$ (μ_0 –molecular mass, T_0 – absolute gas temperature in front of the detonation wave, R – gas constant).
Such a value of q_x corresponds to heat release $\Delta H_x^0 \sim 2$ kcal/mole too low for support of the stationary DW. In reality, under the usual experiment conditions the value of ΔH_x^0 for fuel-air mixtures is some that more 14 and reaches 30 kcal/mole for stoichiometric mixtures of methane or acetylene with oxygen. In such a way although the chemical reaction thermal response is likely important for the phenomenon of the detonation instability, *the reaction exothermicity, consistent with the self-sustained detonation waves, is wittingly enough to produce instability even when the activation energy equals zero*. Hence the most important parameters defined stability of the plane detonation waves are the energy release and the overdrive degree. The contribution of the activation energy appears to be important but not predominant.

The heat capacity behavior of gas taking part in the detonation process.
In the figure 4 the molar capacity dependences of some gases on temperature from [7] are presented. At the near room initial temperature T_0 the vibration and electronic freedom degrees are not excited in the absolute majority of the gas mixtures. Therefore all graphs have tendency to group near $C_p = 7 \div 8$ cal/(mole·K) $\approx 3,5 \div 4$ R. For the diatomic and triatomic molecules this corresponds to the number of the completely excited translational (*tr*) and rotational (*r*) freedom degrees $v^0 = v^0_{tr} + v^0_r = 3 + (2$ or $3)$.

The contribution in the heat capacity of the vibrational freedom degrees becomes noticeable already at the temperature 500 K, and at $T \sim 3000$ K the curves become gently sloping that is to say this freedom
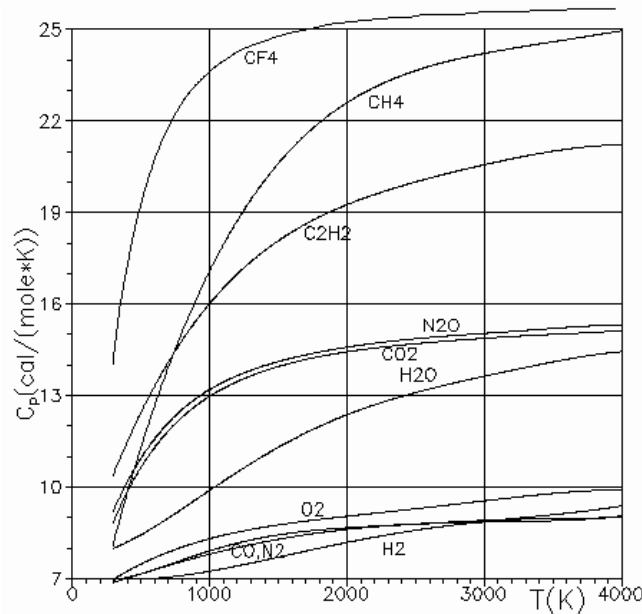
Fig.4.Depedencies of heat capacity of some of
gases from temperature [14]

degrees are "defrosted" almost completely. The heat capacity C_p for the molecules such as methane or acetylene reaches $10 \div 12\ R$. Under this temperature the contribution in the internal energy of the vibrational freedom degrees over quickly relaxed translational and rotational ones may consist no less than $25 \div 40$ kcal/mole. Concretely, at the temperature behind the shock wave of T_s=2000 K the excitation energy E_i of the molecular internal freedom degrees may consist more of 17 kcal/mole.

Behind the shock wave moving with the detonation velocity ($T \sim 2000$ K) the stoichiometric mixtures of these gases with oxygen have C_p of 7,2 R and 6 R, respectively. For the most complex molecules of the reaction products (CO_2, H_2O) C_p does not exceed 7,5 R. As the total mole fraction of triatomic molecules for these mixtures in CJ-point consists about 50%, the product of reaction heat capacity at shock temperature wittingly does not exceed 5 R. The difference in the heat capacities in comparison with the state behind shock wave accounts for 2,5 R through to R. The rough estimation shows that behind the shock wave the mixtures of type "B" have a surplus reserve connected with the vibrational freedom degrees of the thermodynamic part of the internal energy per mole of the gas. This reserve is of 3000 to 7500 kcal/mole or from 5 to 12,5 non-dimensional units (see above and [11, 12]), that also is significant part of the heat release in the CJ-point. Therefore, when the molecular structure in course of the chemical reaction is simplified ($\mu_0 > \mu_e$), the part of the energy accumulated in the internal freedom degrees during the shock transition is released in the form of heat (transfers on to the translational freedom degrees) and provides the positive thermal effect, even if the contribution of the potential energy of the chemical bond becomes moderate negative. At analysis of the detonation and shock wave structure and estimations of the various physical phenomena influence on beginning of the chemical reactions and relaxation processes in comparison with the classic model of Zel'dovitch-Doering-Neumann (ZDN) these facts require more careful treatment.

The first indications on necessity to discus the vibrational freedom degrees excitation and its influence on the energy balance and the initial phases of the chemical reactions in an application to the detonation processes one can find in the works of R.I.Soloukhin and V.V.Voevodsky [15,16]. At present the most complete attempt to classify the physico-chemical processes in the detonation waves according to times and the energy distribution betweenr the freedom degrees seems to be done in the works of S.Tarver [17]. In these works the one-dimension model ZDN and the cellular structure of the detonation wave VMT (of Voitsekhovsky-Mitrofanov-Topchiyan) were extended by including of the vibrational relaxation preceding and following to the main exothermic reactions. Only the CJ-regimes were analyzed for two "exotic" mixtures relevant in our classification to type"B" with comparatively

considerable molecular mass ratios: (for H_2+Cl_2 – $\mu_0=36,465$, $\mu_{CJ}=33,997$ and for O_3 – $\mu_0=48,0$, $\mu_{CJ}=30,781$) and stoichiometric mixture of hydrogen with oxygen, relevant to type "A". For this the latter mixture in [17] the model was developed with the preliminary transition to the equilibrium of the exchange reactions (as, for example, in [18]) and the following the main energy release zone. The phenomena behind the overdriven wave and the degeneration of the detonation in the shock wave remain outside of the cadre. Meanwhile the instability of the shock and the overdriven detonation at $Q<0$ have the same nature.

The instability flow under consideration in any case cannot appear "on the blank space" without the energy feeding to the flow (by any means) or the redistribution energy between internal and translational freedom degrees of the molecules. The instability evolution takes place against the overall decay of pressure and density behind shock wave. This decay can occur both with the drop of the temperature in the adjoining rarefaction wave and with the temperature increase caused by the supply of the energy to the flow at the cost any source, in particular in the detonation wave this takes place in the course of the exothermic chemical reactions.

The experimental detection of shock wave instability under some special conditions [19-21] posed a question about a source of the energy supporting instability against the total endothermic processes behind the shock wave. Although in the following papers of the authors and other [22, 23 a. o.] pointed that the relaxation processes are the possible reason of the instability, there are not the direct indications on the energy source and any transparent quantitative estimation.

It is need to point out some common feature. ***The instability of the shock and detonation waves*** (at zero or even negative balance of the chemical energy ΔH_x^0) ***is observed in the experiments when the initial system contains the molecules involving the relatively great numbers of the freedom degrees*** (CH_4, C_2H_2, фреон-12) ***or atoms having metastable states*** (for example argon). The parameters of the medium and the shock wave must obtain, firstly, the excitation of the vibrational or electronic freedom degrees, or the significant ionization arises and, secondly, relatively low density. The first of ones means the conversion of the part of the flow energy to the internal freedom degrees unconnected with pressure, the second one supplies the different relaxation times of the internal and translational freedom degrees.

During the processes of the shock compression in the transition zone the molecules of the relatively cold immobile medium run against the molecules of the hot gas involved in the ordered motion behind shock wave. Hence the kinetic energy of the ordered motion of gas K_o, which can exceed K_s several times, is added to the kinetic energy of the thermal movement K_s of the "hot" molecules behind shock wave moving "in the direction x". We shall do a rough estimation on an example of perfect gas. One can express K_s and K_o in the known formulas through Mach number of the shock wave relative initial state M_0:

$$K_s = \frac{1}{2}\frac{RT_s}{\mu} = \frac{1}{2}\frac{RT_0}{\mu}\frac{[(\gamma-1)M_0^2+2][2\gamma M_0^2-(\gamma-1)]}{(\gamma+1)^2 M_0^2} , \ K_o = \frac{4\gamma RT_0[M_0^2-1]^2}{\mu(\gamma+1)^2 M_0^2} , .$$

The ratio of these energies is $\xi = \dfrac{K_o}{K_s} = \dfrac{8\gamma[M_0^2-1]^2}{[(\gamma-1)M_0^2+2][2\gamma M_0^2-(\gamma-1)]}$.

At $M_0>1$ this ratio monotonically increases with augmentation of M_0 and at $M_0>>1$ tends to $4/(\gamma-1)$. Hence, at $5/3>\gamma>6/5$ the limit value of ξ changes from 6 for the monatomic molecules to 20 for gas having the sufficiently complex molecules. Already under $M\sim6$ the value of ξ for these value of γ consists of 5,3 and 15. Accordingly the "translation temperature" of collision into the transition layer may be the same times more than the temperature behind the shock wave after Maxwell's distribution being established. Our very rough estimation shows that in the fort shock waves at the exit from the transition zone the population of the high levels of energy can exceed many times the equilibrium population defined by the temperature behind the shock T_s.

The reality of such processes was proved by S.A.Losev's group [24-26]. They have undertaken the experimental study of the argon spectra containing some impurity of hydrogen ($\approx$1%) behind the shock wave with Mach number 6. Under the established temperature behind the shock T_s=3352 K ($\approx$0,35 eV.) from the data of the spectrum measurement the concentration of metastable atoms of argon (energy level is 11,72 eV) consisted of $2 \cdot 10^8$ atoms per cubic centimeter whereas the equilibrium concentration at the given temperature must be 10^7 times less – only 21 atoms per cubic centimeter. Accordingly these data in transition zone of wave front the average "temperature" exceeded T_s 1,7 times.

For the strict theoretical investigation of the structure of the shock and the processes taking place within the front transition zone it is need to solve the Boltzman's equations. The approximate decision of this problem by the introduction of the bimodal distribution function belongs to Mott-Smith [27]. On the ground of this work in [24-26] the formula for calculation of the maximum temperature, which is reached within transition layer, was offered. This finding is more exact than presented here rough estimation and gives the results being in a good agreement with experiments.

At the rather quick temperature drop in the rarefaction wave a "freezing" of the vibrational and electronic freedom degrees can occur and the conditions in which their energy similar to chemical energy appears as "potential" energy in relation to translation ones. In such systems the evident conditions of the instability formation are sufficiently great number of the freedom degrees, lowered density of gas, the significant steepness of the temperature drop and also the existence of the positive feedback: – the pressure augmentation arising in the consequence of fluctuations must lead to acceleration of the heat release the then the small perturbation will be intensified. These conditions are always realized in the detonation waves and in all experiments, in which instability of the shock waves observed, in some way or other.

As we pointed out above in the number of others the instability of the shock waves was also observed in the experiments with the monatomic rare gas argon [20]. The presence of the long-living metastable level 4s(11,72 eV) of argon atom creates possibility of breaking of the equilibrium of flow in the rarefaction wave and hence of instability generation. The suppression of the instability observed in these experiments at addition the impurity of hydrogen points to the same reason. Hydrogen has the short-living level 2p (12,09 eV) near resonant with 4s of argon, that lead to quick (by means of collision) depopulation of the metastable level of argon and the instability suppression.

The discussed phenomena in the transition zone of the shock wave did not take to account in the model [17] but can contribute to accumulation of additional energy in the internal freedom degrees, lead to formation "over-equilibrium" concentrations of free radicals and in a certain condition by which means affect strongly the kinetics of initial stages of the chemical reactions. For the condensed explosives the possibility of these phenomena was discussed in [28].

Conclusions

1. The conservation of instability of the overdriven detonation waves in the mixture of the type "B" when the chemical reaction heat release ΔH_x^0 becomes (already) negative is explained by the additional energy supply appeared in consequence of the reconstitution and "simplification" of the molecules in course of the chemical reactions. Liberation of energy of the internal freedom degrees accumulated during the shock transition supplies additional feed of the translational freedom degrees. The wave becomes stable when it is overdriven so that the feed is fully absorbed by intensification of the detonation products dissociation.

2. The quantity of the energy accumulated during the shock transition by the vibrational and electronic freedom degrees and liberated to the translational freedom degrees can produce the energy supply comparable with the heat release realized in the usual gas detonation.

3. The activation energy is of little significance in the phenomenon of the detonation waves instability as far as it defines manly ignition delay times during of which the energy is not practically released and gasdynamic parameters are nearly constant. The reactions with the triple collision, that manly are responsible for the chemical heat release, have the practically zero activation energy. The rates of them (and therefore temp of the heat release) is defined mainly gas density rather than the exponent of the reverse temperature.

4. An instability of the overdriven detonation at $\Delta H_{ch}^0 \leq 0$ and shock wave have common nature – the energy release during process of non-equilibrium expansion in the rear of front. The existence of the positive feedback – acceleration of the energy release with the pressure increase like it take place in the chemical reaction or in the relaxation zone the shock waves – lead to the gain of fluctuation.

5. The processes taking place within the shock transition can result in the medium having the big number of the free active radicals that leads to sharp changing of the kinetics of the initial stages of the chemical reactions.

Acknowledgements

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