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RADIOLYTIC PROCESSES IN MIXTURE OF WATER WITH CARBON DIOXIDE IN THE PRESENCE OF ORGANIC MATRIX

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Abstract: *The radiolysis of purified water, its mixture with carbon dioxide, and also with the addition of salt of potassium chloride isotope (^{40}KCl) and plant mass to this system were carried out in order to study the mechanism of radiolytic processes taking place in the plant mass in the presence of the natural potassium isotope (^{40}K) contained in it. It revealed that in the presence of ^{40}KCl in the system, the formation of trace amounts of molecular hydrogen is observed (in accord with an insignificant dose of radiation from ^{40}K). A decrease in the rate of formation of molecular hydrogen with an increase in the concentration of CO_2 is observed during the radiolysis of a two-component system (H_2O - CO_2). An increase in the rate of formation of molecular hydrogen, carbon monoxide, methane and relatively heavy hydrocarbons (C6, C7, C8) accompanied by an increase in the absorbed dose of ionizing radiation (at stable concentrations of all components of the system) is observed when studying the kinetics of the formation of radiolysis products of the multicomponent system "H_2O - CO_2 - ^{40}KCl - organic matrix".*

There is a tendency to reduce the rate of formation of molecular hydrogen and methane and increase the rate of formation of CO and relatively heavy (C6, C7, C8) hydrocarbons with an increase in CO_2 concentration. The rates of formation of all products (H_2, CO, CH_4, and relatively heavy hydrocarbons) grow with an increase in the amount of organic matter in the mixture (at stable concentrations of CO_2, H_2O, and ^{40}KCl). İt found that an increase in the mass of the organic matrix in the analyzed multicomponent system stimulates an increase in the formation of all radiolysis products. There is a decrease in the rate of formation of H_2 and CH_4, a relatively slow increase in the rate of formation of carbon monoxide, and an increase in the rate of elementary reactions of the transformation of light radiolysis products (H_2, CO, CH_4) into relatively heavy products (C6, C7, C8) together with an increase in CO_2 concentration. The results obtained show the expediency of taking into account the contribution of ionizing radiation from radionuclides present in the environmental components, when considering multistage biochemical mechanisms of photosynthesis in order to explain the initiation of energy-intensive processes of CO_2 and H_2O decomposition.

Keywords: *radiolysis, ^{40}K isotope, H_2O-CO_2 mixtures, plant mass, hydrocarbons.*
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Introduction

The main part of the soil is formed by chemical compounds in the form of various minerals. The study of various forms of chemical elements occurrence in minerals, organic residues and emissions, soil colloids; the determination of the amount of oxides, hydroxides, carbonates, bicarbonates, nitrates, nitrites, sulfates, and phosphates in soil samples make it possible to assess the ecological state of the soil [1–9].

Carrying out systematic systematic measurements and studies to obtain results on the distribution of radionuclides, heavy metals and other xenobiotics in water, soil and vegetation, information for predicting changes and the rate of changes in the environment are

important for solving many environmental problems.

Systematic studies made it possible to reveal that all environmental objects (water, soil and vegetation) contain natural radionuclides, including ^{40}K and ^{22}Na isotopes [10, 11].

The dose rate of gamma radiation from different parts of the soil are 0.02-0.15 μSv/h. The activities of the ^{40}K isotope in soil minerals and green vegetation are 0.8–2.5 Bq/kg and 0.8–3.0 Bq/kg, respectively. These values for ^{22}Na are 1.0–3.0 Bq/kg and 0.6–2.0 Bq/kg, respectively. The activity values of radionuclides (except for ^{40}K) in vegetation minerals are 1.4-1.7 times higher than their values in water minerals of nearby water sources. However, the activity values of the ^{40}K isotope are 7-10 times higher in vegetation minerals than their values in water minerals. The value of the degree of assimilation of the ^{40}K isotope from water by the vegetation cover is approximately 5–7 times higher than the degree of assimilation of radioactive isotopes of other elements by plants which indicates a higher efficiency of the process of assimilation of ^{40}K by plants from water [11].

The examination of the influence of natural radionuclides on the processes taking place in environmental objects, the role of ionizing radiation from radionuclides in the course of complex multistage processes in the vegetation cover are important sources of arguments for the predictability of possible changes in the habitat of living organisms.

Methodical part

Radiometric measurements were carried out using İnSpector-1000 and Radiagem-2000 (USA, Canberra) radiometers equipped with alpha, beta and gamma radiation detectors. Determination of radionuclides and their activity was carried out by gamma spectroscopy (spectrometer of Canberra company equipped with HPGe detector) using certified point sources for comparative analyzes [10, 11, 12].

The experiments were carried out under static conditions by irradiating glass ampoules filled with the studied mixtures. The filling of ampoules with carbon dioxide, as well as the purification of water from dissolved gases, were carried out using an experimental vacuum installation. The installation consists of a vacuum part, glass volumes for storing initial liquids and gases, a measuring part (pressure gauges) that allows working up to pressures of 2.10^5 Pa. A schematic diagram of a vacuum installation designed to pump out gases dissolved in liquid water is given in previously published works [13, 14].

The glass ampoules intended for filling the studied mixtures were previously connected to the vacuum outlet of the installation and pumped out to $10^{-2} \div 10^{-3}$ Pa (for 30 minutes at T≥500 K, and then for another 30 minutes at room temperature), after which they were disconnected from the vacuum installation and filled with the studied systems: H_2O, H_2O – CO_2, ^{40}KCl - H_2O – CO_2, ^{40}KCl - H_2O – CO_2 – "organic matrix". Various quantities (1.0, 5.0 and 10.0 g) of green leaves of the olive tree (Olea europaea L.) repeatedly washed with distilled water were chosen as the organic matrix.

Gases were evacuated by a vacuum installation equipped with vacuum lamps, traps, oil and mercury manometers, and glass vessels with three-way vacuum cocks to purify water from dissolved gases. The water used is repeatedly (4-5 times) purified according to the cycle “freezing - vacuum filling - pumping out - ice thawing - slow pumping out of dissolved gases" at 293 K.

Qualitative analysis of the studied components was carried out using a gas chromatograph GC-2010 (Shimadzu, Japan) at a temperature of 398 K, with a flame ionization detector (FID) connected to a Supelco Nucol capillary column (30m×0.32mm). Helium (99.995%) was used as a carrier gas. The radiolysis products (H_2, CO, CH_4) were analyzed by chromatography at room temperature on a “Gasochrome 3101” gas analyzer equipped with a thermochemical detector and a packed column (3 m × 3 mm) filled with activated carbon at the factory. The flow of atmospheric air forced by the factory mini-pump of the device was used as a carrier gas. Hydrocarbons were analyzed also, on an Agilent 7890A chromatograph (Agilent, USA) equipped with two detectors for operation in

two modes: for the analysis of hydrocarbons at a temperature of 398 K with a FID connected to a capillary column (30 m × 0.32 mm) GC-Gaspro, as well as for hydrogen analysis at room temperature with a katharometer connected to a capillary column (30 m×0.53 mm) Supelco CarboxenTM. Helium (99.995%) was used as a carrier gas in both modes.

Irradiation of glass ampoules filled with test mixtures was carried out with ionizing gamma radiation from ^{60}Co sources of a powerful gamma installation "УК-120000". The dose rate absorbed in the irradiation zone was 6.6 kRad/hour (0.066 kGy/hour).

Results and discussion

The current analysis of the mineral composition and appearance of vegetation on different areas of green plants, as well as the comparison of the appearance of green spaces grown in experimental areas (on soil with a natural activity of ^{40}K 0.8 Bq / kg and on soil impregnated with an aqueous solution of ^{40}KCl salt with an activity of ^{40}K brought to 2.5 Bq/kg) showed a relatively high growth rate of vegetation in soil areas containing high concentrations of ^{40}KCl (see Fig.1).

a

b

c

Fig. 1. Appearance of sunflower (*Helianthus annuus L.*), corn *(Zea mays)* and coriander *(Coriandrum sativum)* grown on ordinary soil with natural activity ^{40}K 0.8 Bq/kg (*a* – beds on the left and picture *b*) and on soil impregnated with an aqueous solution of chloride salt ^{40}K with activity brought up to 2.5 Bq/kg (*a* – beds on the right and picture *c*).

The course of photosynthesis is described by the overall equation as follows:

$$6CO_2 + 6H_2O \rightarrow C_6H_{12}O_6 + 6O_2 \qquad (1)$$

The high endothermicity (3080 kJ/mol) of this reaction is due to the high values of the dissociation energy of the O–H bond (485–498 kJ/mol or 5.0–5.2 eV/mol) in the water

molecule and the C=O bond (799 kJ/mol or 8.3 eV/mol.) of carbon dioxide molecule [11, 12]. Low energies of visible light quanta (below 5 eV/quantum) are insufficient to break the C=O bond. However, the high energies of gamma radiation quanta from the ^{40}K isotope easily explain the dissociation of carbon dioxide and water molecules into the corresponding radicals and ions. The presence of ^{40}K isotopes in all environmental objects (in water, soil and vegetation), relatively high growth rates of plants in soil areas containing relatively high concentrations of ^{40}KCl, the presence of photosynthesis variants on the cytoplasmic membrane of extreme halobacteria or in the absence of chlorophyll and oxygen, in the depths of large lakes and seas of the planet in the presence of long-wavelength infrared radiation (from volcanic lava) testify in favor of considering the role of ionizing radiation from ^{40}K at the initial energy-intensive stage of photosynthesis, i.e. dissociation of CO_2 and H_2O molecules [11].

The kinetics of the formation of radiolysis products in the systems H_2O, H_2O - CO_2, ^{40}KCl - H_2O - CO_2, ^{40}KCl - H_2O - CO_2 - "organic mass" under the influence of ionizing gamma radiation were studied so as to study the course of chemical processes in a mixture of water with carbon dioxide in the presence of mass green vegetation. Repeatedly washed with distilled water different quantities (1.0, 5.0 and 10.0 g) of green leaves of olive trees (Olea europaea L.) were chosen as the organic matrix. Performed radiometric measurements showed that the activity of radiation from 5.0 gr. ^{40}KCl salt in one ampoule was 40 Bq. Filled with the indicated mixtures ampouyles before irradiation at a powerful gamma installation were stored for 30 days around a vessel containing a weak source of gamma radiation (filled with ^{40}KCl salt) with an activity of 1000 Bq. Figures 2, 3, 4, 5, 6, 7, 8, and 9 are indicative of the kinetics of the formation of radiolysis products of the studied mixtures irradiated at the high-power gamma installation УК-12000.

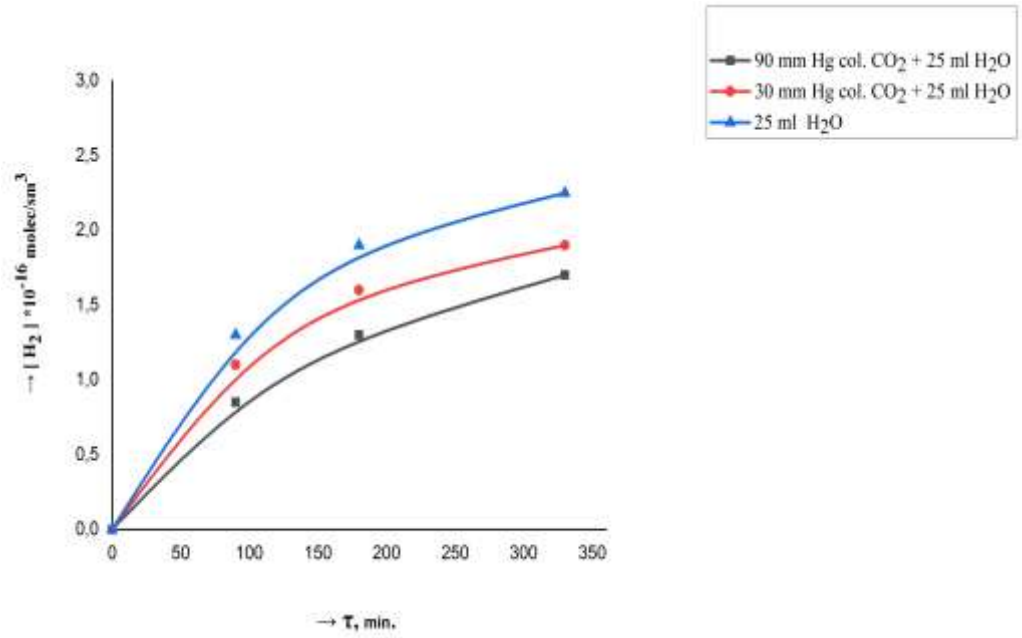

Fig. 2. Kinetics of hydrogen production during radiolysis of water and mixtures of water with carbon dioxide.

Note that no hydrogen formation was observed during the chromatographic analysis in ampoules filled with water and a mixture of water with carbon dioxide, which were stored for a month around a weak source with an activity of 1000 Bq.

However, the presence of a trace amount of hydrogen was detected in ampoules containing multi-component systems stored for 30 days near a weak radiation source. This amount ((0.1-0.2) × 10^{14} molecules/cm3) was much lower (more than 10^6 times) than the values of the hydrogen concentration (shown on kinetic curves in Fig. 4) formed in the ampoule

under the action of ionizing radiation from the powerful gamma installation УК-120000.

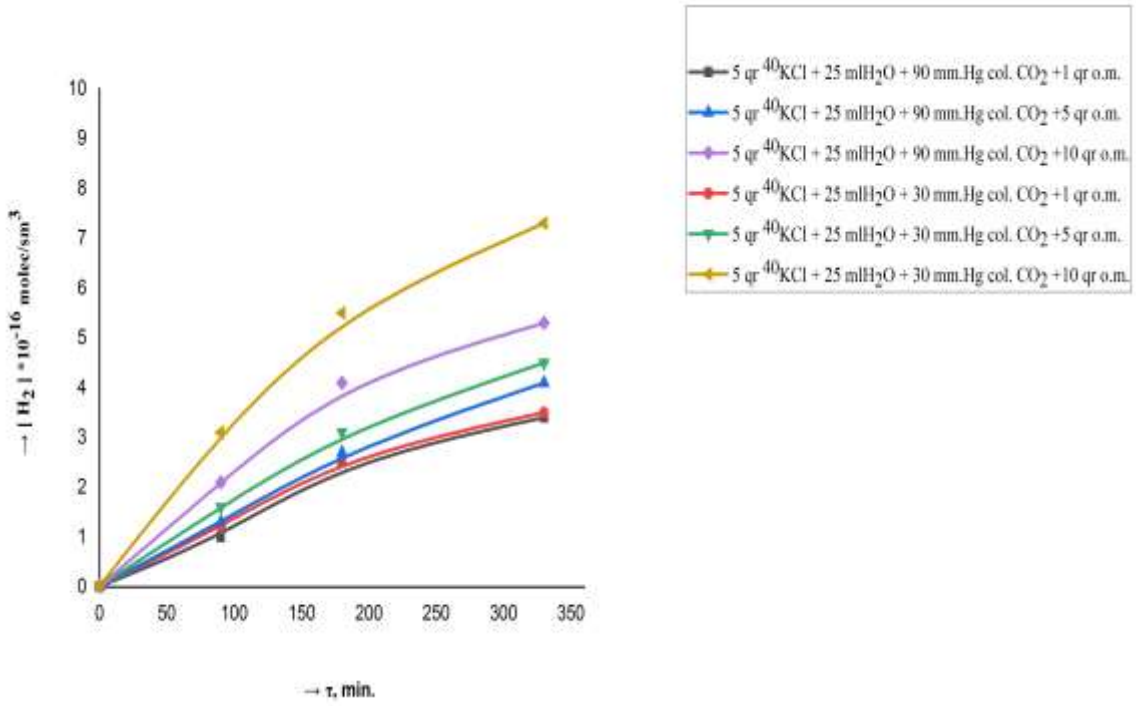

Fig. 3. Kinetics of hydrogen formation during radiolysis of mixtures "5 g ^{40}KCl - 25 ml. H_2O – 30, and 90 mm.Hg.col. CO_2 – 1.0, 5.0 and 10.0 g. organic mass".

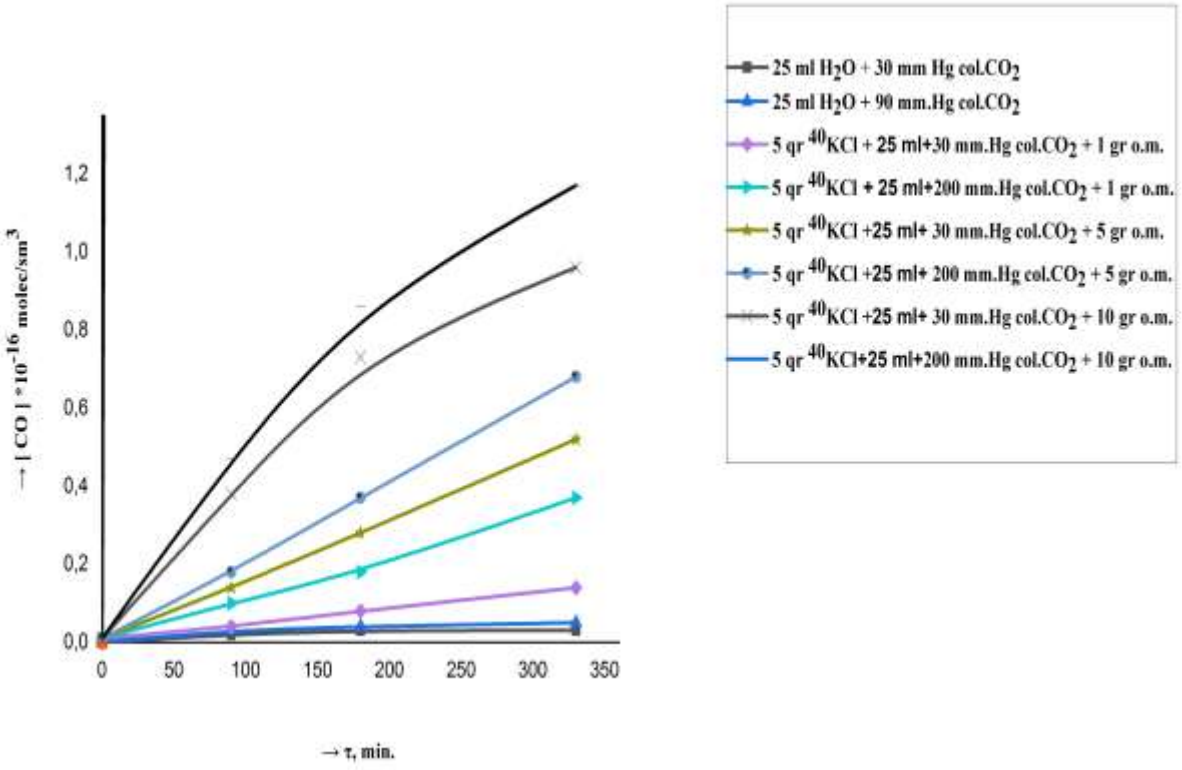

Fig. 4. Kinetics of formation of carbon monoxide during radiolysis of mixtures "25 ml H_2O – 30 and 90 mm.Hg.col. CO_2", "5 g ^{40}KCl - 25 ml H_2O - 30 and 200 mm.Hg.col. CO_2 – 1.0, 5.0 and 10.0 g organic mass".

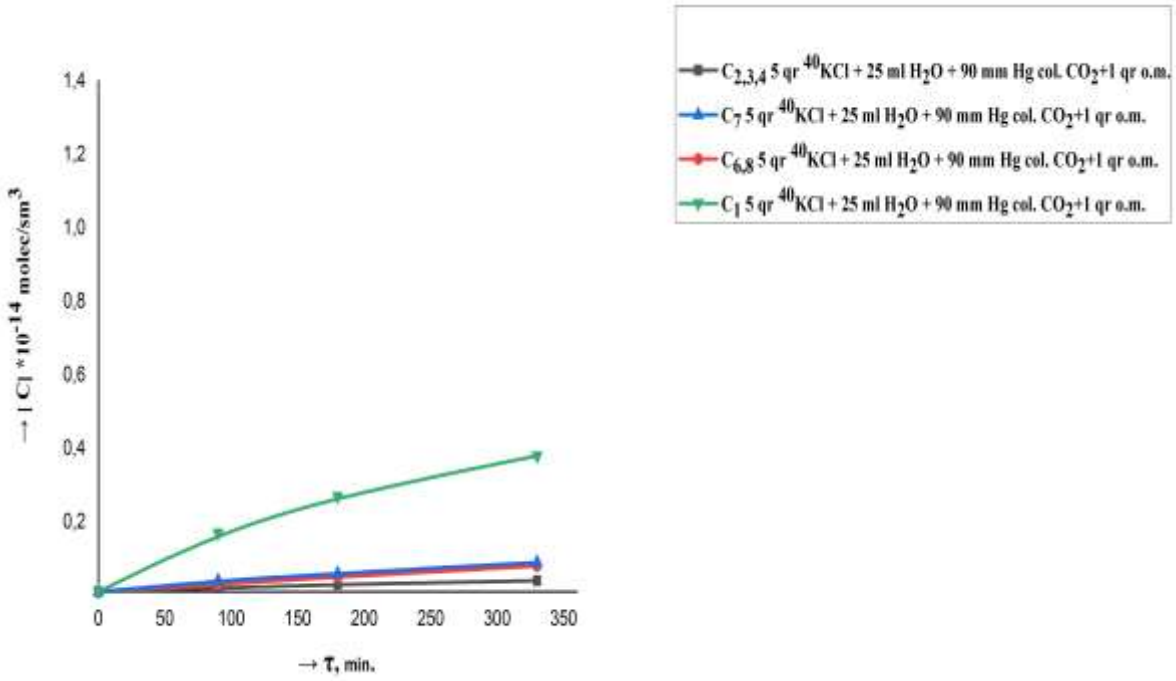

Fig. 5. Kinetics of hydrocarbon formation upon radiolysis of mixtures "5 g ^{40}KCl - 25 ml H_2O - 90 mm.Hg.col. CO_2 - 1 g organic mass".

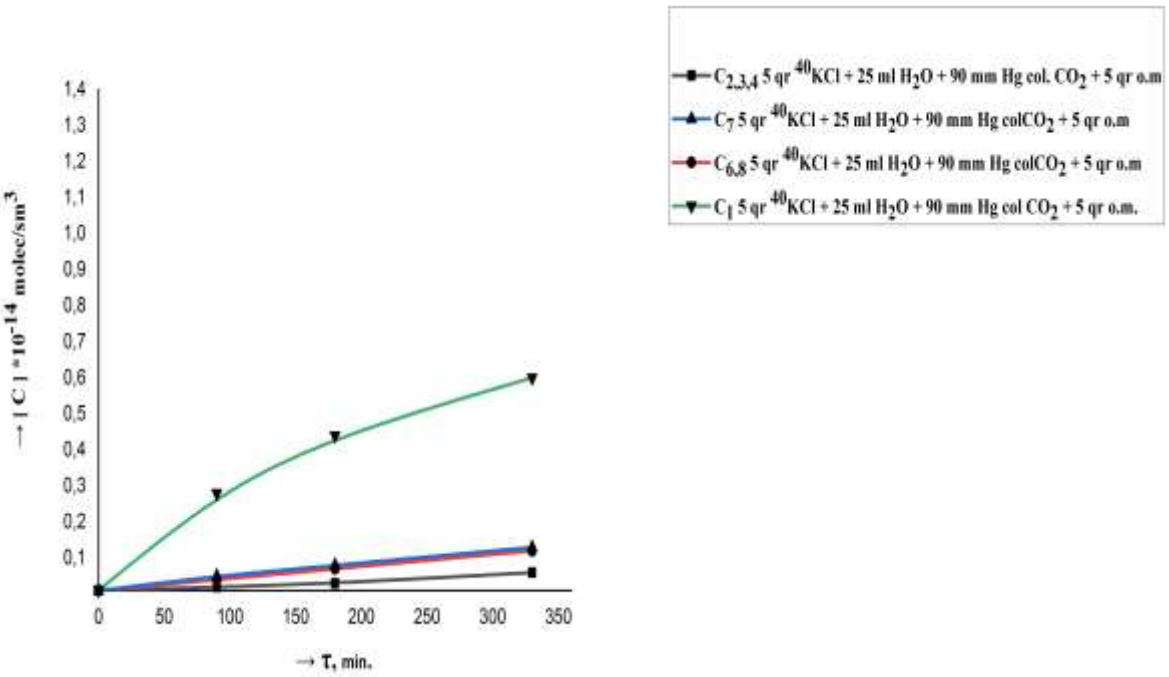

Fig. 6. Kinetics of hydrocarbon formation upon radiolysis of mixtures "5 g ^{40}KCl - 25 ml H_2O - 90 mm.Hg.col. CO_2 - 5 g organic mass".

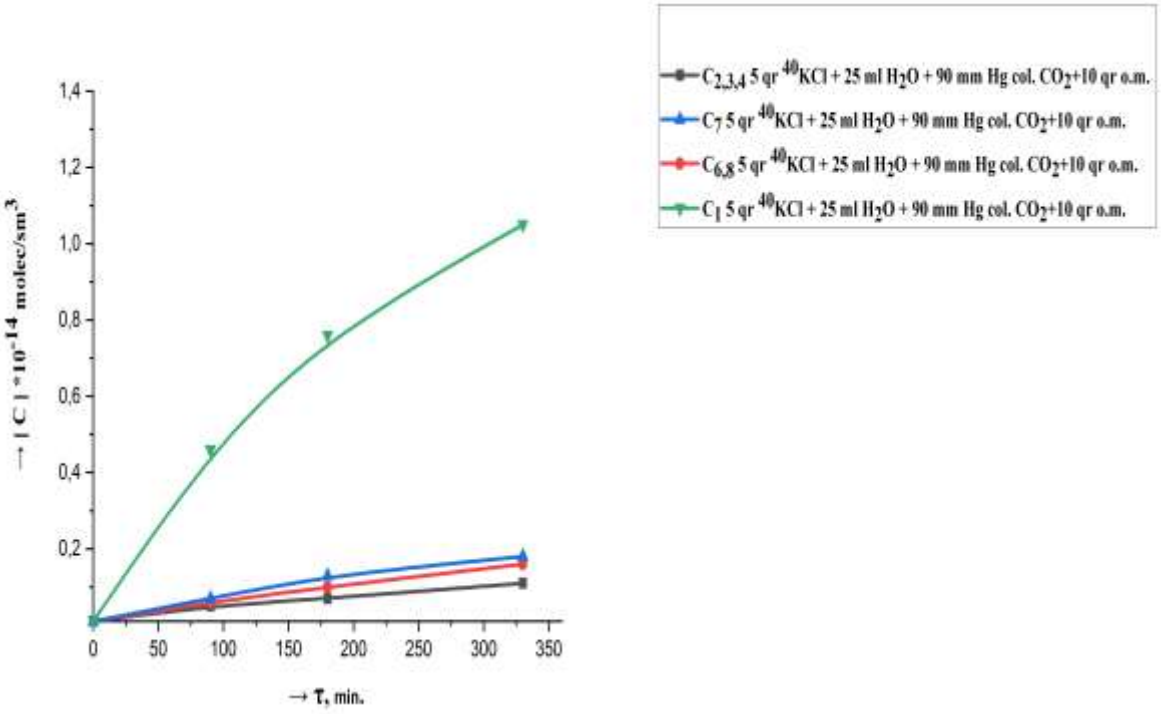

Fig. 7. Kinetics of hydrocarbon formation upon radiolysis of mixtures "5 g ^{40}KCl - 25 ml H_2O - 90 mm.Hg.col. CO_2 - 10 g organic mass".

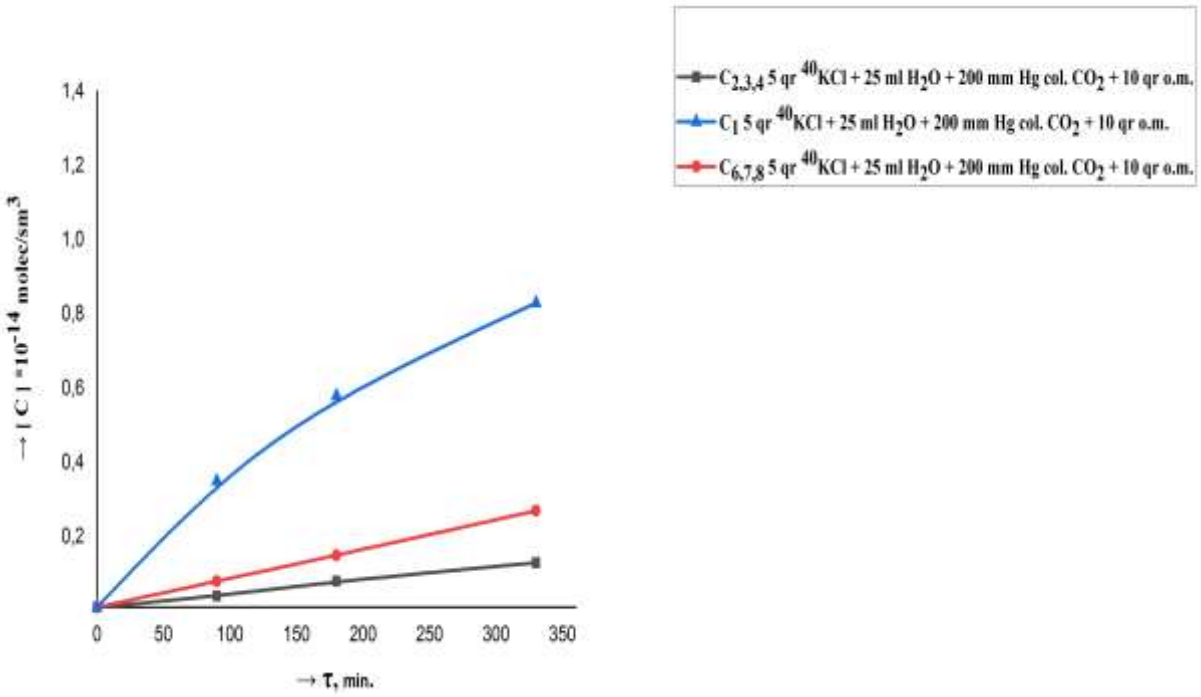

Fig. 8. Kinetics of hydrocarbon formation upon radiolysis of mixtures "5 g ^{40}KCl - 25 ml H_2O - 200 mm.Hg.col. CO_2 - 10 g organic mass".

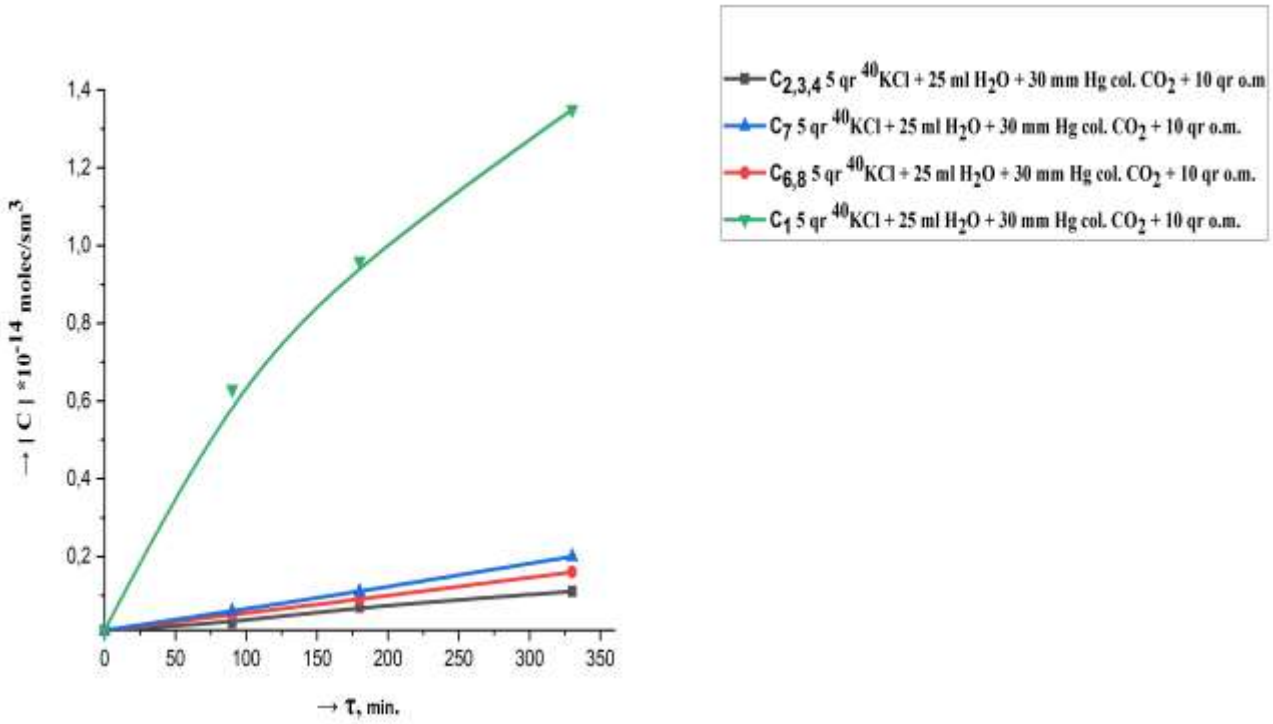

Fig. 9. Kinetics of hydrocarbon formation upon radiolysis of mixtures "5 g ^{40}KCl - 25 ml H_2O - 90 mm.Hg.col. CO_2 - 10 g organic mass".

The energy of a gamma quantum from ^{40}K is 1.45 MeV. The radiation-chemical yield of hydrogen formation from water is 0.3–1.0 molecules/100 eV [12]. Gamma rays emitted by a source with an activity of 1000 Bq emit a total energy over 30 days of 1000 × 30 × 24 × 3600 × 1450000 eV = 3.8×10^{15} eV, which corresponds to the formation of no more than 0.4×10^{14} H_2 molecules from pure water in an ampoule during 30 days. The formation (radiation-chemical yield) of hydrogen during the radiolysis of organic matter is approximately ten times higher, which corresponds to the formation of no more than 4.0×10^{14} molecules or 0.1×10^{14} H_2/cm^3 molecules in the ampoule, taking into account that the volume of the ampoule is 40 cm^3.

The value obtained by calculation explains the detected "dark effect of the formation of trace amounts of hydrogen" in ampoules containing mixtures of "^{40}KCl-H_2O-CO_2-organic mass".

The research into the kinetics of the formation of products during the radiolysis of the above systems led to the following conclusions:

- the rate of CO formation grows with increase in the concentration (pressure) of CO_2 in the H_2O – CO_2 mixture, the rate of H_2 formation decreases and no formation of hydrocarbons is observed in this case. The formation of hydrocarbons during the thermal catalysis of the H_2O – CO mixture in the presence of ionizing rays is considered in [11];

- the rate of formation of CO with increase in the concentration (pressure) of CO_2 from 30 to 200 mm.Hg.col. in mixtures "5 g ^{40}KCl - 25 ml H_2O - CO_2 – 1.0, 5.0 and 10.0 g organic matrix" increases, and the rates of formation of H_2 and CH_4 decrease. At the same time, the total amount of formed relatively heavy hydrocarbons (C2-C8) does not change significantly;

- an increase in the rate of formation of H_2, CO, CH_4 and relatively heavy hydrocarbons (C2-C8) is observed with increase in the amount of organic matter in mixtures "5 g ^{40}KCl - 25 ml H_2O – 90 mm.Hg.col. CO_2 – organic matrix";

- the rate of formation of molecular products with increase in the absorbed dose of radiation in the system "5 g ^{40}KCl - 25 ml H_2O - 30 mm.Hg.col. CO_2 - 10 g organic matrix" do not change significantly and the accumulation of

products is observed in proportion to the value of the absorbed dose.

The revealed regularities prove conclusively the observed high growth rates of vegetation in soil areas containing high concentrations of ^{40}KCl, which are microsources of ionizing radiation that create relatively higher doses than in similar soil with vegetation (with relatively low growth rates) containing relatively low concentrations of ^{40}KCl. The rapid growth and maturation of fruits of figs (*Ficus carica L.*) and white mulberries (*Morus alba L.*), after a certain growth phase, the accumulation of glucose and fructose in them during summer days and nights, testifies in favor of the proposed version.

The formation of heavier hydrocarbons and carbohydrates in the studied multicomponent systems under the influence of ionizing rays can be explained as being due to the mechanism of formation of heavy molecules, described in detail in [13, 14], which consists of a sequence of numerous elementary reactions with a number of values of their rate constants indicated.

Conclusion

The considered and obtained data (presence of ^{40}K isotopes in all environmental objects), the efficiency of the process of assimilation of ^{40}K from water by plants, the relatively high rates of plant development in soil areas containing high concentrations of ^{40}KCl, the presence of photosynthesis and the absence of chlorophyll and oxygen, on the cytoplasmic membrane of extreme halobacteria, in the depths of large lakes and seas of the planet in the presence of long-wavelength infrared radiation /from volcanic lava/, qualitative changes in the composition of fruits of figs /Ficus carica L./, white mulberries /Morus alba L./ both in summer days and at night, the high value of the C=O bond energy in carbon dioxide molecules, which is much higher than the energy of visible light quanta) indicates the expediency of taking into account the role of ionizing radiation from ^{40}K at the initial energy-intensive stage of initiating the dissociation of molecules (CO_2, H_2O, etc.) when considering traditional multistage biochemical mechanisms of photosynthesis.

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РАДИОЛИТИЧЕСКИЕ ПРОЦЕССЫ В СМЕСИ ВОДЫ С ДВУОКИСЬЮ УГЛЕРОДА В ПРИСУТСТВИИ ОРГАНИЧЕСКОЙ МАТРИЦЫ

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Аннотация: С целью изучения механизма радиолитических процессов, протекающих в растительной массе в присутствии содержащейся в ней естественного изотопа калия (^{40}K), был проведен радиолиз очищенной воды, ее смеси с двуокисью углерода, а также с добавлением в эту систему соли хлорида изотопа калия (^{40}KCl) и растительной массы. Было установлено, что в присутствии ^{40}KCl в системе наблюдается (соответствующий незначительной дозе излучения от ^{40}K) образование в следовых количествах молекулярного водорода. При радиолизе двухкомпонентной системы (H_2O - CO_2) с повышением концентрации CO_2 наблюдается снижение скорости образования молекулярного водорода. Изучением кинетики образования продуктов радиолиза многокомпонентной системы «H_2O - CO_2 - ^{40}KCl - органическая матрица” было установлено, что с повышением поглощенной дозы ионизирующего излучения (при стабильных концентрациях всех компонентов системы) наблюдается повышение скорости образования молекулярного водорода, монооксиси углерода, метана и сравнительно тяжелых углеводородов (C6, C7, C8). С увеличением концентрации CO_2 наблюдается тенденция снижения скорости образования молекулярного водорода и метана и повышения скорости образования CO и сравнительно тяжелых (C6, C7, C8) углеводородов. Увеличение количества органической массы (при стабильных концентрациях CO_2, H_2O и ^{40}KCl) сопровождается увеличением скорости образования всех продуктов (H_2, CO, CH_4 и сравнительно тяжелых углеводородов). Было установлено, что увеличение массы органической матрицы в исследуемой многокомпонентной системе стимулирует рост образования всех продуктов радиолиза. С увеличением концентрации CO_2 наблюдается снижение скорости образования H_2 и CH_4, сравнительно медленное повышение скорости образования монооксиси углерода и повышение скорости элементарных реакций

превращения легких продуктов радиолиза (H_2, CO, CH_4) в сравнительно тяжелые продукты (C6, C7, C8). Полученные результаты указывают на целесообразность учета вклада ионизирующего излучения от присутствующих в компонентах окружающей среды радионуклидов, при рассмотрении многоступенчатых биохимических механизмов фотосинтеза, с целью объяснения инициирования энергоемких процессов разложения CO_2 и H_2O.

Ключевые слова: радиолиз, изотоп ^{40}K, смеси H_2O - CO_2, растительная масса, углеводороды.

ÜZVİ MATRİTSA ƏLAVƏ EDİLMİŞ SU VƏ KARBON QAZI QARIŞIĞINDA RADİOLİTİK PROSESLƏR

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Xülasə: Tərkibində kaliumun təbii izotopunun (^{40}K) iştirakı ilə bitki kütləsində baş verən radiolitik proseslərin mexanizmini öyrənmək məqsədilə distillə edilmiş suyun, onun karbon qazı ilə qarışığının, həmçinin kalium xlorid duzu (^{40}KCl) və bitki kütləsi ilə qarışıqlarının radiolizi tədqiq edilmişdir. ^{40}KCl duzu həll edilmiş sistemdə iz miqdarında (^{40}K-dan cüzi şüalanma dozasına uyğun miqdarda) molekulyar hidrogenin əmələ gəlməsi müşahidə olunur. İki komponentli sistemin (H_2O - CO_2) radiolizi zamanı CO_2 konsentrasiyasının artması ilə molekulyar hidrogenin əmələ gəlmə sürətinin azalması müşahidə olunur. "H_2O - CO_2 - ^{40}KCl - üzvi matritsa" çoxkomponentli sistemin radioliz məhsullarının əmələ gəlmə kinetikasını öyrənilməsi ilə müəyyən edilmişdir ki, sistemin bütün komponentlərinin sabit konsentrasiyalarında ionlaşdırıcı şüalanmanın udulmuş dozasının artması ilə molekulyar hidrogenin, dəm qazının, metanın və nisbətən ağır karbohidrogenlərin əmələ gəlmə sürətində artım müşahidə olunur (C6, C7, C8). Karbon qazının konsentrasiyasının artması ilə molekulyar hidrogenin və metanın əmələ gəlmə sürətinin azalması, dəm qazının və C6, C7, C8 karbohidrogenlərin əmələ gəlmə sürətinin artması tendensiyası müşahidə olunur. Üzvi maddələrin miqdarının artması (CO_2, H_2O və ^{40}KCl sabit konsentrasiyalarında) bütün məhsulların (H_2, CO, CH_4 və nisbətən ağır karbohidrogenlərin) əmələ gəlmə sürətinin artması ilə müşayiət olunur. Müəyyən edilmişdir ki, tədqiq olunan çoxkomponentli sistemdə üzvi matritsanın kütləsinin artması bütün radioliz məhsullarının əmələ gəlməsini stimullaşdırır. Sistemdə karbon qazının konsentrasiyasının artması ilə H_2 və CH_4 əmələ gəlmə sürətində azalma, dəm qazının əmələ gəlmə sürətində zəif artım və yüngül radioliz məhsullarının (H_2, CO, CH_4) nisbətən ağır məhsullara (C6, C7, C8) çevrilməsinin elementar reaksiyalarının sürətində əhəmiyyətli artım müşahidə olunur. Alınmış nəticələr çoxmərhələli biokimyəvi fotosintez proseslərinin mexanizmində CO_2 və H_2O molekullarının enerji tutumlu parçalanma proseslərinin izahı məqsədilə ətraf mühit komponentlərində mövcud olan radionuklidlərin ionlaşdırıcı şüalanmasının ümumi prosesin reallaşdırılmasında töhfəsinin nəzərə alınmasının məqsədəuyğunluğunu göstərir.

Açar sözlər: radioliz, ^{40}K izotopu, H_2O-CO_2 qarışıqları, bitki kütləsi, karbohidrogenlər.