Application of V$_2$O$_5$ in NO$_x$ reduction and evaluation of various effects on variation of its activity

S. Mehdi Alavi$^1$, Amir Vahid$^2$, S. Zahra Ebrahimzadeh Ganji, Mehdi Rashidzadeh, Vahid Andaji Garmaroudi, A. Mansour Moezi Azari

$^1$- Thermokinetic Research Laboratory, Chemical Engineering Department, Iran University of Science and Technology, Tehran, Postal Code: 16846-13114, Iran.  
$^2$- Department of Chemical and Biomolecular Engineering, The University of Akron, Akron, Ohio 44325-3906, USA

Abstract
In this research reduction of contaminant NO$_x$ by selective catalytic reduction was investigated. Catalysts were made of a mixture of carrier materials, such as titania or $\gamma$-alumina, and active components such as V$_2$O$_5$ and promoters such as La$_2$O$_3$, Ce$_2$O$_3$, WO$_3$, and MoO$_3$. Active metal and promoters were deposited on the support by adopting a wet impregnation method. For evaluation of factors effects such as V, La, Ce, W, Mo, loading, calcination time, temperature, support type and special manner of accomplish impregnation, 12 samples were made by experimental design method. For evaluation of specification of samples analytical techniques such as XRD, XRF, specific surface area measurements, pore size distribution and reactor test for evaluation of activity were used. It was observed that catalyst with $\gamma$-alumina support converted more NO$_x$ than catalyst with titania support. NO$_x$ conversion varies very low with increasing calcination time and Ce loadings. NO$_x$ conversion decreases with increasing of W and La loadings and also calcination temperature and increases with increasing of V and Mo loadings though (Maximum average of activity for 4% and 1% Vanadium was obtained 29.31 and 21.30 respectively). Finally single-stage operations converted more NO$_x$ than multiple operations.

Keywords: V$_2$O$_5$ catalyst, selective catalytic reduction, wet impregnation, evaluation of activity, NO$_x$ decomposition.

* Corresponding author 
Email address: alavi.m@iust.ac.ir (S. Mehdi Alavi) 
Tel:+98-21-73912715, Fax:+98-21-7896620
1 – Introduction

NOx, as one of the most important environmental pollutants, has always drawn specialist's attention. Some famous organizations like EPA, have worked on various methods of removal and reduction of NOx. Researchers in Germany and Japan have been more active in this field, since 1970 [1].

Control and reduction of NOx through mobile sources is often more difficult. It has been cleared that 60 percent of NOx is produced via stationary sources like fossil fuels and power plants, that in this case, the amount of output NOx can be reduced by improving operating condition.

Different reduction methods have been mentioned for reduction of NOx [3]. Among them, the selective catalytic reduction method (SCR) has more prosperity and fame and also more application. In SCR method, NOx, through a catalyst layer, is exposed to the air that is mixed with a large amount of ammonia or urea. V2O5 catalysts have shown better performance, among others. Recently, Vanadium catalysts (V) on TiO2 support are being used vastly in SCR reaction of NOx. Their high activity in conversion of nitric oxide in presence of oxygen and their resistance against high toxicity of sulfur oxides are the reasons of their extensive usage [3].

The selective catalytic reduction (SCR) is one of the most extensive methods used for reduction of NOx in stack gases. The most used support is TiO2 because of its resistance against sulfur toxicity and complete distribution of V2O5 on its surface are the main reasons of using it. However, titania has some defects such as expensiveness and relatively low surface area. Aluminum-titanium mixture oxides (Ti-Al) have also been examined and it has been cleared that aluminum increases catalyst's resistance against SO2 toxicity and Al2O3 adsorbs the produced surface sulfate species of vanadia which covers the active sites of vanadium. In SCR process, the existing NOx in stack gas reacts with ammonia, in presence of "vanadium on the support of titanium" catalyst, at 370°C, and forms nitrogen and water [4]. In this process, the catalyst is used in the form of V2O5/WO3-MoO3/TiO2. Yield of NOx reduction is directly proportional to NH3: NOx ratio. In high ratios of NH3: NOx, excess ammonia immediately combines with Sulfur Oxides existing in the gas mixture and produces ammonium sulfate that blocks the pores of catalyst [5]. In this research the active component in all of the provided catalysts was vanadium (V). Of course, other elements than vanadium, such as iron, platinum, chromium, manganese, cobalt, nickel, copper, barium, etc. are used by ammonia, in SCR reaction of NOx. But here, because of high resistance against being toxic by sulfur dioxide and high rate of NOx reduction in presence of oxygen vanadium is of great significance.

Also, the VOx species, as active sites, are more used for simplicity of reduction and react with vanadium in both oxidation states, V & IV [6]. In this research lanthanum nitrate, cerium nitrate, ammonium tungstate and ammonium molybdate, were used as promoters. These promoters have following benefits:
1. Lanthanum nitrate (La (NO$_3$)$_3$), a compound that is converted to La$_2$O$_3$ by calcination and improves catalyst performance in low temperature conversion of NO$_x$.

2. Cerium nitrate (Ce (NO$_3$)$_3$) is converted to Ce$_2$O$_3$ after calcination. Cerium, because of reversible conversion between electronic states Ce$^{3+}$ and Ce$^{4+}$, can absorb oxygen in reductive conditions and release it in oxidative conditions, for oxidation. Presence of CeO$_2$ also increases catalyst resistance against sintering, at high temperatures. It also makes the active metal, stable. All of these are influential on conversion of NO$_x$.

3. After calcination, ammonium tungstate is converted to WO$_3$ that is one of the most significant promoters with high efficiency. Activity reduction of SO$_2$ oxidation reaction is one of the most important reasons of using tungsten in commercial catalysts.

4. Ammonium molybdate is converted to MoO$_3$ after calcination. Adding molybdenum to commercial catalysts will increase NO$_x$ conversion, but reduces N$_2$ selectivity, as a result of N$_2$O production. It also shows more resistance against being activated by SO$_2$, and of course, has different characteristics in two cases of titania and vanadia support. It is cleared that activity of MoO$_3$–V$_2$O$_5$/Al$_2$O$_3$ catalyst is much more than V$_2$O$_5$/Al$_2$O$_3$ and it is also true for TiO$_2$ support [7].

3 - Experimental

3.1. Catalyst preparation

Providing catalysts was the first step in all experiments. Different parameters effect on the performance of catalysts used in NO$_x$ reduction, such as supporting materials, calcination time and temperature and impregnation method. Used materials in providing catalysts are given in Table 1.

<table>
<thead>
<tr>
<th>Used compounds</th>
<th>Purity (%)</th>
<th>Manufacturer</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiO$_2$</td>
<td>99.5</td>
<td>Riedel-de Haen</td>
</tr>
<tr>
<td>V$_2$O$_5$</td>
<td>99</td>
<td>Aldrich Chemical Co. Ltd</td>
</tr>
<tr>
<td>Ce (NO$_3$)$_3$.6H$_2$O</td>
<td>98.5</td>
<td>Merck</td>
</tr>
<tr>
<td>La (NO$_3$)$_3$.6H$_2$O</td>
<td>99</td>
<td>Merck</td>
</tr>
<tr>
<td>γ–Al$_2$O$_3$</td>
<td>98.5</td>
<td>Sasol</td>
</tr>
<tr>
<td>(NH$_4$)$_6$Mo$_7$O$_2$4.4H$_2$O</td>
<td>99</td>
<td>Fluka Chemika</td>
</tr>
<tr>
<td>(NH$<em>4$)$<em>6$W$</em>{12}$O$</em>{39}$.xH$_2$O</td>
<td>99.5</td>
<td>Aldrich Chemical Co. Ltd</td>
</tr>
<tr>
<td>NH$_4$VO$_3$</td>
<td>98</td>
<td>Interchem (u.k)</td>
</tr>
<tr>
<td>AlOOH</td>
<td></td>
<td>containing 75% Al$_2$O$_3$</td>
</tr>
<tr>
<td>Oxalic acid</td>
<td>99.5</td>
<td>Merck</td>
</tr>
</tbody>
</table>

According to the experimental design and statistical calculations, 12 samples of catalysts were prepared. Specifications of these catalysts are given in Table 2.
<table>
<thead>
<tr>
<th>Samples</th>
<th>Support</th>
<th>V₂O₃ (wt%)</th>
<th>La₂O₃ (wt%)</th>
<th>Ce₂O₃ (wt%)</th>
<th>WO₃ (wt%)</th>
<th>MoO₃ (wt%)</th>
<th>Calcination temperature (°C)</th>
<th>Calcination time (hr)</th>
<th>Impregnation Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>γ- Al₂O₃</td>
<td>4</td>
<td>10</td>
<td>10</td>
<td>1</td>
<td>11</td>
<td>450</td>
<td>3</td>
<td>Multi-stage</td>
</tr>
<tr>
<td>2</td>
<td>TiO₂</td>
<td>4</td>
<td>10</td>
<td>10</td>
<td>9</td>
<td>11</td>
<td>600</td>
<td>3</td>
<td>Multi-stage</td>
</tr>
<tr>
<td>3</td>
<td>γ- Al₂O₃</td>
<td>4</td>
<td>2</td>
<td>2</td>
<td>1</td>
<td>11</td>
<td>600</td>
<td>6</td>
<td>Single-stage</td>
</tr>
<tr>
<td>4</td>
<td>TiO₂</td>
<td>4</td>
<td>2</td>
<td>10</td>
<td>1</td>
<td>11</td>
<td>600</td>
<td>6</td>
<td>Multi-stage</td>
</tr>
<tr>
<td>5</td>
<td>TiO₂</td>
<td>1</td>
<td>10</td>
<td>2</td>
<td>1</td>
<td>11</td>
<td>450</td>
<td>6</td>
<td>Multi-stage</td>
</tr>
<tr>
<td>6</td>
<td>γ- Al₂O₃</td>
<td>1</td>
<td>10</td>
<td>10</td>
<td>9</td>
<td>1</td>
<td>600</td>
<td>6</td>
<td>Multi-stage</td>
</tr>
<tr>
<td>7</td>
<td>γ- Al₂O₃</td>
<td>1</td>
<td>10</td>
<td>2</td>
<td>1</td>
<td>11</td>
<td>600</td>
<td>3</td>
<td>Single-stage</td>
</tr>
<tr>
<td>8</td>
<td>γ- Al₂O₃</td>
<td>4</td>
<td>2</td>
<td>2</td>
<td>9</td>
<td>1</td>
<td>450</td>
<td>3</td>
<td>Multi-stage</td>
</tr>
<tr>
<td>9</td>
<td>γ- Al₂O₃</td>
<td>1</td>
<td>2</td>
<td>10</td>
<td>9</td>
<td>11</td>
<td>450</td>
<td>6</td>
<td>Single-stage</td>
</tr>
<tr>
<td>10</td>
<td>TiO₂</td>
<td>1</td>
<td>2</td>
<td>2</td>
<td>9</td>
<td>11</td>
<td>600</td>
<td>3</td>
<td>Multi-stage</td>
</tr>
<tr>
<td>11</td>
<td>TiO₂</td>
<td>1</td>
<td>2</td>
<td>2</td>
<td>1</td>
<td>11</td>
<td>450</td>
<td>3</td>
<td>Single-stage</td>
</tr>
<tr>
<td>12</td>
<td>TiO₂</td>
<td>4</td>
<td>10</td>
<td>10</td>
<td>9</td>
<td>1</td>
<td>450</td>
<td>6</td>
<td>Single-stage</td>
</tr>
</tbody>
</table>

Impregnation method was used to provide catalysts. Due to calculated amounts of each element, V, Mo, T, La, and Ce solutions were prepared. After preparing needed solutions, the catalyst can be provided. As an instance, steps of providing catalyst with support, for sample #1 are given in the following sequence:

First, certain amount of lanthanum and cerium solutions were poured in a beaker and needed amount of γ-Alumina powder (support) was added. Then the beaker was put in a sandbath at the fixed temperature of 60-70°C. When the water of the solution approximately vaporized, the sample was put into an oven at 90°C for 16 hours so that its water completely evaporates. The calcination process was performed in every step, in order to avoid having problem with support while adding other solutions. So the sample was then put in a kiln at 500°C for 4 hours with the heating rate of 5K/min. Then the calcined sample was poured into a vessel once more, and needed amount of tungsten and molybdenum solutions were added. After drying, in order to recalcination the sample was put in a kiln at 500°C for 4 hours. Again, the elements were converted to its oxides during calcination. The calcined sample was poured into the beaker and needed amount of Vanadium solution was added. Then it was put in a sandbath at 60-70°C so that its water evaporates. After that, it was put in the oven at 90°C for 16 hours in order to complete vaporization. Then the sample was placed in the kiln at 450°C for 3 hours for final calcination and the final sample was then prepared.

Other 11 samples were provided in the same manner, but the volumetric and gravimetric amounts of solutions and support, also temperature and calcination time were different.

### 3.2. Determining Catalysts' characteristics

XRF method was used for qualitative recognition of samples. The applied apparatus was XR 300, made by Link Co. equipped with Rh lamp to produce beam. First, the
catalyst samples were pulverized and preparation steps such as dissolving in acids and alkali fusion of insoluble materials in inorganic acids were carried out. Existing cations in the provided solutions were measured by atomic absorption apparatus. Gravimetric method was used for measuring W and V. Quantasorb apparatus, made by Quantachrome Co. was used to measure area of the catalyst samples. This apparatus is used for measuring area with BET method. BET method is for measuring specific area on the support of physical absorption of nitrogen in liquid nitrogen environment. Since the temperature of the liquid nitrogen is quite low, (-190°C), the sample absorbs nitrogen after operation. Amount of absorbed nitrogen is determined and BET formula is used for measuring specific area of the sample. X-ray diffraction pattern was provided by PW 1849 apparatus. Porosimeter 2000 apparatus was used for measuring diameter and volume of catalyst's pores, and volume of the pores was determined by plotting diagram.

Specific area of the catalyst samples was measured due to the BET method and the results are given in Table 3.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Specific surface area (m²/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 1</td>
<td>123.6</td>
</tr>
<tr>
<td>Sample 2</td>
<td>3.8</td>
</tr>
<tr>
<td>Sample 3</td>
<td>100.12</td>
</tr>
<tr>
<td>Sample 4</td>
<td>7.3</td>
</tr>
<tr>
<td>Sample 5</td>
<td>9.3</td>
</tr>
<tr>
<td>Sample 6</td>
<td>135.7</td>
</tr>
<tr>
<td>Sample 7</td>
<td>128.4</td>
</tr>
<tr>
<td>Sample 8</td>
<td>143</td>
</tr>
<tr>
<td>Sample 9</td>
<td>109</td>
</tr>
<tr>
<td>Sample 10</td>
<td>4.7</td>
</tr>
<tr>
<td>Sample 11</td>
<td>11.6</td>
</tr>
<tr>
<td>Sample 12</td>
<td>16.3</td>
</tr>
<tr>
<td>Commercial Sample VNX</td>
<td>12.8</td>
</tr>
<tr>
<td>Commercial Sample SK</td>
<td>68</td>
</tr>
</tbody>
</table>

SK and VNX samples are two commercial catalysts which were used to comparison with the prepared catalysts.

XRD experiment was performed with PHILIPS apparatus for determining the existing phases in the catalysts. The results with related spectrums are given in Table 4. XRD spectrums results for six catalysts with alumina support are shown in Fig. 1. VNX and SK commercial catalysts were used for determining elements of XRF experiment and the results are given in Table 5. Also in this table, components of catalyst #8 were determined by XRF, because it was not distinguished in XRD results.
Table 4
Existing phases in the catalyst sample and the commercial catalysts by XRD

<table>
<thead>
<tr>
<th>Sample</th>
<th>Phases</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 1</td>
<td>(\gamma)-Al(_2)O(_3), CeO(_2), La(_6)Mo(<em>2)O(</em>{14}), La(_4)MoO(_8)</td>
</tr>
<tr>
<td>Sample 2</td>
<td>TiO(_2) (Anatase), CeVO(_4), Ce(<em>8)Mo(</em>{12})O(_4), La(_2)O(_3), Ce(_4)W(<em>4)O(</em>{33})</td>
</tr>
<tr>
<td>Sample 3</td>
<td>(\gamma)-Al(_2)O(_3), La(_2)O(_3), Ce(<em>8)Mo(</em>{12})O(_4)</td>
</tr>
<tr>
<td>Sample 4</td>
<td>TiO(_2) (Anatase), CeO(_2)</td>
</tr>
<tr>
<td>Sample 5</td>
<td>(\gamma)-Al(_2)O(_3), CeO(_2), La(_2)O(_3)</td>
</tr>
<tr>
<td>Sample 6</td>
<td>(\gamma)-Al(_2)O(_3), La(_2)O(_3)</td>
</tr>
<tr>
<td>Sample 7</td>
<td>(\gamma)-Al(_2)O(_3)</td>
</tr>
<tr>
<td>Sample 8</td>
<td>(\gamma)-Al(_2)O(_3), CeO(_2), WO(_3)</td>
</tr>
<tr>
<td>Sample 9</td>
<td>TiO(_2) (Anatase), WO(<em>3), Mo(</em>{0.5})O(_3)</td>
</tr>
<tr>
<td>Sample 10</td>
<td>TiO(_2) (Anatase), CeO(_2)</td>
</tr>
<tr>
<td>Sample 11</td>
<td>TiO(_2) (Anatase), WO(_3), CeVO(_4)</td>
</tr>
<tr>
<td>Sample 12</td>
<td>TiO(_2) (Anatase), WO(_3), CeVO(_4)</td>
</tr>
<tr>
<td>Commercial Sample VNX</td>
<td>TiO(_2)</td>
</tr>
<tr>
<td>Commercial Sample SK</td>
<td>TiO(_2), Mg(_2)Al(_4)Si(<em>5)O(</em>{18})</td>
</tr>
</tbody>
</table>

Fig. 1. XRD spectrums for six catalysts with support of alumina

Table 5
Components of commercial catalysts, VNX and SK and catalyst #8 by XRF

<table>
<thead>
<tr>
<th>Sample</th>
<th>Components</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 8</td>
<td>Al, w, Mo, V, La, Ce</td>
</tr>
<tr>
<td>Commercial Sample VNX</td>
<td>Ti, Al, Si, Ca, V, Fe, Ni, W, Zr, Sr, Nb, Mo</td>
</tr>
<tr>
<td>Commercial Sample SK</td>
<td>Al, Si, Ti, V, Fe, W, Zr, Nb</td>
</tr>
</tbody>
</table>

Porosimeter 2000 apparatus was used for determining pore size and volume of the catalyst. The results are given in Table 6.
Table 6
Volume of made and commercial pores (cm\(^2\)/g)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Volume (cm(^2)/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 1</td>
<td>0.25</td>
</tr>
<tr>
<td>Sample 2</td>
<td>0.1</td>
</tr>
<tr>
<td>Sample 3</td>
<td>0.2</td>
</tr>
<tr>
<td>Sample 4</td>
<td>0.2</td>
</tr>
<tr>
<td>Sample 5</td>
<td>0.16</td>
</tr>
<tr>
<td>Sample 6</td>
<td>0.27</td>
</tr>
<tr>
<td>Sample 7</td>
<td>0.35</td>
</tr>
<tr>
<td>Sample 8</td>
<td>0.28</td>
</tr>
<tr>
<td>Sample 9</td>
<td>0.22</td>
</tr>
<tr>
<td>Sample 10</td>
<td>0.1</td>
</tr>
<tr>
<td>Sample 11</td>
<td>0.16</td>
</tr>
<tr>
<td>Sample 12</td>
<td>0.17</td>
</tr>
<tr>
<td>Commercial Sample VNX</td>
<td>0.24</td>
</tr>
<tr>
<td>Commercial Sample SK</td>
<td>0.38</td>
</tr>
</tbody>
</table>

3.3. Reaction tests

A reactor system in pilot scale was used in order to test the samples for controlling air pollutants emission. The reactor system was equipped with controllers and analyzer apparatuses. Flow controllers (MFC) which have been used in the system are from Brooks and High-tech models that, according to the existing situations, all of them were calibrated. A schema of reactor test system is given in Fig. 2. The gaseous feed mixtures (similar to composition of exhaust outlet gas) were provided from related highly pure gas capsules. In order to insert water into the gaseous mixture, an "Orion Sage" model syringe pump was used. Passing a preheater, the water enters the process gas mixture in the form of water vapor. It must be mentioned that the simulated gas mixture includes N\(_2\), O\(_2\), NO and water vapor. A kiln that can provide temperatures up to 1100 °C was used to simulate real conditions. The kiln temperature can be controlled by a TIC with the accuracy of 2 °C. Also a thermocouple was located in the catalyst bed. Other parts like condenser and gas meter were attached to the system.

To start the reactor test, first reactor was loaded with catalyst. The powdered catalyst was pelletized and then crushed and meshed (accumulated granules between 2 and 3 mesh sieves). Then the catalyst was heated (with the rate of 5 °C/min) in presence of N\(_2\) gas (2 lit/min). The kiln was adjusted in a way so that catalyst bed temperature reached 500 °C with the rate of 5 °C/min. After a catalyst residence of an hour in this temperature, process gas was crossed over the catalyst bed. When the operational condition was steady, the analysis (main test) begins and the input feed and output of catalyst bed were injected to the "Rega 7000" apparatus. The first analysis was accomplished at approximately 500 °C. Then bed temperature was declined and when temperature was fixed, test was carried out once more. This was done for each 50 °C of temperature decrease. Experiments were carried out in the range of 250-500 °C. Nitrogen gas (by pass) was crossed over the catalyst bed, while the feed was being analyzed.
Prepared samples with approximate size of 2-3 mm were placed in the reactor with quartz glass tube in the following manner: first, some annular $\alpha - \text{Al}_2\text{O}_3$ was poured into the tube. Then approximately 6 g of catalyst was mixed with 6 g of $\alpha - \text{Al}_2\text{O}_3$ and poured on the rings inside the tube. Again, the empty space above the catalyst bed was packed with cylindrical shape $\alpha - \text{Al}_2\text{O}_3$. Then performance of catalyst (amount of NOx conversion) was studied in a temperature range between 250°C and 500°C. In fact, an analysis of reactor input and output gas was performed, per 50°C temperature changes and conversion was calculated from the following equation:

$$X_{\text{NO}_x} = \frac{C_{\text{NO}_x,\text{in}} - C_{\text{NO}_x,\text{out}}}{C_{\text{NO}_x,\text{in}}} \times 100$$  \hspace{1cm} (1)$$

That $C_{\text{NO}_x,\text{in}}$ is concentration of incoming NOx and $C_{\text{NO}_x,\text{out}}$ is concentration of outlet NOx. GC apparatus has been calibrated several times for feeds and products to reach the essential accuracy for material analysis.
4. Results and discussion

As explained, 12 samples were prepared due to the experimental designs and some experiments were done for determining the characteristics of samples that were: measurement of specific surface area, measurement of pores size and porosity volume, XRD, XRF and reactor test for activity assessment that can be taken as response.

Characteristics of made catalysts are given in table 2 and their responses are given in table 7. The effects of these parameters on responses were studied as follows.

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Surface area (m$^2$/g)</th>
<th>Maximum NO$_x$ conversion</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>123.6</td>
<td>47</td>
</tr>
<tr>
<td>2</td>
<td>3.8</td>
<td>10.3</td>
</tr>
<tr>
<td>3</td>
<td>100.125</td>
<td>45.7</td>
</tr>
<tr>
<td>4</td>
<td>7.3</td>
<td>15.8</td>
</tr>
<tr>
<td>5</td>
<td>9.3</td>
<td>13</td>
</tr>
<tr>
<td>6</td>
<td>135.7</td>
<td>20</td>
</tr>
<tr>
<td>7</td>
<td>128.4</td>
<td>28.8</td>
</tr>
<tr>
<td>8</td>
<td>143</td>
<td>45.1</td>
</tr>
<tr>
<td>9</td>
<td>109</td>
<td>46</td>
</tr>
<tr>
<td>10</td>
<td>4.7</td>
<td>8</td>
</tr>
<tr>
<td>11</td>
<td>11.6</td>
<td>12</td>
</tr>
<tr>
<td>12</td>
<td>16.3</td>
<td>13</td>
</tr>
</tbody>
</table>

4.1. Effect of different parameters on catalyst activity for NO$_x$ conversion

In order to study the effect of support on area, 6 samples with $\alpha$–Al$_2$O$_3$ support and 6 other samples with TiO$_2$ support were considered. The average maximum activity for two cases with $\gamma$–Al$_2$O$_3$ and TiO$_2$ was 38.60 and 12.01 respectively. According to Fig. 3, when the support is $\gamma$–Al$_2$O$_3$, NO$_x$ conversion was more than the case where the support was TiO$_2$.

![Fig. 3 Effect of support on catalyst activity for NOx conversion](image-url)
In order to study vanadium's effect on catalyst activity, average maximum activity for 4% vanadium and for 1% Vanadium were obtained 29.31 and 23.10 respectively. It can be observed from Fig. 4 that with increasing vanadium percentage, catalyst activity for NOₓ conversion increases.

![Graph showing the effect of vanadium on catalyst activity for NOₓ conversion.]

Fig. 4. Vanadium effect on catalyst activity for NOₓ conversion

The average maximum activity for 10% lanthanum was 21.85 and was 28.76 for 2% lanthanum. It shows that catalyst activity and hence NOₓ conversion decreased with increasing lanthanum percentage as shown in Fig. 5.

![Graph showing the effect of lanthanum on catalyst activity for NOₓ conversion.]

Fig. 5. Effect of lanthanum on catalyst activity for NOₓ conversion

Adding cerium also reduced catalyst activity. This decline was observed from 25.60 for 2% cerium to 25.10 for 10% cerium. Fig. 6 shows that with increasing cerium percentage, NOₓ conversion decreased.
Achieved values for effect of tungsten on catalyst activity for 9% tungsten and 1% tungsten were 23.70 and 26.88 respectively. It shows that tungsten changes catalyst activity by 3 percent. With regards to Fig. 7, with increasing tungsten percentage, NOx conversion decreased.

Molybdenum like tungsten has relatively low effect on catalyst activity so that with increasing molybdenum percentage from 1% to 11%, maximum catalyst activity changed only from 28.16 to 22.45. According to Fig. 8, with increasing molybdenum percentage, NOx conversion increased.

Calcination temperature is one of the important factors that affect catalysts activity. In this work, study of effect of calcination temperature on catalyst activity showed that the average maximum activity at 450°C and 600°C were 29.18 and 21.43 respectively.

Fig. 9 shows that with increasing calcination temperature, NOx conversion decreased.
Calcination time did not change catalyst activity a lot. The average maximum activity in times 3 and 6 were 25.03 and 25.58 respectively. According to Fig. 10, it can be observed that with increasing calcination time, NO\textsubscript{x} conversion would increase a little.
Finally the effect of impregnation method on catalyst activity was studied. The average maximum activity for multiple operations was 24.65 and for synchronous or single-stage operations were 25.96.

Fig. 11 shows that in synchronous mode, NO\textsubscript{x} conversion was greater.

![Fig. 11. Effect of impregnation method on catalyst activity for NOx conversion](image)

5. **Conclusion**

In this research, effect of different parameters on NO\textsubscript{x} conversion percentage was studied and due to the entire experiments, we conclude that:

Examining the effect of support on catalyst activity showed that when alumina support was used, in comparison with titania, NO\textsubscript{x} conversion was greater. This can be explained due to the better specifications of alumina than titania, especially its specific area. Vanadium and lanthanum are also effective on catalyst activity; with increasing vanadium percentage, NO\textsubscript{x} conversion increased, whereas with increasing lanthanum percentage, NO\textsubscript{x} conversion decreased. This was because of greater activity of vanadium than lanthanum. With increasing cerium percentage, insignificant changes in NO\textsubscript{x} conversion occurred that can be neglected. With increasing tungsten and molybdenum percentage, NO\textsubscript{x} conversion decreased. Accordingly it seems that cerium did not have a significant effect on the promotion of catalyst; tungsten and molybdenum, not only did not promote catalyst, but also acted as inhibitors. With increasing the calcination temperature, NO\textsubscript{x} conversion decreased. The effect of calcination time on catalyst activity was also studied. With increasing calcinations time, NO\textsubscript{x} conversion changed insignificantly and can be neglected. Since the reactions were performed at the range of 250-500 °C, increasing calcination temperature more than 500 °C leads to reduction of NO\textsubscript{x} conversion, but increasing calcination time did not improve catalyst performance. Finally, effect of impregnation method on catalyst activity for industrial applications was studied. This study showed that NO\textsubscript{x} conversion in synchronous or single operation was a bit more than multistage mode.
References: