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## Radiation-induced isomerization reaction mechanism of hydrocarbons on the surface of solid acid

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A comparative analysis of the edge absorption spectra measured in the temperature range of 290–470 K for two This paper's goal is to investigate how petroleum's n-alkane isomerization reaction occurs on the surface of a solid nanocatalyst. In this experiment, the isomerization of aromatic-rich crude oil is being studied as a result of radiation exposure. It was discovered that sodium-rich bentonite –solid acid, from the Alpoid deposit works well as a catalyst for refining petroleum. Possible mechanisms of the radio-catalytic reaction that converts n-alkanes to branched hydrocarbons have been researched by methods of FT- IR spectroscopy and gas chromatography. The effect of acidity type of the nanostructured bentonite clay on Lewis acid sites is proposed to explain observed results. Additional radiation energy, which is mainly generated due to the nanobentonite volume, results in a more efficient aromatic- rich petroleum radiolysis process. Also, it was investigated the dynamics of dose –dependent changes in the amount of branched hydrocarbons. One of the primary objectives of this research work is increasing the efficiency of transformation of adsorbed beam energy in solid phase to the system and improvement of hydrocarbon isomerization reactions in crude oil. The results of hydrocarbon transformations are discussed in terms of the behavior of excited electrons in solids. Novel eco-friendly nanocatalyst creates new opportunities for hydrocarbons with multi- branched structures.

**Keywords:** gamma irradiation, nanostructures, solid catalyst, multi- branched structures, transformation.

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### Introduction

Petroleum is complex mixture. This is generated by geochemical and chemical processes. Each type of petroleum has unique properties [1]. It impacts on the behavior of petroleum hydrocarbons under gamma radiation. Radiolysis of the petroleum is complicated [2-9]. Recently, it was studied the mechanisms of petroleum's hydrocarbons conversion and role of heterogeneous catalysts application in upgrading processes. Crude oil composition varies widely from one geographical region to another and from field to field. Petroleum recovered from different reservoirs varies

widely in compositional and physical properties [6]. Radiochemical properties in hydrocarbons of different classes differ from each other. Petroleum on platform 8 and 14 of Gunashli fields in X Horizon of Balakhani, Azerbaijan confirmed this fact too. The impact of gamma quantum on transformation mechanism of hydrocarbons from platform 8 in Gunashli oilfields, Azerbaijan studied [2]. But investigations with aromatic- rich petroleum from oil platform 14 at the same reaction conditions brought different results. Isomerization observed under gamma rays. That's why this research became on focus.

A number of authors studied ionizing irradiation application in enhancing branched hydrocarbons in

petroleum fractures [4-6]. Considerable effect in paraffin molecules observed when heavy aromatic hydrocarbons added to relatively lighter feedstock. This composition was irradiated with high-energy  $\gamma$ -rays [4]. Development novel catalyst gives opportunities in formation branched isomers [8, 9]. Presented work deals with investigation of radiocatalytic isomerization mechanism in presence of nanoclay catalyst.

Clays are of considerable interest due to interesting characteristics which make them suitable for industrial applications [10-16]. Among various types of clays, bentonite is more popular. Their physicochemical characteristics depends on montmorillonite content, clay minerals' crystalline structure, also interlayer cations' nature [17, 18]. Sodium - montmorillonite is relatively rare in occurrence in comparison with calcium-montmorillonite [19]. Gamma radiation may also adjust the surface characteristics of bentonite [2, 20, 21]. Natural clays are low-cost natural resource which is nontoxic to ecosystem.

## I. Experimental part

The  $^{60}\text{Co}$  isotope's gamma radiation was used to irradiate the crude oil samples under static conditions in vacuum-sealed quartz tubes at room temperature. 10.5 Rad/sec was the dosage rate. In the 4000-400  $\text{cm}^{-1}$  range, spectrophotometer measurements were made using a VARIAN 640-IR spectrophotometer. Gas chromatography was used on gas products. The chromatographic approach has been used at the "Gaschrom -3101" and "Svet -101" devices to determine the concentration of gases produced as a result of the radiolysis process. The instrument has a hydrogen sensitivity of  $K(\text{H}_2) = 6.0 \times 10^{13}$  molecules / ( $\text{cm}^3 \text{ mm}$ ). Reaction conditions: high-aromatic crude oil (2.0 g) was mixed with a specified amount of catalyst-nanobentonite sample (0.01-0.03 g) in glass ampules, sealed, and then exposed to various radiation energy doses (from 0.7 to 100 kGy).

Between 0.7 and 100 kGy of dose crude oil from Gunashli oilfield samples was irradiated at room temperature, both in presence and absence Na- bentonite clay. While employing raw bentonite clay from the Alpoid deposit of Azerbaijan and gamma rays at room temperature, it was possible to isomerize n-alkanes into branched hydrocarbons of high-aromatic crude oil from platform 14. About 15–25% of the total hydrocarbons in the crude oil from platform 14 may be aromatic hydrocarbons. The bentonite clay sample employed in these tests is nanostructured, with particles ranging in size from 55 d [nm] to 175 nm. The Alpoid deposit contains more than 85% of Na- montmorillonite [2; 21]. Montmorillonite is an aggregate of lamellar platelets, packed together by electrochemical forces and containing interposition water. Each platelet consists of three sandwich-arranged layers: a central octahedral alumina ( $\text{Al}_2\text{O}_3$ ) layer, and two tetrahedral silica ( $\text{SiO}_2$ ) layers. The silicon ion and the aluminium ion often undergo isomorphous substitutions by lower valence metals, such as magnesium and iron. In turn, these substitutions lead to a charge imbalance, compensated by exchangeable

cations, in particular calcium ( $\text{Ca}^{2+}$ ), magnesium ( $\text{Mg}^{2+}$ ) and sodium ( $\text{Na}^+$ ) ions, together with water molecules bonded together by ion-dipole forces. These ions, with no more place inside the reticular structure, migrate to the external silica layers and are the main cause of hydration in the crystal lattice [10;11;15;19]. The chemical formula for nanocrystalline bentonite is  $(\text{Na},0.5\text{Ca})0.7(\text{Al},\text{Mg},\text{Fe})_4(\text{Si},\text{Al})_8\text{O}_{20}(\text{OH})_4 \cdot \text{XH}_2\text{O}$  [2; 21].

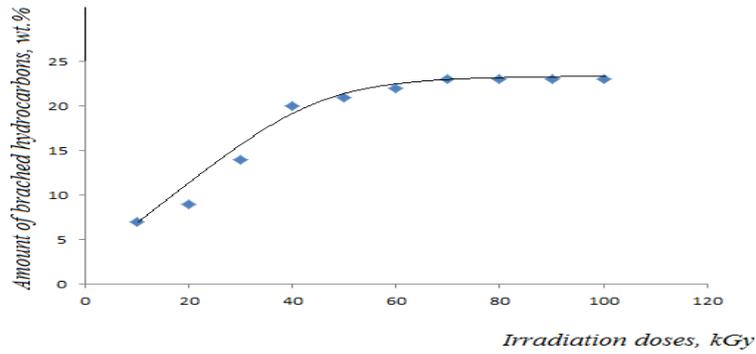
## II. Results and discussion

Crude oil isomerization was conducted with and without nanobentonite at a dose of between 0.7 and 100kGy at room temperature. It was observed that, during radiation of crude oil in presence of clay irradiated hydrocarbons contained much more isomers than without catalyst. The concentration of ( $\Sigma\text{C}_{4-7}$ ) branched hydrocarbons was determined using FT-IR methods. The obtained experimental results are represented in the figure 1(a, b). It has been presented results on the variation of the yield of the radiation – induced isomerization product with the  $\gamma$ -dose and amount of the clay.

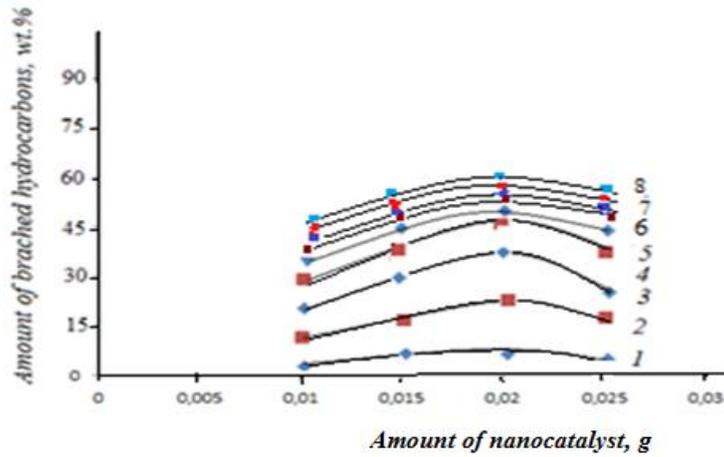
The amount of isoalkane increased significantly, in presence of nanocatalyst, as seen in Fig. 1b. Nanostructured Na-bentonite clay has different surface areas (55 - 175nm). A sharp growth of amount of branched hydrocarbons occurred with an increase in radiation dose ranging from 0.7 to 6.0 kGy and catalyst mass in the range of 0.01-0.02g. The results demonstrate that the number of branched hydrocarbons reaches its highest value (45 weight percent) at 6.0 kGy dose and 0.02g of the nanobentonite in this range of doses of gamma quantum. But the value of isomers began to decrease after adding more than 0.02g catalyst at various doses and it may be due to some of the agglomerations created with nanoparticles of clay. It was not observed significant rise in amount of branched alkanes at doses of 6 - 100 kGy.

The catalytic cycle of the Lewis acid catalyzed radiation-induced isomerization involves chain initiation to form the active carbenium ion species, carbenium ion rearrangement, and chain propagation. However, the value of isomers started to decline after adding more than 0.02g catalyst at various doses (from 0.7 to 100kGy). Both petroleum hydrocarbons and clay are made visible by gamma rays during heterogeneous crude oil radiolysis when it comes into contact with clay (Fig. 2. a,b). Hydrated aluminum silicates combine to make the clay mineral bentonite. As a result of being exposed to gamma rays, they partially lose their pore water and develop the Lewis acid  $\text{H}^+\text{AlO}_3$  [2].

Then the hydrocarbon molecule was dissociatively adsorbed on pairs of coordinatively unsaturated Al sites and acids. Lewis acidity thus undoubtedly plays a role in the radiation-based isomerization of crude oil. The carbocations are produced directly from n-alkane molecules in the presence of an acid catalyst, according to the theory put forward by G. Ola. The initial protonation was brought about by the production of the carbonium ion and the bond, which was predicated on the preexisting 3 central and 2 electron bonds. Following the release of a hydrogen molecule, carbonium ions decompose to create

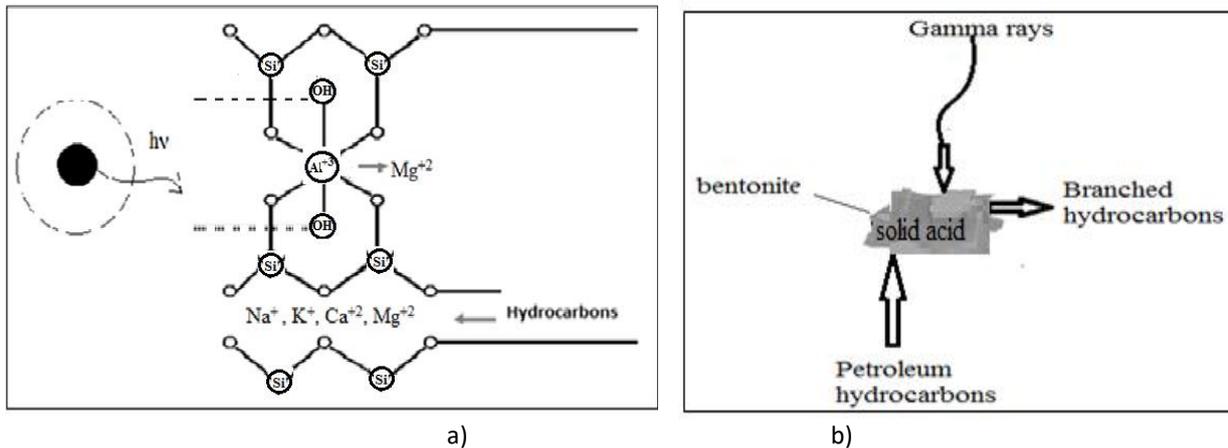


a)



b)

**Fig. 1.** a) - dependence of quantity of branched hydrocarbons and gamma quantum dose (irradiation without nanostructured sodium - bentonite clay); b) - dependence of quantity of branched hydrocarbons and the catalyst mass at doses: 1- 0.7 kGy; 2- 2.5kGy; 3- 4.5 kGy; 4- 6.0 kGy; 5- 20 kGy; 6- 40kGy; 7-70kGy; 8- 100kGy,  $m_{cat.} = 0.01-0.025$  g.



**Fig.2.** Transformation of hydrocarbons on the nanocatalyst (Na-bentonite): a) Hydrocarbons penetration in clay's pores; b) Formation of branched hydrocarbons on the surface of solid acid – bentonite.

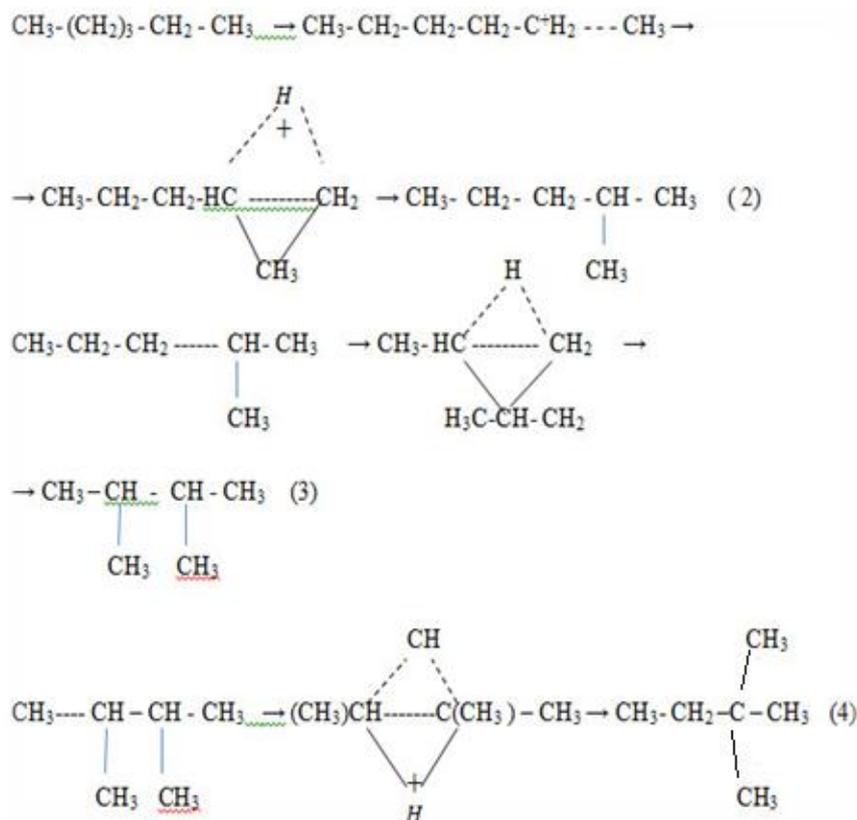
carbenium ions [22].

By using FTIR spectroscopy, it was shown that the C (CH<sub>3</sub>)<sub>2</sub> dimethyl substituted products predominated in the liquid mixture among the isomers. Where these kinds of

isomers come from was explored in the current scientific effort. The rearrangements, or 1, 2-shifts of alkyl groups, are typical for carbenium ions:



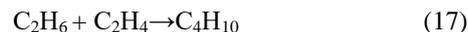
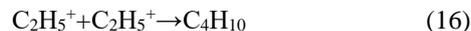
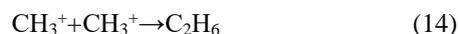
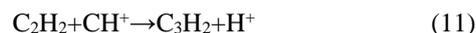
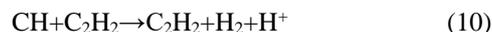
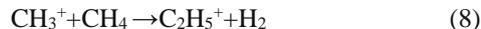
Equation 2- 4 could demonstrate one of the possible ways of isomerization process:



The decomposition products in the gas phase consist predominantly of hydrogen, which is likewise characteristic for low-temperature radiolysis of alkanes (table). Alkanes are subjected to radiolysis, which results in some of their dehydrogenation, the release of free hydrogen, the production of free radicals, and the generation of ions. These radical and ion products transformed into gases:  $\text{CH}_4$ ,  $\text{C}_2\text{H}_4$ ,  $\text{C}_2\text{H}_6$ ,  $\text{C}_3\text{H}_8$ ,  $\text{C}_4\text{H}_{10}$  (Fig.3. and Fig.4). The table shows that when crude oil was exposed to gamma rays, there was greater cracking than when bentonite was present. When isomerizing occurred with nanocatalyst, the yield of  $\text{CH}_4$ ,  $\text{C}_2\text{H}_4$ ,  $\text{C}_2\text{H}_6$ ,  $\sum\text{C}_3$  reduced,  $\sum\text{C}_5$ ,  $\sum\text{C}_6$ ,  $\sum\text{C}_7$  increased, and  $\text{C}_4$  amount very little changed.

As can be seen from the table more cracking occurs under gamma irradiation in crude oil than in presence of bentonite. In the case of nanobentonite, during isomerization, the yield of  $\text{CH}_4$ ,  $\text{C}_2\text{H}_4$ ,  $\text{C}_2\text{H}_6$ ,  $\sum\text{C}_3$  decreased,  $\sum\text{C}_5$ ,  $\sum\text{C}_6$ ,  $\sum\text{C}_7$  increased, only amount of  $\sum\text{C}_4$  changed slightly. On the other hand, it should be noted that isomerization on clay occurred due to alkanes rather than olefins, since the radiolysis of oil in the presence of nanoclay produces a relatively low yield of gaseous products.

The development of unbalanced molecular states required for the progression of chain reactions at lower temperatures is considerably aided by radiation-induced isomerization [23–25]. Different molecular fragments are produced by nuclear processes. Their concentration is relatively low, and they dissipate quickly (5–17):



and others.

Results from platform 14 of Gunashli crude oil point to the possibility that a significant number of aromatics in the feed may have a substantial negative impact on aromatization; yet, this chemical functions as a catalyst for the isomerization of n-alkanes in branched hydrocarbons. The synergism effect is produced by combining this kind of crude oil with nanostructured Na-bentonite clay.

Table.

Results of gas chromatographic studies at 6.0 kGy dose

Samples	Amount of components, g		Amount of formed gases output $N_i \cdot 10^{-16} \text{ molec. / cm}^3$								
	Aromatic-rich petroleum	Nanobentonite	H <sub>2</sub>	CH <sub>4</sub>	C <sub>2</sub> H <sub>6</sub>	C <sub>2</sub> H <sub>4</sub>	$\Sigma C_3$	$\Sigma C_4$	$\Sigma C_5$	$\Sigma C_6$	$\Sigma C_7$
1	2.0	-	8.45	0.057	0.010	0.048	0.007	0.005	0.045	0.195	0.056
2	2.0	0.02	10.96	0.036	0.009	0.023	0.004	0.005	0.067	0.278	0.082

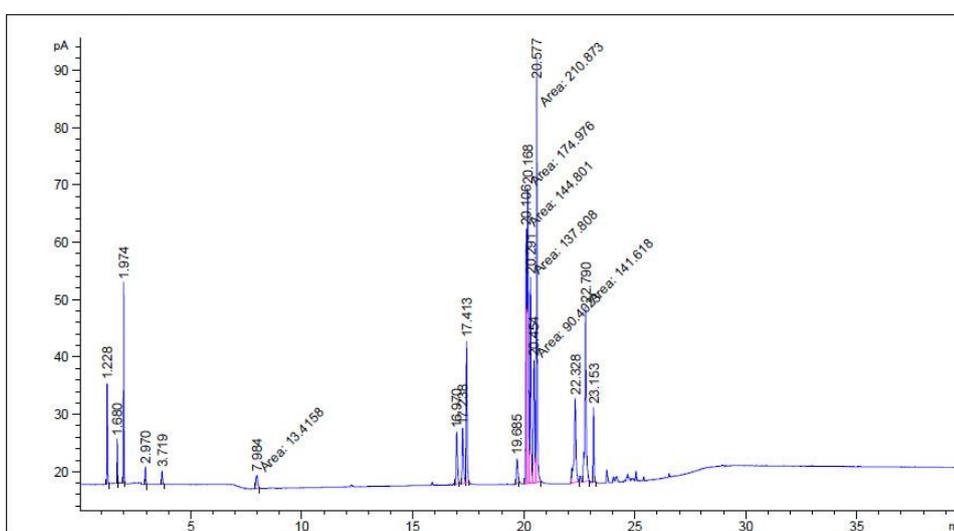


Fig. 3. Chromatogram of formed gases at petroleum radiolysis at 6.0 kGy dose.

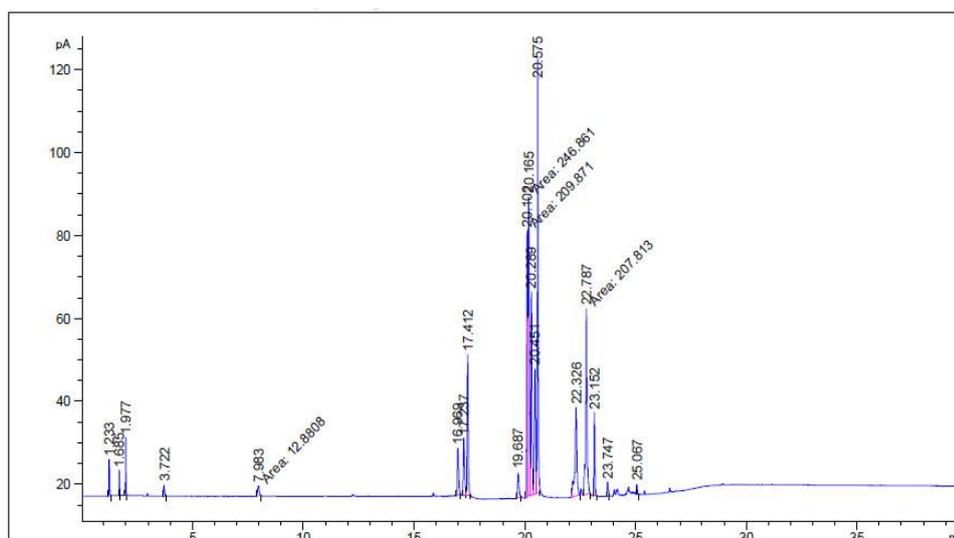


Fig. 4. Chromatogram of formed gases at petroleum radiolysis in presence of nanobentonite at 6.0 kGy dose.

## Conclusions

The study suggests that enhanced isomerization by ionizing radiation should take place when crude oil

contains considerable content of aromatics (about 20% wt.). This is a practical way to raise the oil's quality.

Presented work shows the possibility of using a cheap nanostructured catalyst in the low-temperature isomerization process.

The possible application of Lewis acidity-containing

Na-bentonite clay with a range of 55–175nm for isomerization is discussed.

The current study determined the optimal amount of catalyst, the optimal dose of irradiation in hydrocarbon isomerization reactions.

The mechanism of radiation-induced isomerization reaction was proposed using G. Ola's theory. The results of hydrocarbon transformations are discussed in terms of the behavior of excited electron solids.

#### Data Availability

Date is not shared because information based on date is already available in the paper.

#### Conflicts of Interest

The author fully declare that no conflicts of interest.

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## Механізми реакції радіаційної ізомеризації вуглеводнів на поверхні твердої кислоти

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У роботі виконано порівняльний аналіз крайових спектрів поглинання, виміряних у температурному діапазоні 290–470 К. Метою цієї статті є дослідження процесу реакції ізомеризації n-алканів нафти на поверхні твердої нанокаталізатора. У експерименті вивчається ізомеризація насиченої ароматичними речовинами сирої нафти в результаті радіаційного опромінення. Виявлено, що багатий натрієм бентоніт – тверда кислота з родовища Альпоїд – добре працює як каталізатор для переробки нафти. Методами ІЧ-Фур’є спектроскопії та газової хроматографії досліджено можливі механізми радіокаталітичної реакції, що перетворює n-алкани на розгалужені вуглеводні. Вплив типу кислотності наноструктурованої бентонітової глини на кислоти Льюїса пропонується пояснювати спостережувані результати. Додаткова енергія випромінювання, яка в основному генерується завдяки об’єму нанобентоніту, призводить до більш ефективного процесу радіолізу нафти, насиченого ароматичними речовинами. Також досліджено динаміку дозозалежних змін кількості розгалужених вуглеводнів. Однією із основних задач цієї наукової роботи є підвищення ефективності перетворення енергії адсорбованого пучка твердої фази в систему та вдосконалення реакцій ізомеризації вуглеводнів у сирій нафті. Результати вуглеводневих перетворень обговорено з точки зору поведінки збуджених електронів у твердих тілах. Новий екологічно чистий нанокаталізатор створює нові можливості для вуглеводнів з багаторозгалуженими структурами.

**Ключові слова:** гамма випромінювання, наноструктури, твердий каталізатор, багаторозгалужені структури, перетворення.