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Active Fe species of Fe2O3/Fe-Silicalite-1 nanowires in N₂O decomposition

1、存在的问题(研究背景)	Recently, we reported the hydrothermal conversion of Fe_2O_3/SiO_2 spheres (Fe_2O_3 nanoparticles sup-ported on SiO_2 spheres) into Fe_2O_3/Fe -Silicalite-1 nanowires in the presence of ethylenediamine and triethylamine (Micropor. Mesopor. Mater. 200 (2014) 52–60). The catalytic performance of Fe_2O_3/Fe -Silicalite-1 nanowires in CO oxidation, N ₂ O decomposition, and catalytic reduction of NO by NH ₃ was evaluated . However, the nature of active Fe species of Fe_2O_3/Fe -Silicalite-1 nanowires in N ₂ O decom-position was not clear .
2、我们的策略	In the current work, additional samples (i.e., Fe-Silicalite-1, Fe_2O_3 /Silicalite-1,and Fe_2O_3 /Fe-Silicalite-1) were prepared by using conventional Silicalite-1 as a precursor, and tested in N ₂ O decomposition.
3、结果是什么	Fe_2O_3 /Silicalite-1 and Fe_2O_3 /Fe-Silicalite-1 prepared by impregnation showed higher activities than Fe-Silicalite-1 prepared by isomorphous substitution, and Fe_2O_3 /Fe-Silicalite-1nanowires showed the highest activity among these samples.
4、原因原理是什么	Through relevant characterization employing XRD, XRF, N ₂ adsorption, TEM, UV–vis, and 57Fe Mössbauer spectroscopy, we conclude that small Fe ₂ O ₃ nanoparticles with size of 1–2 nm are the main active Fe species for N ₂ O decomposition, whereas framework Fe ³⁺ and larger Fe ₂ O ₃ particles are not so active .



Flower-like Ag₂O/Bi₂MoO₆ p-n heterojunction with enhancedphotocatalytic activity under visible light irradiation

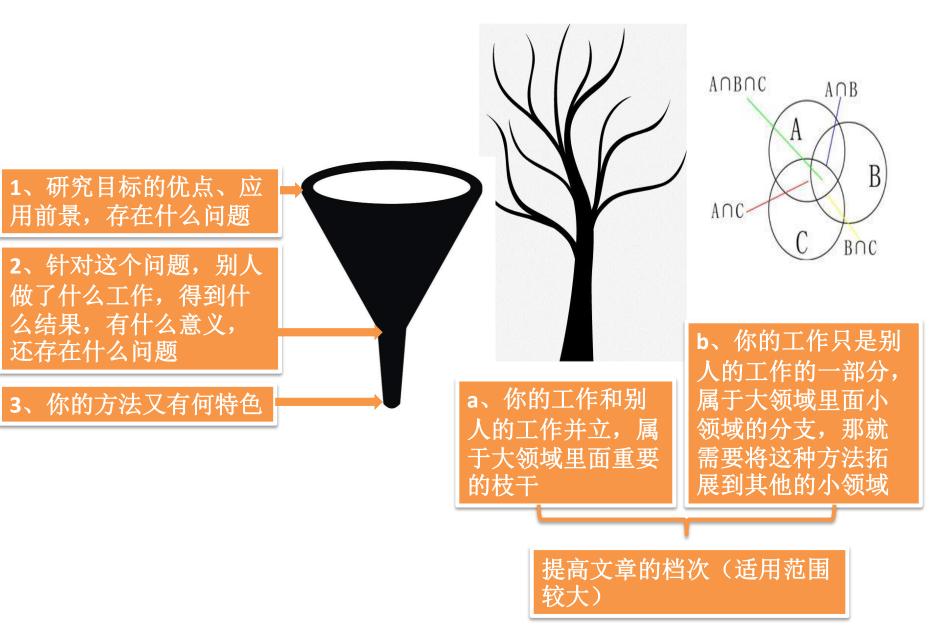
1、我们的策略(合成、表征)	Ag_2O/Bi_2MoO_6 composed of flower-like Bi_2MoO_6 microspheres (diameter: 1–3 m) and well dispersed Ag_2O nanoparticles (size: ~10 nm) was prepared via solvothermal deposition-precipitation. The catalytic performance of Ag_2O/Bi_2MoO_6 and other samples in photocatalytic degradation of Rhodamine B (RhB) and methyl orange (MO) dye under visible light irradiation (> 420 nm) was studied
2、结果是什么	Ag_2O / Bi_2MoO_6 p-n heterojunction exhibited higher activity than Bi_2MoO_6 or Ag_2O with the same weight of the corresponding component in Ag_2O / Bi_2MoO_6 . It was even more active than a mixture of Bi_2MoO_6 and Ag_2O .
3、原因是什么	The high photocatalytic activity of Ag_2O/Bi_2MoO_6 was due to the efficient separation of photogenerated electron-hole pairs. The photogenerated holes (h+) and superoxide radical anions ($\cdot O^{2-}$) were found to be the primary reactive species for the degradation of RhB dye in aqueous solution.



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2、引言结构-示例

1、这个领域应用前景及存 在的问题

2、别人和自己课题组以前 采取什么行动或者方法, 还有什么问题遗留。

3、针对这个问题你采用什 么(巧妙的、聪明的、系 统的)方法;最后你得到 什么结果(包括结果的内 容和意义) Supported Fe₂O₃ catalysts **have demonstrated applications in** phenol hydroxylation [1,2], dehydrogenation of propane [3,4], CO oxidation [5,6], and catalytic reduction of NO by NH3[7–9]. On the other hand, Fe-containing zeolites also have many applications in the selective oxidation of methane to methanol [10–12], selective oxidation of benzene to phenol by N2O [13–16], oxidative dehydrogenation of alkanes [17–19], Fenton oxidation [20,21], selective reduction of Nox with hydrocarbons [22–24] or NH3[25–27], and decomposition of N2O [28–37]. The catalytic activities of these catalysts **are critically influenced by many factors, such as** preparation methods/conditions, nature of Fe species, and FeOx-support interactions, etc. **The nature of active Fe species is complicated, especially when** a catalyst contains several different

In a previous communication [38], Tang and co-workers proposed a new strategy for the synthesis of single-crystalline SiO₂ nanowires. In their synthesis, $Fe_2O_3/SiO2$ spheres (prepared by impregnation and spray drying) were placed on top of an ethylene-diamine solution in an autoclave and heated. The amorphous SiO2 support transformed into crystalline SiO2 nanowires, and Fe_2O_3 nanoparticles on the support surface was leached by an acid afterwards, leaving behind almost pure SiO2 nanowires for blue light emission.

We continued that work [38], and found that $Fe_2O_3/SiO2spheres$ can be transformed into Fe2O3/Fe-Silicalite-1 nanowires via hydrothermal treatment in a mixture of ethylenediamine and trimethylamine [39]. This synthesis is interesting, because (i) amorphous SiO2 can be transformed into crystalline Silicalite-1, a zeolitic microporous material; (ii) SiO2 spheres can be transformed into Silicalite-1 nanowires; (iii) Fe_2O_3 nanoparticles on the support sur-face become smaller due to the leaching of some Fe species during the synthesis; (iv) a small portion of Fe3+ species enter the Silicalite-1 framework. The resulting catalyst showed 100% conversion in N2O decomposition at 500 °C. Considering that (i) a small portion of Fe3+ incorporated into the Silicalite-1 framework during the hydrothermal synthesis and (ii) Fe2O3/SiO2 spheres (prepared by impregnation of Fe(NO₃)₃ onto SiO₂ spheres followed by calcination) showed very low activity, we tentatively ascribed the activity of $Fe_2O_3/Fe-Silicalite-1$ nanowires to framework Fe^{3+} [39]. Nevertheless, clear elucidation of the nature of active Fe species is still needed.

framework Fe³⁺ species; (ii) Fe₂O₃/Silicalite-1 (prepared by impregnation using conventional Silicalite-1 as a support) containing only supported Fe₂O₃; (iii) Fe₂O₃/Fe-Silicalite-1 (prepared by impregnation using conventional Fe-Silicalite-1 as a support) containing both frame-work Fe3+and supported Fe₂O₃. Through detailed characterization and a comparison of the catalytic performance, we conclude that small Fe2O3nanoparticles of 1–2 nm are the main active Fe species, whereas framework Fe³⁺ and larger Fe₂O₃ particles are not so active.

2、引言-段落结构-1

1、应用前景 (研究价值)	Supported Fe ₂ O ₃ catalysts have demonstrated applications in phenol hydroxylation [1,2], dehydrogenation of propane [3,4], CO oxidation [5,6], and catalytic reduction of NO by NH ₃ [7–9]. On the other hand, Fe-containing zeolites also have many applications in the selective oxidation of methane to methanol [10–12], selective oxidation of benzene to phenol by N ₂ O [13–16], oxidative dehydrogenation of alkanes [17–19], Fenton oxidation [20,21], selective reduction of NO _x with hydrocarbons [22–24] or NH ₃ [25–27], and decomposition of N ₂ O [28–37].
2、性能的影响 因素	The catalytic activities of these catalysts are critically influenced by many factors, such as preparation methods/conditions, nature of Fe species, and FeO _x -support interactions, etc.
3、存在的问题	The nature of active Fe species is complicated, especially when a

The nature of active Fe species is complicated, especially when a catalyst contains several different Fe species, e.g., framework Fe³⁺, isolated Fe³⁺ or Fe²⁺ anchored to zeolite frame-work, dinuclear Fe-O-Fe species, oligomeric Fe oxo species, small FeO_x nanoparticles, and bulk FeO_x particles.

2、引言-段落结构-2

1、方案1

In a previous communication [38], Tang and co-workers **proposed a new strategy for** the synthesis of single-crystalline SiO₂ nanowires. In their synthesis, $Fe_2O_3/SiO2$ spheres (prepared by impregnation and spray drying) were placed on top of an ethylene-diamine solution in an autoclave and heated. The amorphous SiO2 support transformed into crystalline SiO2 nanowires, and Fe_2O_3 nanoparticles on the support surface was leached by an acid afterwards, leaving behind almost pure SiO2 nanowires for blue light emission.

2、方案二

We continued that work [38], and found that $Fe_2O_3/SiO2$ spheres can be to via hydrothermal treatment in a mixture of ethylenediamine and trim because (i) amorphous SiO2 can be transformed into crystalline Silicalit spheres can be transformed into Silicalite-1 nanowires; (iii) Fe_2O_3 nanopa due to the leaching of some Fe species during the synthesis; (iv) a smal framework. The resulting catalyst showed 100% conversion in N2O decomportion of Fe3+ incorporated into the Silicalite-1 framework during the hyd (prepared by impregnation of $Fe(NO_3)_3$ onto SiO_2 spheres followed by calci **ascribed** the activity of Fe_2O_3/Fe -Silicalite-1 nanowires to framework $Fe^{3+}[39]$

3、仍然存在的 问题

<u>Nevertheless</u>, clear elucidation of the nature of active Fe species is still needed.

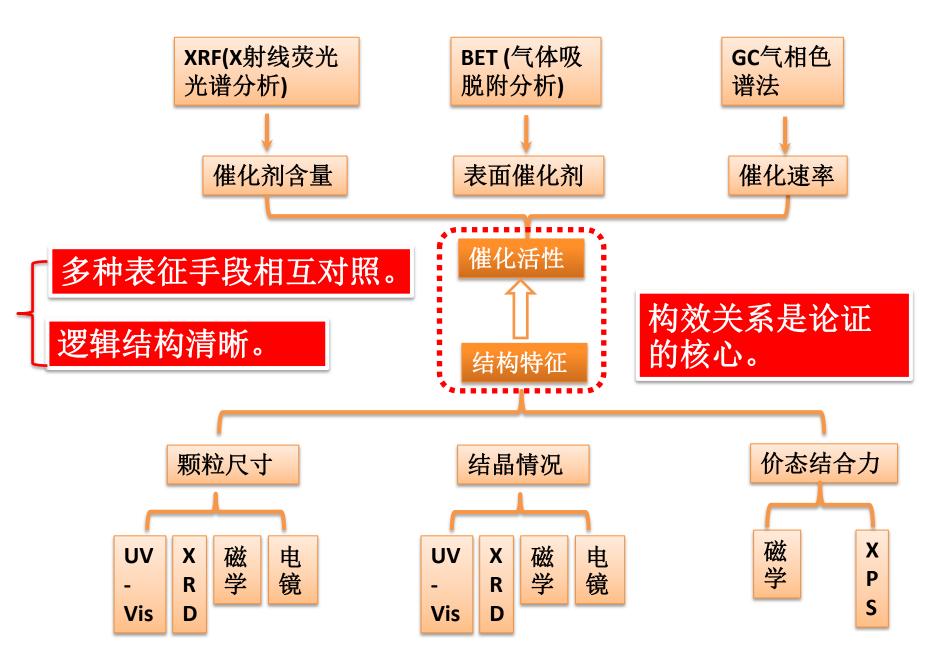
2、引言-段落结构-3

我们的方案 In the current work, we additionally prepared three different Fecontaining Silicalite-1 samples: (i) Fe-Silicalite-1 (prepared by isomorphous substitution) containing only framework Fe³⁺ species; (ii) Fe₂O₃/Silicalite-1 (prepared by impregnation using conventional Silicalite-1 as a support) containing only supported Fe₂O₃; (iii) Fe₂O₃/Fe-Silicalite-1 (prepared by impregnation using conventional Fe-Silicalite-1 as a support) containing both frame-work Fe³⁺ and supported Fe₂O₃.

b、结论

Through detailed characterization and a comparison of the catalytic performance, we conclude that small Fe_2O_3 nanoparticles of 1–2 nm are the main active Fe species, whereas framework Fe^{3+} and larger Fe_2O_3 particles are not so active.





3、讨论部分段落结构

1、主题句(为什么做这个实验或者 表征)

2、表征结果以什么形式给出

3、观察到什么现象,说明什么(浅 层次)

4、所有现象汇总,物理本质是什么 (深层次)

充分挖掘表征结果背后的结构 信息。

XRD and FTIR **are applied to further investigate structural variation** from M-CA complex to O-doped g-C3N4.

Figure 2a shows the XRD patterns of M-CA complex, M-CA mixture, and their polycondensation products.

The appearance of new peaks for M-CA complex compared with M-CA mixture can be viewed as a **solid evidence for** the occurrence of new arrangement. The high crystallinity noted in the XRD pattern of M-CA complex strongly suggests that the M-CA complex possesses a hexagonal structure. According to the hexagonal structure model of M-CA supramolecular complex proposed by Rao, a three obvious peaks at 10.67, 11.90 and 18.41 may be assigned to (100), (010), and (110) of the in-planar pattern, respectively. Apparently, another two peaks at 21.41 and 23.80 are precisely corresponding to crystal faces of (200) and (020), respectively. Furthermore, the strongest diffraction peaks observed at 27.92 is well assigned to the lamellar stacking peak of (002) crystal plane, supporting the observed sheet-like structure (see Figure 1a). In addition, no peaks of melamine or cyanuric acid have been detected in M-CA complex, indicating a complete reaction of melamine and cyanuric acid through the solution mixing process. **Obviously**, the XRD pattern of M-CA mixture presents no other diffraction peaks except for those of melamine and cyanuric acid, confirming a simple physical mixing of melamine and cvanuric acid. For both These results could be interpreted as the lattice distortion and the nd delocalization of π -conjugated bonds due to the introduction of oxygen heteroatoms.

sheets, respectively. It is notable that, with a close observation (Figure 2b), the full width at half maximum (FWHM) of (110) peak for O-gCN enlarges from 0.45 to 0.6 compared with gCN. Moreover, the (002) peak in O-gCN shifts from 27.43 to 27.23



1、做了什么	This paper described the synthesis, characterization, and catalytic behavior of Au/Cab-O-Sil fumed SiO2 and Au/MOx/SiO2 catalysts synthesized using Au(en)2Cl3 as the precursor.
2、结果是什么	The novel preparation method <u>resulted in</u> small and welldispersed gold nanoparticles on SiO2 supports. These Au/SiO2 catalysts <u>were</u> highly active for CO oxidation below room temperature.
3、原因原理是什么	The pretreatment of as-synthesized Au/SiO2 in H2-Ar at 150 8C and in O2- He at 500 8C is beneficial for high activity. The optimum gold loading was in the range of 1.1 and 2.5 wt.%. The post-treatment of calcined (and activated) Au/SiO2 in different media influenced the activity in CO oxidation. The calcined Au/SiO2 might be deactivated during the storage in ambient environments, but could be regenerated via recalcination. The addition of metal oxide dopants can be used to tune the catalytic performance as well.
┗ 4、有什么意义(评价)	Considering the fact that highly active Au/SiO2 catalysts are scarce, this report <u>furnishes</u> <u>new perspective for</u> this subject, and provides new possibilities for following X-ray absorption, spectroscopic, in situ HRTEM experiments and first-principle calculations.

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