WOVEN STAINLESS STEEL WIRE MESH SUPPORTED CATALYST FOR NO_X REDUCTION IN MUNICIPAL SOLID WASTE FLUE (MSW) GAS: SYNTHESIS AND CHARACTERIZATION

Kamariah Noor Ismail*, Ku Halim Ku Hamid, Sharifah Aishah S. A. Kadir, Mohibah Musa and Robert Mikhail Savory

Faculty of Chemical Engineering, Universiti Teknologi MARA (UiTM), 40450 Shah Alam, Selangor, Malaysia

**Corresponding author email: knoor@salam.uitm.edu.my*

Keywords: wire mesh, Al_2O_3-SiO_2, impregnation, dip-coating, NO_x reduction, flue gas

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_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_2O_3-SiO_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_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_x reduction.

Abstrak

Kertas kerja ini membentangkan sintesis dan pencirian mangkin yang mempunyai keporosan tinggi dan tahap rekatan baik yang dipegunkan di atas permukaan jejaring dawai tahan karat. Mangkin ini digunakan untuk proses penurunan sebatian nitrogen oksida, NO_x dalam gas serombong insinerator sisa pepejal bandaran. Jejaring dawai berbentuk bulat yang bergaris pusat 90.00 mm mengandungi seratus sel setiap inci persegi digunakan sebagai bahan pegun mangkin. Proses pra-rawatan dilakukan ke atas sampel-sampel jejaring dawai dimana ianya digetarkan dalam asid inorganik berkepekatan 1.0 M selama 30 minit untuk menyah-aktifkan permukaannya, diikuti dengan bilasan air suling dan dikeringkan pada suhu 50°C selama 3 jam. Proses penyalutan-celup di dalam buburan Al_2O_3-SiO_2 diulang beberapa kali sehingga jumlah endapan yang diperlukan dicapai. Pemegunan mangkin vanadium oksida dilakukan dalam larutan vanadia diikuti dengan pengeringan pada suhu 102°C selama 12 jam dan seterusnya dikalsinkan pada 500°C selama 5 jam pada kadar 4°C/min menghasilkan mangkin untuk penurunan sebatian nitrogen oksida, NO_x . Analisis terhadap keporosan dan penghabluran secara kualitatif dikaji dengan menggunakan Mikroskop Pengimbasan Elektron (SEM) dan Pembelauan sinar-x (XRD). Darjah kekuatan rekatan mangkin pada permukaan jejaring dawai dikaji menggunakan alat yang dibangunkan sendiri ditermakan sebagai *Kaedah Uji Rekatan-KH*. Keputusan pencirian yang dipeolehi menunjukkan mangkin yang dibangunkan mempunyai potensi untuk digunakan sebagai mangkin penurunan sebatian nitrogen oksida, NO_x dalam gas serombong insinerator.

Introduction

Innovation in industrial NO_x reduction catalysis (deNO_x) technology has grown tremendously in recent years due to increasingly restrictive environmental regulations on mobile and stationary NO_x sources [1]. The currently preferred methods employed for controlling air pollution from hydrocarbon post-combustion and selective catalytic reduction (SCR) of NO_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_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_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_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 fromdata acquired from SEM and XRD.

Experimental

Washcoat Preparation
A mixture consisting of industrial technical grade silica quartz, SiO_2, and alumina, Al_2O_3 was used as the catalyst support, *washcoat*. The Al_2O_3-SiO_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 perfomed 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^2/g.

The *washcoat* slurry was prepared from solid sodium metasilicate nonahydrate (SMS), $Na_2SiO_3.9H_2O$ 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_2O_3-SiO_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 ultrasonicater 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.

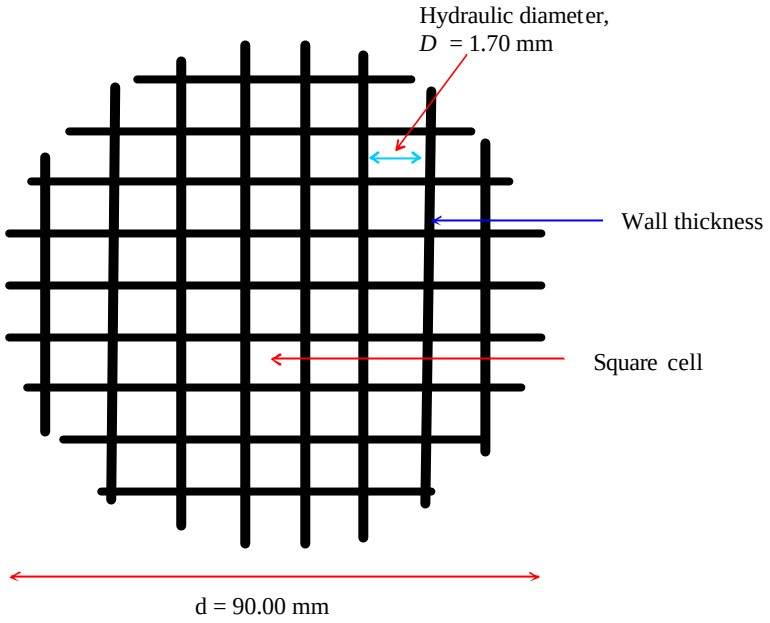

Figure 1: Circular woven stainless steel wire mesh substrate

Coating procedure

The WSSWM substrate was coated with slurry using an in-house Dip-Coater Machine at a rate of 12cm/min. In order to ensure homogeneity of the coating the Al_2O_3-SiO_2 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 precursory vanadium oxide catalyst, V_2O_5, was prepared by dissolving solid ammonium monovanadate, NH_4VO_3 (Fisher Scientific) in oxalic acid, $H_2C_2O_4$ (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 V_2O_5/Al_2O_3-SiO_2 (x*I*), where Al_2O_3-SiO_2 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-K_α radiation source; wave length 1.5406Å. Catalyst samples were analysed from 3º to 90º using a step size of 3º 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.

The *washcoat* surface morphology was investigated using magnified images and elemental analysis acquired using a Jeol JSM-6360LA scanning electron microscope. SEM was performed on 2.5cm square catalyst samples using an accelerating voltage and current of 15kV and 30mA, respectively.

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.

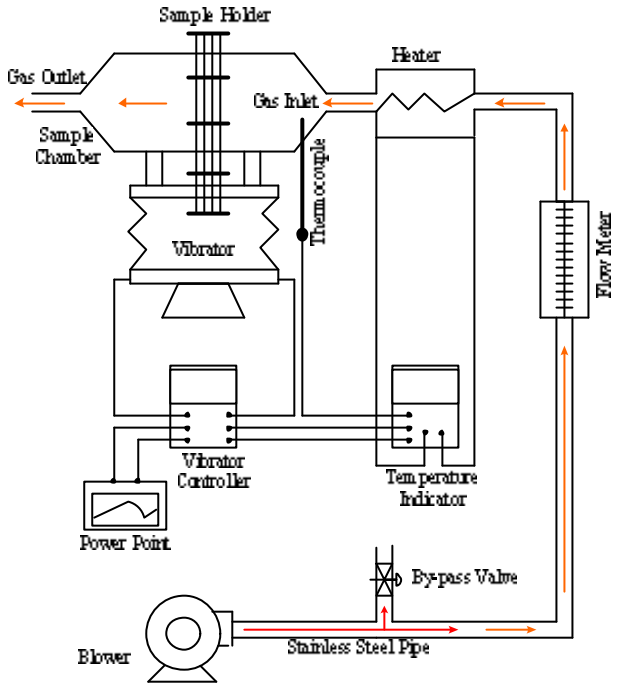

Figure 2 Schematic diagram of the Adhesion Testing Rig

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.

Results and Discussion

Adhesion Strength

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

Table 1: Experimental results for the adhesion strength tests

Samples	Initial Weight, W_i (g)	Final Weight, W_f (g) Operating Air Flow Rate (l/min) x 100, T = 250° C					Stdev	Avg % Weight Loss
		0.2	0.4	0.6	0.8	1.0		
1	11.4861	11.4857	11.4853	11.4852	11.4853	11.4847	3.58E-04	7.49E-03
2	12.0237	12.0232	12.0229	12.0227	12.0228	12.0224	2.92E-04	7.49E-03
3	11.0802	11.0800	11.0797	11.0795	11.0796	11.0794	2.30E-04	5.05E-03
4	10.9747	10.9743	10.9742	10.9739	10.9737	10.9735	3.35E-04	7.11E-03
5	11.7562	11.7557	11.7554	11.7553	11.7551	11.7549	3.03E-04	7.83E-03

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

$$\% \text{ weight loss} = \frac{W_f - W_i}{W_i} \times 100\,\% \qquad \text{(Eq 1.0)}$$

where Wi is the initial catalyst sample weight and Wf is the final catalyst sample weight.

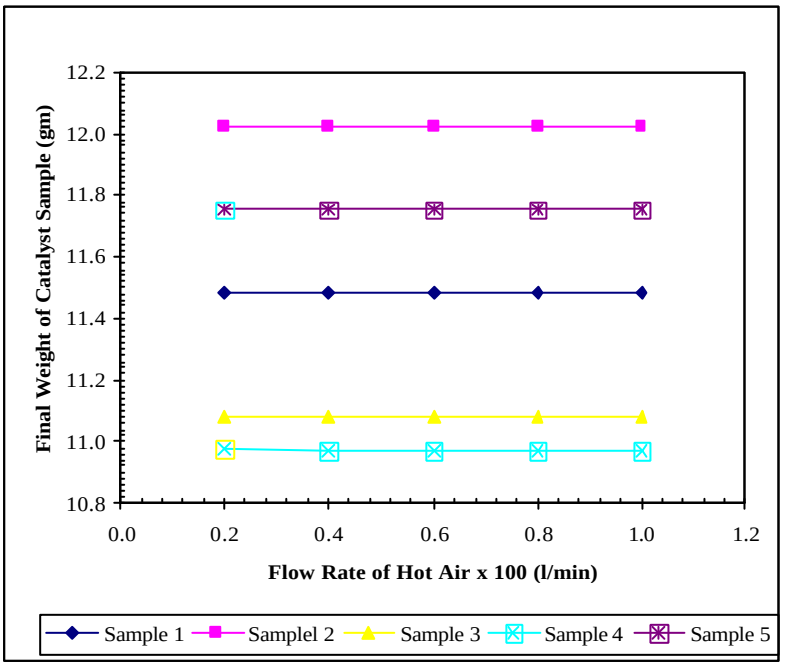

Figure 3: Plot of final weight after adhesion test verses hot air flowrate

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.

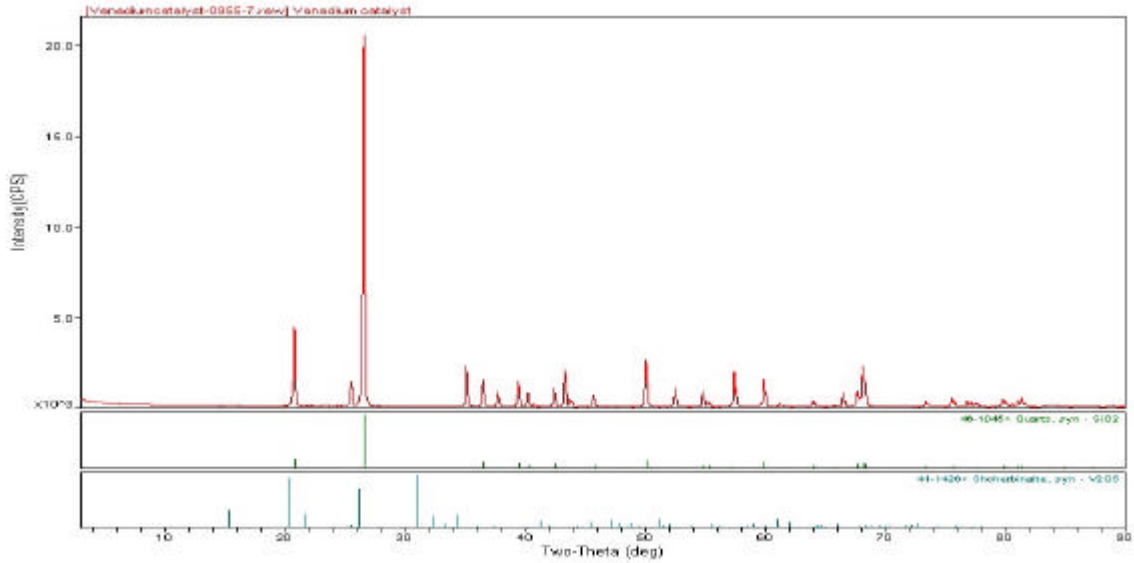

Figure 4: Diffractogram V_2O_5/Al_2O_3-SiO_2 (24 hours Impregnation)

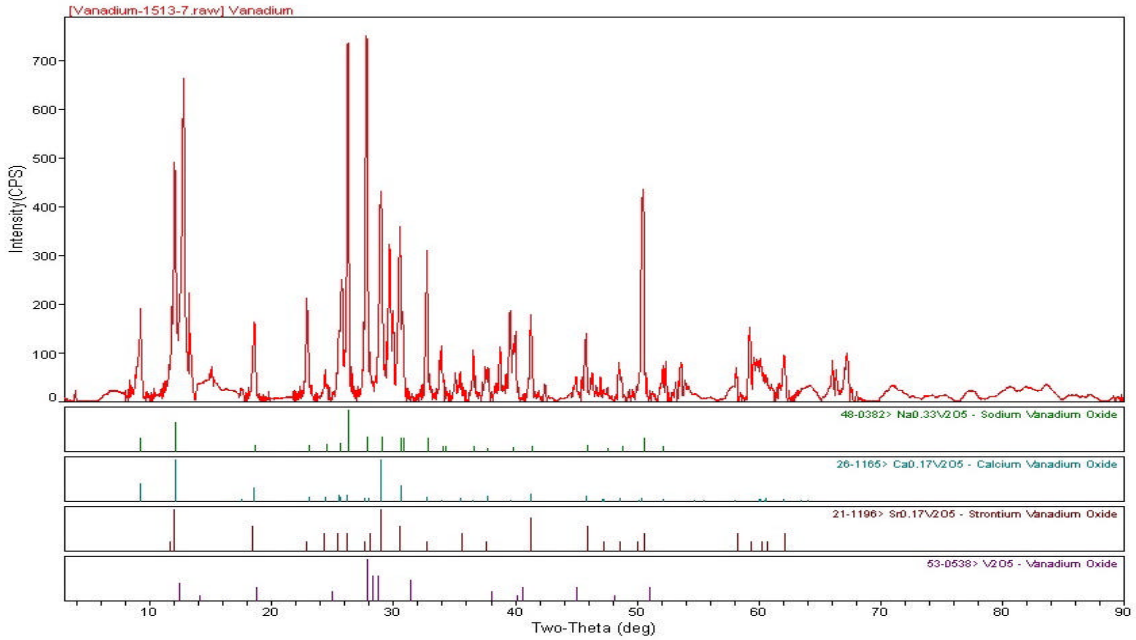

Figure 5: Diffractogram of pure V_2O_5

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_2O_5/Al_2O_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.

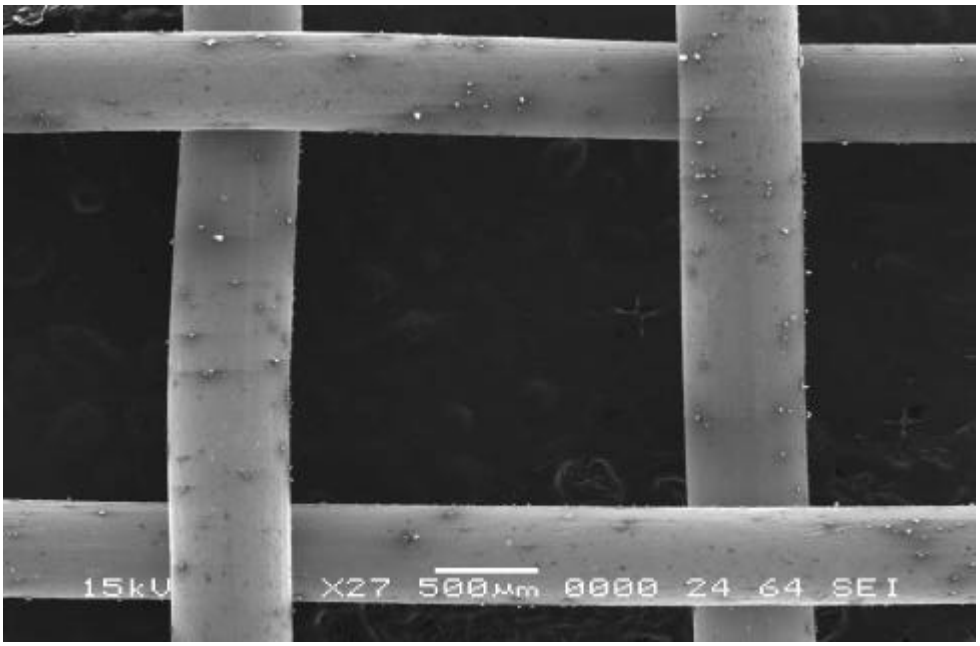

Figure 6: Treated WSSWM prior to coating

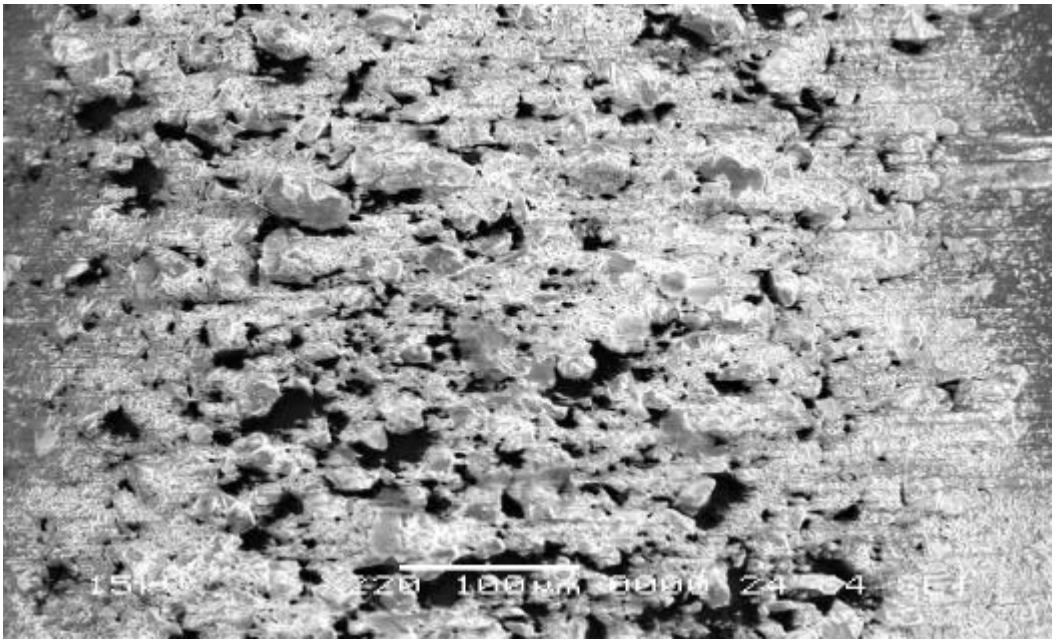

Figure 7: Micrograph of V_2O_5/Al_2O_3-SiO_2 impregnated for 1 hour

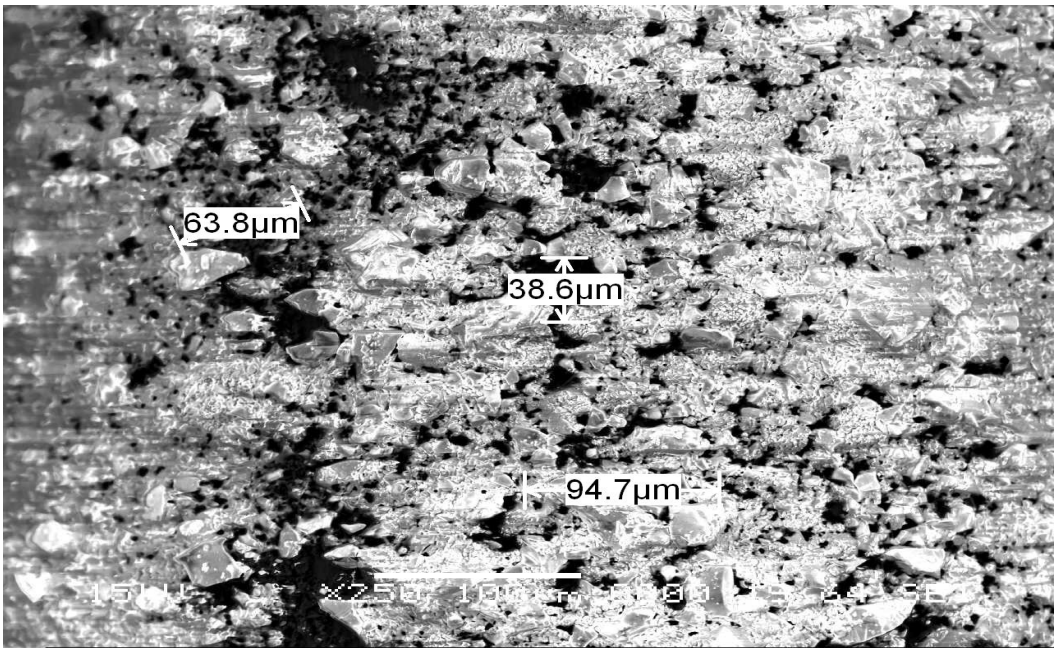

Figure 8: Micrograph of V_2O_5/Al_2O_3-SiO_2 impregnated for 24 hours

Conclusion

A WSSWM substrate was impregnated with V_2O_5/Al_2O_3-SiO_2 using dip-coating and calcination; the morphological and degree of crystallinity of the resulting catalysed surface was characterised using SEM and XRD. Analysis of the results indicates that the catalysed *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 100 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 V_2O_5 loading should be performed in order to determine the optimum loading prior to catalytic implementation.

Acknowledgement

The authors gratefully acknowledged the financial supports of the Institute of Research and Commercialization (IRDC) of Universiti Teknologi MARA (UiTM), research grant: 600-IRDC/ST.5/3/717

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