Physical and chemical processes in the contact zone when processing metals with a blade tool

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Abstract. The article considers the theory of radical-chain mechanism of formation of lubricant films on tribos-connected metal surfaces when cutting metals. It is shown that chemically active particles formed by the destruction of molecules of lubricant and cooling technological means (LCTM) in the contact zone play a decisive role in the synthesis of lubricants. Mechanisms of formation of chemically active atoms, radicals and ions and factors influencing these processes are presented. It has been established that the main of them can include the temperatures that occur in the contact zone, exoelectrons issued by freshly opened metal surfaces, as well as the catalytic effect of the juvenile surfaces themselves. On this basis, various methods of pre-activation of LCTM are proposed in order to obtain the required quantity and type of chemically active particles.

1. Introduction

This scientific work is dedicated to the memory of Latyshev Vladimir Nikolaevich (28.09.1929 – 03.05.2018), who was a major Soviet and Russian scientist, a public figure, an honorary figure of science and technology of the Russian Federation, a Doctor of Technical Sciences, a Professor, a Rector of Ivanovo State University (1974–2000), a creator of the scientific school for the study of lubricant and cooling technological means in the processing of materials cutting, an author of more than 400 scientific articles, monographs and educational manuals, more than 100 author's certificates and patents for inventions. The positive solution to the problem of synthesis and use of lubricant and cooling technological means (LCTM) is largely determined by the level of theoretical researches in this area. The cutting process imposes specific features on the mechanism of action of the external environment, which is determined, first of all, by high unit pressures and temperatures on the working surfaces of the cutting tool, the presence of juvenile non-oxidized metal surfaces, the flow of chemical reactions and electrochemical processes on the edge of the section of the tool-processed material, the high deformation rate of the cut metal and the short contact time between the chips and the machined surface and the cutter.

2. Results and discussion

The results of experimental and theoretical studies allow us to describe the mechanism of the lubricating action of the environment when cutting metals based on modern representations of physics, chemistry and mechanics of the blade cutting process and to outline ways of synthesis of effective LCTM. This is
primarily due to the study of a complex and, at the same time, unique mechanism of interaction of the external environment with metal surfaces. The main reason for the specific mechanism of action of the environment in metal cutting is the presence in the contact area of the cutter with the processed material of physically pure neo-oxidized surfaces, extremely active chemically, high temperatures and pressures, as well as unusual catalytic properties of juvenile surfaces, formed when metal bonds break.

This surface comes into contact while cutting either with natural environment (air with different physical and chemical properties) or with artificially brought external environments. At the same time, the interaction of the juvenile surface with components of the external environment can be characterized by various sorption processes, first of all, physical and chemical adsorption, as well as absorption. The characteristics of the contact zone are such that the molecules of the external environment, undergoing destruction, break down into atoms, ions and radicals, that are also chemically active particles and are able to enter into chemical interaction with freshly opened metal surfaces.

Taking into account the above-mentioned features of the contact zone, it is likely that at the contact boundary the tool-processed material will be dominated by the formation of new chemical compounds as a result of chemical reactions between juvenile metal surfaces and components of the external environment, i.e. iono- and chemosorption processes. At the same time, the physical and the chemical activity of these surfaces is such that with their participation it is possible to initiate the course of chemical reactions with components of the external environment, the thermodynamic possibility of which is unlikely under normal conditions. It is known that the number of product reactions in the cutting zone is more than 2.5 times higher than the same figure obtained under the same conditions but outside the contact zone (figure 1).

Thus, when considering the mechanism of lubricating chemical action LCTM should take into account the process of formation of surface radicals, the speed of chemical reactions, catalytic phenomena on micro-sites of incisor and shavings, the consequences of chemical reactions (film formation, the warmth of exothermic effects of reactions).

In accordance with modern views on the mechanism of heterogeneous chemical reactions, the interaction of metal surfaces with the external environment depends on a number of conditions, including the electronic structure of reacting molecules, the presence of unpaired electrons, the type and strength of bonds in molecules of various LCTM compounds. It is known that the chemical reaction and the process of the formation of lubricants on the contact sites of the incisor and shavings includes several successive stages: the impact of molecules and atoms with electrons, the formation of surface radicals, the period of their existence in a free active state, the elementary act of chemical reaction. Consider the factors that influence the kinetics of chemical reactions in metal cutting.

The first and necessary condition for chemical reactions is the collision of molecules or atoms of reacting components with juvenile surfaces. When cutting metals at high speeds, atoms or LCTM molecules are in a gas-like state, and metal surfaces are in an elastoplastic, chemically activated state. Chemical reactions between them will be possible if the colliding molecules or atoms have an excess of free energy.
It is known from physical chemistry that the smaller the difference in the level of direct and reverse reactions is, that is, the less energy activation will be and the greater the proportion of molecules and atoms will collide with the surface and between each other, the higher the rate of chemical reaction. Thus, one of the means of intensification of chemical reactions in friction and cutting is the selection of reacting substances that have low activation energy.

The speed of the chemical reaction can be increased by catalysts. Catalysts reduce the energy of activation of chemical reactions and thus contribute to an increase in the number of active molecules and atoms. Metals and their oxides, as well as carbides, borids, nitrids and other compounds can be used as catalysts. In addition to positive catalysts that accelerate the speed of chemical reactions, there are also negative catalysts, inhibitors, that slow down the speed of chemical reactions.

The catalytic effect of various metals on the decomposition process of LCTM and oils has been shown in a number of our earlier researches. Thus, the method of differential-thermal analysis (DTA) has established that when LCTM are heated, these substances during decomposition form intermediate chemical compounds, which differ in their properties from the original components. Metals present during thermal decomposition of LCTM in the cutting process can have a significant impact on the physical and chemical properties of the formed intermediate substances, on the speed of their formation (figure 2).

![Figure 2. DTA results on the interaction of crystalline iodine with various metals: a) VT1-0; b) Amg-2; c) 12H18N10T; d) U10A.](image)
Calculating the order of chemical reactions in the interaction of different metals with microcapsulated LCTM showed that for all peaks on DTA-curves studied samples of microcapsules (MC, MMC) and their constituent parts (Table 1) this indicator turned out to be fractional. Here: microcapsule – particle LCTM, which has a size of 0.1–50 microns, surrounded by an impenetrable shell, MC – microcapsule, MMC – microcapsule, which has a magnetic and sensitive component. Therefore, all processes with thermal destruction of LCTM occur not sequentially but in complex parallel paths. Moreover, the increase in the heating temperature often complicated the nature of destructive processes, which indicates the reactions that occur both inside the polymer shells of microcapsules, and between the polymer and the nucleus, polymer and metal, the nucleus and metal, as well as all components with the atmosphere of the surrounding air.

**Table 1.** Estimated order of chemical reactions of MC and their individual components in the presence of different metals.

| Metal sample | MC shells | MC magnetic shells | MCI-40A | MMC-50I-40A | MC CCl₄ | MMC-50 CCl₄ |
|--------------|-----------|--------------------|--------|-------------|---------|-------------|
| 1            | 2         | 3                  | 4      | 5           | 6       | 7           |
| –            | 0.806     | 0.714              | 1.096  | 0.545       | 1.430   | 0.548       | 0.870       | 0.596       |
| MVP          | 0.708     | 1.333              | 0.625  |             |         |             |             |             |
| 12H18N10T    | 1.275     | 1.211              | 0.1031 | 1.235       | 1.529   | –           | –           | 0.595       | 0.676       |
| Steel 45     | 1.080     | 1.714              | 1.367  | 0.486       | 1.140   | –           | –           |             |             |
| VT-6         | 1.048     | 1.167              | 1.385  | 1.478       | 3.640   | 1.265       | –           | –           |             |
| R-9          | 0.923     | 1.973              | 0.560  | 0.720       | 2.333   | 0.755       | 0.537       | 1.682       |

* the fraction numerator indicates the order of the low-temperature peak reaction on the DTA curve, in the denominator – of the high-temperature.

Thus, the task of fusion of effective LCTM for mechanical processing of metals depends to a large extent on the intermediate substances generated during the thermal decomposition of LCTM, free radicals and applicable catalysts. In most cases, the cutting tool or the material being processed represents a complex set of catalysts, including oxides, carbides, nitrides and other compounds.

Aggregate state of the applied environment and temperature is the other factor on which the activity of chemical reactions depends. The gas and gas-shaped atmosphere contributes to the intensification of chemical processes, as they contain a greater number of potential active molecules and radicals involved in interaction with contact surfaces of metals. In the study of the effectiveness of the sprayed LCTM, it has been established that the specific surface of the air-liquid high-dispersal mixture with the size of the LCTM particles of 12–25 microns is approximately 700 times greater than the specific surface of the unsprayed liquid (1 g of sprayed liquid has the same surface as 27 kg of unsprayed). Thus, the gas and the locomotive atmospheres are the most favorable in terms of acceleration and intensity of chemical reactions and the formation of protective films.

The temperature effect on the speed of chemical reactions is also very large. In cutting conditions, due to the uneven distribution of temperature by width and length of shaving contact with the incisor,
as well as the presence of zones of local flares, chemical reactions can occur on separate areas of the incisor. This was recorded in our studies when cutting steel 45 using different LCTM. In most cases, the temperature in the cutting zone is almost always sufficient for thermal destruction of technological means and chemical reactions. As a result of uneven distribution of temperature and specific pressures along the length of contact on different parts of the front surface, different compounds are formed: oxides, sulfides, chlorides, metal diiodides. X-ray analysis and electronic microscopic studies have shown that in the areas directly adjacent to the cutting edge, films of sulfides, chlorides, diiodides are formed, and metal oxides – at the points of separation of shavings from the cutter (figure 3).

The next factor to consider when compiling LCTM formulations for cutting metals is the relationship between cutting speed and chemical reaction rate. The deformation speed while cutting metals is $10^5 - 10^6 \text{ sec}^{-1}$ in average, and the rate of impact of reacting particles or the time of the elementary act of chemical reaction is $10^{18} \text{ sec}^{-1}$; the speed of oxidative processes is even higher. From the comparison it follows that the rate of chemical reaction is much higher than the rate of deformation when cutting metals, that is, the act of elementary chemical reaction has time to occur.

An extremely important factor determining the chemical activity of LCTM is the ability of its molecules to break down into reaction particles – atoms, ions and radicals, as only with continuous generation of chemically active particles is possibly branching, continuous, chemical reaction, and, therefore, maintaining on the contact sites stable chemical films, sufficient for a tangible impact on the tribological environment. The process of surface radicals in cutting is carried out in a variety of ways, including by breaking the bonds in the LCTM molecule with free valences, as a result of interaction with electrons emitted by juvenile surfaces, with environment molecules, thermal pyrolysis, etc.

In the first case, the free valences formed on the surface of the shavings are similar to free radicals, as they can interact with LCTM molecules. At the same time, the LCTM molecule, which has a small activation energy, is torn apart and the resulting radical saturates valence, i.e. chemosorbs. A distinctive feature of the cutting process is that it continuously forms free valences, that is, generates "superficial" radicals. As a result of the chemical interaction between radicals and LCTM atoms with the juvenile surface, its unbalanced force field (free valence) is extinguished, a lubricated chemical film is formed and additional heat from the exothermal reaction is released.

The described case of radical formation seems to be typical in cutting with low speeds of structural steels, as well as in the processing of light and colored alloys. The process of generating radicals depends in this case on the force field of the juvenile surface, the ratio of geometric parameters of crystal lattice of deformable metal and the size of LCTM molecules.

The second case of destruction of neutral molecules of the external environment on atoms and radicals is carried out as a result of their interaction with electrons or other energy particles, including
quantums of light, issued by juvenile surfaces as a result of thermoelectronic or photoelectronic emission. For example, as a result of the interaction of exoelectrons with neutral LCTM molecules, energy is transferred from electrons to LCTM molecules, activation of the latter and their disintegration with the formation of radicals and radical groups, the activity of which is caused by the presence of an electron with uncompensated spin on the outer shell. In general, such processes in the formation of peroxide radicals can be described as follows

$$H_2O + hv(e) \rightarrow H_2O^* + hv(e)$$

$$↓$$

$$H. + OH.$$

In the interaction of hydroxyl radicals, hydrogen peroxide is formed, the effect of which while cutting is caused by the ability to release active oxygen

$$OH. + OH. \rightarrow H_2O_2$$

$$2H_2O_2 \rightarrow 2H_2O^* + O_2$$

$$O_2 + hv(e) \rightarrow O_2^* + hv(e)$$

$$↓$$

$$O. + O.$$ where $hv(e)$ is the energy (electron) emitted by the juvenile surface; $H_2O^*$, $O^*$ - excited water and oxygen molecules; $H.$, $OH.$, $O.$ – chemical radicals.

Temperature, light irradiation (electromagnetic waves), the absolute magnitude of the electrons' output and its energy are the main characteristics determining the intensity of the emission of electrons, and therefore the process of generating radicals. The electron output is several electron-volts and is minimal for metals such as potassium, sodium, lithium, and for stainless and heat-resistant materials containing, for example, nickel output of the electron 2–2.5 times higher.

The energy of the electron is one of the main criteria for the formation of radicals. The results of theoretical and experimental studies have shown that electrons simulated from the juvenile surface of metal accelerate in the electrostatic field generated by friction. At the same time, their energy increases and becomes enough to destroy neutral lubricant molecules into radicals. B.I. Kostetsky and his collaborators, using the electronic paramagnetic resonance method (EPR), found that complex organic radicals are formed when metals are frictioned in organic lubricants. Studies have established (figure 4) that an increase in the spectrum of the reference was observed during the joint heating of nitroxil and magnetic microcapsules (MMK) This indicated a decrease in the free bonds of nitroxyl as a result of chemical reactions. In the study of the mixture of nitroxyl-ozone-containing magnetic microcapsules (OMMK) spectrum was a triplet characteristic of a low concentration of nitroxyl radicals, due to the increased number of radicals, released during thermodestruction of OMMK.

Thus, the less electron output is and the higher its energy is, the higher the probability of LCTM molecule decay with the formation of radical and chemical film synthesis.

The third way to get radicals is to heat the components of LCTM to high temperatures, i.e. in thermal pyrolysis. This method of obtaining radicals is especially convenient for the decomposition of organic liquids: oils, hydrocarbon mixtures, surface-active substances. In this case, when heated to high temperatures, the vibrational movement of atoms in molecules increases and at high enough amplitudes of vibrations the bonds between atoms are broken. This case of radical generation is typical, perhaps when cutting heat-resistant and stainless steels, as well as cutting at higher speeds.

Once the radicals have formed, two things must be taken into account: their life expectancy and the type of radical. Researches carried out by Science Schools of N.N. Semenova and V.II. Kondratieva show that the life expectancy of radicals is different and depends on the temperature, type of radical, surrounding atmosphere and compiles on average 0.01–0.4 seconds for hydroxyl, which is several
orders of magnitude longer than the time needed for the formation of a chemical compound. Depending on what type of compounds break down into atoms and radicals, the latter can be of two types: simple and complex. Inorganic aquatic CF (cutting fluid is a special case of LCTM) form simple radicals when cutting (monoradicals, biradicals and trnradicals) and organic lubricants form complex radicals. Simple radicals studied in this work include chlorine, sulfur, phosphorus, iodine, and complex - radicals formed in pyrolysis surfactant, oils, microcapsules.

| Structural formula nitroxy1 | Spectra EPR |
|-----------------------------|-------------|
|                             | Nitroxy1    | Nitroxy1 and MMK | Nitroxy1 and OMMK |
| ![Structural formula nitroxy1](image1) | ![Spectra EPR nitroxy1](image2) | ![Spectra EPR nitroxy1 and MMK](image3) | ![Spectra EPR nitroxy1 and OMMK](image4) |

**Figure 4.** The ability of ozone-containing magnetic microcapsules to initiate radical chain reactions: a – a structural formula of nitroxy1; b-d – spectrums of electronic paramagnetic resonance: b – nitroxy1; c – nitroxy1 with magnetic microcapsules: d – nitroxy1 and ozone-containing magnetic microcapsules.

Chemical reactions that occur when cutting using CF and gas are, in most cases, spontaneous and radically chained. This has been shown in our works by the example of spontaneous formation and decomposition of hydrogen peroxide when cutting alloys on an iron basis, autocatalytic reactions of nitrogen oxides with titanium and solid alloys, chemical interaction of iodine and sulfur vapors respectively with titanium and nickel alloys.

The study of the mechanism of chemical interaction of radicals of the environment with metal surfaces, first of all, oxygen and water with juvenile shavings and cutters, allows to outline ways of purposeful regulation of chemical reactions in cutting, the possibility of creating defensive films at the expense of these reactions on the cutting tool and shavings. Here are some things to consider:

First, it is necessary to keep in mind the mechanism by which the lubricant molecule spontaneously collapses in the cutting zone on atoms and radicals of free "superficial" valence, electron or electromagnetic wave, thermal pyrolysis. The first mechanism of action (destruction of free valencies) is mainly typical in cutting with low speeds (no thermostimulated emission of electrons), and the destruction of molecules of lubricant by electron (quantum of light) or pyrolysis occurs at medium to high cutting speeds. The destruction of LCTM molecules by electrons is most intense when cutting steel with a ferritic structure (maximum emission) and minimal when cutting heat-resistant alloys that have a solid solution structure (minimum emission). In the latter case, the breakdown of LCTM molecules into radicals is mainly due to thermal pyrolysis.

Secondly, it is necessary to rationally select components of LCTM, keeping in mind, first of all, two parameters of the molecule: the type and strength of the connection between atoms. These statistical characteristics can be judged in advance on the possibility of the formation of free radicals and atoms and their reactionary ability. Compounds of type - 0 - 0 -, I-I, NO, CN are the most favorable from the point of view of chemical connections.
Thirdly, consider the possibility of activating the necessary (positive) chemical reactions by various methods: ionization of the air-liquid mixture by electric field, activation of the gas mixture by light of optimal wavelength, activation of reactions by ultrasonic vibrations, electric current, etc. (figure 5). The correct selection of environmental components and the appropriate activation method can trigger the necessary chemical reaction and obtain chemical compounds with predetermined properties on tribo-conjugated metal surfaces.

![Diagram of Activation Methods](image)

**Figure 5.** A physical model that describes activation.

In our previously published works, the influence of various films and individual chemical compounds (oxides, sulfides, chlorides, iodides, etc.) on friction and wearout of the cutting instrument, established a correlation between the type of chemical compound and the friction factor, found out the effect of protective films on fluctuations in the forces and temperature of the cutting, identified the optimal components of LCTM for various contact pairs of metals.

The theory of radical-chain mechanism of lubricating films formation is tested in the cutting of metals with cooling – lubricant with various substances, including atomic oxygen, peroxide compounds, ozone, surface-active substances with additives of haloid compounds, other compounds [1–3]. The mechanism of spontaneous formation of hydrogen peroxide in metal cutting has been studied in detail, the conditions of its formation and decomposition have been revealed, catalytic processes of hydrogen peroxide decay on juvenile surfaces of various metals have been studied and stabilizers of $H_2O_2$ decomposition have been proposed in order to maintain some excess of CF. From the point of view of the mechanism of radical-chain reactions, a theoretical explanation of the effective action of sprayed and ionized CF and oils when cutting some steels and alloys is given.

Based on experimental data, the first approximation established the connection of the internal structure of the environmental molecules with the parameters of the cutting process: the durability of the cutters and the cutting temperature (figure 6).
Figure 6. Effect of the bond length in the LCTM molecule on the instrument durability.

For various chemical compounds formed on heavily loaded contact sites of the cutter and shavings, a crystal and chemical parameter is calculated, which determines the ease of sliding of interatomic planes and is actually a characteristic of frictional properties of films. The resulting experimental data allowed in general form to draw up a scheme of LCTM action during the cutting process, to assess the impact of their individual species (oxidizers, emulsions, oils, chemically active liquids, etc.) on the durability of fast-cutting and solid-alloy instruments (figure 7).

Figure 7. A generalized chart of the impact of LCTM on the durability of cutting tools: 1 – oxidized oils with high pressure additives, 2 – oxygen-containing emulsions, 3 – mineral oils, 4 – oil emulsions, 5 – solid lubricants (MoS₂, S, C), 6 – oxygen-containing LC (O, O₂, H₂O₂), 7 – LC with chemically active radicals (S, P, J, Cl), 8 – diffuse environments.
In general, the theory presented in this paper about the lubricating chemical action of the environment in metal cutting is in following: the successive stages of the mechanism of destruction of cutting fluid molecules into atoms and radicals have been revealed, a radical-chain mechanism of chemical reactions in cutting (on the example of hydrogen peroxide), a general scheme of selection of components of cutting fluid (based on the internal structure of molecules and atoms) is established and ways of activating and regulating the necessary chemical reactions are outlined, followed by the formation of protective films on the contact surfaces of the cutters and shavings.

3. Conclusion
The obtained results provide a number of considerations about the scientific and technical basis for the synthesis of effective lubricants and cooling environments.

1) In the process of cutting lubricant functions are performed not by the original components of LCTM, but by intermediate reaction particles (atoms, ions and radicals), formed by the interaction of neutral molecules with the juvenile surface. At the same time, the development of a radical-chain reaction requires a small concentration of reactionary particles. Therefore, when formulating LCTM should assume that its basis should be a neutral liquid (for performing transport functions, cooling, detergent action) and a small percentage of reaction additive. As it was established in this work, peroxide and inorganic oxidizers can be such additives for iron alloys, in the processing of titanium alloys – iodine additives, for nickel alloys sulphur additives are optimal.

2) The introduction of reaction additives in LCTM should not always be carried out during the process of its manufacture. For example, the half-life of such a reactionary additive as benzoyl peroxide at 95 °C is 1 hour, so it can break down into radicals even before cutting. Similarly, ozone and hydrogen peroxide have high rate of decomposition in water and water cutting fluids, hence zoning of cutting fluid and dissolution of peroxide is advisable to produce immediately before the use of cutting fluid, i.e. in the workplace.

Broming, chlorinating, iodation and sulphation of technological means can be done in advance, as the lubricating functions of these additives are manifested at elevated temperatures and in contact with juvenile metal surfaces. When introducing such additives into the LCTM, it is necessary that they are organically linked to the molecules of the main substance and they aren’t a mechanical impurity.

3) Synthesized LCTM can be activated to quickly decompose their molecules in the cutting area into chemically active particles. These methods include different methods of physical, chemical and physicochemical effects (see figure 4).

4) LCTM can move from one aggregate state to another when cutting. Studies show that the effectiveness of lubricating chemical action of the environment depends on the intensity of the molecule's decay into radicals and the speed at which they move to the juvenile surface. As a rule, in viscous, dense liquids, the disintegration of molecules into radicals is difficult, as the latter can not move away from each other, that is, their recombination occurs. In this regard, the gas or locomotive atmosphere is more favorable for chemical radical reactions, because the rate of movement of free radicals in gases is 160 times higher than in liquids.

Thus, in view of the foregoing, the following indicative scheme for developing effective LCTM can be offered. A contact pair of metals is set – a contact cutting tool and a processed part (chemical composition, physical-mechanical and chemical properties, changes in properties under the influence of temperature). Next, they determine the work of the electron output, its possible energy, the estimated temperature range of the technology, the ratio of geometric parameters of the crystal lattice of the processed metal and the parameters of the internal structure of LCTM. The original components of the LCTM are installed – the base and additive responsible for the lubricating action. The basic principles of choosing such an additive are following: the distance between atoms in the molecule, the strength of
the connection between them, the possibility of destruction on radicals due to electronic impact, pyrolysis, the force field of the juvenile surface, chemical activity in relation to the material being processed. In conclusion the LCTM test takes place: the fundamental possibility of chemical reactions, critical temperatures, heat reaction effect, i.e. the behavior of instrumental and processed materials in relation to the components of LCTM is assessed by its chemical activity, as well as the change in the tribological situation of the contact zone is assessed.

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