用于 NH₃ 选择催化还原 NO 反应的新型 Ce-P-O 催化剂李 飞^{1,2}, 肖德海^{1,3}, 张一波^{1,2}, 王德强^{1,2}, 潘喜强^{1,2}, 杨向光^{1,3}¹中国科学院长春应用化学研究所稀土资源利用国家重点实验室, 吉林长春 130022²中国科学院研究生院, 北京 100049³中国科学院长春应用化学研究所绿色化学与过程实验室, 吉林长春 130022

摘要: 报道了一种新型的可用于氨选择催化还原氮氧化物的 Ce-P-O 催化剂. 在空速为 20 000 h⁻¹, 较宽的温度范围内经 500 °C 焙烧的 Ce-P-O 催化剂上表现出了较高的 NO 转化率(可达到 90% 以上). 与 V-W-Ti 催化剂相比, 该催化剂具有一定的耐水蒸气和 SO₂ 能力以及较好的抗碱中毒性能.

关键词: 氮氧化物; 选择催化还原; 氨; 铈; 磷酸盐

中图分类号: O643

文献标识码: A

A Novel Ce-P-O Catalyst for Selective Catalytic Reduction of NO with NH₃LI Fei^{1,2}, XIAO Dehai^{1,3}, ZHANG Yibo^{1,2}, WANG Deqiang^{1,2}, PAN Xiqiang^{1,2}, YANG Xiangguang^{1,3,*}

¹State Key Laboratory of Rare Earth Resource Utilization, Changchun Institute of Applied Chemistry,
Chinese Academy of Sciences, Changchun 130022, Jilin, China

²Graduate University of Chinese Academy of Sciences, Beijing 100049, China

³Laboratory of Green Chemistry and Process, Changchun Institute of Applied Chemistry,
Chinese Academy of Sciences, Changchun 130022, Jilin, China

Abstract: A novel Ce-P-O catalyst for the selective catalytic reduction of NO with NH₃ was synthesized. The Ce-P-O catalyst was calcined at 500 °C and showed higher deNO_x activity (NO conversion > 90%) within the temperature range of 300–500 °C and at GHSV = 20 000 h⁻¹. The NO conversion was not significantly affected by the presence of H₂O and SO₂. By comparison with the commercial V-W-Ti catalyst, the Ce-P-O catalyst exhibited better resistance to deactivation due to K₂O poisoning.

Key words: nitrogen oxide; selective catalytic reduction; ammonia; cerium; phosphate

化石燃料燃烧产生的 NO_x(NO, NO₂) 是主要的大气污染源之一. 氨选择催化还原 (NH₃-SCR) 消除 NO_x 是一种应用广泛的烟道气脱硝技术. 近年来, 它还被用于重型柴油机尾气净化^[1~3]. NH₃-SCR 消除 NO_x 是基于反应 4NO + 4NH₃ + O₂ → 4N₂ + 6H₂O. 尽管用于该反应的 V₂O₅-WO₃ 或 MoO₃/TiO₂ (V-W-Ti) 催化剂已在烟道气脱硝中得到应用^[4,5]; 但由于 V 基催化剂具有较高的氧化 SO₂ 为 SO₃ 能力, 从而导致较高温度 (> 400 °C) 下还原 NO 的活性迅速下降. 虽然 V-W-Ti 催化剂对 SO₃ 不特别敏感, 但仍存在失活现

象^[5,6]. 同时, 活性组分 V₂O₅ 因其高挥发性导致在使用过程中不可避免地排放到大气中, 从而对人体和环境造成伤害^[7].

近年来, 非 V 基催化剂已成为研究热点, 其中两类催化剂广受关注: (1) 过渡金属氧化物 (如 MnO_x-CeO₂, MnO_x/TiO₂ 和 Fe-MnO_x 等). 这类催化剂主要用于低温条件下 NH₃-SCR 反应^[8~13], 但或多或少存在 N₂ 选择性低、易水蒸气或 SO₂ 中毒和硝酸铵沉积等问题. (2) 分子筛类催化剂. Long 等^[14]用离子交换法制备的 Fe-ZSM-5 催化剂, 表现出非常高温消除

收稿日期: 2010-03-15.

联系人: 杨向光. Tel/Fax: (0431)85262228; E-mail: xgyang@ciac.jl.cn

基金来源: 国家重点基础研究发展计划 (973 计划, 2004CB719502).

本文的英文电子版由 Elsevier 出版社在 ScienceDirect 上出版 (<http://www.sciencedirect.com/science/journal/18722067>).

NO_x 能力,但仍存在易硫中毒问题,而且高温下水热稳定性问题还有待于提高。

过渡金属的磷酸盐或焦磷酸盐具有特殊的表面酸性和氧化还原性,因而广泛被应用于多种催化反应中,如环己烷选择氧化^[15-17]、甘油脱水生成丙烯醛^[18]、气相 Beckmann 重排^[19]和对甲酚选择氧化^[20]等。众所周知,对于 NH_3 -SCR 反应,催化剂必须具有酸性位和氧化还原中心:酸性位有利于还原剂 NH_3 的吸附活化,而氧化还原中心可以促使氧化剂和还原剂之间发生反应。氧化还原中心通常由可变价过渡金属离子构成,如 Cu^{2+} , Fe^{3+} , Mn^{2+} 和 Ce^{3+} 等。考虑到过渡金属的磷酸盐或焦磷酸盐表面同时具有酸性位和氧化还原中心,因而可用于 NH_3 -SCR 反应。本文制备了一种无毒的 Ce-P-O 催化剂,该催化剂在较宽的温度范围内 (300~550 °C) 表现出较高的 NO 转化能力。

采用共沉淀法制备 Ce-P-O 催化剂。将 0.01 mol 硝酸亚铈 (AR) 溶于 200 ml 去离子水中配制成溶液,然后迅速倒入含有 7.5 mmol 焦磷酸 (45%) 的烧杯中并搅拌,再加入 0.10 mol 尿素,完全溶解后加热至 95 °C 左右,并保持 2 h。然后在室温持续搅拌 8 h,过滤、洗涤并在 110 °C 干燥 12 h,最后在 500 °C 焙烧 4 h,压片,破碎,筛分取 0.180~0.425 mm 颗粒备用。V-W-Ti 催化剂 (V_2O_5 1.5%, WO_3 10%) 的制备方法参照文献 [21]。掺杂 K_2O 的 Ce-P-O 或 V-W-Ti 催化剂通过等体积浸渍 KNO_3 溶液制得,所得样品在 110 °C 干燥并在 400 °C 焙烧 4 h。催化剂的评价在固定床流动反应体系中进行,反应条件为: 0.3 ml 催化剂, 0.1% NO , 0.1% NH_3 , 10% O_2 , 5% 水蒸气, 0.004% 或 0.05% SO_2 , Ar 平衡气,总流速为 100 ml/min。尾气各组分的含量采用四级杆质谱仪在线检测 (Hiden QIC-20)。

图 1 为 Ce-P-O 和 V-W-Ti 催化剂上 NH_3 -SCR-NO 反应活性。可以看出,与 V-W-Ti 催化剂相比, Ce-P-O 催化剂在较宽的温度范围表现出更高的活性。当反应温度超过 250 °C 时, Ce-P-O 催化剂上 NO 的转化率超过 90%; 其活性超过 V-W-Ti 催化剂。尤其在 450 °C 以上时, Ce-P-O 比 V-W-Ti 表现出更高的高温活性。氨程序升温脱附 (NH_3 -TPD) 和 H_2 程序升温还原 (H_2 -TPR) (图略) 结果表明, Ce-P-O 上同时具有酸性位和氧化还原中心。Ce-P-O 的 NH_3 -TPD 谱出现较宽的 NH_3 脱附峰,即使在 500 °C 以上仍存在。尽

图 1 Ce-P-O 和 V-W-Ti 催化剂上 NH_3 -SCR-NO 反应活性
Fig. 1. Activity of NH_3 -SCR- NO_x over Ce-P-O and V-W-Ti catalysts. Reaction conditions: 0.3 ml catalyst, $[\text{NO}] = [\text{NH}_3] = 0.1\%$, $[\text{O}_2] = 10\%$, Ar balance, 100 ml/min, GHSV = 20000 h^{-1} . SCR—selective catalytic reduction.

管 V-W-Ti 催化剂的 NH_3 的脱附峰也很宽,甚至峰面积大于 Ce-P-O 催化剂,但是 NH_3 -SCR 消除 NO_x 活性还与催化剂的氧化还原性能紧密相关。 H_2 -TPR 谱表明, Ce-P-O 催化剂的耗 H_2 量仅为 V-W-Ti 的 0.17 倍。由此推断, Ce-P-O 的氧化性能较弱,因而在高温时表现出较高的 NO 转化效率。Chen 等^[22]认为, V-Ti 催化剂高温活性的下降是由于 NH_3 过量氧化所致。

图 2 为水蒸气及 SO_2 对 Ce-P-O 催化剂上 NH_3 -SCR-NO 活性的影响。在原料气中加入 5% 水蒸气

图 2 水蒸气及 SO_2 对 Ce-P-O 和 V-W-Ti 催化剂上 NH_3 -SCR-NO 活性的影响

Fig. 2. Effect of water vapor and SO_2 on NH_3 -SCR-NO activity over Ce-P-O (closed symbols) and V-W-Ti (open symbols) catalysts. (1) Without SO_2 and H_2O ; (2) With 0.004% SO_2 ; (3) With 0.05% SO_2 ; (4) After removal of 0.05% SO_2 ; (5) With 0.004% SO_2 and 5% H_2O ; (6) With 5% H_2O ; (7) With 0.05% SO_2 over V-W-Ti. Reaction conditions: 0.3 ml catalyst, $[\text{NO}] = [\text{NH}_3] = 0.1\%$, $[\text{O}_2] = 10\%$, $[\text{H}_2\text{O}] = 5\%$ (when used), $[\text{SO}_2] = 0.004\%$ or 0.05% (when used), Ar balance, 100 ml/min, GHSV = 20000 h^{-1} .

后, 250 °C 以上时, 催化剂活性几乎不受影响. 250 °C 时 NO 转化率由 88.5% 下降至 66.5%. 而 0.004% SO₂ 的加入只是明显抑制了催化剂的高温活性. 当 SO₂ 的加入量为 0.05% 时, Ce-P-O 催化剂上 NO 转化率约为 50%, 而 V-W-Ti 催化剂上约为 40%. 当停止加入 SO₂ 后, 催化剂活性基本恢复. 此外, 0.004% SO₂ 和 5% 水蒸气同时存在时, 催化剂活性与仅有 5% 水蒸气时略有差别. 这是由于催化剂吸附硫后催化剂表面的酸性不同所致^[22]. Saur 等^[23]认为, 硫酸盐化的催化剂可以形成新的表面酸性位 (如 S-OH).

由于烟道气中含有较高浓度的灰分, 其中部分灰分是碱性化合物 (如 K₂O 或 CaO 等), 所以脱硝催化剂的耐碱中毒能力也是非常重要的技术指标. 如图 3 所示, 当 Ce-P-O 和 V-W-Ti 负载 2.8%K₂O 时, 活性都明显降低. 由于 K₂O 的负载量较高, V-W-Ti 催化剂上 NO 转化率都低于 20%. 这与 Kamata 等^[24]报道的结果一致. 他们认为, K 的存在影响了氨活性中间体 (如 NH₄⁺) 的形成, 从而导致催化剂失活. 碱金属化合物可以与氨的吸附位键合, 且这种键合是不可逆的, 会导致催化剂永久失活. 由图 3 可见, Ce-P-O 催化剂抗 K₂O 中毒的能力远强于 V-W-Ti 催化剂, 尤其在 300~400 °C 间这一能力更加明显, 此时 Ce-P-O 催化剂上 NO 转化率约为 V-W-Ti 催化剂上的 4 倍. 可见, Ce-P-O 有可能代替 V-W-Ti 催化剂.

尽管针对固定源低温 NH₃-SCR 催化剂已引起广泛关注, 但由于这类催化剂的比活性低而不能用于

于重型柴油机尾气净化. 和 V₂O₅ 一样, Ce-P-O 催化剂也可被负载在载体上, 甚至可以直接涂覆到蜂窝陶瓷上, 从而达到提高催化效率的目的. 由于 Ce-P-O 催化剂具有较宽的操作窗口, 通过负载提高效率后有望应用到重型柴油机尾气净化中.

综上所述, 本文合成的 Ce-P-O 催化剂在 NH₃-SCR 反应中具有较高的高温活性和较强的抗碱中毒能力, 以及一定的耐 SO₂ 和水蒸气能力, 显示出一定的应用前景.

参 考 文 献

- 1 Keim W. Handbook of Heterogeneous Catalysis. Weinheim: Wiley-VCH, 2008. 2274
- 2 Kröcher O. Past and Present in DeNO_x Catalysis. Oxford: Elsevier, 2007. 262
- 3 Curtin T. Environmental Catalysis. Boca Raton: Taylor & Francis, 2005. 197
- 4 Bosch H, Janssen F. *Catal Today*, 1988, **2**: 369
- 5 Busca G, Lietti L, Ramis G, Berti F. *Appl Catal B*, 1998, **18**: 1 (and references therein)
- 6 Brandenberger S, Kröcher O, Tissler A, Althoff R. *Catal Rev-Sci Eng*, 2008, **50**: 492 (and references therein)
- 7 Costigan M, Cary R, Dobson S. Concise International Chemical Assessment. Geneva: WHO, 2001. 29
- 8 Liu F, He H, Ding Y, Zhang C. *Appl Catal B*, 2009, **93**: 194
- 9 Liu F, He H, Zhang C. *Chem Commun*, 2008: 2043
- 10 Roy S, Viswanath B, Hegde M S, Madras G. *J Phys Chem C*, 2008, **112**: 6002
- 11 Qi G, Yang R T. *Chem Commun*, 2003: 848
- 12 Long R Q, Yang R T, Chang R. *Chem Commun*, 2002: 452
- 13 Smirniotis P G, Pena D A, Uphade B S. *Angew Chem, Int Ed*, 2001, **40**: 2479
- 14 Long R Q, Yang R T. *J Am Chem Soc*, 1999, **121**: 5595
- 15 Du Y, Xiong Y L, Li J, Yang X G. *J Mol Catal A*, 2009, **298**: 12
- 16 Zhu Y J, Li J, Yang X G, Wu Y. *Chem Lett*, 2004, **33**: 822
- 17 Zhu Y J, Li J, Xie X F, Yang X G, Wu Y. *J Mol Catal A*, 2006, **246**: 185
- 18 Liu Q B, Zhang Z, Du Y, Li J, Yang X G. *Catal Lett*, 2009, **127**: 419
- 19 Zhang Z, Li J, Yang X G. *Catal Lett*, 2007, **118**: 300
- 20 朱宇君, 李静, 杨向光, 吴越. 催化学报 (Zhu Y J, Li J, Yang X G, Wu Y. *Chin J Catal*), 2005, **26**: 261
- 21 Alemany L J, Lietti L, Ferlazzo N, Forzatti P, Busca G, Giamello E, Bregani F. *J Catal*, 1995, **155**: 117
- 22 Chen J P, Yang R T. *J Catal*, 1993, **139**: 277
- 23 Saur O, Bensitel M, Saad A B M, Lavalley J C, Tripp C P, Morrow B A. *J Catal*, 1986, **99**: 104
- 24 Kamata H, Takahashi K, Odenbrand C U I. *J Mol Catal A*, 1999, **139**: 189

图 3 K₂O 对 Ce-P-O 和 V-W-Ti 催化剂上 NH₃-SCR-NO 活性的影响

Fig. 3. Effect of 2.8% K₂O on NH₃-SCR-NO activity over Ce-P-O and V-W-Ti catalysts. Reaction conditions: 0.3 ml catalyst, [NO] = [NH₃] = 0.1%, [O₂] = 10%, Ar balance, 100 ml/min, GHSV = 20000 h⁻¹.

英 译 文

English Text

Nitrogen oxides (NO_x : NO and NO_2) present in exhaust gases from the combustion of fossil fuels remains a major source of air pollution and acid rain. The selective catalytic reduction of NO_x with ammonia (NH_3 -SCR) is a wide-spread process for the abatement of NO_x from the flue gas of stationary emission sources. NH_3 -SCR has also been used to remove NO_x in exhaust gases from diesel trucks [1–3]. The selective catalytic reduction (SCR) process is based on the reaction between NO and NH_3 to produce water and nitrogen according to the reaction $4\text{NO} + 4\text{NH}_3 + \text{O}_2 \rightarrow 4\text{N}_2 + 6\text{H}_2\text{O}$.

The well-known commercial catalyst used for this process is based on $\text{V}_2\text{O}_5/\text{TiO}_2$ and is promoted with either WO_3 or MO_3 [4,5]. However, these catalysts suffer from many disadvantages [5,6]. For example, the vanadium-based catalysts have a high activity for the oxidation of SO_2 into SO_3 , which results in a rapid decrease in activity and selectivity for the reduction of NO_x above 400 °C. Despite the low sensitivity of this catalyst to SO_2 poisoning, deactivation does occur. Furthermore, the emission of vanadium can pose a health problem because of vanadium toxicity [7]. The emission of vanadium from the V-based catalyst in the flue gas cannot be strictly excluded because of the higher volatility of vanadium when the V_2O_5 is near or above the monolayer surface in the industrial V-based catalysts. Therefore, an alternative NH_3 -SCR catalyst that only contains harmless substances and can survive the harsh operating conditions is required.

Recently, two main types of non-vanadium catalyst have been developed for different purposes. Supported or unsupported transitional metal oxide catalysts have been widely investigated for the low-temperature de NO_x reaction by NH_3 [8–13]. These catalysts include MnO_x - CeO_2 mixed oxides, MnO_x loaded on TiO_2 , and Fe-Mn based catalysts, but these catalysts suffer from low N_2 selectivity, deactivation by H_2O and SO_2 , and ammonium nitrate deposition, etc. Many researchers have also investigated zeolite-type SCR catalysts [6]. For example, Long et al. [14] have reported that Fe-ZSM-5 catalysts prepared by specific ion-exchange procedures show high activities for the NH_3 -SCR reaction. However, the durability of the corresponding catalysts, especially their high-temperature hydrothermal stability and their resistance against sulfur poisoning, was not adequate under their boundary conditions.

Because of their specific surface acidity and redox properties, we have recently used many transition metal pyrophosphates or phosphates for various reactions such as the selective oxidation of cyclohexane [15–17], the vapor-phase dehydration of glycerol to produce acrolein [18], the va-

por-phase Beckmann rearrangement of cyclohexanone oxime [19], and the gas-phase selective oxidation of *p*-cresol [20]. It is known that surface acidity and certain redox properties are necessary for catalysts in the NH_3 -SCR reaction of NO. Surface acidity facilitates the activation of NH_3 and/or NO while the acid sites are supplied by the catalyst support or by active components. The redox properties benefit the reaction between the reductants and the oxidants, and the redox sites are usually supplied with multivalent ions or relevant oxides such as Cu^{2+} , Fe^{3+} , Mn^{2+} , Ce^{3+} , etc. Therefore, some transition metal phosphates or pyrophosphates should be active in NH_3 -SCR reactions. Based on this idea, in this paper we present a novel non-toxic catalyst using a Ce-P-O compound as the active component working in a broad operation window in a medium-high temperature range (300–550 °C).

The Ce-P-O catalyst was synthesized by the precipitation method. In a typical preparation procedure, 0.01 mol cerium(III) nitrate (AR) was dissolved in 200 ml distilled water at room temperature. Then 7.5 mmol pyrophosphoric acid (45%) was dropped into the cerium(III) nitrate solution with stirring and the solution was stirred for several minutes. To clarify the solution a small amount of HNO_3 was added. Urea (0.10 mol) was added to the solution until it became clear. The solution was then heated to ~95 °C and kept at this temperature for 2 h. The solution was then cooled to room temperature and stirred for another 8 h. The precipitate was recovered by filtration, washing, and drying at 110 °C for 12 h. After calcination at 500 °C for 4 h, the samples were pressed, crushed, and sieved to 0.18–0.425 mm before use. The V-W-Ti catalyst (V_2O_5 1.5%, WO_3 10%, similar to the commercial de NO_x catalyst) was prepared according to Ref. [21]. The K_2O loaded catalysts were prepared by impregnation of the V-W-Ti catalyst and the Ce-P-O catalyst with aqueous solutions of KNO_3 . The samples were then dried at 110 °C and calcined at 400 °C for 4 h in air. SCR activity measurements were carried out in a fixed-bed quartz reactor. The reaction conditions were as follows: 0.3 ml (0.4 g) catalyst, 0.1% NO, 0.1% NH_3 , 10% O_2 , 5% water vapor (when used), 0.004% or 0.05% SO_2 (when used), and the balance was Ar, 100 ml/min total flow rate and GHSV = 20 000 h^{-1} (ambient conditions). Water vapor was generated by passing the Ar stream through a gas-wash bottle containing deionized water at a suitable temperature. The reactants and products were analyzed on-line using a mass spectrometer (Hiden QIC-20).

Figure 1 shows the catalytic activities for NO reduction by NH_3 over Ce-P-O and V-W-Ti catalysts. By comparison with the V-W-Ti catalyst in Fig. 1, the Ce-P-O catalyst exhibited excellent catalytic properties over a broad temperature range. With increasing temperature, NO conversion over the Ce-P-O catalyst increased and reached nearly 90% above

250 °C. At temperatures above 300 °C, NO conversions over the Ce-P-O catalyst exceeded 95%, which was higher than that of the V-W-Ti catalyst, especially at 500 °C. The Ce-P-O catalyst, therefore, showed better high-temperature deNO_x performance than the V-W-Ti catalyst. The surface acidity and redox properties of Ce-P-O were characterized by temperature-programmed desorption of NH₃ (NH₃-TPD) and temperature-programmed reduction of H₂ (H₂-TPR) (not shown). The NH₃-TPD spectra of the Ce-P-O catalyst showed a wide ammonia desorption peak even at higher temperatures (> 500 °C) and this is related to higher-temperature deNO_x activity. Although V-W-Ti also shows a wider NH₃-TPD peak, the oxidation ability of the catalysts is another key factor for deNO_x activity. By comparison with the V-W-Ti catalyst, the Ce-P-O catalyst showed a much lower H₂-uptake between 300 and 600 °C. The ratio of H₂-uptake area on V-W-Ti to that on Ce-P-O is 5.8, which implies that the Ce-P-O catalyst has a lower oxidation ability than V-W-Ti. The lower oxidation ability of Ce-P-O can prevent the excessive oxidation of NH₃ in the SCR reaction especially at higher temperatures. As discussed by Chen et al. [22], the high-temperature decline of NO SCR activity over the V-Ti catalysts was caused by the oxidation of NH₃. Therefore, the Ce-P-O catalyst shows better activity above 450 °C. In view of this catalyst's surface acidity and lower oxidation ability over a wide temperature range, the high deNO_x activity of the Ce-P-O catalyst at higher temperature can be explained.

The effect of water vapor and SO₂ on the catalytic behavior of Ce-P-O is shown in Fig. 2. When 5% H₂O was added to the feed gas, the NO conversion was influenced slightly over the whole temperature range except at 250 °C, where the NO conversion decreased from 88.5% to 66.5%. The addition of 0.004% SO₂ did not change the NO conversion significantly at lower temperatures (≤ 450 °C). At 500–550 °C, more than 75% NO conversion was obtained over the Ce-P-O catalyst. The effects of a high SO₂ concentration (0.05%) on SCR activity were also investigated and are shown in Fig. 2. When 0.05% SO₂ was added to the reaction gases and the space velocity was kept at 20 000 h⁻¹, the NO conversion over the Ce-P-O catalyst decreased significantly to ~50% at 300–550 °C under steady state conditions. Over the V-W-Ti catalyst, the conversions decreased much more severely than the case of the Ce-P-O catalyst, which decreased to ~40% under the same conditions. However, the deNO_x activity of the Ce-P-O catalyst was restored when the 0.05% SO₂ was removed from the feed gas. In addition, the results for the 0.004% SO₂ plus 5% H₂O feed gas are also shown in Fig. 2. The catalytic behavior was not affected at 400–550 °C. More than 90% NO conversion was obtained at higher temperatures (≥ 350 °C) in the presence of

0.004% SO₂ and 5% H₂O and this was a little higher than that in the presence of 5% H₂O. As described in the literature, sulfation of the Ce-P-O surface increases the acidity of the catalysts [22]. When water vapor is fed into the reaction system, a considerable amount of Lewis acid sites are converted to Brønsted acid sites. Saur et al. [23] suggested that hydroxylation of surface sulfated catalysts leads to the formation of surface S-OH groups. These new acid sites may be responsible for the higher SCR activity in the presence of 0.004% SO₂ and 5% H₂O above 450 °C.

The high ash content (K₂O, CaO, and As₂O₃) in the flue gas reduces the performance and longevity of the deNO_x catalysts and, therefore, it is necessary to evaluate the resistance to deactivation of the catalyst by poisoning with alkaline metal compounds. As shown in Fig. 3, the catalytic activity decreases over the Ce-P-O and V-W-Ti catalysts after loading with 2.8% K₂O. However, the deactivation of NO conversion by K₂O over the V-W-Ti catalyst is consistent with the results of Kamata and co-workers [24], and it was much more severe than that of Ce-P-O. As Kamata suggested, potassium disturbs the formation of the active ammonia intermediate, NH₄⁺, resulting in deactivation of the catalyst. The alkali binds irreversibly to the ammonia adsorption sites and causes a permanent loss of activity. By comparison with the V-W-Ti catalyst, the Ce-P-O catalyst shows better resistance to deactivation by poisoning with K₂O, especially between 300 and 400 °C where the conversion is three times higher than that of the V-W-Ti catalyst. Ce-P-O is, therefore, a promising catalyst for the reduction of NO_x emissions from power plants compared with the V-W-Ti catalyst at high temperatures.

Although many low temperature SCR catalysts have been studied for the treatment of flue gas from stationary sources [5,6], these catalysts cannot be used for mobile sources under highly dynamic operating conditions and space restrictions, especially for the exhaust gas of diesel trucks. The Ce-P-O catalyst is an active ingredient like V₂O₅ and can be supported on different catalyst supports. It can also be coated directly onto the honeycomb monolith. Therefore, the Ce-P-O catalyst has potential application in the abatement of NO_x from mobile sources because of its broad operating temperature window.

In conclusion, a new highly active Ce-P-O catalyst with water, SO₂, and alkaline metal compounds resistance was developed for the SCR of NO with NH₃ in the presence of excess O₂. Further studies are in progress to better understand the mechanistic aspects of our catalytic system.