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MAGNETIC STIMULATION OF CHEMICAL REACTIONS IN COAL

Purpose. To identify a mechanism in terms of which a signal of weak magnetic field is transformed into response of some components of nanostructure of a carbonaceous matter with further formation of chemical bonds.

Methodology. Physical and mathematical modelling procedures of elementary chemical acts have been applied. Regularities of quantum mechanics and dynamics of chemical bonds were also used in addition to a magnetic isotope theory, a diffusion theory of recombination of radical pairs taking into consideration triplet-singlet transit, and nuclear-spin selectivity of chemical reactions.

Findings. The physical mechanism of the magnetic scenario of interradsial reactions is considered from the viewpoint of their energy stimulation through magnetic fields, i.e. the idea has been implemented according to which the number of radical pairs, able to be recombined into stable molecules, increases significantly if the weak magnetic field exercises certain influence. In addition to stimulation of interradsial reactions, the magnetic field impact on *organic coal mass-radicals* results in stabilization of carbonaceous structures with regular atomic arrangement (being two-dimensional matrices, chains etc.) and their increase.

Originality. A physical model of structural and phase changes in coal, depending upon the effect by external weak magnetic field, has been developed. It has been shown that the weak magnetic field signal, being incomparably smaller energetically to compare with the energy of thermal molecular motion, can initiate triplet-singlet transitions, i.e. transform radicals into reactive state. A mechanism has been proposed to form chemical bonds between movable radicals and non-complete bonds of atoms at the surfaces of solid phases of carbonaceous matter.

Practical value. Implementation of the obtained results, concerning magnetic coal processing, may be connected with the development of a new procedure to avoid explosive conditions in coal seams. Progress in the field of magnetic processing should involve studies concerning calculation and selection of coal processing parameters (i.e. magnetic field density, frequency, energy, and a processing period) to control efficiently the chemical reactions in the carbonaceous matter. A substantiation degree of coal processing will influence greatly both efficiency of chemical processes and expediency of practical use of the results.

Keywords: coal, Lande factor, radical pair, spin, splitting, singlet state, electron, unstable balance state

Introduction. Influence of weak electric and magnetic fields on microstructure of carbonaceous matter stimulates demonstration of numerous unexpected physicochemical effects. For instance, coal samples, processed by electric field, have obtained electret potential whose charge relaxation took place during several weeks [1]. A characteristic feature of each fossil fuel grade is concentration of defects of crystalline structure and internal specific structure, i.e. increase in coal reactivity. After coal samples were processed using electric field, all their parts of the initial range became smaller [2, 3]. The first methane-saturated coal samples have demonstrated an increase in methane desorption interval along with an increase in a carbonization degree; the interval turns out to be shorter for coal processed by means of electric field. After coal processing by electric field, Raman spectra (RS) are split into six components; at the same time, RS of the initial coal (starting from the initial mineral of carbonization rank up to anthracite) are split maximally into five components; the fact coincides with RS data of explosive coal [1]. As a result, electric field effect on the fragmented coal sample, RS has no fundamental differences to compare with the explosive coal spectra [4, 5]. When impacted by a weak electric field (coal sample heating up to 318–320 K), the coal generates methane as well as other hydrocarbon gases inclusive of certain heavy hydrocarbons, argon, and carbon dioxin [2]. Weak thermal and electric actions factor into significant changes in coal microstructure, which may heavily influence the expected effect of harmful gas decrease from the improved blasting procedures [6], use of techniques preventing from manifestations of gas-dynamic phenomena [7], etc.

Statement of the problem. The experiments and theoretical studies have helped understand that the magnetic fields, being of low density, influence biological systems [8, 9]; in this regard, over the last 30 years, such a branch of knowledge as

magnetic biology has experienced its tremendous upgrowth [10]. Interest in the effects of the magnetic field influence on biological systems looks logical since the reason of biological structure destabilization through a weak magnetic field is not obvious. Such systems have no magnetic receptor units able to respond to the magnetic field; there are only biomagnetite particles in some bacteria [11]. It is possible to assume that the Earth's magnetic field, being one of the external physical factors, influences heavily both living organisms and physicochemical processes of rock forming and carbonization.

In the 1970s, bright experimental results were obtained as for magnetic and spin effects in radical chemical reactions. In 1967, a phenomenon of nuclear spin polarization during chemical transformation was discovered. In 1972, experiments helped discover the external magnetic field impact on the output of products of radical pair recombination. In 1976, a magnetic isotopic effect was discovered. All the results could be explained, relying upon consideration of a selection rule in terms of a spin to recombine two radicals and the detailed description of dynamics of the radical pair spins.

According to the data of studies represented in papers by I. Sokolyk and Ye. Frankevych (1973); R. Sahdeiev and A. Buchachenko (1974); K. Salikhov, et al. (1975, 1977); A. Buchachenko, et al. (1978), it becomes obvious that within the weak magnetic fields, both velocity and completeness of chemical reactions between radicals in gases and liquid increase significantly. The identified regularities have been involved while interpreting experimental results obtained during studies on the weak magnetic field influence on physicochemical processes as well as coal characteristics [12].

It should be mentioned that coal processing by means of a weak pulsating magnetic field (up to 50 Oe and 50 Hz frequency) as well as a weak electric field [13] was used in the experiments for the first time. An increase in C/H, being a phenomenon of continuous transition of the analysed samples to coal with the characteristics of the following higher carbonization

degree, has been determined [12, 14]. Essentially, the effect simulates action by one of physical factors (i. e. natural magnetic field) on a carbonization process expressed among other things by the increased content of crystalline coal component, decreased sorption surface, and increased C/H ratio as well as thermal effect during combustion [15] which coincides smoothly with the characteristics of the initial fossil coal samples [16, 17].

It is common knowledge that an individual act of molecular transformation needs the thermal-scale energy ($\sim kT$) [18, 19]. The energy is several orders greater than that of a weak magnetic field. In other words, field energy cannot initiate any chemical reaction. Nevertheless, the experimental results support the opposite idea, which is the classic scientific contradiction. It has been proved convincingly that if an external magnetic field is not available, no other physical effect can initiate the system of effects, shown by [1, 2, 12–14], in the carbonaceous matter. An interesting situation arises when impacts by a weak magnetic field cannot be suppressed through incompatible competitive thermal disturbances.

In terms of carbonates [22], rocks and fossil coal [20, 21], and minerals after mechanical activation correspond to the unstable balance state similarly to outburst coal [23, 24]; hence, there are no objects in such a transition state which could be characterized by average energy of thermal motion kT (Yu. Butiagin, 1984). Laws of energetic and motion are not able to foresee phenomena for such systems and describe them.

The analysis of papers in the sphere of magnetic field influence on chemical reactions in solutions and solid polymers supports the idea that studies on potential transformations in carbonaceous matter have never been carried out.

Thus, the purpose is to propose a physicochemical mechanism in terms of which a weak magnetic field signal is transformed into response of certain nanostructure components of a carbonaceous matter with further formation of chemical bonds.

Results and their discussion. Vector atomic model. The obtained experimental results on magnetic processing of fossil coal samples [1, 12–14] may be explained just from the viewpoint of spin effects. Probably, chemical reactions take place at open surfaces of coal particles and at the closed surfaces of pores and fractures. Such an assumption is provoked first of all relying upon the consideration of necessary even if restricted movability of reaction systems.

Studies focus on the use of a weak magnetic fields. Why do weak fields favour acceleration of chemical reactions? The matter is that recombination (interaction) of two molecules is possible only if spin restriction is not available. Generally, a recombination is implemented from S states; in the context of such states, concentration of molecules depends heavily upon the available weak magnetic field at the expense of the complex (abnormal) Zeeman effect being explained by the dependence of an energy level split value on g factor, i. e. the Lande factor [25].

A strong magnetic field factors into Russel-Souder bond breaking. It breaks interaction between the magnetic orbital and spin moments. In such a way, a contact typical for heavy atoms is made if an external magnetic field is non-available. In this case, the radical pair is transferred into a state differing greatly from the resonance one. Thus, if Russel-Souder couplings are available, then Zeeman effect calculation is g-factor determination; in other words, determination of the total magnetic moment projection $\bar{\mu}_j = \bar{\mu}_e + \bar{\mu}_s$ on the direction of magnetic orientation \vec{B} (where $\bar{\mu}_e$ and $\bar{\mu}_s$ are magnetic orbital spin effect, and magnetic spin effect, respectively).

As a whole, the angular momentum consists of momenta of electrons composing the atom. Quantum number l defines a value of the resulting angular momentum. Letter symbols correspond to numerical L values coinciding with those for one electron. In this context, capitals are applied to differentiate the atom moment L from angular moment of a separate electron.

Samuel A. Goudsmit and George E. Uhlenbeck (1925) assumed that an electron has its own angular and magnetic mo-

menta, and $M = \hbar\sqrt{l(l+1)}$ is an angular momentum. A spin momentum is identified through a spin number using $M_s = \hbar\sqrt{s(s+1)}$ formula where s is a spin quantum number. The spin availability arises from Dirak equation, i. e. relativistic wave equation. In such a way, spin is both quantum and relativistic characteristic. M_s cannot be tilted; the number of the projection is $2s+1$. The experiments by Stern result only in two M_s orientations; hence, $2s+1=2$ ($s=1/2$).

Consequently, magnitude of the spin momentum is

$$M_s = \hbar\sqrt{\frac{1}{2} \cdot \frac{3}{2}} = \frac{1}{2}\hbar\sqrt{3},$$

and its projection is $M_{sz} = m_s\hbar$ ($m_s = \pm s = \pm \frac{1}{2}$).

The magnetic momentum-mechanical moment ratio should be equal to a gyromagnetic ratio

$$\frac{\mu}{M} = -\frac{l}{2m_e}.$$

Nevertheless, both compound Zeeman effect and other experimental data indicate the following

$$\frac{\mu_s}{M_s} = -\frac{l}{2m_e},$$

i. e. magnetic-orbital momenta ratio is twice more. It means that the spin has two-fold magnetism. Thus,

$$\mu_s = -\frac{e}{m_e}M_s = -\frac{e\hbar}{m_e}\sqrt{s(s+1)} = -2\mu_B\sqrt{s(s+1)},$$

where $\mu_B = \frac{e\hbar}{2m_e}$ is Bohr magneton.

Magnetic momentum projection on the specified z direction may have $\mu_{sz} = \pm\mu_B$, values; and a proper mechanical moment projection becomes $3/2 \cdot \hbar, -1/2 \cdot \hbar$.

Magnetic momentum μ to mechanical moment M is a gyromagnetic ratio μ/M . Owing to the two-fold magnetism, the gyromagnetic ratio between the complete μ_j and M_j turns out to be a function of quantum numbers L, S , and J . It should be mentioned that L and S numbers identify the ratio between lengths of \vec{M}_L and \vec{M}_S vectors; and quantum number J defines their mutual spatial orientation. Corresponding quantum-and-mechanical calculation provides a formula for magnetic moment of the atom

$$\mu_J = -\mu_B \cdot g\sqrt{J(J+1)}, \quad (1)$$

where $g = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)}$.

Derive the formula using a vector model. Fig. 1 shows $\vec{M}_L, \vec{M}_S, \vec{M}_J$ vectors as well $\bar{\mu}_L, \bar{\mu}_S, \bar{\mu}_J$ vectors corresponding to them. Scales are selected in such a way to represent \vec{M}_L and $\bar{\mu}_L$ as the equal-length sectors. In this context, $\bar{\mu}_S$ vector will be a sector being twice as long as a sector represented by \vec{M}_S vector.

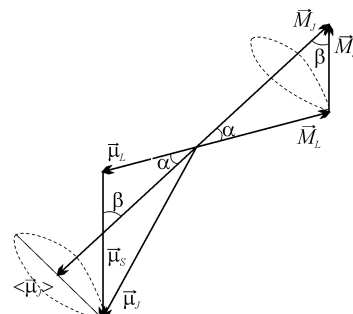


Fig. 1. Vector atomic model

As a result of the doubled magnetism, $\vec{\mu}_j$ vector cannot be collinear with \vec{M}_j vector. \vec{M}_L and \vec{M}_S precess around \vec{M}_j direction. The average value of $\langle \vec{\mu}_j \rangle$ vector will be recorded during rather a long observation period (Fig. 1). $\vec{\mu}_j$ vector projection on \vec{M}_j direction is

$$\mu_j = -\mu_L \cos \alpha - \mu_S \cos \beta, \quad (2)$$

where $\mu_L = \mu_B \sqrt{L(L+1)}$; $\mu_S = \mu_B \sqrt{S(S+1)}$.

To define the resulting magnetic momentum μ_j , determine $\cos \alpha$ and $\cos \beta$ using a cosine theorem.

$\vec{M}_S = \vec{M}_j - \vec{M}_L$ ratio helps identify

$$M_S^2 = M_j^2 + M_L^2 - 2M_j M_L \cos \alpha.$$

From which

$$\cos \alpha = \frac{M_j^2 + M_L^2 - M_S^2}{2M_j M_L} = \frac{J(J+1) + L(L+1) - S(S+1)}{2\sqrt{J(J+1)} \cdot S(S+1)}; \quad (3)$$

$$\cos \beta = \frac{M_j^2 + M_S^2 - M_L^2}{2M_j M_S} = \frac{J(J+1) + S(S+1) - L(L+1)}{2\sqrt{J(J+1)} S(S+1)}. \quad (4)$$

Insert (3 and 4) into (2). Simple transformations factor into

$$\begin{aligned} \mu_j &= -\mu_B \sqrt{J(J+1)} \frac{3J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} = \\ &= -\mu_B \cdot g \sqrt{J(J+1)}. \end{aligned}$$

In such a way, formula (1) has been proved.

Let some atom, having its magnetic momentum, get into the magnetic field with magnetic induction \vec{B} . Let directions of the vectors shape α angle (Fig. 2). Divide \vec{B} vector into two components: \vec{B}_{\parallel} being parallel and \vec{B}_{\perp} being perpendicular to the vector. \vec{B}_{\parallel} component will stipulate the forces either extending or compressing the loop. \vec{B}_{\perp} component will initiate $\vec{N} = [\vec{\mu}_j \vec{B}_{\perp}]$ torque.

\vec{N} vector modulus is equal to $N = \mu_j B \sin \alpha$. Increase in α angle up to $d\alpha$ should involve action being opposite to forces influencing the loop within the magnetic field

$$dA = Nd\alpha = \mu_j B \sin \alpha \cdot d\alpha. \quad (5)$$

While returning to the starting position, the loop may compensate efforts using them for any body. Hence, efforts (5) are required to increase potential atomic energy (i.e. a current loop)

$$dE = \mu_j B \sin \alpha d\alpha. \quad (6)$$

Define $E = -\mu_j B \cos \alpha + \text{const}$ in the process of (6) integration

Assume the state when vector $\vec{\mu}_j \perp \vec{B}$ as a zero electric potential. Then, $\text{const} = 0$.

Thus, the atom obtains extra energy within the magnetic field

$$\Delta E = -\mu_j B \cos \alpha = -\vec{\mu}_j \cdot \vec{B} = -\mu_{jB} \cdot B, \quad (7)$$

$\mu_j \cos \alpha$ value is vector $\vec{\mu}_j$ projection on the \vec{B} field direction, i.e. $\mu_{jB} = \mu_j \cos \alpha$. Hence, under the effect of the magnetic field, a magnetic momentum vector is oriented quantumly in such a way that the angle decreases. Therethrough, the atom

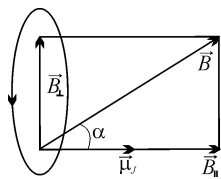


Fig. 2. \vec{B} vector division into components

obtains extra forces determined by means of (7) formula. In other words, the magnetic field has such an orienting action on the electron motion that $\vec{\mu}_j$ vector lines up towards the field action, i.e. towards \vec{B} .

Use a simple substitution for m_j in formula (1) (by analogy with orbital and spin momenta) to have

$$\mu_{jB} = -\mu_B g m_j, \quad (8)$$

where the quantum number m_j obtains the following values $m_j = -J, -J+1, \dots, J-1, J$.

Insertion of (8) into (7) generates

$$\Delta E = \mu_B g B m_j. \quad (9)$$

It follows from the formula that energy level, corresponding to $2S+1L_J$ term, splits into $2J+1$ equidistant sublevels; in this context, the split value depends upon the Lande factor, i.e. upon quantum numbers L, S and J . Consequently, a magnetic field removes degradation of the energy level in terms of quantum number m_j .

Consider a transition between $L=1$ and $L=0$ states ($P \rightarrow S$ transition). If a field is not available, the only line is observed with ω_0 frequency. A field inclusion results in the P level split into three sublevels corresponding to such quantum numbers as $m_j = 1, 0$ and -1 . That is why, the lines with $\omega_0, \omega_0 - \Delta\omega, \omega_0 + \Delta\omega$ frequencies are observed.

Similar situation is typical for more complicated $D \rightarrow P$ transition. Field inclusion factors into the P term split into three sublevels; D term splits into five sublevels. Seemingly, the initial line should split into seven components. However, only three components are available: a line with ω_0 frequency; and two lines symmetrical to it with $\omega_0 - \Delta\omega$ and $\omega_0 + \Delta\omega$ frequencies (Fig. 3).

The abovementioned is explained by the fact that the following is a selection rule for the magnetic quantum number m_j

$$\Delta m_j = 0, \pm 1.$$

ω_0 displacement is either normal or the Lorentz transformation. According to (9) formula, it is

$$\Delta\omega_0 = \frac{\Delta E}{\hbar} = \frac{\mu_B B}{\hbar} = \frac{l}{2m_e} \frac{\hbar B}{\hbar} = \frac{l}{2m_e} B.$$

Split into the three lines, two of which are shaped from the non-transformed line by the amount of the standard ω_0 displacement is the normal Zeeman effect. In this case, $S=0$; hence, $g=1$.

Consequently, the simple Zeeman effect is observed if the initial lines do not have any thin structure; thus, they are singlets. The lines, differing in thin structures, have more than three components; their split value is a rational fracture of normal splitting

$$\Delta\omega = \Delta\omega_0 \cdot \frac{r}{q},$$

where r and q are small whole numbers. Such splitting is anomalous Zeeman effect.

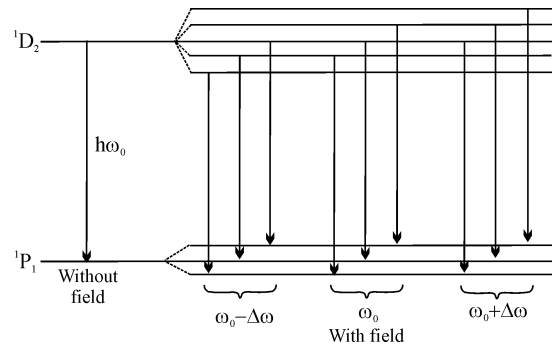


Fig. 3. Split of levels and transitions for ${}^1D_2 \rightarrow {}^1P_1$ line

The anomalous Zeeman effect is explained by splitting dependence upon the Lande factor g , i. e. ultimately upon availability of an electron spin as well as its doubled magnetism.

Fig. 4 demonstrates splitting of levels, and transitions for ${}^2P_{1/2} \rightarrow {}^2S_{1/2}$ lines, permitted by $\Delta m_j = 0, \pm 1$ rule.

In terms of ${}^2S_{1/2}$ level, energy increment is

$$\Delta E' = \mu_B B g' m'_j,$$

where $g' = 2 = 6/3$.

In terms of ${}^2P_{1/2}$ level, energy increment is

$$\Delta E'' = \mu_B B g'' m''_j,$$

where $g'' = 2/3$.

Transformation of the lines is identified through the expression

$$\Delta\omega = \frac{\Delta E'' - \Delta E'}{\hbar} = \frac{\mu_B B}{\hbar} (g'' m''_j - g' m'_j).$$

In Fig. 4, line gaps show $(g'' m''_j - g' m'_j)$ values. It follows from Fig. 4 that there is no initial line in terms of field inclusion. Instead, four lines arise whose displacements, expressed through the normal transformation units, are $-4/3, -2/3, +2/3$ and $+4/3$. The aforementioned can be recorder as follows $\Delta\omega = \Delta\omega_0 [\pm 2/3, \pm 4/3]$.

In the context of more complicated case (i. e. for ${}^1D_2 \rightarrow {}^1P_1$ transition), splitting of energy levels results in six lines whose transformations are

$$\Delta\omega = \Delta\omega_0 [\pm 1/3, \pm 3/2, \pm 5/2].$$

The initial line is not available as well.

All the above mentioned is true for a weak magnetic field.

Within a strong magnetic field, \vec{M}_L and \vec{M}_S decoupling takes place; hence, they are projected on a field direction independently.

A physical mechanism of the magnetic scenario of intraradical responses [26, 27] is considered from the viewpoint of their energy stimulation by magnetic fields. It means that the idea has been implemented experimentally according to which the circle of radical pairs, able to recombine into the stable molecules, expands significantly if a weak magnetic field exerts influence (in terms of biological systems [28, 29]). Magnetic field effect on the *organic rock mass-radicals* system stimulates intraradical responses while resulting in stimulation of carbonaceous structures with a regular atomic arrangement and their increase inclusive of two-dimensional crystals as well as carbon and hydrocarbon chains [1, 2, 12–14].

It has been known since Wigner times that electronic spin of reagents should be preserved in the course of chemical transformations (I. Sokolyk and Ye. Frankevych, 1973; R. Sahdeiev and A. Buchachenko, 1974; K. Salikhov, et al., 1975, 1977; A. Buchachenko, et al. 1978; 2006, 2007, 2010; Ya. Zeldovich, et al., 1988). Thus, recombination of radical pairs (RPs) must depend upon the mutual orientation of the un-

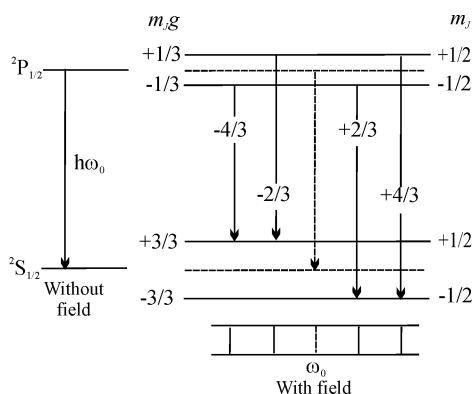


Fig. 4. Split of levels and transitions for line ${}^2P_{1/2} \rightarrow {}^2S_{1/2}$

paired electrons. For a long time, no experiments have demonstrated convincing facts showing a spin state role in the relaxation of RPs; and the problem of recombination dependence upon a spin state did not attract serious attention of researchers. It was predicted at the best that owing to the dependence upon spins, extra factor of the steric multiplier type might be added to the rate constant of a chemical reaction.

The authors apply the term *recombination* in a narrow sense; namely, in a sense of formation of reaction chemical compounds which decomposition initiate development of the chemical reaction products.

Description of magnetic and spin effects in the recombination of radical pairs needs data of twofold nature. It is required to solve a problem of the molecular motion dynamics within a reaction zone and dynamics of reagent spins. Issue one has experienced rather detailed study in the traditional, spinless theory of recombination of radicals. Statistics of contacts of reagents in a gas phase have been well developed. Kinetics of liquid-phase reaction differs from the gas-phase one. In the context of radical liquid-phase reactions, impact by medium is extremely diverse. However, the developed physical models describe real effects rather adequately. The so-called cellular effect is one of the brightest manifestation of medium impact in the radical reactions. Description of recombination of radicals in the 'cell' is based upon the two models – diffusion model (R. Noyes, 1956) and exponential model (R. S. Johnson, et al., 1967). In terms of a diffusion model, the 'cell' is an area with 10^{-9} m radius; life of the radicals is defined as a period during which they pass through the area. Their life in the 'cell' will depend upon viscosity: if it is high, the period will exceed greatly $10^{-9} - 10^{-10}$ s. If the radicals are in a singlet state and have high reaction ability, then they recombine each other at the first contact. If the radicals have not high reaction ability, then their recombination potential will increase in the process of the repeated collisions of the pairs. Under a diffusion model, S-T transitions take place in the intervals between collisions.

It is known that in terms of a gas phase, fragments of the damaged molecules disperse. The idea of cellular effect is as follows: in a liquid medium, molecular fragments do not have to disperse. Daughter radicals may either recombine and generate the mother molecule again or undergo disproportional reaction and generate molecular products of other structure. The condensed medium prevents partners from dispersion keeping them in a unique cell; the fact has been verified experimentally. Thus, the collisions take place serially increasing the chemical reaction potential. Moreover, the repeated series of collisions are possible even if the partners are beyond the coordination sphere. Such a possibility arises owing to diffusion processes; hence, sometimes terms of secondary, tertiary etc. cell for reagent pairs are introduced. In some cases, reaction has no need in the direct contact of partners. For instance, electrons may be transferred to large distances, ~ 10 Å. Then, the secondary, tertiary, ... cells differ from the primary one in the transfer potential.

As for neutral radical reaction, 10 Å value is that one assessing the 'cell' radius. Therefore, 'in-cell' period for nonviscous liquid with $D = 10^{-5} \text{ cm}^2/\text{s}$ is $\tau_c \approx 10^{-9}$ s.

A quantity theory of cellular effect is based upon certain ideas of a liquid structure and nature of thermal motion of the liquid molecules.

A continuous medium model in the form of a quasi-crystalline structure is the most popular one. Molecular mobility in a liquid lattice has been considered thoroughly in papers by Frenkel according to which molecules spend much time near nodes of a quasi-crystalline lattice performing elastic oscillations with $\sim 10^{13} \text{ s}^{-1}$ frequency. If liquid viscosity is $\sim 1 \text{ Pa} \cdot \text{s}$, then 'sedentary' life in the neighbourhood of the 'node' is $\tau \approx 10^{-11}$ s (Buchachenko, 2010). Resulting from the oscillations, reagents, being within the nearby nodes, may collide and recombine (E. Pestryaev, 2018).

To date, it has been proved experimentally and theoretically that a magnetic field is a stimulating factor of the course of chemical transformations between free radicals in oil-gas and liquid media. Moreover, kinetics of such reactions in liquids depends heavily upon dynamics of molecular movements; it is defined through the liquid structure as well as characteristics of radical pairs. Compared to gases, dynamic features of molecular movements are more diverse; thus, description of radical reactions becomes more complicated. Despite the fact, research in the field progresses successfully being widely implemented while performing chemical transformations with spin manifestations (during magnetic field action) and without them (a traditional approach).

Currently, no scientific sources contain information on spin manifestations in the context of solid-phase reactions. Theoretical studies intended to demonstrate influence by weak magnetic fields on the solid-state chemical reactions are also unavailable. Our experimental results, concerning thermo-magnetic processing of rocks (inclusive of coal), may be explained specifically from the viewpoint of spin effects in the solid-state reactions. Most likely, such chemical transformations take place mainly at free surfaces of materials (i.e. surfaces of powder particles; surfaces of pores, fractures, etc.). First of all, such a conclusion follows from the necessary, even if limited, mobility of reaction complexes. Progress of the new scientific tendency, being spin effects in chemical transformations of the solid condensed media, needs solving such problems as: dynamics of molecular complexes at free surfaces of materials during the action by thermal and magnetic fields; and interaction dynamics between radical and non-radical within a field of surface electrostatic impacts.

It is rather difficult to compute accurately the magnetic field influence on the dynamics of radical pairs since the process should involve complex calculations of orbital, spinal, and resulting magnetic momenta taking into consideration such fundamental quantum mechanical principles and characteristics of atomic orbitals as the Pauli exclusion principle, Hund's rules, characteristics of a molecular symmetry, etc. Only when the calculations are performed, accurate g factor magnitude is derived.

Exemplify use of the Pauli exclusion principle and Hund's rules while determining the basic term of a carbon atom which knowledge is required to describe carbon-containing radicals of CH_3 and CH_2 type. In its ground state, a carbon atom has the following electronic configuration: $1s^2 2s^2 2p^2$.

Orbital and spin momenta of impulse of $1s^2$ and $2s^2$ subshells are equal to zero. Hence, the basic term is determined by means of two p electrons being at the second energy level $n = 2$. Orbital number may be $l = 0, 1$; and spin number may be $S = \pm 1/2$. Consequently, the total orbital momentum is characterized by such quantum numbers as $m_L = 0, 1$, and 2 ; in turn, the total spin momentum is characterized by $m_S = 0, 1$. Formally, the following terms are possible

$$^1S, ^1P, ^1D, ^3S, ^3P, ^3D.$$

Actually, only 1S , 1D , and 3P terms correspond to the Pauli exclusion principle.

For instance, D^3 is characterized by similar $m_l = 1$ and $m_s = 1/2$ values for both electrons.

3P ($L = 1, S = 1$) term is the ($J = 2, 1, 0$) triplet. It is subdivided into the components

$$^3P_2, ^3P_1, ^3P_0.$$

According to Hund's rule, a state with the largest S , i.e. $S = 1$, corresponds to the minimum of atomic energy. In terms of the second Hund's rule, if the shell is open, then the minimal $J = 0$ magnitude corresponds to the energy minimum. In such a way, 3P_0 is the basic carbon atom term.

Spin prohibition against chemical reaction means that only such reactions are permitted where electron spin is con-

served; i.e. the total spin of reagents is conserved with the spin of reaction products. An example of CH_3 radicals, formed in coal, helps demonstrate that merger of two of them (being chemical reaction of their recombination) originates ethane molecule C_2H_6 with zero electronic spin. In the context of the reaction, the spin is conserved if only a molecule with zero spin originates from a radical pair having zero spin, i.e. from a singlet spin state of the radical pair. Recombination of radicals is prohibited from a triplet state of the pair (where spin is a unit) since it needs the spin change.

Thus, the prohibition because of a spin in the recombination of radicals results in the fact that of the four potential spin states of a radical pair (being one singlet and three triplet states), only one singlet state turns out to be reactive.

For instance, while facing, two triplet CH_2 polycarbons initiate a pair with nine spin states; the only of them (that very singlet where the total electronic spin is equal to zero) is permitted to recombine the polycarbons into ethylene molecule C_2H_4 . The principle is adhered while hydrocarbon recombining with CH_3 radical into methane molecule CH_4 .

Electron is an elementary spin particle; it is its magnetic momentum, and it reacts sensitively to a magnetic field even if it is of low intensity, which may vary orientation of the electronic spins, spin state of reagents, and abolish the spin prohibition. As papers by A. Buchachenko (1973–1978) define, a magnetic field can transform radical pair from a triplet (nonreactive) spin state into the singlet (reactive) one; hence, it can change recombinational potential as well as output of corresponding products.

Consider an example with radical pairs H and CH_3 ; and CH_2 and CH_3 being the most probable for coal. Fig. 5 demonstrates an orientation scheme of their total spin of their vector total. The radical pairs can compensate each other as follows: zero spin, singlet state S (Fig. 5, a); zero spin, triplet state T_0 (Fig. 5, b); plus one, triplet state T_{+1} (Fig. 5, c); minus one, triplet state T_{-1} (Fig. 5, d).

Fig. 6 shows schematically magnetic energy of the four states. If magnetic field is not available ($H = 0$), then all the states have similar energy. Increase in magnetic fields results in the increase in T_{+1} state energy according to the law

$$E = 1/2(g_1 + g_2)\beta_B H,$$

in turn, T_{-1} state energy decreases in accordance with the law

$$E = -1/2(g_1 + g_2)\beta_B H,$$

where β_B is the Bohr magneton (i.e. a quasiparticle describing collective oscillations of atomic magnetic momenta within the magnetically ordered media); $\beta_B = 9.27 \cdot 10^{-24} \text{J/T}$; g_1 and g_2 are g factors of electrons on the partner radicals, they characterize magnetism of the radicals.

Energy of S and T_0 states does not depend upon the field since in terms of them spin projection (as well as magnetic momentum) on the field direction is equal to zero. S state is reactive. As for T_0 state, a magnetic field transforms it (S and T_0) as follows. It is known that electrons, belonging to partner radicals, differ in energy as well as precession frequencies

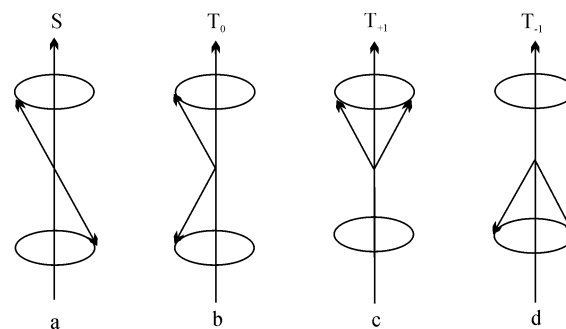


Fig. 5. Spins of two unpaired electrons of radical pairs $\text{H} + \text{CH}_3$ and $\text{CH}_2 + \text{CH}_3$

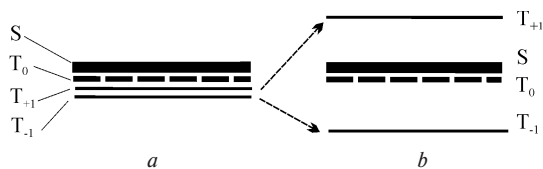


Fig. 6. Illustration of the spin states:

a – if a field is not available ($H=0$); b – under the magnetic field influence ($H>0$)

$$W_1 = 1/2g_1\beta_B H \quad \text{and} \quad W_2 = 1/2g_2\beta_B H$$

for τ time being

$$\tau[W_1 - W_2] = \pi.$$

Hence, the two spins, precessing at different rates, will turn to 180° relative to each other; i. e. T_0 spin state transforms to S state (Fig. 5). In other words, magnetic field induces triplet-singlet evolution of the radical pair transforming it from non-reactive T_0 state to chemically reactive S state. Since the frequency difference of $W_1 - W_2$ precession depends upon field intensity H as well as τ time, $T_0 \rightarrow S$ evolution rate depends on the magnetic field intensity.

The chemical reaction can be performed through the spin transitions $T_+ \rightarrow S$ and $T_- \rightarrow S$. Such a scenario is possible under interaction of identical radical pairs, e. g. CH_3 and CH_3 , CH_2 and CH_2 , etc. In this case, the mentioned spin transitions can be involved at the expense of 180° turn of one of the spin momenta, i. e. if the spin quantum number changes by one, e. g. $\Delta m_S = \frac{1}{2} - (-\frac{1}{2}) = 1$. The ‘turn’ implementation should involve the following. Induction of the external magnetic field B must exceed magnetic induction of a field developed by one of valence electrons of the radical pair. In other words, strength criterion of the transition has to be involved.

Assess magnetic induction of the field, developed by a valence electron, using the classic ideas of an electron motion as the charged particle motion in a circular orbit with almost $5 \cdot 10^{-10}$ Å radius. The value of orbit radius corresponds to the traditional RP radius. Take $R = 8 \text{Å} = 8 \cdot 10^{-10}$ m for certainty. Assess magnetic induction of a field, developed by a valence electron, using a formula for a circular current loop

$$B_e \approx \frac{\mu_0}{4\pi} \cdot \frac{2\bar{\mu}_S}{R^3}, \quad (10)$$

where is spin magnetic momentum being equal to $\mu_S = -\frac{e}{m_e} M_S = -\frac{e\hbar}{m_e} \sqrt{s(s+1)} = -\mu_B \sqrt{3}$.

Replace the letters with numbers in (10) and obtain $B_e = 6 \cdot 10^{-3}$ T.

Taking into consideration the fact that while processing magnetically coal, our experiments applied alternating magnetic field with maximum $B \approx 1.8 \cdot 10^{-2}$ T magnetic induction one could state that the strength criterion of the ‘turning’ action was performed.

Assess the magnetic field work under $T_\pm \rightarrow S$ transitions

$$A = \Delta E = \mu_B B (g_2 m_{S2} - g_1 m_{S1}),$$

where g_2 , g_1 , m_{S2} and m_{S1} are final and initial values of the Lande factor as well as the spin projections on the vector \vec{B} direction.

Considering the fact that $m_{S2} = 1/2$ and $m_{S1} = -1/2$, obtain

$$A = \mu_B \cdot B \cdot \frac{1}{2} (g_2 + g_1).$$

Assume as a rough estimation of g factor that the final g_2 value corresponds to oppositely directed electron spins providing a covalent link of S state; and initial g_1 value corresponds to one-directional electron spins of T_\pm state. The estimation

toughness is that we assume $L = S = J = 0$ for $1s^2$ electrons as well as for covalent interacting radical links.

Hence, in the approaching process of components of the pair, both orbital and spin characteristics will be defined through electrons of the covalent link being formed. Consequently, g_2 state will be identified with the help of such quantum numbers (Hund’s rule is involved) as $L = 2$, $S = 0$, $J = 2$ and $L = 2$, $S = 1$, $J = 3, 2, 1$ quantum numbers will define g_1 state.

Taking into consideration formula (1), obtain

$$g_2(J=3) = 1 + \frac{3 \cdot 4 + 1 \cdot 2 - 2 \cdot 3}{2 \cdot 3 \cdot 4} = 1.33;$$

$$g_2(J=2) = 1.17; \quad g_2(J=1) = 0;$$

$$g_1 = 1 + \frac{2 \cdot 3 - 2 \cdot 3}{2 \cdot 2 \cdot 3} = 1.$$

Having assumed the maximum $g_2 = 1.33$ and $g_1 = 1$ values, we obtain

$$A = 9.27 \cdot 10^{-24} \cdot 1.38 \cdot 10^{-2} \cdot 1/2(1.33 + 1) \approx 0.15 \cdot 10^{-24} \text{ J}.$$

At a room temperature, RT magnitude is $4 \cdot 10^{-24}$ J, i. e. it is 26 times more than the magnetic field work. Actually, A evaluation is overestimated.

Hence, despite the fact that energy of weak magnetic fields is much less than the one of thermal molecular motion, they are the power factor of chemical transformations of radical pairs.

Dwell briefly on the features of magnetic coal processing implemented by our experiments. First, it should be mentioned that coal was processed using variable magnetic field with $f = 50$ Hz frequency (single semi-periodic current rectification of the induced coil). In this context, magnetic action time significantly exceeds a typical interaction period of radical pairs. Consequently, a quasi-stationary mode was applied to relax the radical pairs.

In such a way, achievement of a certain intensity level of a magnetic field creates favourable conditions for $T_\pm \rightarrow S$ transitions with further chemical RP transformations; at the same time, those RP components, capable of chemical transformations if a field is not available, become nonreactive after transfer of one of the radical pair spins.

If a field is deactivated at the expense of relaxation processes, such pairs become reactive again. In such a way, deep ‘development’ of RPs, being both in a singlet state and triplet ones, takes place.

Nonmagnetic radical pairs (containing nonmagnetic isotopic nuclei of ^{12}C , ^{16}O , ^{18}O and other types) have no magnetic field of nuclei; hence, spin prohibition remains. Actually, the reaction performs sorting: radicals with magnetic nuclei recombine; and radicals with nonmagnetic isotopic nuclei, having no such possibility, pass into other products transforming to other molecules.

Chemical interaction of radicals, being of selective nature in terms of electron spin, turns out to be of selective nature in terms of nuclear spin as well since both of the spin systems are connected through magnetic electron-nuclear interaction.

It is known [26] that recombination of two radicals depends upon the fact which of isotopic nuclei (i. e. magnetic or nonmagnetic) reacting radicals contain. For example, CH_3 radical, containing nonmagnetic carbon nucleus 12, responds slower than a radical with magnetic carbon nucleus 13 (in connection with the data it would be helpful to understand, whether isotopic composition methane, released from coal, was analysed).

Chemical reactions between radicals may take place similarly to those in free radicals as well as between movable and immovable ones. Carbon atom inserting in carbon chains of carbonaceous matter increases concentration of the structures characterized by a periodic atomic arrangement (two-dimensional formations of carbon-based grids).

Fragments of the remained radicals (i.e. oxygen, hydrogen) shape stable H₂O molecules or molecules of OH group. In this case, chemical reactions result in the carbonaceous matter deactivation. The effect may be rather useful for outburst coal from the viewpoint of prevention from the potential destructive processes in organic coal mass and decrease in outburst intensity.

Influence by a weak magnetic field may stimulate chemical reactions intended to improve stability of the destabilized microstructure and unstable balance between the shaped radicals and active centres (stress and strain bonds) located at the surfaces of particles of an organic coal matter.

Conclusions. Physicochemical mechanism of a magnetic scenario of intraradical bonds is considered from the viewpoint of their stimulation by means of magnetic fields; i.e. the idea has been implemented according to which the number of radical pairs, able to recombine in the stable molecules, increases significantly if a weak magnetic field exerts certain influence. In addition to stimulation of intraradical bonds, the magnetic field impact on *organic rock mass-radicals* factors into stabilization as well as increase in carbon structures with regular atomic arrangement (i.e. two-dimensional grids, chains etc.).

Electric stimulation of phase transformations in coal as well as their chemical activity on the whole is the most important tendency of research focused mainly on the potential use for such processes as gasification and liquefaction of low-grade coal. Analogy of processing results through two physical actions (i.e. mechanical activation and electric field) is of significant fundamental importance for coal science; it especially concerns the connection between its electric characteristics and outburst tendency.

Progress of the activities in the field of magnetic processing should involve studies connected with calculation and selection of magnetic field parameters to control efficiently chemical reactions between radicals in coal matter. Substantiation degree of the methods for coal processing will depend heavily upon the efficiency of chemical processes as well as expediency of the result implementation. Among other things, it concerns a technique to prevent formation of unstably stable states characterized in mining as outburst ones [30].

Further experiments should apply the fact of magnetic stimulation of crystalline phase increment for scientific research which would support (or refute) the potential of experimental imitation of carbonization process. It should be mentioned that possible thermal disturbances at 320 K have not been recorded; thus, they cannot factor into such physicochemical changes in coal as it results from the impact by a weak magnetic field. The findings are important scientifically in the field of coal genesis as well as evolution features of coal characteristics from long-flame grades up to anthracite.

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Магнітне стимулювання хімічних реакцій у кам'яному вугіллі

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Мета. Встановити механізм, за яким сигнал слабого магнітного поля трансформується у відгук деяких компонентів наноструктури вугільної речовини з подальшим утворенням хімічних зв'язків.

Методика. Використані методики фізичного й математичного моделювання елементарних хімічних актів. Використані закономірності квантової механіки та динаміки хімічних зв'язків, теорія магнітної ізотопії, дифузійна теорія рекомбінації радикальних пар з урахуванням триплет-синглетних переходів, ядерно-спінової селективності хімічних реакцій.

Результати. Фізичний механізм магнітного сценарію міжрадикальних реакцій розглядається з точки зору їх енергетичного стимулювання магнітними полями, тобто експериментально реалізована ідея, згідно з якою коло радикальних пар, здатних рекомбінувати у стійкі молекули, значно розширюється у випадку впливу слабого

магнітного поля. Вплив магнітного поля на систему «органічна маса вугілля – радикали» окрім стимулювання міжрадикальних реакцій призводить до стабілізації та зростання вуглецевих структур із регулярним розташуванням атомів (двовимірних сіток, ланцюжків і т. п.).

Наукова новизна. Розроблена фізична модель структурно-фазових змінювань у вугіллі під дією зовнішнього магнітного поля слабкої напруженості. Показано, що сигнал слабого магнітного поля, незрівнянно меншого за енергією відносно енергії теплового руху молекул, здатен ініціювати триплет-синглетні переходи, тобто переводити радикали до реакційно спроможного стану. Запропоновано механізм утворення хімічних зв'язків між рухомими радикалами й незавершеними зв'язками поверхневих атомів на поверхнях твердих фаз вугільної речовини.

Практична значимість. Практичне застосування отриманих результатів із магнітної обробки вугілля може бути пов'язане зі створенням нового способу запобігання викидонебезпечного стану у вугільних пластах. Для розвитку робіт в області магнітної обробки необхідно проводити дослідження, пов'язані з розрахунком і вибором параметрів обробки вугілля (напруженості магнітного поля, частоти, енергії та часу обробки), з метою ефективного управління хімічними реакціями у вугільній речовині. Від того, наскільки будуть обґрунтовані методи обробок вугілля, багато в чому залежатиме не тільки ефективність хімічних процесів, зокрема, але й доцільність практичного використання отриманих результатів у цілому.

Ключові слова: *вугілля, множник Ланде, радикальна пара, спин, розщеплення, синглетний стан, електрон, стан нестійкої рівноваги*

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