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# A novel technique for quantifying the cohesive strength of washcoat

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ABSTRACT

The washcoat used as the catalyst carrier in catalytic converters is designed to enlarge the contact area between exhaust gas and catalysts. The cohesive and adhesive strength of a washcoat layer is therefore highly important for the service life of catalytic converters. However, in the present literature, there is no direct method to measure these strengths distinctively. Given the current knowledge gap, this paper was written to present a novel technique which was able to separately quantify the cohesive strength of a washcoat layer based on stress measurements. In this method, a washcoat sample made of wet-milled γ-alumina was formed in the shape of a tablet. The cohesive strength of a washcoat layer can be obtained by measuring the tensile strength of these tablets. The tablets prepared using a suspension with pH higher than the isoelectric point of γ-alumina and under low drying rates were found to be the strongest. The reason behind this observation was determined to be that at high pH the particles in suspension were more mobile to travel to favourable packing sites, to form a layer, and low drying rates granted more time for the packing to complete. The fact that the cohesive strength obtained can be logically explained supported the validity of the new method.



## 1. Introduction

Catalytic converters are widely employed in vehicles with the aim of reducing harmful emissions from engines. Catalysts employed for these conversion reactions are fixed onto catalytic converters using a carrier. The catalyst carrier is a coated layer, and its role is to distribute catalyst particles, so that the contact area between catalyst particles and exhaust gas is enlarged. The increased contact area is known to effectively raise the rate of reaction (Degenstein et al., 2006). A large geometric area is also achieved by using a monolith which has a honeycomb structure. The monolith can be made of either metals or ceramics and a washcoated catalyst carrier layer (usually of a thickness less than 200 μm) that contains a catalyst is applied on the surface of the honeycomb channels.

In respect of this, the strength of the coated layer becomes a key parameter for the performance of catalytic converters. The activity of catalysts will be lowered if the washcoat layer suffers from mechanical failure due to any one or a combination of high temperature, high velocity gas flow in the monolith channels and vehicle vibration. Two types of strength are important for a washcoat layer and these are cohesion and adhesion. The cohesive strength of a washcoat layer describes the bonding between particles in the layer; the adhesive strength is determined by the bonding between these particles and the substrate (in this case, the monolith).

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**Table 1 – Summary of existing techniques to measure the strength of a washcoated layer (part 1).**

| Method | Operating principle |
|---|---|
| Ultrasonic bath | A substrate coated with a washcoat layer is immersed in an ultrasonic bath. The strength of the washcoated layer is measured as the mass loss from the ultrasonic vibration |
| Simulated environment | A coated substrate is placed in a chamber that simulates the high temperature and high velocity gas flow conditions in a catalytic converter. The mass loss observed is again used to indicate the strength of the washcoated layer |
| Drop test | A washcoat layer that is coated on a chosen substrate is released from a set height. The percentage of mass lost from the washcoat upon impact is registered as the strength of the washcoated layer |
| Abrasive test | A coated layer is rubbed against an abrasive of a chosen grade. The mass of the washcoat removed by friction is used to describe the strength of the washcoat layer |
| Pull-off method | A washcoat layer fixed on a substrate is subjected to a normal pulling force. The work done to remove the coating is used as a measure of the strength of the layer |
| Scratch test | A stylus with a known dimension is employed to scratch a washcoated layer. The amount of force applied, the displacement travelled by the stylus and the resultant failure pattern can be used to calculate the strength of the layer |

However all existing methods published in the literature are either not able to give reproducible measurements for the strengths or differentiate between them; this is a significant obstacle in building a predictive model for the strength of a coated layer. The poor reproducibility arises from the operating principles of the measurement techniques. The strength of a washcoat layer is reported in mass loss after the coated layer has been subjected to a destructive environment such as immersion in an ultrasonic bath. Given that mass loss is not a direct measurement of the inter-particle bonding in a layer, its value will be highly sensitive to the environment in which the coated layer is tested, e.g. the magnitude of ultrasound used and the relative position of the sample to the source of ultrasound. Therefore without using the same testing environment, the mass loss based strength of a washcoated layer is not comparable between different publications. Another weakness in the existing measurement techniques is their inability to separately quantify cohesive and adhesive strength. This is caused by the uncontrolled breakage pattern of a washcoat layer in these methods. For example, in the scratch test, a washcoated layer is often found to fail in some parts from the layer itself and in some parts from the substrate in a single test when the stylus scratches across the coating, in which case a separate measurement of the two distinctive strengths cannot be obtained. A list of the current techniques to measure the strength of a washcoated layer is presented in Tables 1 and 2. A brief description of the operating principles of each method and the associated limitations are also provided.

In recognition of the limitations in the current strength measurement techniques, this paper aims to present a novel way of separately quantifying the cohesive strength of a coated layer. Measurements obtained from this method will also be based on a physically derived quantity, allowing different authors employing this method to be able to compare their results. In order to prove the validity of the method, two process parameters in the manufacturing process of the coated layers were chosen to be varied to see if the new method was able to produce meaningful results with respect to variation in the chosen parameters.

## 2. Experimental methods

### 2.1. *Preparation of γ-alumina suspension*

The washcoat was prepared by wet milling anhydrous γ-alumina. The milling was performed using an Eiger mini mill at the following conditions (Table 3).

The result of wet-milling was a suspension of alumina particles at a pH value of approximately 5.5. The particle size distribution in the alumina suspension after milling was measured by laser diffraction using a Malvern Mastersizer 2000

**Table 2 – Summary of existing methods to determine the strength of a washcoated layer (part 2).**

| Publication | Ultrasonic bath | Simulated environment | Drop test | Abrasive test | Pull-off method | Scratch test |
|---|---|---|---|---|---|---|
| Giani et al. (2006) | √ | | | | | |
| Jia et al. (2007) | √ | | | | | |
| Valentini et al. (2001) | √ | | | | | |
| Zhao et al. (2003) | √ | √ | | | √ | |
| Germani et al. (2007) | | | √ | | | |
| Roth et al. (1987) | | | | | | √ |
| Ruhi et al. (2006) | | | | √ | | |
| Jiang et al. (2005) | √ | √ | | | | |
| Sun et al. (2007) | √ | √ | | | | |
| Yang et al. (2003) | √ | √ | | | | |
| Limitations | Unable to provide the strength of a washcoated layer as a physically derived quantity which is comparable between different authors | | | | Unable to provide distinctive measurement between cohesive and adhesive strength | |

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