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ХАБАРЛАРЫ

ИЗВЕСТИЯ

НАЦИОНАЛЬНОЙ АКАДЕМИИ НАУК
РЕСПУБЛИКИ КАЗАХСТАН
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Қазақстан Республикасы Ұлттық ғылым академиясы "ҚР ҰҒА Хабарлары. Химия және технология сериясы" ғылыми журналының Web of Science-тің жаңаланған нұсқасы Emerging Sources Citation Index-те индекстелуге қабылданғанын хабарлайды. Бұл индекстелу барысында Clarivate Analytics компаниясы журналды одан әрі the Science Citation Index Expanded, the Social Sciences Citation Index және the Arts & Humanities Citation Index-ке қабылдау мәселесін қарастыруда. Webof Science зерттеушілер, авторлар, баспашылар мен мекемелерге контент тереңдігі мен сапасын ұсынады. ҚР ҰҒА Хабарлары. Химия және технология сериясы Emerging Sources Citation Index-ке енуі біздің қоғамдастық үшін ең өзекті және беделді химиялық ғылымдар бойынша контентке адалдығымызды білдіреді.

НАН РК сообщает, что научный журнал «Известия НАН РК. Серия химии и технологий» был принят для индексирования в Emerging Sources Citation Index, обновленной версии Web of Science. Содержание в этом индексировании находится в стадии рассмотрения компанией Clarivate Analytics для дальнейшего принятия журнала в the Science Citation Index Expanded, the Social Sciences Citation Index и the Arts & Humanities Citation Index. Web of Science предлагает качество и глубину контента для исследователей, авторов, издателей и учреждений. Включение Известия НАН РК в Emerging Sources Citation Index демонстрирует нашу приверженность к наиболее актуальному и влиятельному контенту по химическим наукам для нашего сообщества.

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Co-Mg-Mn COMPOSITE CATALYSTS FOR PARTIAL OXIDATION OF NATURAL GAS

Abstract. The problem of rational utilization of natural and associated petroleum gases and the cessation of their flaring is one of the acute and unresolved environmental problems. The aim of this work is to develop effective thermally stable catalysts of a new generation for the processes of oxidative conversion of light alkanes of natural and associated gas into synthesis gas. The results of partial oxidation of the methane of natural gas on the catalysts prepared by solution combustion synthesis are presented. Investigation of the activity of catalysts produced from initial mixture of $Co(NO_3)_2$ - $Al(NO_3)_3$ - $Mg(NO_3)_2$ - $Mn(NO_3)_2$ + urea of different composition was carried out for the production of synthesis-gas. It was found that the optimal conditions for producing of synthesis-gas are: CH_4 conversion - 98%, yield of target products: H_2 - 98 - 99% and CO - 40 - 43%, T = 900°C, space velocity - 2500 h^{-1}. The catalysts were studied by X-ray diffraction, transmission electron microscopy, specific surface area, pore volume and average pore diameter. The presence in the catalysts of simple and mixed oxides, metal aluminates and spinel-type structures, the presence of which contributes to the active work of catalysts for oxidative conversion of CH_4, has been established.

Key words: catalytic oxidation, methane, synthesis-gas, cobalt, magnesium.

Introduction. In the twenty-first century, humanity is faced with a global problem – Earth's climate change as a result of an increase in the concentration of greenhouse gases in atmosphere. The problem of greenhouse gases and their reduction is closely related to energy security, the stability of global energy markets and the sustainable development of each state and the world community as a whole. It is generally accepted that full reduction and mitigation of the effects of climate change is currently impossible, but measures must be taken to reduce the emissions of anthropogenic greenhouse gases. The study of the selective oxidative conversion of methane is consistent with Kazakhstan's development priorities in the use of its own hydrocarbons. According to forecasts, saturated hydrocarbons will not only preserve, but also strengthen their position as raw materials for the production of valuable organic compounds and fuel compositions, which will contribute to the revival of the national petrochemical industry. The activation of natural and associated petroleum gas (containing from 65 to 98% methane) for the targeted single-stage synthesis using nanoscale catalysts is one of the most relevant and important tasks in the field of organic catalysis. The gas processing plants of Kazakhstan are currently engaged mainly in the purification of gases from water, impurities of carbon dioxide and hydrogen sulfide for their use in domestic purposes. Therefore, there is a great economic incentive to develop effective catalysts for converting of natural gas into valuable products. So far, the production of synthesis gas is the only economically feasible way of converting methane into more valuable chemicals. The production of synthesis-gas from methane using active and stable catalysts plays an important role in the chemical and petrochemical industries. Prospects for the development of the global gas processing industry are associated with the creation and implementation of new catalytic environmentally friendly technologies for the production of synthesis-gas and hydrocarbons based on the production of fuel mixtures. Often the targeted production of synthesis-gas ($CO + H_2$) as an environmentally friendly modern fuel, as well as expensive olefins, is the first step in converting of natural gas.

For the above processes, oxide catalysts [1-3], their mixtures, and composites based on them [4,5] have been recently used instead of the noble metals [6,7] that were used previously. The method of self-propagating high-temperature synthesis has become widespread in recent years [8], especially its modification - the solution combustion synthesis (SCS) [9-11], as a result of which finely dispersed oxides and spinels are synthesized [12,13].

Previously, we investigated the process of oxidative conversion of light alkanes into synthesis-gas in the presence of oxygen on different types of catalysts: noble metals [14-16], oxides [17,18] and catalysts prepared by solution combustion synthesis [19-23].

The paper presents the data of the activity of the developed SCS catalyst based on Co - Al - Mg - Mn, capable of carrying out the process of oxidative conversion of methane into synthesis-gas.

Experimental

Catalyst preparation

SCS was used to prepare catalysts. A mixture of salts and distilled water was placed in a quartz glass. The content of the glass was heated to 80 - 100°C. Then the beaker with the solution was placed in a preheated muffle furnace, where the catalysts were prepared at various temperatures. At the beginning of the reaction, a large amount of heat is generated, which ensures a rapid spread of the combustion front and a sharp increase in temperature. After several minutes, structural catalysts are formed, the formation of which is one of the reasons for the high activity of the prepared samples.

Characterization techniques

The initial mixture and reaction products were analyzed using a Chromos GC-1000 chromatograph with the Chromos software. Chromatographic peaks were calculated using calibration curves constructed for the corresponding products using the Chromos software for pure substances. The catalysts were studied by XRD on a Siemens Spellman DF3 spectrometer with Cu-Kα ($\lambda = 1.5406E$) radiation in steps of 0.03°/1" in the 2θ range from 5° to 100°. For semi-quantitative X-ray analysis, 5% KCl was added to the analyzed samples as an internal standard. Transmission electron microscopy (TEM) was used to determine the morphology of the developed catalysts. The electron-microscopic characteristics of the catalysts were obtained on an EMK-125 K microscope (Sumy, Ukraine) at an accelerating voltage of 75 kV. The specific surface area was determined and the pore distribution in the catalysts was measured by the BET method (Brunauer–Emmett–Teller) using a GAPPV-Sorb 2800 analyzer. Nitrogen (99%) with helium (99%) was used as the carrier gas. Pore volume and average pore diameter were calculated by the BJH method using desorption isotherm curves.

Results and discussion. Catalysts of the Co - Al - Mg - Mn + urea series were prepared in a muffle furnace heated to 500°C. Three thermocouples were installed on top. All thermocouples were in a glass, which was located inside the muffle furnace. The first thermocouple was in the lower layer of the solution, the second thermocouple was in the middle layer and the third thermocouple was in the surface layer of the solution. Two combustion modes are carried out during the synthesis of catalysts by the solution combustion method: volumetric explosion and self-propagating mode. In the volume of the explosive mode, the solution is heated and the water evaporates. The gel is formed after the evaporation of water. The temperature in the muffle furnace gradually rises to a critical temperature. As soon as the temperature reaches critical, an exothermic reaction is carried out throughout the volume of the catalyst. Figure 1 shows the temperature-time profile of the volumetric combustion regime of the system 12.5% $Co(NO_3)_2$ + 12.5% $Al(NO_3)_3$ + 12.5% $Mg(NO_3)_2$ + 12.5% $Mn(NO_3)_2$ + 50% urea.

In the process of synthesis, the solution evaporates at T_o = 100°C and a gel is formed starting from temperature T_1 = 167°C, and a volumetric explosion occurs with T_2 = 270°C. The maximum temperature in the lower part reaches 564°C, in the middle part - up to 825°C, in the upper part reaches 1007°C. Table 1 shows the temperature at different levels of the solution in the synthesis of Al - Co - Mg - Mn catalysts with different contents of active components.

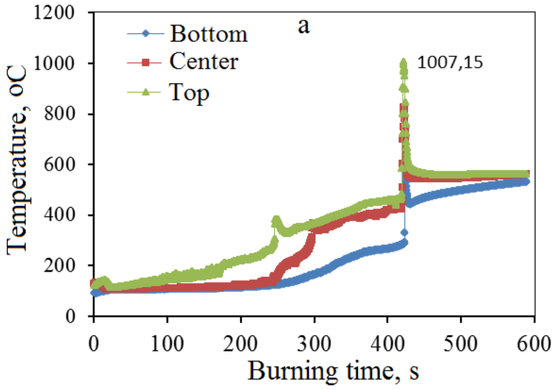

Figure 1 – Temperature-time profile of the volumetric combustion mode of the 12.5% Al - 12.5% Co - 12.5% Mg - 12.5% Mn + 50% urea system

Table 1 – Temperature in various layers during the synthesis of Al - Co - Mg - Mn catalysts with different contents of active components

The initial composition of the catalyst	T_o	T_1	T_2
12.5% $Al(NO_3)_3$ - 12.5% $Co(NO_3)_2$ - 12.5% $Mg(NO_3)_2$ - 12.5% $Mn(NO_3)_2$ + 50% $CO(NH_2)_2$	92.6	286.1	564.2
3% $Al(NO_3)_3$ - 3% $Co(NO_3)_2$ - 41% $Mg(NO_3)_2$ - 3% $Mn(NO_3)_2$ + 50% $CO(NH_2)_2$	97.4	280.4	659.98
3% $Al(NO_3)_3$ - 3% $Co(NO_3)_2$ - 3% $Mg(NO_3)_2$ - 41% $Mn(NO_3)_2$ + 50% $CO(NH_2)_2$	95.9	287.6	755.7
41% $Al(NO_3)_3$ - 3% $Co(NO_3)_2$ - 3% $Mg(NO_3)_2$ - 3% $Mn(NO_3)_2$ + 50% $CO(NH_2)_2$	97.3	284.0	523.6
3% $Al(NO_3)_3$ - 41% $Co(NO_3)_2$ - 3% $Mg(NO_3)_2$ - 3% $Mn(NO_3)_2$ + 50% $CO(NH_2)_2$	100.7	276.0	491.7

Table 1 shows the temperatures in the solution (only the lower part of the solution) during the synthesis of Al - Co - Mg - Mn catalysts with different element contents and shows the initial temperature (T_o), the ignition temperature (T_1), and the adiabatic flame temperature (T_2). The adiabatic flame temperature indicates the maximum combustion temperature achieved under adiabatic conditions.

Study of the activity of 12.5% Al - 12.5% Co - 12.5% Mg - 12.5% Mn + 50% urea catalysts prepared at 300 - 600°C by solution combustion synthesis in a flow catalytic installation was carried out at 900°C, GHSV = 2500 h^{-1}, 34% CH_4 + 17% O_2 + 49% Ar. The results of studies carried out during the synthesis of catalysts at temperatures from 300 to 600°C are presented in Figure 2.

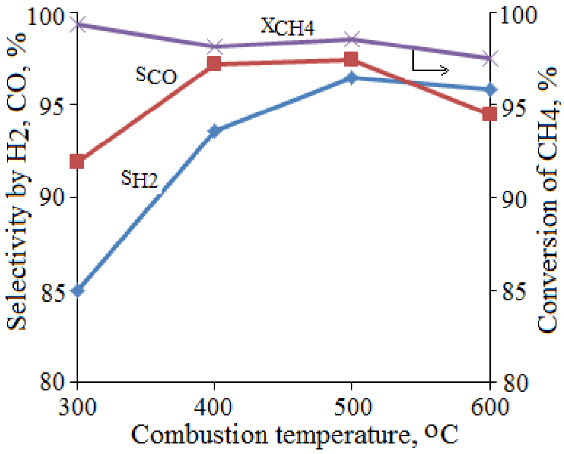

Figure 2 – Dependence of methane conversion and selectivity by H_2 and CO from the catalyst preparation temperature

The catalyst 12.5% Al - 12.5% Co - 12.5% Mg - 12.5% Mn + 50% urea prepared at 500°C showed high selectivity by H_2 (96.5%) and CO (97.4%). The methane conversion was 98.5%. Further, to determine the activity of the samples, the composition of the catalyst was varied. A series of catalysts prepared at 500°C by the solution combustion synthesis was studied using a PKU-1 apparatus. The catalytic reaction was carried out at atmospheric pressure and a temperature of 900°C. The experimental results are presented in Table 2.

Table 2 – Methane conversion and selectivity by H_2 and CO on catalysts of various compositions during the oxidation of methane to synthesis gas

Catalysts	X_{CH4}, %	Selectivity, %	
		H_2	CO
12.5% Al – 12.5% Co – 12.5% Mg – 12.5% Mn + 50% $CO(NH_2)_2$	94.7	95.4	99.4
41% Al - 3% Co - 3% Mg - 3% Mn + 50% $CO(NH_2)_2$	42.1	63.8	74.5
3% Al - 41% Co - 3% Mg - 3% Mn + 50% $CO(NH_2)_2$	99.7	87.9	96.9
3% Al - 3% Co - 41% Mg - 3% Mn + 50% $CO(NH_2)_2$	14.7	19.5	22.6
3% Al - 3% Co - 3% Mg - 41% Mn + 50% $CO(NH_2)_2$	32.4	42.9	46.9

As can be seen from Table 2, as a result of varying the content of elements in Al - Co - Mg - Mn catalysts, it was found that the catalysts exhibit the highest activity in the case of the highest content of aluminum and especially cobalt. Samples of 12.5% Al - 12.5% Co - 12.5% Mg - 12.5% Mn and 3% Al - 41% Co - 3% Mg - 3% Mn possess the maximum catalytic activity, while 41% Al A - 3% Co - 3% Mg - 3% Mn sample has lower activity. As a result of the analysis of the obtained results, a catalyst of the composition 20% Al - 20% Co - 5% Mg - 5% Mn was synthesized.

An analysis of the data indicates that the methane conversion values, as well as the H_2/CO ratios, are almost identical both at 1500 h^{-1} and at 2500 h^{-1}. The methane conversion reaches 94 - 98% in both cases. The $H_2/CO = 2$ is achieved at a space velocity of 1500 h^{-1}, starting from 800°C, which is favorable for syntheses of alcohols and olefins. The ratio slightly exceeds 2 at 2500 h^{-1}. At a space velocity of 2500 h^{-1}, the highest values were obtained for the yields of H_2 and CO. When implementing the process at 1500 h^{-1}, the hydrogen yield is reduced to 80%.

The phase composition of the Al - Co - Mg - Mn series of catalysts was studied by XRD. Figure 3 shows the XRD spectra of the catalysts.

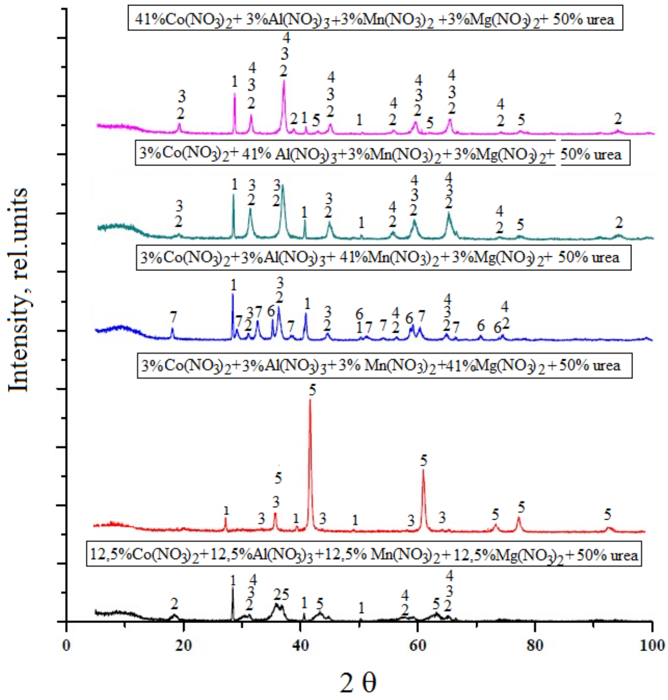

Figure 3 – X-ray phase analysis of catalysts of various compositions based on Al - Co - Mg - Mn + urea. 1 - KCl, 2 – Co_3O_4, 3 – $MgAl_2O_4$, 4 – $CoAl_2O_4$, 5 – MgO, 6 – MnO, 7 – Mn_5O_8

As a result of X-ray phase studies, it was found that the samples contained: Co_3O_4, $MgAl_2O_4$, $CoAl_2O_4$, MgO Periclase, MnO, and Mn_5O_8.

Table 3 presents the texture characteristics of Al - Co - Mg - Mn + urea catalysts with different element contents.

Table 3 – Textural characteristics of Al - Co - Mg - Mn + urea catalysts with different content of elements

Catalysts	Textural characteristics		
	Surface area, m^2/g	Pore volume, cm^3/g	The average pore size, nm
12,5% Al - 12,5% Co - 12,5% Mg - 12,5% Mn + urea	7.6	0.02	11.8
41% Al - 3% Co - 3% Mg - 3% Mn + urea	5.2	0.02	11.4
3% Al - 41% Co - 3% Mg - 3% Mn + urea	6.1	0.02	13.7
3% Al - 3% Co - 41% Mg - 3% Mn + urea	7.8	0.03	16.8
3% Al - 3% Co - 3% Mg - 41% Mn + urea	5.4	0.02	14.3
20% Al - 20% Co - 5% Mg - 5% Mn + 50% urea	22.2	0.03	18.5

The specific surface area of the catalysts is low. This is due to high combustion temperatures during preparation of catalysts. Despite this, the synthesized catalysts have a high specific activity, which allows them to compete even with catalysts of the Pt group.

Electron microscopy studies were carried out for the developed series of catalysts. For example, electron microscopic photographs of 3% Al - 41% Co - 3% Mg - 3% Mn + urea catalyst were obtained (Figure 4). The microdiffraction pattern of particles with a size of 20 - 40 nm, located on a thin film, is represented by reflections and corresponds to a mixture of phases: β-MnO_2, возможно, Co_2O_3. For samples with a size of 30 - 50 nm or more, the microdiffraction pattern is represented by reflections located along the rings and corresponds to a mixture of phases: MgO, (Co, Mg) O, ϵ-MnO_2, $MgH_{0.85}$, CoO.

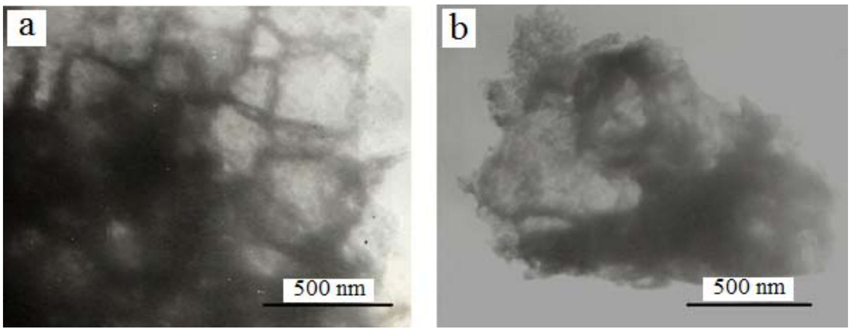

Figure 4 – TEM photographs of 3% Al - 41% Co - 3% Mg - 3% Mn + urea catalysts

From the obtained results it was found that the optimal conditions for producing of synthesis-gas are: CH_4 conversion - 98%, yield of target products: H_2 - 98 - 99% and CO - 40 - 43%, T = 900ºC, GHSV = 2500 h^{-1}.

Conclusion. Thus, it was found that the synthesized Al - Co - Mg - Mn + urea catalysts are active in the reaction of partial conversion of methane into synthesis-gas. The optimal conditions for the maximum operation of this catalyst are: a space velocity of 2500 h^{-1} at a temperature of 900°C. The presence in the catalysts of simple and mixed oxides, metal aluminates and spinel-type structures, the presence of which contributes to the active work of the catalysts for the oxidative conversion of methane, has been established.

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ТАБИҒИ ГАЗДЫ КОМПОЗИТТІ Co-Mg-Mn КАТАЛИЗАТОРЛАРЫНДА ПАРЦИАЛДЫ ТОТЫҚТЫРУ

Аннотация. Жиырма бірінші ғасырда адамзат атмосферада парниктік газдардың шоғырлану мөлшерінің өсуі нәтижесінде Жердегі климаттың өзгеруінің жаһандық проблемасына тап болды. Парниктік газдар және олардың таралу жағдайын бәсеңдету мәселесі энергетикалық қауіпсіздігі, жаһандық энергетикалық нарықтың тұрақтылығымен және мемлекеттер мен жалпы әлемдік қоғамдастықтың тұрақты дамуымен тығыз байланысты. Қазіргі уақытта климаттың өзгеру салдарын толық қалпына келтіру және оны жеңілдету жеңіл тимейді, дегенмен антропогендік парниктік газ шығарындысын азайту жөніндегі шараларды қабылдау қажет. Метанды талғамды тотықтыра айналдыруды зерттеу Қазақстанның өзіндік көмірсутектерін пайдаланудың даму басымдықтарына сәйкес келеді. Жасалынған болжамдар бойынша қаныққан көмірсутектер шикізат көзі ретінде ғана сақталмай, ұлттық мұнай-химия өнеркәсібін қайта жандандыруға ықпал ететін құнды органикалық қосылыстар мен отын композицияларын алу үшін өзіндік ұстанымдарын нығайтады. Нанөлшемді катализаторларды пайдалана отырып, мақсатты бір сатылы синтез үшін табиғи және мұнайға ілеспе газдарды (құрамында 65-тен 98%-ға дейін метан бар) активтендіру органикалық катализ саласындағы ең өзекті және маңызды міндеттердің бірі. Қазіргі уақытта Қазақстанның газ өңдеу зауыттары негізінен газды тұрмыстық мақсатта пайдалану үшін судан, көмірқышқыл газы мен күкіртсутегі қоспаларынан тазартумен айналысады. Сондықтан табиғи газды құнды өнімдерге айналдыру үшін тиімді катализаторларды әзірлеуге деген үлкен экономикалық қажеттілік бар. Әлі күнге дейін метанды анағұрлым құнды химиялық заттарға айналдырудың жалғыз экономикалық және қолжетімді жолы – синтез-газ өндірісі. Белсенді және тұрақты катализаторларды пайдалана отырып, метаннан синтез-газды алу химия және мұнай-химия өнеркәсібі үшін маңызды рөл атқарады. Әлемдік газ өңдеу өнеркәсібін дамытудың келешегі отын қоспаларын өндіруге негізделген синтез-газ бен көмірсутектерді алудың жаңа экологиялық таза катализдік технологияларын құруға және оны енгізуге байланысты. Қазіргі заманғы экологиялық таза отын ретінде синтез-газды ($CO+H_2$), сондай-ақ қымбат тұратын олефиндерді мақсатты өндіру табиғи газды түрлендірудегі басты қадам болып саналады.

Жоғарыда аталған үрдістер үшін бұрын пайдаланылған асыл металдардың орнына соңғы уақытта оксидті катализатор мен оның қоспалары негізіндегі композиттер қолданылуда. Соңғы жылдары өздігінен таралатын жоғары температуралы синтез әдісі, әсіресе оның модификациясы – ерітіндіде жану әдісі (SCS) арқылы жұқа дисперсті оксидтер мен шпинельдерді синтездеу кеңінен таралуда.

Ерітіндіде жану әдісімен Co - Al - Mg - Mn + несепнәр катализаторларының сериясы муфельдік пеште дайындалды. Катализаторларды ерітіндіде жану әдісімен синтездеу үрдісінде жанудың екі режимі жүзеге асырылады: көлемдік жарылыс және өздігінен таралу режимі. Көлемдік жарылыс режимі кезінде ерітінді алдымен біртіндеп қызады және құрамындағы су буланады. Су буланғаннан кейін гель пайда болады. Муфель пешіндегі температура біртіндеп шектік температураға дейін жоғарылайды. Температура шегіне жеткенде экзотермиялық реакция катализатордың барлық көлемі бойынша жүзеге асырылады.

Ерітіндіде жану әдісі арқылы 300 - 600°C температурада дайындалған 12,5% Al - 12,5% Co - 12,5% Mg - 12,5% Mn + 50% несепнәр катализаторларының белсенділігі ағымды катализдік қондырғыда зерттелді. Реакцияның шарты: 34% CH_4 + 17% O_2 + 49% Ar, көлемдік жылдамдық 2500 $сағ^{-1}$, ал реакцияның температурасы – 900°C. Температура 500°C кезінде дайындалған 12,5% Al - 12,5% Co - 12,5% Mg - 12,5% Mn + 50% несепнәр катализаторы H_2 (96,5%) және CO (97.4%) бойынша жоғары селективтілікті көрсетті. Метанның конверсиясы 98,5% көрсетті. Одан әрі дайындалған үлгілердің белсенділігін анықтау үшін катализатор құрамына түрлендіру жүргізілді. Құрамындағы элементтерді түрлендіру нәтижесінде дайындалған Al-Co-Mg-Mn катализаторларда ең көп мөлшерде алюминий, сонымен қатар кобальттың мөлшері көп болған жағдайда катализаторлар ең жоғары белсенділік танытатыны анықталды.

Синтез-газ алу үшін оңтайлы жағдай: T = 900°C пен W = 2500 $сағ^{-1}$ болғанда, CH_4-тің конверсиясы 98%, ал мақсатты өнімдердің шығымы: H_2 - 98 - 99% және CO - 40 - 43%, болатыны айқындалды. Сондай-ақ, катализатор құрамында метанның тотыға айналу жұмысына белсенді ықпал ететін қарапайым және аралас оксидтердің, металл алюминаттары мен шпинель түріндегі құрылымдар анықталды.

Түйін сөздер: катализдік тотығу, метан, синтез-газ, кобальт, магний.

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Co-Mg-Mn КОМПОЗИТНЫЕ КАТАЛИЗАТОРЫ ПАРЦИАЛЬНОГО ОКИСЛЕНИЯ ПРИРОДНОГО ГАЗА

Аннотация. В двадцать первом веке человечество сталкивается с глобальной проблемой изменения климата Земли в результате роста концентрации парниковых газов в атмосфере. Проблема парниковых газов и их сокращение тесно связаны с энергетической безопасностью, стабильностью глобальных энергетических рынков и устойчивого развития каждого государства и мирового сообщества в целом. Принято считать, что полное восстановление и смягчение последствий изменения климата в настоящее время невозможно, но необходимо принять меры по сокращению выбросов антропогенных парниковых газов. Изучение селективного окислительного превращения метана соответствует приоритетам развития Казахстана в использовании собственных углеводородов. По прогнозам, насыщенные углеводороды не только сохранят, но и укрепят свои позиции в качестве сырья для получения ценных органических соединений и топливных композиций, которые будут способствовать возрождению национальной нефтехимической промышленности. Активация природного и попутного нефтяного газа (содержащего от 65 до 98% метана) для целевого одностадийного синтеза с использованием наноразмерных катализаторов является одной из наиболее актуальных и важных задач в области органического катализа. Газоперерабатывающие заводы Казахстана в настоящее время занимаются в основном только очисткой газов от воды, примесей углекислого газа и сероводорода для их использования в бытовых целях. Поэтому существует большой экономический стимул в разработке эффективных катализаторов для превращения природного газа в ценные продукты. До сих пор единственным экономически доступным путем преобразования метана в более ценные химические вещества является производство синтез-газа. Получение синтез-газа из метана с использованием активных и стабильных катализаторов играет важную роль в химической и нефтехимической промышленности. Перспективы развития мировой газоперерабатывающей промышленности связаны с созданием и внедрением новых каталитических экологически чистых технологий получения синтез-газа и углеводородов, основанных на производстве топливных смесей. Первым шагом в преобразовании природного газа часто является целевое производство синтез-газа ($CO+H_2$) как экологически чистого современного топлива, а также дорогостоящих олефинов.

Для вышеперечисленных процессов в последнее время используют оксидные катализаторы, их смеси, композиты на их основе вместо благородных металлов, которые использовали ранее. Метод самораспространяющегося высокотемпературного синтеза получил распространение в последние годы, особенно его модификация – метод растворного горения (SCS), в результате которых синтезируются тонкодисперсные оксиды и шпинели.

Катализаторы серии Co - Al - Mg - Mn + мочевина были приготовлены в муфельной печи методом растворного горения. В процессе синтеза катализаторов методом горения в растворе осуществляется два режима горения: объемный взрыв и самораспространяющийся режим. В объеме взрывного режима раствор нагревается и вода испаряется. После испарения воды образуется гель. Температура в муфельной печи постепенно растет до критической температуры. Как только температура доходит до критической, экзотермическая реакция осуществляется по всему объему катализатора.

Было проведено исследование активности 12,5% Al - 12,5% Co - 12,5% Mg - Mn + 50% мочевина катализаторов, приготовленных при 300 - 600°C методом горения в растворе, в проточной каталитической установке. Условия реакции: 34% CH_4 + 17% O_2 + 49% Ar, объемная скорость 2500 $ч^{-1}$, температура реакции - 900°C. Катализатор 12,5% Al - 12,5% Co - 12,5% Mg - 12,5% Mn + 50% мочевина, приготовленный при 500°C, показал высокие селективности по H_2 (96,5%) и CO (97.4%). Конверсия метана составила 98,5%. Далее для определения активности образцов было проведено варьирование состава катализатора. В результате варьирования содержания элементов в Al - Co - Mg - Mn катализаторах было установлено, что в случае наибольшего содержания алюминия и в особенности кобальта катализаторы проявляют наивысшую активность.

Оптимальными условиями для получения синтез-газа являются: 98% конверсия CH_4, выход целевых продуктов: H_2 - 98 - 99% и CO - 40 - 43%, T = 900°C и W = 2500 $ч^{-1}$. Установлено присутствие в катализаторах простых и смешанных оксидов, алюминатов металлов и структур шпинельного типа, присутствие которых способствует активной работе катализаторов окислительного превращения метана.

Ключевые слова: каталитическое окисление, метан, синтез-газ, кобальт, магний.

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