WOVEN STAINLESS STEEL WIRE MESH SUPPORTED CATALYST FOR NO\textsubscript{x} REDUCTION IN MUNICIPAL SOLID WASTE FLUE (MSW) GAS: SYNTHESIS AND CHARACTERIZATION

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Abstract

This paper presents the synthesis and characterization of a highly porous and well-adhere catalyst immobilised on a woven stainless steel wire mesh (WSSWM). The prepared catalyst is used to reduce oxide nitrogen, NO\textsubscript{x}, in MSW incineration flue gas. A circular wire mesh 90mm in diameter consisting of a hundred cells per square inch (cpsi) was used as the catalyst support. The surface of the WSSWM was pre-treated and passivated by sonicating with 1M inorganic acid for 30 minutes followed by rinsing with distilled water and drying at 50°C for 3 hours. The WSSWM was repeatedly dip-coated in a rheologically modified Al\textsubscript{2}O\textsubscript{3}-SiO\textsubscript{2} slurry until desired loading was attained. Impregnation of vanadium oxide on the wire mesh followed by drying at 102°C for 12 hours and calcining at 500°C for 5 hours, with a heating rate of 4°C/min, yielded the final NO\textsubscript{x} reduction catalyst. Qualitative porous analysis and the crystallinity of the impregnated catalyst were investigated using Scanning Electron Microscopy (SEM) and X-Ray Diffraction (XRD), respectively. The adhesion strength of the catalyst was examined using the in-house KH-Adhesion Test Method.

Results from catalyst characterization indicate its potential to be used for incinerator flue gas NO\textsubscript{x} reduction.

Introduction

Innovation in industrial NO\textsubscript{x} reduction catalysis (deNO\textsubscript{x}) technology has grown tremendously in recent years due to increasingly restrictive environmental regulations on mobile and stationary NO\textsubscript{x} sources [1]. The currently preferred methods employed for controlling air pollution from hydrocarbon post-combustion and selective catalytic reduction (SCR) of NO\textsubscript{x} from stationary and mobile sources use monoliths and pellet-packed beds [2].
Monoliths are typically made of ceramic or metallic substrate and shaped as honeycombs, which offer a larger surface area than pellet-packed beds, and are characterised by low cross-bed pressure drops even at high gas flow rates [3, 4]. Several recent studies have shown that wire mesh catalyst substrates are able to provide excellent mass and heat transfer rates coupled with higher mechanical strength and lower pressure drop than typical ceramic monolith honeycomb catalysts. Catalyst clogging and fouling, a major source of catalyst failure, is readily resolved since wire mesh catalysts can be disassembled and cleaned with relative ease. Wire mesh catalysts also offer greater conformational and loading flexibility, which makes industrial application easier in unit operations such as boilers, furnaces, power plants and incinerators. Implementation of catalytically impregnated wire meshes is a viable and cost-effective method for air pollution control [2, 3, 5-7]. Since the early 1990s wire mesh catalysts have been used commercially in the production of nitric acid from ammonia using Pt/Rh-catalysts for selective catalytic reduction (SCR) of NO\textsubscript{x} and the silver catalysed production of formaldehyde from methanol [3]. These catalysts possess low surface areas and are considerably more expensive, because they consist of homogenous bulk metal wires [6]. Ahlstrom-Silversand investigated the use of wire mesh gauze as a catalyst substrate for the treatment of flue gas from small-scale biofuel combustors and discovered mono and/or multi-layers of wire mesh catalyst gave better performance than commercial monoliths in the mass transfer controlled domain [8].

The objective of this work is to develop a deNO\textsubscript{x} catalyst impregnated on a WSSWM substrate and focuses on catalyst preparation and characterization. The primary goals of this study are to develop a highly porous and adhesive catalyst washcoat on the WSSWM surfaces using a specially formulated alumina-silica slurry, which includes SiO\textsubscript{2} binder and a rheological agent to enhance chemical bonds between the washcoat catalyst and WSSWM surface. The adhesive strength of the prepared catalyst was examined using an in-house ‘Adhesion Testing Device’, which employs the KH-Adhesion Test Method. Physical and chemical characteristics of the newly developed catalysts were determined and appraised from data acquired from SEM and XRD.

**Experimental**

**Washcoat Preparation**

A mixture consisting of industrial technical grade silica quartz, SiO\textsubscript{2}, and alumina, Al\textsubscript{2}O\textsubscript{3} was used as the catalyst support, washcoat. The Al\textsubscript{2}O\textsubscript{3}-SiO\textsubscript{2} mixture was ball milled for a minimum of 2 hours in order to reduce particle size and generate a suitable rheology for use in the subsequent coating process [4]. Pre-calcination was then performed on this mixture to remove any impurities. The washcoat particle size distribution was determined using a MALVERN Particle Size Analyser; the pre-calcined mixture has particle diameters in the range 5-160µm with a specific surface area of 428m\textsuperscript{2}/g.

The washcoat slurry was prepared from solid sodium metasilicate nonahydrate (SMS), Na\textsubscript{2}SiO\textsubscript{3}.9H\textsubscript{2}O from ACROS ORGANIC, which acted as an inorganic binder and rheological agent. An appropriate amount of SMS was completely dissolved using a mechanical stirrer in a specific quantity of deionised water in a one litre beaker. The Al\textsubscript{2}O\textsubscript{3}-SiO\textsubscript{2} mixture was gradually added to the SMS solution and continuously stirred for another 3 hours to obtain a homogeneous slurry. The slurry viscosity was continuously measured using a Brookfield Viscometer to determine the optimum slurry coating viscosity.

**WSSWM substrate preparation**

A circular WSSWM of average diameter 90mm and consisting of hundred cells per square inch (cpsi), Figure 1, was constructed from 0.9-1.0mm wire with a hydraulic diameter, \(D_h\), of 1.7mm. The WSSWM was pre-treated to clean and roughen the surface prior to coating. Multiple WSSWMs were immersed in an ultrasonicating containing five litres of 1M inorganic acid and sonicated at 30°C for 30 minutes. The sonicated WSSWMs were then thoroughly rinsed with distilled water and dried at 50°C for 3 hours.
Coating procedure
The WSSWM substrate was coated with slurry using an in-house Dip-Coater Machine at a rate of 12 cm/min. In order to ensure homogeneity of the coating the Al₂O₃-SiO₂ slurry was continuously agitated for the duration of the coating process. Any excess slurry on the WSSWM surface was immediately air-blown to evacuate the square dimension cells and prevent agglomeration at the woven site of the cells [4]. The coated WSSWM was left to dry at room temperature for 12 hours to elude rapid heat-up since water trapped in the coating is capable of developing sufficient pressure to crack the coating layer on the substrate [4]. The WSSWM was then dried at 102°C for 12 hours to remove excess water and other volatile species. Further careful thermal treatment was performed during calcination at 500°C for 5 hours at a heating rate of 4°C/min, which enables development of strong bonding between the washcoat and WSSWM surfaces.

Catalyst preparation
A precursor vanadium oxide catalyst, V₂O₅, was prepared by dissolving solid ammonium monovanadate, NH₄VO₃ (Fisher Scientific) in oxalic acid, H₂C₂O₄ (Fisher Scientific), at specific molar ratio [9]. The catalyst was loaded using wet impregnation, which involved immersing the washcoat in the ammonium monovanadate solution for 1 hour and then a further 24 hours. It was then dried at room temperature for 12 hours followed by further heat treatment at 102°C for 12 hour. The impregnated washcoat was then calcined under air flow at 500°C for 5 hours at heating rate of 4°C/min. The finished catalyst is henceforth referred to in the following manner V₂O₅/Al₂O₃-SiO₂ (xI), where Al₂O₃-SiO₂ corresponds to the alumina-silica support, x is the impregnation duration and I indicates the impregnation method.

Catalyst Characterisation
X-ray diffraction (XRD) of the alumina-silica loaded vanadium oxide catalyst was conducted using a Rigaku diffractometer equipped with a monochromatic Cu-Kα radiation source; wave length 1.5406Å. Catalyst samples were analysed from 3° to 90° using a step size of 3° and time step of 1 second to determine the degree of crystallinity. The XRD phases present in the catalyst samples were identified using PDF powder data files.

Adhesion Test Procedure
The most common technique to study the adhesion strength between a washcoat containing catalyst particles and the substrate surface is by applying high velocity air that simulates linear velocities inside the catalyst.
chamber [10]. The catalyst coating adhesion strength is determined by the catalyst percentage weight loss caused by erosion, degradation and vibration. In this study an in-house ‘Adhesion Testing Device’ referred to as the KH-Adhesion Test Method located in the UiTM Catalysis Laboratory was used to evaluate the strength of the bonds between the washcoat and the catalyst. According to Heck et al. [4] the adhesion strength of a catalysed washcoat can be evaluated by exposing the washcoat to high gaseous flowrates and rapid temperature fluctuations, which consequently leads to attrition. By measuring the amount of attrited particles the adhesion strength can be inferred; a large attrited particle mass means weak adhesion. The KH-Adhesion Test Method presented in Figure 2 consists of an air compressor, air flow meter, heating chamber employing a finned heating element, sample chamber, speed control vibrator and digitally controlled thermocouple, and has been used to infer the adhesive strength between the catalysed washcoat and WSSWM.

A series of experiments were designed to investigate the adhesion between the WSSWM and the vanadium oxide catalyst, Table 1. Five randomly chosen catalyst samples were independently tested as follows: the initial sample weight was recorded and the sample was then placed on washers and attached to the sample holder. The air was heated to the desired temperature, 250°C, and the sample was then vibrated for 30 minutes. The catalyst sample was then removed from the holder and reweighed; the weight difference represents the catalyst-WSSWM adhesion strength. Results were acquired at different hot air flow rates of hot air as tabulated in Table 1.

Figure 2 Schematic diagram of the Adhesion Testing Rig
Results and Discussion

Adhesion Strength

The following Table 1 presents the adhesion test results for the five randomly chosen catalyst samples:

<table>
<thead>
<tr>
<th>Samples</th>
<th>Initial Weight, $W_i$ (g)</th>
<th>Final Weight, $W_f$ (g)</th>
<th>Operating Air Flow Rate (l/min) x 100, $T = 250°$ C</th>
<th>Stdev</th>
<th>Avg % Weight Loss</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>11.4861</td>
<td>11.4857</td>
<td>11.4852</td>
<td>11.4853</td>
<td>11.4847</td>
</tr>
<tr>
<td>3</td>
<td>11.0802</td>
<td>11.0800</td>
<td>11.0797</td>
<td>11.0795</td>
<td>11.0794</td>
</tr>
<tr>
<td>4</td>
<td>10.9747</td>
<td>10.9743</td>
<td>10.9742</td>
<td>10.9739</td>
<td>10.9737</td>
</tr>
<tr>
<td>5</td>
<td>11.7562</td>
<td>11.7557</td>
<td>11.7554</td>
<td>11.7553</td>
<td>11.7549</td>
</tr>
</tbody>
</table>

The catalyst weight loss percentages are calculated according to the following equation:

\[
\% \text{ weight loss} = \frac{W_f - W_i}{W_i} \times 100\% \quad (\text{Eq 1.0})
\]

where $W_i$ is the initial catalyst sample weight and $W_f$ is the final catalyst sample weight.

According to the results presented in Table 1, little or no attrition took place even with increasing air flow rate, because there was no significant difference between the initial and final sample weights, Figure 3. These results are strongly supported by the value of standard deviation (Stdev) and average percent weight loss which is nearly zero. Almost constant pattern in the final weight of all samples revealed that a substantial strong bonding has been developed between catalysed washcoat and WSSWM substrate even though at considerably high hot air flow rate of 100 l/min. The lack of attrition indicates a strong degree of bonding between the catalysed washcoat and the WSSWM substrate.
XRD analysis
The $V_2O_5/Al_2O_3-SiO_2$ XRD patterns after 24 hours of impregnation is shown in Figure 4.

![Diffractogram $V_2O_5/Al_2O_3-SiO_2$ (24 hours Impregnation)](image1)

Figure 4: Diffractogram $V_2O_5/Al_2O_3-SiO_2$ (24 hours Impregnation)

![Diffractogram of pure $V_2O_5$](image2)

Figure 5: Diffractogram of pure $V_2O_5$

The $V_2O_5$ phase, which appears at very low intensity, corresponds to scherbinaite, syn (41-1426). The diffractogram of pure $V_2O_5$ contains a substantially greater number of sharp peaks between 10° and 50°, as shown in Figure 5, which indicates that $V_2O_5$ is dispersed on the $Al_2O_3-SiO_2$ support and as such is of insufficient quantity to be detected by XRD. The low intensity peaks in Figure 4 could be due a number of factors; pH, concentration, support surface properties of support, catalyst carrier content and pre-treatment conditions, which may contribute to a poor degree of impregnation [9].

Scanning Electron Microscopy (SEM)
Figure 6 presents a SEM micrograph showing the surface roughness of the uncoated WSSWM substrate; a major physical requirement for catalysed washcoat adhesion. Figures 7 and 8 present two further micrographs
of the V$_2$O$_5$/Al$_2$O$_3$-SiO$_2$ impregnated WSSWM surface after 1 and 24 hours, respectively. It is apparent that particles of irregular and varying sizes are unevenly distributed on the WSSWM surface, which leads to a high degree of porosity. This degree of porosity coupled with a rough catalytic surface, presented in Figures 7 and 8, provides a higher specific surface area, which is beneficial for catalytic activity. It also can be observed that the presence of varieties of pores on the surface as indicated by the black spot.

Figure 6: Treated WSSWM prior to coating

Figure 7: Micrograph of V$_2$O$_5$/Al$_2$O$_3$-SiO$_2$ impregnated for 1 hour
Figure 8: Micrograph of $\text{V}_2\text{O}_5/\text{Al}_2\text{O}_3-\text{SiO}_2$ impregnated for 24 hours

**Conclusion**

A WSSWM substrate was impregnated with $\text{V}_2\text{O}_5/\text{Al}_2\text{O}_3-\text{SiO}_2$ using dip-coating and calcination; the morphological and degree of crystallinity of the resulting catalyst surface was characterised using SEM and XRD. Analysis of the results indicates that the catalyst washcoat surface is highly porous, due to the presence of crystals of irregular and varying sizes, which also results in a high specific surface area. The strength of adhesion between the WSSWM substrate and the catalyst has been determined to be sufficient to withstand air flow rates of 50 l/min without attrition. The synthesized catalyst has potential application in the NO$_x$ abatement for large-scale stationary air pollution sources, such as municipal solid waste incinerators, boilers and power plants. Further investigation on $\text{V}_2\text{O}_5$ loading should be performed in order to determine the optimum loading prior to catalytic implementation.

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**References**

