



Short Communication

Application of glass soot catalysts on metal supports to achieve low soot oxidation temperature



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ABSTRACT

K–Ca–Si–O glass was applied to metal supports for use as a catalyst for diesel soot combustion. Glasses were processed from the melt and by a sol–gel route. Catalyst activity for the oxidation of diesel exhaust soot and flame soot from an oil lamp was compared by thermogravimetric analysis (TGA). The results show that a K-based catalytic glass coating on metal substrates can reduce the temperature where half of the engine soot is oxidized (T_{50}) to as low as 360 °C under loose contact conditions, and offers catalytic stability for long term combustion cycling. Scanning electron microscopy observations show that sol–gel glass processing is effective for coating complex wire mesh shapes without pore clogging.

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1. Introduction

Increasing interest is being shown in using metallic substrates for diesel particulate filter (DPF) applications primarily due to their higher mechanical resistance and thermal conductivity, as well as the possibility of thinner walls allowing higher cell density and lower pressure drop [1–5]. A number of metal filters have been proposed, including a porous metal substrate made of Ni–Cr–Al alloy foam [1,2], spirally wound flat sheets of metallic fabric alternated with corrugated sheets of metal foil [3], a metal wool filter consisting of a housing filled with compressed metal wool, and a flow through metallic wire filter [4]. Among these designs, the most popular metal filter design is based on a stainless steel wire mesh sheet, which can be coated with metal powder mixed with additives [5]. Wire mesh DPFs were reported to achieve 60–90% filtration efficiency based on the design.

In practice, the most efficient way to maintain low backpressure during engine operation and minimize fuel penalty due to filter regeneration is through continuous soot oxidation [6]. Most continuous regeneration systems, e.g. commercially developed CRT® from Johnson Matthey, require that the engine remains at conditions to give a certain NO_x–PM (particulate matter) ratio out of the engine. The disadvantage is that, thus far, all of these catalyst systems cannot avoid using noble metals to provide sufficient NO₂ upstream for soot oxidation, because NO₂ is a much more effective oxidant than O₂.

Because of economical and environmental concerns, potassium-containing catalysts have attracted much interest for soot oxidation due to their high activity in the absence of NO₂. Unfortunately, potassium-containing catalysts usually tend to degrade after repeated thermal cycles due to the loss of potassium. This is especially the case for K₂CO₃ or KNO₃ [7–9]. This deactivation is thought to occur due to loss of potassium through sublimation. Thus the challenge in using potassium and other alkali based catalysts is minimizing, if not eliminating, the loss of the potassium or alkali metal. Glass catalysts have been recently developed as an effective solution to mitigate the effect of potassium loss by providing new ions over time [10,11]. The concept is shown schematically in Fig. 1. Briefly, ion exchange promoted by ambient water vapor leads to K⁺ accumulation on the glass surface, and subsequently to formation of KOH and then K₂CO₃. The surface layer of K₂CO₃ oxidizes soot, and if any K⁺ is lost over time, it can be replaced by additional ion exchange interactions.

We report here the feasibility of applying a K-containing glass catalyst onto metallic DPFs to achieve soot oxidation temperatures that are in the typical diesel exhaust temperature window, thereby avoiding the need for auxiliary devices or high noble metal loading.

2. Experimental methods

2.1. Application of silicate glass powder to stainless steel sheets

Before attempting to coat wire mesh, powder catalysts were applied to stainless steel sheets (SS420). Glass powder was synthesized from SiO₂, K₂CO₃ and CaCO₃ in a weight ratio of 52SiO₂–35K₂O–13CaO by a solid state synthesis method described elsewhere [10,11]. The obtained glass mass was then ground and sieved into powders for further study.

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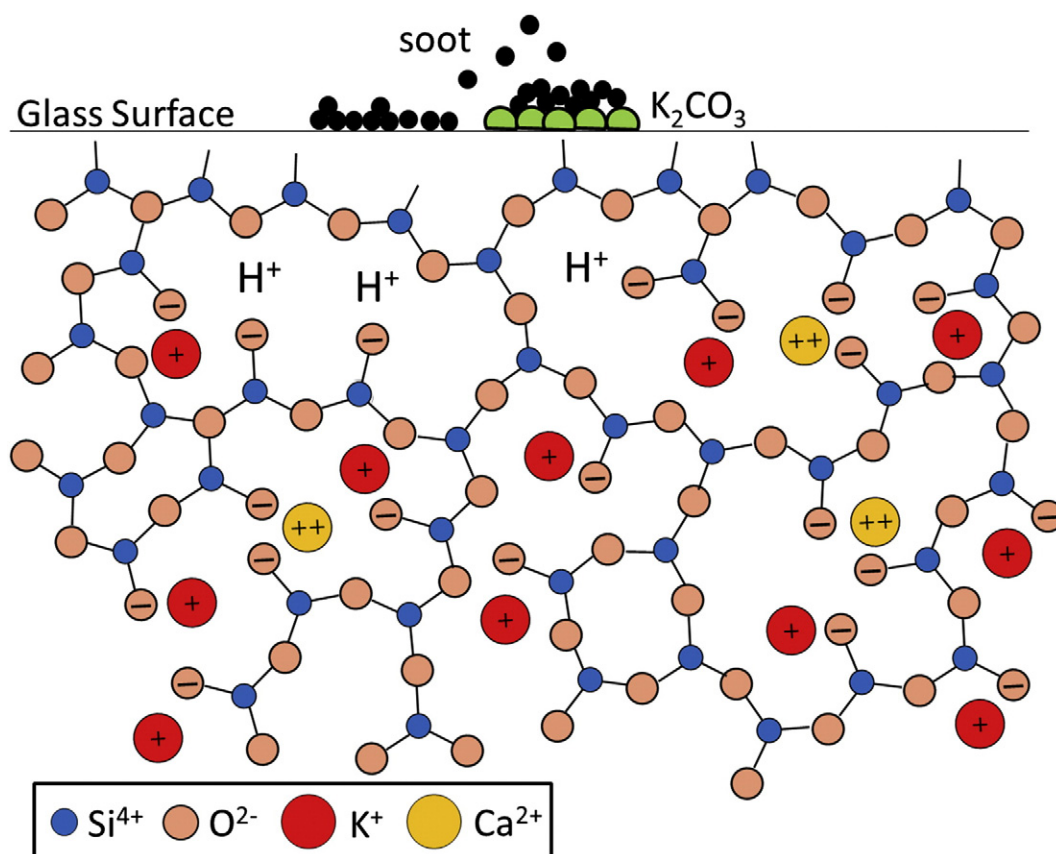


Fig. 1. Schematic diagram of the surface of a catalytic K–Ca–Si–O glass. Reaction with the atmosphere causes ion exchange and surface accumulation of K^+ ions. After reaction to form K_2CO_3 , potassium carbonate facilitates soot oxidation.

A thick particulate-based film of catalytic glass did not adhere well to stainless steel, likely because of the mismatch of coefficient of thermal expansion (CTE). As a remedy, a commercial glass frit (SCU-1 from SEM-COM Inc., $TiO_2 < 25\%$, Al_2O_3 , CaO , $MgO < 5\%$, $SiO_2 < 50\%$ in wt%) was applied to the metal substrate to act as an intermediate layer between the metal and the catalytic glass. The SCU-1 glass has a CTE of $\sim 120 \times 10^{-7}/^\circ C$, which matches the CTE of the 420 stainless steel ($\sim 116 \times 10^{-7}/^\circ C$). In comparison, the CTE of potassium silicate catalysts is typically $\sim 70 \times 10^{-7}/^\circ C$. Tape casting slurries were prepared with approximately 20 wt% glass powder in a binder solution (18 wt% poly(vinyl butyral), 64 wt% toluene, and 18 wt% other additives).

SCU glass powder was added to the binder solution and both were mixed with ZrO_2 milling media for 12 h. The slurry was cast on the substrates using a blade set to ~ 0.3 mm above the metal surface. The parts were then heated in air to $450^\circ C$ at $5^\circ C/min$, held for one hour for binder burn off, before ramping to $600^\circ C$ and holding for 30 min to fuse the SCU glass to the substrate. The sample was then furnace cooled to room temperature. Catalytic glass powder was applied to the SCU-coated substrate by painting on a glass/binder slurry layer, and applying the same heating cycle as described above.

2.2. Sol–gel coating process

Tetraethylorthosilicate (TEOS, $Si(OC_2H_5)_4$, 98%), calcium nitrate tetrahydrate ($Ca(NO_3)_2 \cdot 4H_2O$, ACS reagent) and potassium nitrate (KNO_3 , ACS reagent) were used as starting materials. A composition equivalent to the powdered silicate catalyst was synthesized using a common approach [12]. Briefly, 2 M nitric acid was added to deionized (DI) water and gently stirred for 5 min. TEOS was dissolved in a similar volume of ethanol and then added to the nitric acid. Mixing was continued to ensure hydrolysis and the progression of

condensation. Calcium and potassium nitrate were dissolved in DI water and added to this mixture of nitric acid and TEOS.

A viscometer (Brookfield DVIII Ultra) was used to monitor the sol solution viscosity change. After development of a suitable sol–gel solution, wire mesh (SS316-McMaster) was coated by dipping. The catalytic coatings on the wire substrates were characterized by SEM/EDS.

2.3. Catalytic activity and realistic soot contact

The soot oxidation of powder catalysts was characterized by TGA (TA Instruments 2950). Typically, 0.5 mg of soot was combusted with 5.0 mg of catalyst (10:1 catalyst/soot ratio). This soot mass is low enough to avoid concerns about heat and mass transfer limitations. A pre-mixed gas (10% O_2 –5% CO_2 – N_2) was used to simulate diesel exhaust. The catalytic activity was evaluated based on the ignition temperature (T_{ig}) or when half of the soot was combusted (T_{50}). To examine the catalysts' soot oxidation stability, the same coatings were subjected to repeated soot application and TGA combustion cycles.

Two kinds of soot were used in this study: 1) flame soot collected from a TIKI-brand oil lamp; 2) soot collected from a diesel engine. Flame soot from a TIKI torch was used to apply soot to TGA compatible catalyzed substrates as described elsewhere [13]. Briefly, a 2" diameter stainless steel tube was held 10 cm above a steady and consistent TIKI fuel flame to direct the flame soot onto the sample.

To assess and validate the catalytic performance of the glass catalyst for oxidation of soot applied by the flame-deposit method, it was compared with soot applied from a diesel engine exhaust system. A specially designed holder with multiple TGA compatible samples was placed in the exhaust pipe of a heavy duty diesel engine test stand. This produced samples with realistic loose contact and actual engine soot for further comparative study by TGA.

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