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Determination of the Efficiency of High-Entropy Cutting Tool Materials

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Abstract—Modern approaches to determination of the durability of cutting tool materials taking into account the effect of their entropy, Thermo-EMF, and the functional relationships between them are presented. It is confirmed that the tribological properties of complex alloyed high-speed steels and experimental cemented carbide hard alloys (ECCs) with modified cobalt binder as high-entropy materials can be improved. The results of a study of wear resistance, oxidation resistance, and optimum cutting conditions of ECCs are presented.

Keywords: cutting tool materials, wear resistance, Thermo-EMF, entropy, high-entropy materials

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INTRODUCTION

In the course of development of mechanical engineering and metalworking, there has been a constant increase in the requirements for the quality of produced machines using the current equipment, technologies, tools, and cutting process management systems.

An important role in providing the efficiency of the metalworking and the reliability of tool adjustment is played by blade cutting tools (CTs). Currently, the main trends in improving the performance characteristics of CTs are the development of advanced designs using multiblade indexable inserts, aging of working surfaces of tools by wear-resistant coatings of various compositions, implantation of thin surface layers using elements forming wear-resistant structures, a decrease in the carbide grain sizes in hard alloys, the application of enhancing technologies, increasing complication of the chemical composition of high-speed alloys (HSSs) by introducing new alloying elements, and upgrading of the carbide and the binding phases in hard alloys (HAs) with the purpose of increasing the consumption of unavailable tungsten and cobalt without decreasing the physico-mechanical characteristics.

It is worth noting the necessity for further increasing of the specific weight of HAs for the equipment of cutting tools because the level of cutting speeds with hard alloy tools used in turning operations performed using the current CNC machine tools, FMM, and FMS can reach 300–500 m/min. In such conditions, hard alloy tools work at temperatures reaching the heat resistance limit and are subject to mainly the diffusion and the oxidation types of wear.

Currently, the performance characteristics of both HAs and HSSs are found largely on the basis of experimental data; in the development of new compositions thereof, metallurgical, technological, and economic factors are mainly considered. Thus, it is advisable to a priori prognose the properties of current and newly developed compositions for cutting tool materials (CTMs) based on studies of the complex of physical or, more precisely, the thermodynamic processes that occur in the friction zone upon cutting and the analytical dependences for evaluation of the friction and wear characteristics. In this area, the main techniques for the wear decrease and the wear resistance management at friction were determined, focused on providing the thermodynamic state with the minimum density of the accumulated thermal entropy in CTMs, applying materials with high entropy values, and taking into account the other inhomogeneities of thermodynamic nature (diffusion, oxidation, and triboelectric effects) [1], which are macrolevel problems, and the evaluation of the influence of a current CTMs on wear as heterogeneous entropy structures and of the absolute Thermo-EMF as a structurally sensitive characteristics of materials [1] on the macrolevel.

The aim of this study is, based on the relations between the entropy and structural characteristics of CTMs, to develop high-entropy alloys and evaluate their tribological characteristics.

Table 1. Compositions of binders of experimental cemented carbide hard alloys

No	Alloy	Binder
1	2.1	6.23% Co + 2.08% Mo
2	2.2	5.6% Co + 3% Mo
3	2.3	3.7% Co + 6.02% Mo
4	2.19	1.52% Co + 5.03% Fe + 0.82% Cu
5	2.20	3.6% Co + 3.2% Fe + 0.82% Cu
6	2.21	5.4% Co + 1.43% Fe + 0.82% Cu
7	2.22	5.65% Co + 1.8% Mo + 0.6% Ti
8	2.23	5.1% Co + 2.7% Mo + 0.61% Ti
9	2.24	3.34% Co + 5.44% Mo + 0.67% Ti
10	VK8 (by specification)	[7.5–8]% Co; no more than 0.3% Fe; O_2 no more than 0.5%

MATERIALS AND METHODS

The experiments for the determination of the resistance were carried out at longitudinal turning of steel 45, without cooling, using a 1K620 tool with the stepless control of rotational speed of a spindle; the cutting rate was varied in the range of 0.5–2.9 m/s, the cross section of the cut was assumed as constant $ts = (0.5 \times 0.23) \times 10^{-6}$ m^2. The 0229 shape plates of the experimental cemented carbide hard alloys (ECC) were mechanically fixed in the holder and had the following geometry: $\gamma = 8°$, $\alpha = 6°$, $\varphi = 45°$.

The determination of the heat resistance of HAs (Table 1) was carried out in accordance with GOST (State Standard) 6130–71 at temperatures of 300 and 500°C for 0.5–4 h. After cooling, the pots were weighed. The weight gain was calculated by the difference of masses before and after the oxidation.

Qualitative X-ray diffraction analysis (XRD) of HAs was carried out by the powder method using an ARLXTRA diffractometer.

RESULTS AND DISCUSSION

CTM wear evaluation using the entropy approach at friction and cutting of a tool–element pair as for an open thermodynamic system exchanging energy and mass with the environment permits using the entropy balance equation for the description of its behavior [1]:

$$\frac{d}{d\tau}\int_V S\,dV = \int_V \sigma[S]\,dV - \int_V \operatorname{div} J[S]\,dV \equiv P[S] - \Phi[S], \tag{1}$$

where $P[S]$ is the entropy production, $\Phi[S]$ is the entropy flow, dV is the volume, and $d\tau$ is the time.

For evaluation of the wear rate, it was assumed that wear volume V of a material is caused not by the whole density of the accumulated entropy, but only by a part of it taken into account by the coefficient k:

$$k\left(\int_V \sigma[S]\,dV - \int_V \operatorname{div} J[S]\,dV\right). \tag{2}$$

The adopted measure of the efficiency of a cutting tool material (CTM) at cutting was the entropy criterion of failure; the influence of the thermal, mechanical, chemical, electrical, and convective processes can be taken into account based on their contribution to entropy production and flow [1, 2].

The initial equation for the calculation of the wear rate taking (1) and (2) into account is as follows:

$$\int_V [S]\,dV_u = k\left\{\int_0^\tau\int_V (\sigma[S] - \operatorname{div} J[S])\,dV d\tau\right\}. \tag{3}$$

The volumetric wear rate is determined as follows:

$$\frac{dV_u}{d\tau} = \frac{k\int_V (\sigma[S] - \operatorname{div} J[S])\,dV}{S}. \tag{4}$$

If q is the nominal contact surface area, then the wear rate is as follows:

$$\frac{dV_u}{d\tau} = \frac{kq\int_0^h (\sigma[S] - \operatorname{div} J[S])\,dx}{S}. \tag{5}$$

Taking into account that the wear rate is determined as

$$J = \frac{dV_u}{d\tau}\frac{1}{q\upsilon}, \tag{6}$$

where υ is the friction (cutting) speed, then, taking into account (5) and (6), the following results:

$$J = \frac{\int_0^h (\sigma[S] - \operatorname{div} J[S])\,dx}{\upsilon S} k, \quad (7)$$

where h is the linear wear.

Limit density of entropy S in the worn TM taking into account the standard (zero) value and the temperature [3] has the following form:

$$S = S_{298}^0 + \int_{298}^{T} \frac{C_p}{T} dT, \quad (8)$$

where C_p is the heat capacity of TM and T is the absolute temperature, K.

Because the hard alloys are used during cutting in conditions of intense heat release and are exposed to oxidative wear, it is necessary to take into account the contribution of the chemical reactions of the oxidation of HA components to the entropy production using the corresponding parameter $\sigma_x[S]$ in (7) [2]:

$$\sigma_x[S] = \sum_1^k G_i \frac{dh_i}{d\tau} \eta_i \frac{1}{T} = \sum_1^k (H_i - TS_i) \frac{dh_i}{d\tau} \eta_i \frac{1}{T}, \quad (9)$$

where G_i, H_i, and S_i are the Gibbs potential, the enthalpy, and the entropy of the formation of ith grade oxide, respectively; $\frac{dh_i}{d\tau}$ is the film growth rate of the ith grade oxide; and η_i is the fraction of the ith oxide in the oxide layer.

Taking into account the prevailing role of the thermal processes and the oxidation in the HA wear yields the following:

$$J = \frac{\int_0^h \lambda\left[\left(\frac{dT}{dx}\frac{1}{T}\right)^2 - \frac{d^2T}{dx^2}\frac{1}{T}\right]dx + \sum_1^k \left[(H_i - TS_i)\frac{dh_i}{d\tau}\eta_i \frac{1}{T}\right]}{\upsilon S} k, \quad (10)$$

and the oxide film growth rate is determined by the expression [2]

$$\frac{dh}{d\tau} = \frac{1}{F\sqrt{2\tau}} \sqrt{\sum_1^k \left(\frac{M_i}{\gamma_i m_i n_i}\right) \eta_i \sum_1^k \left(\frac{H_i - TS_i}{m_i n_i p_i}\right) \eta_i}, \quad (11)$$

where F is the Faraday constant, M_i is the molar weight, γ_i is the density, m_i is the number of moles, n_i is the valence, p_i is the resistivity, and η_i is the mass fraction of the ith grade oxide.

From (10) and (11), an important conclusion follows that the wear at friction and cutting is lower in tool alloy materials with the highest entropy value. The decrease of the fraction of the oxidative wear in the total wear is contributed by oxides with the maximum value of the entropies of their formation S_i (expression (9)).

In dependences (10), to determine the wear rate, it was assumed that the material is compositionally homogeneous. However, the current tool materials, like all the metal alloys, are multicomponent heterogeneous structures consisting of at least two phases: hard carbide alloys and a metal binder, while high-speed steels are alloyed cementite, martensite, and retained austenite. These components differ both by the mechanical and by the physico-chemical properties. For multicomponent systems, if they are considered ideal solutions, all the thermodynamic potentials possess the additivity property [3]. Thus, the entropy of CTM can be calculated as the sum of the product of the molar fraction of each component on the value of its standard entropy; i.e., $S_{ij} = \eta_1 S_i + \eta_2 S_j$, where $\eta_1 + \eta_2 = 1$, S_i and S_j are the entropies of each component of an alloy.

On the other hand, to evaluate the wear resistance of a CTM, a structure-sensitive microlevel parameter, namely, the absolute Thermo-EMF ε of a material, can be used, which, taking into account the diffusion component, can be evaluated through the dependence [4]

$$\varepsilon = -\frac{\pi^2 k^2}{3e} T\left(3/2W_F - N_9'/N_9\right) W_F, \quad (12)$$

where k is the Boltzmann constant; e is the electron charge; T is the absolute temperature; W_F is the Fermi energy; and N_9 is the density of the electronic states in the valence band, $N_9' = dN/dT$.

Using the classic dependence of M.M. Khrushchev for evaluation of the wear resistance in conditions of the abrasive wear

$$\mathrm{WR} = bH, \quad (13)$$

where H is the hardness of a material and b is the dimensional coefficient of proportionality, m^2/N, express the hardness value if the following form [5]

$$H = (E + K_{H1})^2 + K_{H2}, \quad (14)$$

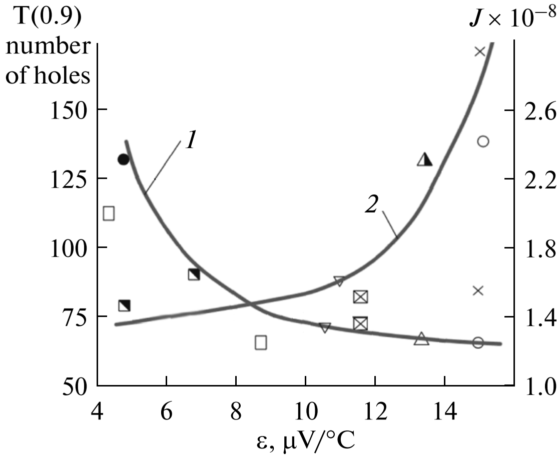

Fig. 1. The relation between the absolute Thermo-EMF and the wear rate of HSSs at friction (*2*) and the resistance warranty of drill bits at cutting (*1*) for various high-speed steel grades. Curves:(◪) R6F2K8M5; (●) R12F2K8M3; (○) R18; (□) R4M4F4; (△) R6M4F4; (×) R6M5; (▲) R6M5K5; (▽) R8M3F4; (⊠) R9F5

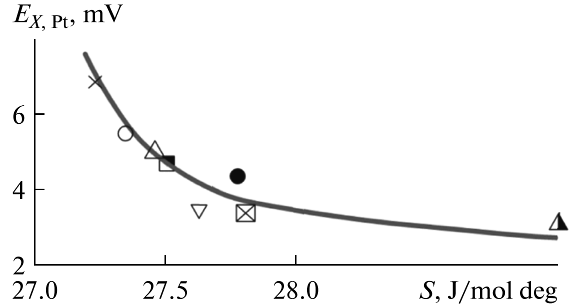

Fig. 2. The relation between the entropy and Thermo-EMF for high-speed steels of various grades (GOST (State Standard) 19265–73): (▽) R6F2K8M5; (▲) R12F2K8M3; (○) R18; (△) R4M4F4; (×) R6M5; (⊠) R6M5K5; (◪) R8M3F4; (●) R9F5.

where E is the binding energy and K_{H1} and K_{H2} are the varying kinetic coefficients, and taking into account that

$$E = \int_{W_{\mathrm{F}}}^{W_{\mathrm{F}-\Delta}} W_{\mathrm{F}} N(W_{\mathrm{F}})\, dE + K_f, \tag{15}$$

where K_f is the energy of atoms in the free state, taking into account (12), the resulting dependence for the calculation of the wear resistance of a material is obtained [5]:

$$\mathrm{WR} = b\left\{\left(A + W_{\mathrm{F}} e^{-\frac{\varepsilon W_{\mathrm{F}}}{K_\varepsilon T}} + K_f + K_{H1}\right)\right\}^2 + K_{H2}. \tag{16}$$

$$\text{Or}\quad \mathrm{WR} = b\left(A + Be^{-\frac{\varepsilon W_{\mathrm{F}}}{K_\varepsilon T}} + C\right)^2 + D. \tag{17}$$

Comparison of calculation wear resistance values WR by (16) with the known experimental data on the relation of the wear resistance with the Thermo-EMF of the group of hard alloy materials shows that, with a decrease in the Thermo-EMF, the wear resistance of HA increases.

The obtained calculated data on the wear resistance agree with the experimental results of the resistance tests of drill bits made of high-speed steels of various grades and the wear rate of these steels at friction. From Fig. 1, it follows that the resistance warranty T(0.9) of a batch of drill bits of the diameter of 13.0 mm (curve *1*) and the wear rate of high-speed steels under friction (curve *2*) correlated with the value of the absolute Thermo-EMF of the steels: the most efficient are the steels having the lowest Thermo-EMF values.

From the theory of thermoelectric phenomena, it is known [5] that the absolute Thermo-EMF of a material is entropy of the moving charge carriers (electrons) S_e, which is determined as the relation of heat transfer Q_e to absolute temperature T:

$$S_e = Q_e/T. \tag{18}$$

Thermal entropy S as a function of the thermodynamic state of a material is calculated using a similar dependence. However, it can be assumed that there is a functional relation between the thermal entropy of a material S and its absolute Thermo-EMF. For the evaluation of the change of Gibbs potential ΔG of the reaction, electromotive force (EMF) of an electric cell E and the analytical dependence for the determination of ΔG are used:

$$\Delta G = -nFE. \tag{19}$$

However, the use of Gibbs potential ΔG of a material is determined using the dependence

$$\Delta G = \Delta H - T\Delta S. \tag{20}$$

The result is as follows:

$$\Delta H - T\Delta S = -nFE, \tag{21}$$

$$\text{thus,}\quad E = -\frac{\Delta H - T\Delta S}{Fn}, \tag{22}$$

where F is the Faraday constant and n is the number of electrons.

The obtained dependence is a relation between the change of the entropy of a material ΔS and its Thermo-EMF: higher values of the entropy of a material correspond to lower values of its EMF assuming that the process occurs at constant enthalpy ΔH and temperature T.

Theoretical dependence (22) and the resulting conclusions were proven for cutting tool materials. In Fig. 2, the results of the calculated values of the entropy for various grades of high-speed steels and the

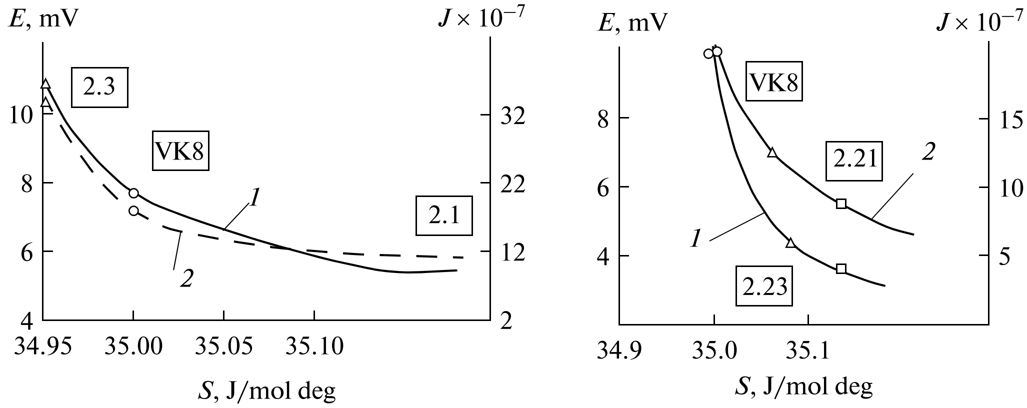

Fig. 3. The relation between the entropy and the Thermo-EMF for experimental HAs of various compositions shown in Table 1. Curves: (*1*) $E = f(S)$; (*2*) $I = f(S)$.

comparison with the values of their relative EMF toward platinum are shown. From Fig. 2, one can see that there is a relation between the entropy of CTM as a thermodynamic characteristic and the relative (absolute) Thermo-EMF, which is approximated by a hyperbolic curve.

Similar results were obtained for hard alloys of various compositions (Fig. 3). From Figs. 2 and 3, it follows that there is a relation between the entropy of CTM as a thermodynamic characteristic and the relative (absolute) Thermo-EMF, which is approximated by a hyperbolic curve, which proves dependence (22).

Hereinafter, entropy is used as the parameter characterizing the efficiency of CTM in conditions of cutting because this parameter can be calculated at a known chemical composition of a material; thus, in CTM design, high-entropy cutting tool materials (HECTMs) should be aimed at. Currently, in the field of materials science, works exist [6, 7] on the development and study of properties, including thermodynamic ones, of multicomponent high-entropy systems as a new class of promising materials possessing high strength, wear resistance, and heat resistance, as well as satisfactory plasticity [7].

In the global practice of cutting treatment, the application of more than 100 grades of hard alloys is known [8]. Based on analysis of literature data, from the HA compositions, some conclusions can be drawn concerning the compositional evolution of hard alloy cutting tool materials: there is a tendency for introduction into the structure of two, three, and four carbide HA; nitrides, carbonitrides, and oxides are introduced into the composition of carbide phases of a HA; and the composition of the binding phase varies in a wide range (pure cobalt; a mixture of elements of the iron group (Fe, Co, Ni) or with Mo, Ti, and Cu additives; and carbon stainless heat-resistant steels and alloys for cutting tools containing, in addition to iron, Cr, Mn, Ti, Ni, Mo, and Cu in their structure [9]).

The known studies do not consider the heterogeneity of HAs' structures as multiphase systems that differ from each other by mechanical, thermophysical, physico-chemical, and thermodynamic properties and by the electronic structure or the mutual influence of carbides (their mixtures) and binders on the physical, first of all, the thermodynamic, characteristics of HAs taking the structural factor into account. As was already mentioned, the HA structures are multiphase compounds, and the carbide phase is seen as consisting of maximum four individual carbides or their hard solutions; a binder can also be multiphase, especially if it is based on steel. Thus, the HA surface can be presented as "spotted", and the "spots" are characterized by different output work of electrons. As was shown earlier [1], this parameter determines the value and the sign of the Thermo-EMF of an element. Thus, the composition of the carbide and the binding phases should be chosen so that the difference of the absolute Thermo-EMF values of the carbide phase and a binder were as low as possible (ideally $\Delta E = 0$) or a binder were electropositive toward the carbide phase.

At the Department of Tool Production of Don State Technical University, more than 40 alloys of pure metals have been developed as potential binders for HAs. Of the tested HA binders, the most electropositive toward carbides or those having the lowest Thermo-EMF by the absolute value were chosen. The experimental compositions of hard alloys based on WC have three types of binders: Co–Mo, Co–Fe–Cu, and Co–Mo–Ti, which differ by the percent ratio of these phases (Table 1).

The alloy samples were exposed to standard resistance tests at turning of steel 45 as compared to the basic VK8 alloy. In Fig. 4, the data on the calculated entropy values of HAs of the three composition groups, their resistance coefficients k_r, and the mechanical properties are shown. From the comparison of the curves of the alloys entropy dynamics and the resistance coefficients, it can be seen that the high-

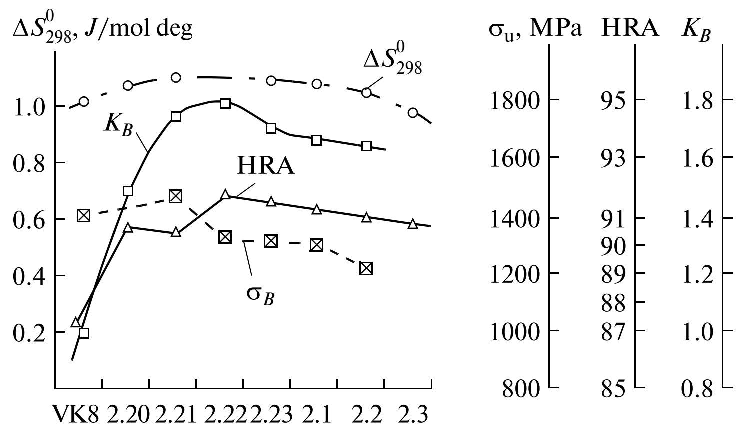

Fig. 4. The characteristics of the experimental hard alloys with a modified cobalt binder.

est resistance is shown by the alloys 2.20, 2.21, 2.22, and 2.23, which have the highest entropy value. These data agree with each other and prove the conclusions of the analysis of the theoretical dependence for the calculation of the wear rate (the formula (10)): the increase in the HA entropy contributes to the decrease of the wear rate J. It is noteworthy that the alloys 2.3 and 2.24, which have more than 5% Mo in their structure, proved non-efficient and cracked at the first seconds of turning due to their low flexural strength: for the alloy 2.3, $\sigma_b = 668$ MPa, and for the alloy 2.24, $\sigma_b = 646$ MPa.

The efficiency of ECC was also evaluated at longitudinal turning of 12Kh18N10T heat-resistant steel. With flank wear $h_f = 0.4 \times 10^{-3}$ m accepted as a criterion, at each speed in the chosen range ($V = 0.5$, 0.83, 1.25, 1.66, 2.49, and 2.9 m/s), wear curves h_3 depending on the operation time were developed (Fig. 5).

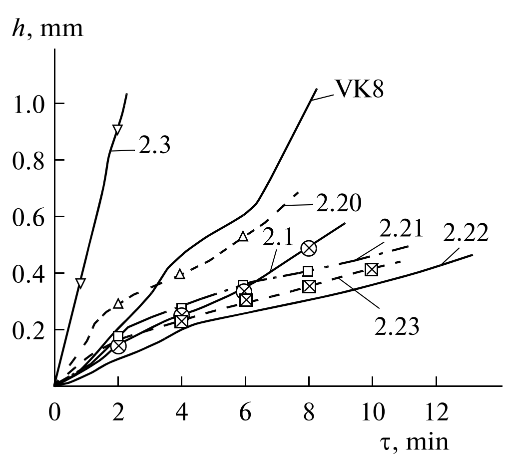

Fig. 5. The wear of plates made of the experimental hard alloys at turning of 12Kh18N10T heat-resistant steel (υ = 1.25 m/s, $t = 0.5 \times 10^{-3}$ m, $S = 0.23 \times 10^{-3}$ m/rev).

As a result, source data for developing the dependences of the resistance of ECC on the cutting speed were obtained. The logarithmic anamorphosis of the dependences $T = f(\upsilon)$ for all the ECC and the VK8 alloy for all the studied compositions have a break point corresponding to the optimal cutting speeds in the range of 0.83–1.25 m/s. The maximum resistance was demonstrated by the alloys 2.22, 2.23, 2.20, 2.21, and 2.1, which demonstrate the resistance 2–2.5 times higher as compared to the plates made of VK8 (like at turning of steel 45). The alloys 2.3 and 2.24 containing more than 5% Mo in a binder proved inefficient due to their increased brittleness; for the alloy 2.3, over the whole range of the cutting speed, a catastrophic wear 1.5 min after the beginning of cutting was observed.

A series of experiments for evaluation of wear rate J by measuring the wear increase of plates from the way of cutting established that, for all the ECC compositions, there is an optimal cutting speed at which wear rate J is minimum (Fig. 6). The ECC compositions (except 2.3 and 2.24) at the optimal cutting speeds show 2.4–3.0 times higher wear resistance than the basic VK8 alloy; at high cutting speeds ($\upsilon > 2.5$ m/s), the alloys 2.22 and 2.23 are still advantageous, as are the alloys 2.21 and 2.1; in the zone of low cutting speeds υ = 0.5–0.8 m/s, as compared to VK8, the alloys 2.1, 2.20, 2.22, and 2.23 are advantageous due to their lower tendency for hardening and adhesion wear and lower outgrowth formation. In addition, the application of ECC with a modified cobalt binder widens the range of optimal cutting speeds (υ_o = 1.25–2.00 m/s).

A probable reason of the improved wear resistance of ECC and widening the range of optimal cutting speeds can be their higher resistance to the oxidative wear. The evaluation of the phase composition of oxide layers after the exposure at 300°C for 30 min proves that the scale mainly consists of the WO_3 and $CoO \cdot WO_3$ oxides. The maximum resistance to gas

Table 2. ECC gas corrosion resistance

Alloy	Weight gain of samples at heating until the temperature, g		Wear rate, $J \times 10^{-7}$, ($V = 1.25$ m/s)	Entropy S^0_{298}, J/mol °C
	300°C	500°C		
VK8	1×10^{-4}	23×10^{-3}	18.0	35.00
2.1	5×10^{-4}	1.7×10^{-3}	9.0	35.12
2.19	1×10^{-4}	1.1×10^{-3}	6.0	35.16
2.21	4×10^{-4}	1.0×10^{-3}	0.5	35.18
2.22	2×10^{-4}	0.6×10^{-3}	0.45	35.26

corrosion at $T = 500$°C is shown by the alloys 2.1, 2.19, 2.21, and 2.22 (Table 2).

From Table 2, it follows that improved resistance of ECC to the gas corrosion provides an increase of the wear resistance, apparently due to a decrease in the fraction of the oxidative wear, and this parameter is higher in ECCs having higher entropy values.

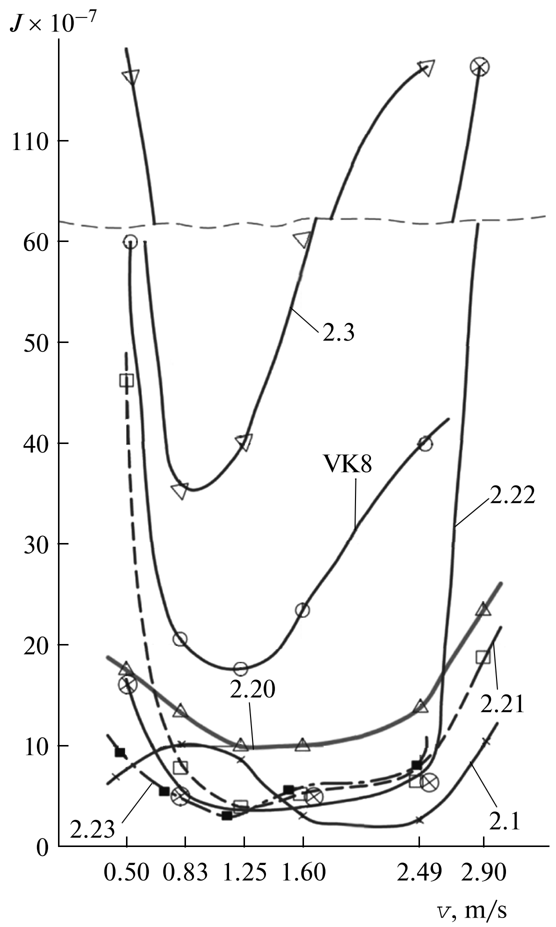

Fig. 6. The wear rate of the experimental hard alloys at turning of 12Kh18N10T steel.

CONCLUSIONS

Theoretical evaluation of the wear rate of CTM as heterogeneous structures showed that, to decrease the wear at friction and cutting, it is necessary to increase their entropy or to decrease their absolute Thermo-EMF. For high-speed steels and hard alloys, a functional relation between the structural characteristic of the materials (Thermo-EMF) and the entropy was established.

Experimental hard alloys (ECCs) were developed based on the VK8 alloy with a modified cobalt binder, the Thermo-EMF of which toward the carbide phase is positive and the entropy of which is higher than that of the VK8 alloy, which increases their electrochemical stability.

The effectiveness of the use of a high-entropy HA instead of VK8 alloy was experimentally proven: in treatment of steel 45, their resistance in the zone of the optimal speed increases by up to two times, while for 12Kh18N10T heat-resistant steel the increase is 2.4–3.0 times. A probable reason of the increase of the resistance is a high scale resistance of these alloys and the decrease of the oxidative wear.

To improve the wear resistance of CTMs, it is advisable to increase the entropy in their development by varying the composition of both a binder and the carbide phase.

ACKNOWLEDGMENTS

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NOTATION

CTMs	cutting tool materials
HSSs	high-speed steels
ECCs	experimental cemented carbide hard alloys
HAs	hard alloys
EMF	electromotive force

$P[S]$	entropy production
$\Phi[S]$	entropy flow
dV	volume
$d\tau$	time
υ	friction (cutting) speed
h	linear wear
C_p	heat capacity of TM
T	absolute temperature, K
G_i, H_i, and S_i	Gibbs potential, enthalpy, and entropy of formation of *i*th grade oxide
$\frac{dh_i}{d\tau}$	film growth rate for *i*th grade oxide
η_i	fraction of *i*th oxide in oxide layer
F	Faraday constant
M_i	molar weight
γ_i	density
m_i	number of moles
n_i	valence
ρ_i	resistivity
η_i	mass fraction of *i*th grade oxide
k	Boltzmann constant
e	electron charge
W_F	Fermi energy
N_9	density of electron states in the valence band
H	hardness of a material
b	dimensional coefficient of proportionality, m^2/N
E	binding energy
K_{H1} and K_{H2}	varying kinetic coefficients, GPa
K_f	energy of atoms in the free state

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