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MOLECULAR THERMODYNAMIC METHOD OF CORRELATING HEAT TRANSFER IN LIQUID BOILING

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A correlation is obtained for heat transfer in boiling of various liquids. A classification of materials in similarity groups is proposed.

The present thermodynamic method for analyzing the vapor-generation process has been used successfully to develop important aspects of theory and to calculate many thermal processes. Boiling involves very complex physical processes which depend on thermodynamic, hydrodynamic, and molecular factors.

The well-established correlations of Kruzhilin, Kutateladze, Borishanskii, Tolubinskii, Labunstov, et al., based on the thermodynamic method of analyzing the process, do not allow correlation of test data on heat transfer in bubble boiling of liquids with sharply differing physical properties and process conditions.

The method proposed by I. I. Novikov and V. M. Borishanskii for obtaining correlations, based on using the thermodynamic law for the respective states, although it enlarges the capability to calculate the influence of physical properties of the boiling medium, does not however provide a broad correlation of heat transfer in boiling of liquids under different hydrodynamic process conditions.

The reasons for the unsatisfactory correlation with methods of thermodynamic analysis of test data on heat transfer in bubble boiling, in our opinion, lie in the deviation of individual properties of substances from the general thermodynamic law for the respective states [8, 10, 52, 53], in the incomplete allowance for hydrodynamic process conditions and the effect of the molecular properties of the material, particularly the intermolecular interactions in phase transition, and also in the fact that existing methods for allocating substances into thermodynamically similar groups do not take into account certain properties which characterize the behavior of different substances in the vapor-generation process.

It is evident that the thermodynamic method of analysis, while remaining important, has lost its exclusive feature and must be supplemented. The supplement may take the form of analysis of the boiling process allowing for the molecular characteristics of the system.

The main content of the molecular thermodynamic method is the fact that the boiling process is considered from the viewpoint of simultaneous interaction of macro- and microparticles of material; the allocation of boiling substances to similarity groups is carried out on the basis of the influence of the thermodynamic properties of the substance in the corresponding process states, and also the nature of interaction of molecules

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and their quantum-mechanical structure; a more complete system of original equations and boundary conditions is used, including the previously determined initial systems of equations of Kruzhilin, Kutateladze, Borishanskii, as well as additional boundary conditions [13].

In [13] a correlation is derived for heat transfer in boiling, and it also gives the thermodynamic basis and an interpretation of the basic parameters of the equation

$$\mathrm{Nu} = c\mathrm{M}^{n}\mathrm{Pr}^{n_1}\mathrm{W}^{n_2}\mathrm{H}^{-m}T_{\tau}^{k}. \qquad (1)$$

Below we give a molecular basis for Eq. (1) and describe the method of analysis of the boiling process.

The aggregate state of an actual substance, as is known, is determined by intermolecular bonds, on which depend properties of the substance such as capillary phenomena, thermal capacity, thermal expansion, viscosity, thermal conductivity, latent heat of vaporization, vapor pressure, work function, the amount of work done by the vapor, and the internal energy of vaporization. All these properties are interrelated and are determined by the intermolecular interaction. However, it is as yet difficult to relate these quantities uniquely with the energy of collective intermolecular interaction [8, 9, 52, 55, 58]. This is probably why there has not yet in fact been a theoretical investigation of heat transfer of boiling of liquids, allowing for the molecular properties of the system. All the known correlations have described the process only with the help of thermodynamic parameters without direct calculation of the basis molecular properties of the material. No laws have been established describing the heat transfer process in boiling with the molecular conditions. In this paper we use the molecular thermodynamic method to analyze and describe the heat transfer process in boiling of liquids of various kinds.

Any change in the intermolecular interactions with combination or decomposition of molecules during phase transformations is associated with a rearrangement of the electronic shells and a variation of distances in the molecules [2, 4, 9, 17]. Therefore, the energy expended in the change of aggregate state of the material can be determined, with sufficient accuracy, only if one takes into account the molecular structure of the substance [1, 2, 7, 8, 59]. However, the thermodynamic parameters T_s and L, and also the Lennard–Jones potential cannot account for the molecular structure of the substance and do not accurately describe the energy of intermolecular interaction in the state [1, 7, 8, 10-12, 53, 56].

The energy characteristic of intermolecular interactions depends in a complex way on the nature, mass, and size of the molecules, the shape of their lattice structure, the polarity, the polarizability, and other effects. The change in the aggregate state of the liquid and vapor is connected with the complex intermolecular interaction and as yet there is no method by which one could calculate these interactions numerically [2, 8, 9, 18, 57].

Because there is almost no potential energy of interaction between molecules in a gas, compared to molecules in a liquid, it appears possible to determine the energy of potential interaction of liquid molecules from the kinetic energy communicated by the molecules in transition of material from the liquid to the vapor state, i.e., from the amount of heat expended in complete breakdown of the bonds between molecules in the liquid–vapor phase transition. To do this, one must consider the change in energy of molecules of different substances in the boiling process in rigorously corresponding states, and allocate materials to similarity groups, allowing for the structure of molecules and the nature of the interactions.

Different methods are known for allocating substances to thermodynamically similar groups [10, 12, 53, 56, et al.]. However all these methods have a number of defects. The use of the principle of equality of critical coefficients, of Truton number, and also Guldberg number does not give positive results, since the values do not prove to be constant for the materials, referenced to a particular group (or even a homologous) [1, 8, 10-12, 56]. These methods do not provide a unique comprehensive characteristic for the substance, probably because the parameters which they use do not take into account the molecular properties of the substance.

Below we describe the molecular thermodynamic method of classifying heat transfer agent materials by similarity groups, accounting for the molecular and thermodynamic properties of the boiling substances.

As a generalized parameter describing, as will be shown below, the level of intermolecular interaction in the above-mentioned process, we take the molecular mass, which, according to Avogadro's law, $M = (\rho/\rho_C)M_C$ or $M = (\rho/\rho_{O_2})M_{O_2}$. The number M was first used in the heat transfer equation in boiling by V. M. Borishanskii [58].

TABLE 1. The Energy System of Molecules of Boiling Substances

Group	Substance	M	$B = \frac{NuH^m}{Pr^{0.35} W^{0.7} T_k}$	L, kcal/kg	Lr, J/mole	Nuclear charge	Electronic configuration of the molecules: K σ	L σ	L π	M σ	M π	M δ	N σ	N π	N δ	N φ	O σ	O π	O δ	O φ	P σ
First	H_2	2	920	108,9	916	2	2														
	He^4	4	780	485	81	2	2														
	CH_4	16	540	122	8200	10	6	2	2												
	Ne	20	520	20,8	1755	10	2	2	6												
	N_2	28	470	47	5530	14	4	4	6												
	O_2	32	460	50,9	6800	16	4	4	8												
	F_2	38	440	41,1	6560	18	4	4	10												
	Ar	39,94	430	39	6500	18	2	2	6	2	6										
	O_3	48	400	75,6	15200	24	6	6	12												
	Kr	83,8	355	25,8	9050	36	2	2	6	2	6	10	2	6							
	Xe	131,4	320	23	12600	54	2	2	6	2	6	10	2	6	10	—	2	6			
Second	NH_3	17	835	327	23300	10	5	2	3												
	H_2O	18	830	539	40000	10	4	2	4												
	CO_2	44	750	83	15250	22	6	6	10												
	C_2H_5OH	46	730	205	40000	26	12	6	8												
	C_6H_6	78	710	76	24700	42	18	12	12												
	CF_4	88	700	33	12000	42	10	10	12												
	CF_2Cl_2	120,9	680	40	20000	58	10	10	24	4	10										
Third	Li	6,94	180	4680	136500	3	2	1													
	Na	23	240	1005	96700	11	2	2	6	1											
	K	39	300	496	80700	19	2	2	6	2	6	—	1								
	Cs	132	380	119	66100	55	2	2	6	2	6	10	2	6	10	—	2	6	—	—	1
	Hg	200,6	425	70	58500	80	2	2	6	2	6	10	2	6	10	14	2	6	10	—	2

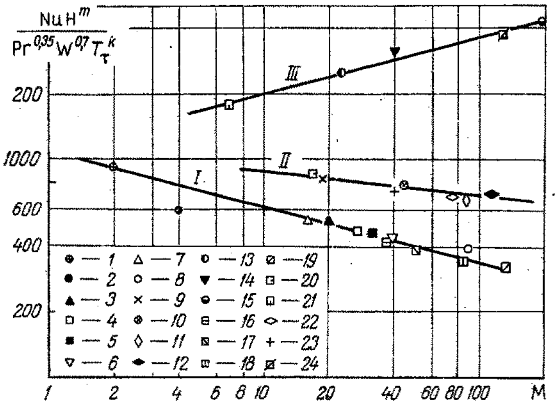

Fig. 1. Influence of the parameter M on the classification of boiling liquids in similarity groups: 1) H_2; 2) He^4; 3) Ne; 4) N_2; 5) O_2; 6) Ar; 7) CH_4; 8) Φ_{-22}; 9) H_2O; 10) C_2H_5OH; 11) Φ_{-14}; 12) Φ_{-12}; 13) Na; 14) K; 15) Hg; 16) F_2; 17) O_3; 18) Kr; 19) Xe; 20) NH_3; 21) Li; 22) C_6H_6; 23) CO_2; 24) C_S.

It is known [7, 8, 12] that the molecular mass M is a constitutive quantity, i.e., it is related to the molecular structure. We therefore postulated [19, 54] that the molecular mass of a substance, if one takes into account the nature of the molecule, can describe in a complex way the activity of molecular interactions in a process like phase transition of a substance during vapor formation. For this kind of characteristic it is necessary that all the substances considered be classified in physically similar groups, allowing for the molecular structure and their interactions.

On the basis of an energy table (Table 1) of molecules for cryogenic, high-boiling, and metallic liquids we have given: the relative molecular mass of the substance, the molecular structure, the similarity of the shape (or nature) of intermolecular bonds, and the interaction energy between molecules.

It can be seen from Table 1 that materials which are widely known, and for which the boiling process has been studied in detail experimentally, are divided into several groups according to similarity in the nature of the intermolecular bonds. In each group the substances are ordered by sequential increase in the relative molecular mass and by the complexity of the system of electronic configuration of the molecules and their nuclei.

The first group contains mainly liquids with nonpolar mono- and diatomic nonassociated molecules with a covalent bond, the outer orbital of which is populated by only P-electrons (except for H_2 and He^4), and whose nuclear charge is even. This group contains all the cryogenic liquids.

The second group contains predominantly liquids with polar multiatomic associative molecules with an ionic bond, whose outer orbital is also populated only by P electrons with an even value of nuclear charge. This group contains liquids with a high boiling temperature and completely halogenized Freons.

The third group contains liquids with a metallic bond for the molecules (or atoms), where the outer orbital contains only S electrons, and the nuclear charge has an odd value (except for Hg).

It is typical that in each group of Table 1 substances are located in the order of sequential increase in the number of electrons in the external quantum level or in the two last levels, which indicates a connection between the relative molecular mass and the electron configuration of the molecules and with the structure of their nuclei. A similar bond also exists between the atomic mass and its electronic and nuclear structure [4, 7, 10].

To determine the energy bond between groups of molecules for different substances (graph 4 and Table 1) experimental data from more than 70 investigators on the boiling of 20 liquids was processed. Figure 1 shows the results of processing in the coordinates of the correlation equation Eq. (1). It can be seen in Fig. 1 that all the liquids considered fall into three physically similar groups, as was foreshadowed by Table 1. On the basis of graph 4 of Fig. 1, Table 1 shows the numerical values of the interaction energy between molecules in the vapor-generation process.

TABLE 2. Cryogenic Liquids

Liquid	Point No.	P, kg/cm²	T_s, °K	$\frac{T_s}{T_{cr}}$	Boiling conditions	Literature source
Helium	1	1	4,25	0,811	Free volume	[20]*
	2	1	4,25	0,811	In a tube l/d=86	[20]*
	3	0,71	3,84	0,733	Free volume	[20]*
	4	1,83	4,91	0,446	The same	[20]*
	5	1	4,21	0,811	»	[21]
	6	1	4,21	0,811	»	[22]
	7	1	4,21	0,811	»	[23]
	8	1	4,21	0,811	»	[24]
	9	1	4,21	0,811	»	[25]
Hydrogen	10	1	20,27	0,61	Free volume	[26]
	11	5,1	27,5	0,83	The same	[27]
	12	8,5	31	0,94	»	[27]
Neon	13	1	27,07	0,61	»	[22]
	14	1	27,07	0,61	»	[26]
	15	1	27,07	0,61	»	[28]
	16	4	32,5	0,73	»	[28]
	17	10	37,5	0,84	»	[28]
Nitrogen	18	1	77,3	0,61	In a tube l/d=560, H=0,4	[29–33]
	19	1	77,3	0,61	Free volume	[34]
	20	1	77,3	0,61	The same	[21]
	21	1	77,3	0,61	»	[26]
	22	0,42	70	0,55	»	[23]
	23	7,72	100	0,79	»	[23]
	24	15,7	111,2	0,88	»	[23]
Liquid air 50% N_2+O_2	25	1	83,8	0,59	In a tube l/d=560, H=0,6	[29–33]
Argon	26	1	87,3	0,58	Free volume	[26]
	27	4	103,4	0,68	The same	[35]
	28	16	125	0,85	»	[35]
	29	32,9	141	0,94	»	[35]
Oxygen	30	1	90,2	0,53	In a tube l/d=106, H=0,4	[29—33]
	31	1	90,2	0,53	The same H=0,6	[29—33]
	32	1	90,2	0,53	» H=0,9	[29—33]
	33	1	90,2	0,53	l/d=330, H=0,8	[29—33]
	34	1	90,2	0,53	l/d=560, H=0,6	[29—33]
	35	1	90,2	0,53	Free volume	[36]
Oxygen	36	1	90,2	0,53	The same	[36]
	37	10	120	0,776	»	[37]
	38	32,2	143	0,93	»	
Methane	39	1	111,7	0,58	Free volume	[35]
	40	1	111,7	0,58	The same	[38]
	41	14,1	156,5	0,82	»	[38]
	42	37,8	183,7	0,96	»	[38]

*The thermal conductivities used are for the metal [39].

The parameter B expresses in dimensionless quantities the energy expended in breaking down the bonds between molecules of the liquid in the vapor-formation process. From Table 1 and Fig. 1 it can be seen that the energy of interaction between molecules in a phase transformation of different substances belonging to the same similarity group varies with increase in the relative molecular mass of the substance. In a transition from substances of the first group (line I) to substances of the second and third groups (lines II and III), the effect of change in energy with increase of M is repeated periodically. We shall call this energy change in a phase transition quasiperiodic. This nature of the relationship continues through Table 1, depending on the electronic configuration of the molecules. We note that for substances whose external orbital is populated by P electrons and whose nuclear charge is even (cryogenic and high-boiling liquids), the energy in breakdown of molecules in vapor formation decrease with increase of the molecular mass of the material. Conversely, for substances consisting of molecules whose outer orbital is populated by S electrons and whose nuclear charge is odd (metallic liquids), the total energy in breakdown of molecules during vapor generation increases with increase of molecular mass of the substance.

The cause of variation of energy expended in breakdown of molecules during vapor formation of different substances is the effect of screening of the nuclei of the molecules by their own electrons.

Thus, the kinetic energy expended in overcoming the intermolecular interactions in the boiling process depends on the quantum-mechanical structure and the nature of the bond between the interacting molecules.

The effects of molecular thermodynamic interaction described permit us to formulate the following law: the energy properties of materials during vapor formation depend in a quasiperiodic manner on the nature of the intermolecular bonds and on the relative molecular masses of the substance.

TABLE 3. High-Temperature Liquids

Liquid	Point No.	P, kg/cm^2	T_s, °K	$\frac{T_s}{T_{cr}}$	Boiling conditions	Literature source
Water	*1*	1	373	0,577	In a tube l/d=46, H=1	[40]
	2	1	373	0,577	The same H=0,7	[40]
	3	1	373	0,577	The same H=0,5	[40]
	4	1	373	0,577	l/d=58, H=0,25	[41]
	5	10	453	0,7	Free volume	[42]
	6	50	536	0,83	The same	[42]
	7	100	584	0,9	»	[42]
	8	199,9	638	0,99	»	[42]
Freon-14	*9*	1	145	0,64	Free volume	[35]
Freon-12	*10*	1,72	256	0,668	Free volume	[43]
	11	5,76	293	0,76	The same	[43]
	12	5,05	290	0,752	»	[44]
Freon-22	*13*	3,62	263	0,713	Free volume	[43]
	14	6,95	283	0,767	The same	[43]
	15	9,38	293	0,795	»	[43]
	16	8,5	289	0,784	»	[44]
Ethyl alcohol	*17*	1	351	0,68	Free volume	[42]
	18	40,2	486	0,94	The same	[42]
	19	59,6	513	0,995	»	[42]
Benzene	*20*	0,987	348	0,62	Free volume	[45]
	21	2,72	381,2	0,68	The same	[45]
	22	22,5	501	0,893	»	[45]
	23	36,2	537	0,957	»	[45]
Carbon dioxide	*24*	44,8	283	0,93	Free volume	[46]
	25	64,4	298,1	0,96	The same	[46]
	26	65,9	298,3	0,98	»	[47]

TABLE 4. Liquid Metals

Liquid	Point No.	P, kg/m^2	T_s, °K	$\frac{T_s}{T_{cr}}$	Boiling conditions	Literature source
Sodium	*1*	0,147	973	0,385	Free volume	[48]
Sodium	*2*	0,922	1133	0,448	Free volume	[48]
Potassium	*3*	1	1033	0,486	Free volume	[49]
	4	2,04	1095	0,515	The same	[49]
	5	1	1033	0,486	In a tube l/d=60	[51]
Cesium	*6*	0,006	613	0,322	Free volume	[48]
Mercury	*7*	1	629	0,431	Free volume	[50]
	8	4,5	718	0,432	The same	[48]

What has been said above is evidence that one of the important dimensionless parameters governing the boiling process of liquids of different types is the number M, the molecular mass. However, no laws were established in a previous paper [58] for the influence of M on the boiling process for different liquids, and the physical meaning of this was not identified, taking into account the nature of interaction and the structure of the molecules of the boiling substance.

The validity of the above classification of materials by similarity groups in the molecular thermodynamic method of analysis, and of the law established is confirmed by the fact that by using these one can correlate the experimental results of bubble boiling of different liquids in a single technique, from helium to liquid metals at low and near-critical pressures (Fig. 2), and obtain correlations for numerical evaluation of heat transfer during boiling in a free volume and in tubes with natural flow circulation: for cryogenic liquids we have

$$\mathrm{Nu} = 9\mathrm{M}^{-0.25}\mathrm{Pr}^{0.35}\mathrm{W}^{0.7}\mathrm{H}^{-m}T_{\tau}^{7},$$

for high-temperature boiling liquids we have

$$\mathrm{Nu} = 9\mathrm{M}^{-0.1}\mathrm{Pr}^{0.35}\mathrm{W}^{0.7}\mathrm{H}^{-m}T_{\tau}^{10},$$

and for liquid metals we have

$$\mathrm{Nu} = 0.8\mathrm{M}^{0.25}\mathrm{Pr}^{0.35}\mathrm{W}^{0.7}T_{\tau}^{8}.$$

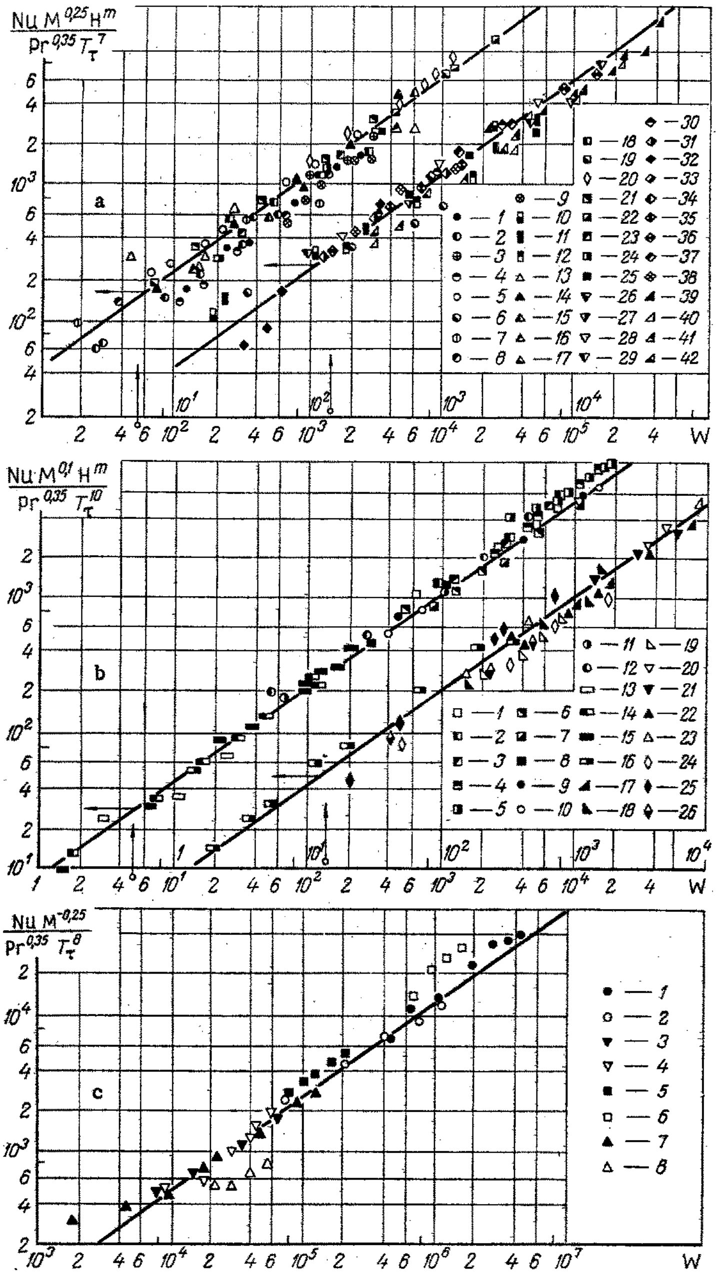

Fig. 2. Heat-transfer in bubble boiling: a) cryogenic liquids (the parameters of the experimental points are given in Table 2); b) high-temperature liquids (the parameters of the experimental points are given in Table 3); c) liquid metals (the parameters of the experimental points are given in Table 4).

These formulas have been checked, and with them we can calculate to an accuracy up to ±20% the heat transfer coefficients in bubble boiling of the liquids listed in Table 1 in the following range of parameters: $W = 0.15\text{-}10^6$; $M = 2\text{-}200$; $H = 0.25\text{-}1$; $T_\tau = 0.32\text{-}0.99$; $Pr = 2.41^{-3}\text{-}7.9$*; $l/d = 1\text{-}560$; $d = 1\text{-}40$ mm with circulating velocities up to 1 m/sec.

These formulas describe heat transfer on different heating surfaces of thickness less than 0.5-1 mm with a technical process purity of $\delta \leq \mu$m.

NOTATION

$Nu = (q/\lambda\Delta t)(\sigma/\gamma-\gamma_v)^{0.5}$ is the Nusselt number for boiling;
$Pr = \nu/a$ is the Prandtl number;

*As in Russian Original – Publisher.

$H = h/l$ is the apparent liquid level in a vertical tube;
$W = [q(l/d)^{0.65}/L\gamma_v\nu](\sigma/(\gamma-\gamma_v))^{0.5}]$ is the analogue of the Reynolds number for boiling, where the reduced velocity of motion of the vapor is taken as the governing parameter;
$M = (\gamma/\gamma_C)M_C$ is the relative molecular mass of the substance;
$T_\tau = T_s/T_{cr}$ is the thermodynamic similarity parameter for the substance, describing the relative kinetic energy of thermal motion of the molecules;
q, q_{cr1} are the specific and critical heat flux density;
c is the specific heat of the liquid;
γ, γ_v are the specific weight of liquid and vapor;
γ_C, γ_{O_2}, M_C, M_{O_2} are the specific weight and the molecular mass of carbon dioxide and oxygen;
λ is the thermal conductivity of the liquid;
σ is the surface tension;
Δt is the temperature difference;
ν is the kinematic viscosity of the liquid;
$a = \lambda/c\gamma$ is the thermal diffusivity;
h is the absolute level or the hydrostatic head of the liquid in the tube;
d, l are the inner diameter and the length of the tube;
L is the heat of vapor formation;
T_s is the boiling temperature;
T_{cr} is the critical temperature;
$m = [2900\cdot(l/d)^{-1.65}]^{1-k}\cdot 10^{-k}$;
$m_1 = [1300\cdot(l/d)^{-1.65}]^{1-k}\cdot 10^{-1.35k}$;
$k = q/0.1q_{cr1}$
For boiling in a free volume $H=1$, $l/d = 80$.

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