



Gold and Ceria as Catalysts for VOC Abatement: A Review

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Abstract: Due to its excellent oxygen storage capacity, ceria is a well-known oxidation catalyst. However, its performance in the oxidation of volatile organic compounds can be improved by the introduction of gold. Depending on the type of VOC to be oxidized, the surface of gold nanoparticles and the gold/ceria interface may contribute to enhanced activity and/or selectivity. Choosing a proper preparation method is crucial to obtain optimal gold particle size. Deposition–precipitation was found to be more suitable than coprecipitation or impregnation. For industrial applications, monolithic catalysts are needed to minimize the pressure drop in the reactor and reduce mass and heat transfer limitations. In addition to the approach used with powder catalysts, the method employed to introduce gold in/on the washcoat has to be considered.

Keywords: volatile organic compounds; Au; CeO₂; VOC total oxidation; noble metal catalysts

1. Introduction

Volatile organic compounds (VOCs) are organic compounds having an initial boiling point of less than 250 °C measured at a standard atmospheric pressure of 101.3 kPa, which participate in atmospheric photochemical reactions [1].

VOCs are harmful to the environment and human health. They are emitted into the atmosphere from thousands of sources and recognized as one of the major factors responsible for the increase in global air pollution. They can also react with NO_x and oxygen to form ozone: VOC + NO_x + O₂ + $h\nu \rightarrow O_3$ + other products. Ozone causes human health problems, affects DNA and enzyme functions, is toxic for vegetation, and impairs the quality of some materials. Therefore, an abatement of VOC emissions is considered desirable. Anthropogenic VOC emissions are regulated by law [2].

The best available techniques for reducing VOC emissions include the measures focusing on process design (primary measures) and post-process removal (secondary measures) [3]. Among secondary measures, catalytic oxidation is the most effective for low concentrations of VOCs [1,3–9]. It is an efficient, cost-effective and environmentally sound way to treat these harmful emissions [10,11]. The mechanism of total catalytic oxidation depends on the type of catalytic material used. Two types of catalysts are employed for oxidation reactions [3]: (i) metal oxides and (ii) noble metals (supported or not).

Metal oxide catalysts (in particular Cu, Mn, Cr, Co, Mn, Ni, Mo and V oxides) show appreciable activity; they are cheap but not sufficiently stable [12–16]. In general, noble metal catalysts possess greater activity than oxide catalysts, contributing to the reduction of operating costs despite their higher manufacturing cost. Their catalytic activity depends on the noble metal and varies with the nature of VOC [17]. The mainstream of commercial catalysts belongs to this group and consists of Pt or Pd or both, supported on high-surfacearea γ -Al₂O₃ [3,18–27]. Palladium or bimetallic Pt–Pd active phase supported on alumina or CeO₂–Al₂O₃ mixed oxides are still probably the most studied group of catalysts [28–30].

For a long time, the noticeable exception of noble metals was gold. Au, in the bulk form, is considered the most noble of all metals [31]. The low adsorption energy of gases and high energy barriers for their dissociation disadvantage reactivity on gold. Thus, for



Citation: Gaálová, J.; Topka, P. Gold and Ceria as Catalysts for VOC Abatement: A Review. *Catalysts* 2021, 11, 789. https://doi.org/10.3390/ catal11070789

Academic Editor: Stefano Cimino

Received: 1 June 2021 Accepted: 26 June 2021 Published: 29 June 2021

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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). years, it was also the most abundant noble metal in catalysis. Nowadays, gold-based catalysts attract increasing attention [32]. They have proved a beneficial role in various catalytic reactions of commercial and environmental status, e.g., exhaust gas purification, oxidation of glucose, water gas shift, CO oxidation, water or hydrogen purification, air cleaning, fuel cells and VOC oxidation.

2. Gold Catalysts for VOC Oxidation

The preparation of Au catalysts for VOC oxidation has been widely investigated. The studies on reaction mechanisms enhance the understanding of the oxidation process using Au catalysts [33]. To achieve the best performance of the catalyst in the oxidation process, diverse approaches to design the gold catalysts have been applied. The preparation method, the loading of gold, the nature of VOC to be oxidized are just some of them [34–38].

According to many authors, the role of support is crucial. It contributes to the control of the final amount, the size and the shape of gold particles, influencing the activity of material in VOC oxidation. Typically, different oxide supports are employed, e.g., oxides of iron [39–47], titan [48–54], aluminum [55–57], vanadium [58–60], manganese [61,62] or cobalt [56,63–66]. Figure 1 shows the example of gold nanoparticles supported on ceria–zirconia mixed oxide. According to Scirèand Liotta [67], VOC deep oxidation over Au/metal oxide catalysts is governed by both the support and gold properties, which often amplify their effects due to a synergistic action. However, there is still debate on how different variables affect support and gold properties and thus the catalytic performance.

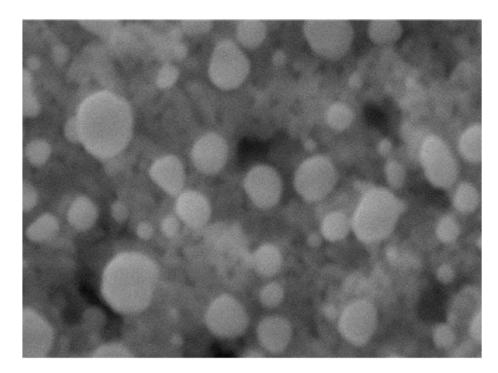


Figure 1. The TEM image of the $Au/Ce_{0.5}Zr_{0.5}O_2$ powder catalyst prepared by impregnation studied in Ref. [68].

A real catalyst usually contains several components, and its surface composition and structure may change during the catalytic process. Corma's group [69] reported that extremely low concentrations of mononuclear gold complexes in impregnation solution resulted in catalytically active gold clusters (TOF ca. 10⁷). Thus, new properties of supported gold catalysts can be reached using nanometer and sub-nanometer structures [70]. Recently, the group of Keiski showed that Au/Al₂O₃ was more active and selective to HCl in dichloromethane oxidation than the Pt/Al₂O₃ catalyst due to small Au particle size (~5 nm) with narrow size distribution (\pm 5 nm) and good metal dispersion [71].

It is possible to modify the monometallic gold catalyst by adding a second metal to improve the dispersion, adsorption and activity towards the oxidation of hydrocarbons [72]. For example, neighboring Pd atoms change Au's electronic structure, leading to high activity. Preparation and characterization of the bimetallic catalysts containing Pt and Au for VOC oxidation were studied by Kim et al. [73]. Influence of Pt-Au molar ratio on the catalytic activity for toluene oxidation revealed that Pt₇₅Au₂₅ and Pt₆₇Au₃₃ catalysts concurrently coated with Pt and Au precursors by impregnation method showed higher activity than monometallic Pt and Au catalyst for toluene oxidation. Kim et al. also studied Au_{0.5}Ag_{0.5} alloy films and highlighted that VOCs affect only the surfaces of metal nanoparticles [74]. Noble metals (Pd, Au) were deposited onto macro-mesoporous metaloxide supports to form mono and bimetallic catalysts by Barakat et al. [75]. The catalytic materials were tested in the oxidation of selected VOCs. PdAu/TiO₂ and PdAu/TiO₂-ZrO₂ 80/20 catalysts demonstrated the best activity and life span in the oxidation of toluene and propene; the lowest coke content after catalytic testing was observed for them too. An effective method of enhancing catalytic activity is the core-shell construction of nanosized bimetallic particles [76]. The Au–Pd is one of the core–shell systems that have gained considerable attention [53,77].

3. CeO₂ and Ceria-Supported Noble Metal Catalysts for VOC Oxidation

Cerium oxide can be used as a catalyst or an active phase/support of more complex catalytic material for VOCs oxidation. However, ceria is rarely used alone in the oxidation process [78], usually for the matter of comparison [79,80]. The possibilities of mixed oxides forms [81,82] and combining ceria with other metals (noble or other supported metals [30,83]) are multiples, as demonstrated in the selected publications gathered in Table 1 (excluding gold catalysts, which are included in Table 2). The nature of catalytic components, how they influence each other, a preparation process, and many others determine the properties of final catalysts; e.g., a preference of reduced centers for surface positions, stability and activity in the oxidation of different VOCs, etc.

The study of Pitkäaho et al. [84] reports total oxidation of perchloroethylene over Pt, Pd, Rh and V_2O_5 metallic monolith catalysts supported on Al_2O_3 , CeO₂ and TiO₂-doped Al_2O_3 . It shows that the redox properties of the catalyst and the amount of activated oxygen may play a bigger role than the acidity in the oxidation of certain VOCs.

1st Author Buhl Vern	Ref. —	Catalyst Containing			
1st Author, Publ. Year		Noble Metal	Ce and:	- VOCs to Oxidize	
Zheng, 2021	[85]	Pt	-	Benzene	
Zeng, 2021	[86]	-	Cu	Toluene	
Jiang, 2020	[87]	Pt	-	Methanol	
Li, 2020	[88]	-	Со	Methanol, Acetone, Toluene, O-xylene	
Sophiana, 2020	[89]		Cu, Mn, Ni, Zr	Benzene	
Al-Aani, 2020	[90]	-	Cu, Mg, Al	Methane	
Figueredo, 2020	[91]	-	Mn, Cu	Ethylene, Propylene	
Jiang, 2020	[92]	-	Fe	Toluene	
Zheng, 2020	[93]	-	Со	Acetone	
Jiang, 2019	[94]	-	Mn	Ethyl acetate	
Wang, 2019	[95]	-	Cu	Methanol, Acetone, Toluene, O-xylene	
Genty, 2019	[96]	-	Co, Al	Toluene	
Shah, 2019	[97]	-	Zr	Propane, Naphthalene	
Chen, 2019	[98]	Pd-Pt	Al	Benzene	
Gołąbek, 2019	[99]	-	zeolite β	Trichloroethylene	
Okal, 2018	[100]	-	Ru	Iso-butane, N-butane, Propane	
Hosseini, 2017	[101]	Pd	Al-clinoptilolite	Toluene	

Table 1. Recently studied ceria-based catalysts for VOCs oxidation.

1st Author, Publ. Year	Ref. —	Catalyst Containing		VOCs to Oxidize	
ist Aution, I ubi. Tear		Noble Metal	Ce and:	vocs to oxidize	
Konsolakis, 2017	[102]	-	Со	Ethyl acetate	
Piumetti, 2017	[103]	-	Cu	Ethylene	
Saedy, 2017	[104]	-	Al	Toluene	
Issa, 2017	[105]	-	Ti	Ethyl acetate	
He, 2016	[106]	-	Y, Sm, La	Methanethiol	
Carabineiro, 2016	[107]	-	Gd, La, Pr, Nd, Sm	Ethyl acetate	
Topka, 2016	[108]	Pt	Zr	Ethanol, Toluene	
Dinh, 2015	[109]	-	Mn	Trichloroethylene	
Abdelouahab-Reddam, 2015	[110]	Pt	С	Ethanol	
Xue, 2015	[111]	-	Y, Zr, montmorillonite	Acetone, Toluene, Ethyl acetate	
Yosefi, 2015	[112]	-	Cu, clinoptilolite	Toluene	
Nousir, 2015	[113]	-	Zr	Ethyl acetate	
Yang, 2014	[114]	-	Cr	1,2-dichloroethane	
Deng, 2014	[115]	-	Cu, Zr	Toluene	
Perez, 2014	[116]	-	Co, Mg, Al	Toluene	
Sedjame, 2014	[83]	Pt	Al	N-butanol, Acetic acid	
Urbutis, 2014	[117]	-	Cu, NaX	Benzene, Toluene, O-xylene	
Barakat, 2014			Fe, Ni, Ti	Toluene	
Ozawa, 2013	[81]	-	Zr	Toluene	
Konsolakis, 2013	[118]	-	Cu, Sm	Ethyl acetate	
Yue, 2013	[119]	Pd	ZSM 5	Methyl ethyl ketone	
Dziembaj, 2013	[120]	-	Cu	Methanol, Acetone	
Shi, 2012	[121]	Ag	Mn	Formaldehyde	
Pitkäaho, 2012	[84]	Pt, Pd, Rh	Al, Ti, V	Perchloroethylene	
Matějová, 2012	[122]	Pt, Pd	Al	Dichloromethane, Toluene, Ethanol	
Aranda, 2012	[80]	-	Cu	Naphthalene	
Abbasi, 2011	[123]	Pt	Al	Toluene	
Pérez, 2011	[124]	-	Al, Co, Cu, Mg, Pr	Ethanol	
Weng, 2011	[125]	-	Cr, Cu, Mn, Ti	Toluene	
Mayernick, 2011	[126]	Pd	-	Methane	
Liao, 2011	[127]	-	Al, La, Mn, Zr	benzene	
He, 2011	[78]	-	-	O-xylene	
Yu, 2010	[128]	-	Mn, Ti	Toluene	
Masui, 2010	[129]	Pt	Al, Bi, Zr	Toluene	
Ferreira, 2010	[130]	-	Cu	Methane	
Azalim, 2010	[131]	-	Mn, Zr	n-buthanol	
Puertolas, 2010	[79]	-	mesoporous siliceous KIT-6	Naphthalene	
Delimaris, 2008	[132]	-	Mn	Ethyl acetate, ethanol, toluene	
Yan, 2008	[133]	-	Al, Mn, Y, Zr	benzene	
Zhi Min, 2007	[134]	Pt	Mn, Y, Zr,	Toluene	
Gutiérrez-Ortiz, 2004	[82]	-	Zr	Aliphatic C2 chlorohydrocarbons	

Table 1. Cont.

Similarly, Aranda et al. [80] reported that oxygen defects are the key parameter controlling the activity and selectivity of mesoporous copper-doped ceria for the total oxidation of naphthalene. In perchloroethylene oxidation, the Pt/Al_2O_3 -CeO₂ and Pd/Al_2O_3 -CeO₂ catalysts exhibited lower amounts of by-products (trichloroethylene, ethylene and CO) than their analogues without ceria, confirming its positive effect also on the selectivity of the catalysts [84]. Sedjame et al. [83] showed that the addition of ceria to Pt catalysts leads to the formation of numerous intermediate products that have mainly been attributed to ceria-active sites. Nevertheless, this addition improves the catalytic performance for nbutanol and acetic acid oxidations. The authors highlight that the catalytic activity of ceria depends on the nature of the VOCs. Recently, Li et al. [88] and Jiang et al. [94] confirmed the importance of oxygen vacancies that correlated with catalytic activity in the oxidation of different types of VOCs (Table 1).

4. Au/CeO₂ Catalysts for VOC Oxidation

Scirèet al. [135] studied the catalytic oxidation of volatile organic compounds over gold/cerium oxide catalysts. Catalytic combustion of 2-propanol, methanol and toluene was investigated over catalysts prepared by coprecipitation (CP) and deposition-precipitation (DP). DP has been found to be more suitable than CP to obtain highly active Au/CeO₂ catalysts because DP leads to gold nanoparticles, which are preferentially located on the surface of ceria [135]. The effect of the preparation method on the catalytic activity of Au/CeO₂ for VOC oxidation was also investigated by Li and Li [136]. The Au/CeO₂ catalysts were synthesized by CP, DP and metallic colloids deposition (MCD) method, and tested in the total oxidation of toluene. The best results were obtained with 3 wt.% Au/CeO₂ catalyst prepared by the DP method. The catalytic activity of Au/CeO₂ was related to the interaction between gold particles and supports, and the capability of gold particles to weaken the surface Ce–O bonds adjacent to Au atoms. The Ce–O bonds are weakened with the reduction of Ce^{4+} to Ce^{3+} (Figure 2) due to charge transfer to Ce during Au adsorption, which is accompanied by an electron charge transfer between the neutral metal atom and neighboring Ce⁴⁺ cation. Thus, the mobility of the surface lattice oxygen-involved in the VOC oxidation through a Mars-van Krevelen reaction mechanism—increases, which then leads to higher catalytic performance (Figure 3).

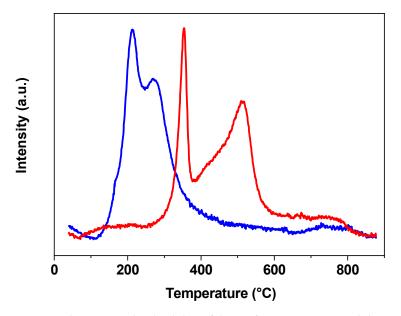


Figure 2. The improved reducibility of the Au/Ce_{0.5}Zr_{0.5}O₂ monolithic catalyst studied in Ref. [137] (blue) compared to the Ce_{0.5}Zr_{0.5}O₂ monolithic catalyst (red) as shown by their H₂-TPR profiles.

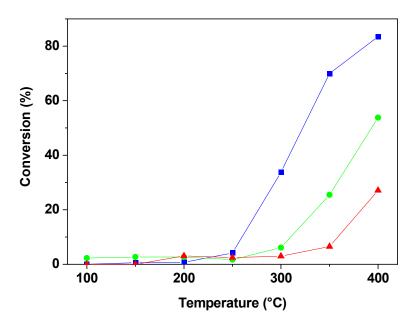


Figure 3. A shift of chlorobenzene conversion curve over the Au/Ce_{0.5}Zr_{0.5}O₂ powder catalysts studied in Ref. [138] with 0.3 wt.% Au (green) and 2.8 wt.% Au (blue) compared to the Ce_{0.5}Zr_{0.5}O₂ support (red).

The importance of the interface between gold and ceria was underlined in the operando IR study of methanol oxidation [139]. The comparison of deposition–precipitation and impregnation method was studied by Aboukaïs et al. [140] for catalytic oxidation of propylene and toluene. They confirmed that the catalysts prepared by deposition–precipitation were more active than those prepared by impregnation due to the high number of gold nanoparticles and Au⁺ species and the low concentration of Cl⁻ ions present on its surface. Generally, the impregnation method requires the right choice of gold source and suitable treatment needed for preparing NPs; e.g., when popular source HAICl₄ is used, gold particles grow rapidly as a consequence of chlorine contamination. A better selection of gold source during impregnation can be Au(PPh₃)(NO₃), [Au₉(PPh₃)₈](NO₃)₃ or [Au₆(PPh₃)₆](BF₄)₂ [141]. Similar preparations can lead to catalytic material offering activities of a different magnitude. Precursor, methodology or support additives could be restrictive, and a more general approach is still needed [142].

Small gold nanoparticles (NPs > 5 nm in diameter) with homogeneous dispersions on the supports seem, so far, the most advantageous for catalytic activity. However, for a good understanding of the effects of gold particle size on catalytic properties, the samples with monodisperse size distributions are preferred; for Au/TiO₂ cathodic plasma deposition method was successfully used [143]. On the other hand, it should be noted that the contribution of large gold agglomerates to the catalytic activity in VOC oxidation was observed as well (Figure 3) [144]. High-resolution electron microscopy is crucial for estimating the size of Au nanoparticles (Figure 4). It should be noted that is difficult to distinguish Au supported on CeO₂ in the TEM image mainly due to low diffraction contrast; HAADF-STEM is more suitable [141]. Moreover, every Au nanoparticle counted in the particle analysis should be confirmed by EDX (Figure 5). Examples of prepared Au catalysts containing ceria (and possibly other compounds) for VOC oxidation are listed in Table 2. The patent of Rhodia company on Au/CeO₂-ZrO₂ catalysts for the treatment of tobacco smoke and polluted air is well known.

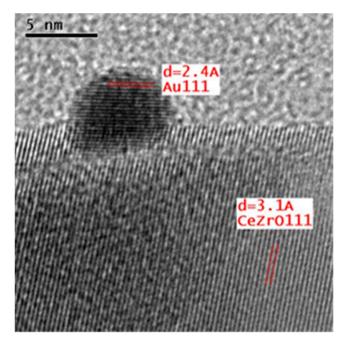


Figure 4. The identification of gold nanoparticles on the ceria–zirconia surface by the spacing of lattice fringes using HRTEM (the Au/Ce_{0.5}Zr_{0.5}O₂ monolithic catalyst studied in Ref. [137]).

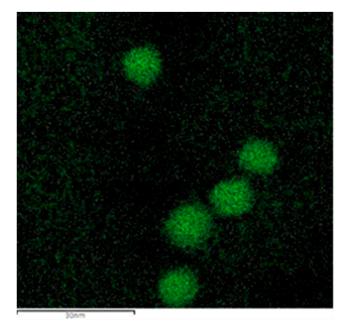


Figure 5. Confirmation of gold nanoparticles by EDX spectroscopy (the $Au/Ce_{0.5}Zr_{0.5}O_2$ monolithic catalyst studied in Ref. [137]).

	-	Catalyst Containing			
st Author, Publ. Year	Ref.	Au loading (wt.%)/Composition	Ce and:	VOCs to Oxidize	
Liu, 2021	[145]	1	-, Nb	Benzene	
Bu, 2020	[146]	1	-	Formaldehyde	
Gaálová, 2019	[144]	0.20-3.35	Zr	Ethanol, Toluene	
Nevanpera, 2019	[147]	Pt–Au, Cu–Au	-, Al	Dimethyl disulfide	
Aboukaïs, 2016	[148]	4	-	Propylene, Toluene	
Wang, 2015	[149]	0.91–1.11	Hydroxyapatite	Formaldehyde, Benzene	
Fiorenza, 2015	[150]	Au–Ag, Au–Cu	-	2-propanol, Ethanol and Toluene	
Idakiev, 2015	[151]	1.36–3.21	Ti, Zr	Methanol, Dimethylether	
Topka, 2015	[138]	0.3 and 2.8	Zr	Chlorobenzene	
Barakat, 2014	[152]	2.36-2.99	Ti, Fe, Ni	Toluene	
Tabakova, 2013	[153]	3	Mn, Fe, Co	Methanol	
Aboukaïs, 2013	[140]	0.5 and 4	-	Propylene and toluene	
Mandal, 2013	[154]	3.5	Mn	Benzyl alcohol	
Matějová, 2013	[155]	0.3 and 2.8	Zr	Ethanol and dichloromethane	
Petrova, 2013	[156]	3	Со	Benzene	
Ciftci, 2013	[157]	<0.01 to 2.14	-	Ethanol and formic acid	
Bastos, 2012	[158]	0.75 and 0.77	(compared to Mn)	Ethyl acetate, ethanol and toluene	
Bazin, 2012	[159]	1	-	Methanol	
Ilieva, 2012	[160]	3	Со	Benzene	
Li, 2011	[161]	2.5-3	-	Formaldehyde	
Solsona, 2011	[162]	2.2	Ni	Propane	
Gaálová, 2011	[68]	0.3 and 2.5	Zr	Ethanol and toluene	
Tana, 2011	[163]	3.2	-	Carbon monoxide	
Bonelli, 2011	[164]	1, 2.2 and 2.7	Fe	Methanol and toluene	
Ousmane, 2011	[165]	1.1	Al	Propene and toluene	
Ying, 2011	[166]	1.1 and 1.7	-	Benzene	
Scirè, 2010	[167]	4.3–4.6	-	Methanol, Acetone, Toluene	
Lakshmanan, 2010	[168]	1	Al	Propene	
Delannoy, 2010	[169]	0.25–4	-	Propene	
Rousseau, 2010	[139]	1	-	Methanol	
Zhang, 2009	[170]	0.42-0.56	-	Formaldehyde	
Gennequin, 2009	[171]	2.64	Ti, Zr	Propene and toluene	
Shen, 2008	[172]	0-0.85	-	Formaldehyde	
Lamallem, 2008	[173]	1.5–3.3	Ti	Propene	
Jia, 2008	[174]	1	-	Formaldehyde	
Gennequin, 2007	[175]	2.7–3.5	Ti	Propene	
Pina, 2006	[176]	1	Zr	Benzene, hexane, chlorobenzene	
Andreeva, 2006	[177]	2.8, 2.9 and 3	Mo, Al	Benzene	
Gluhoi, 2005	[177]	4.5	Al (compared to Co, Mn, Fe)	Propene	
Andreeva, 2004	[179]	2.3 and 3	V, Al	Benzene	
Centeno, 2002	[180]	1.5 and 2.5	Al	n-hexane, benzene and 2-propanol	

Table 2. Recently studied ceria-based Au catalysts for VOC oxidation.

Bonelli et al. [164] reported the design of nano-sized Au/FeO_x catalysts supported on CeO₂ for total oxidation of methanol and toluene. The presence of iron in the CeO₂ structure was found to increase Au dispersion by creating sites with an increased electronic density, which act in a similar way to oxygen vacancies. The deposition of gold nanoparticles was favored at these sites. A strong influence of the nature of dopant was observed by Tabakova et al. [181] as well. The same group evaluated the activity of different doped AuCe catalysts towards the oxidation of CH₃OH; the activity decreased in the order: AuCeCo > AuCe > AuCeFe > AuCeMn [153].

The different behaviour of noble metals [152,163,167] offers the possibility of designing bimetallic catalysts. Lee and Chen reported on the mutual promotional effect of Au–Pd/CeO₂ bimetallic catalysts on the destruction of toluene [182]. The addition of a suitable amount of gold to Pd/CeO₂ increased the proportion of metallic palladium and the catalytic activity.

5. Cordierite Monolithic Catalysts for VOC Oxidation

For practical applications, macro-structured catalysts are preferred over powdered catalysts. A low-pressure drop in the exhaust system, good thermal resistance, refractoriness, good washcoat adherence and compatibility between the washcoat and the catalyst are only a few excellent operational properties of cordierite monoliths, which are more than suitable for gas phase applications [183]. A catalytic layer can be deposited onto cordierite monolith by numerous techniques, selected accordingly to the VOCs to be oxidized. Similar to powder catalysts, the selectivity to carbon dioxide is important, as the by-products formed (Figure 6) may be more detrimental than the original VOC.

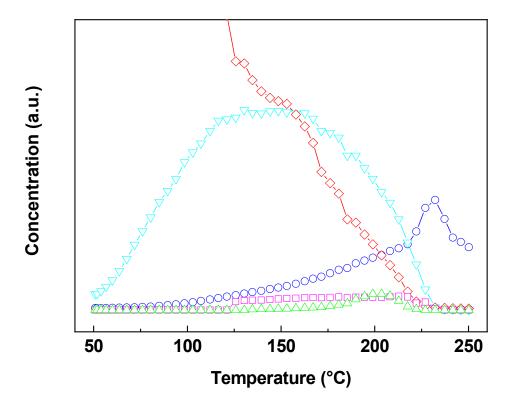


Figure 6. The evolution of by-products during ethanol oxidation over the powder Au/Ce_{0.5}Zr_{0.5}O₂ catalyst studied in Ref. [68]: ethanol (red), ethylene (blue), acetaldehyde (cyan), acetic acid (magenta) and ethyl acetate (green).

The catalytic combustion of toluene over Fe–Mn mixed oxides supported on cordierite monolith was investigated by Ma et al. [184]. The high dispersion of the mixed oxides on the surface of the cordierite support was the most important factor for the catalytic combustion of VOCs. Total oxidation of naphthalene at low temperatures using palladium nanoparticles

supported on inorganic oxide-coated cordierite honeycomb monoliths was the subject of the research of Varela-Gandía et al. [28]. Thin films of ZSM-5 and BETA zeolites, and a SAPO-5 silicoaluminophosphate (by in situ synthesis), then γ -Al₂O₃ (by a dip-coating), were coated onto a cordierite honeycomb monolith. The coated monoliths were impregnated with polymer-protected Pd nanoparticles and tested for the total oxidation of naphthalene. From the combined use of the zeolite with polymer-protected nanoparticles, enhanced catalytic properties were found for the total abatement of the representative of VOCs. A high degree of stability remained even after undergoing accelerated ageing experiments.

Aguero et al. reported the catalytic combustion of different VOCs: ethyl acetate, n-hexane, and its mixture [185]. Cordierite monolith was dipped in colloidal alumina slurry, dried and calcined. After that, impregnation with aqueous solutions of $(CH_3CO_2)_2Mn \times 4 H_2O$ and $(CH_3CO_2)_2Cu \times 4 H_2O$, drying and calcination was carried out. Prepared MnCu-mixed oxide catalysts with homogeneous and well-adhered coatings revealed very good activity and stability in the oxidation of VOCs.

Zhao et al. tested $Cu_xCo_{1-x}/Al_2O_3/cordierite$ monolithic catalysts (x = 0–1) in the catalytic combustion of toluene [186]. A boehmite primer sol was the first washcoat layer, and copper with cobalt oxides was the activity washcoat layer. The $Cu_{0.5}Co_{0.5}/Al_2O_3/cordierite$ catalyst with Co^{2+} and Co^{3+} on its surface showed the best catalytic activity; toluene was completely oxidized at 315 °C.

Zhou et al. [187] studied MnO_x –CeO₂/La–Al₂O₃ monolithic catalysts prepared by the deposition–precipitation method, impregnation method and a combination of the two methods in toluene oxidation. The results showed that the catalyst prepared by the deposition–precipitation method showed the best catalytic activity and possessed richer surface oxygen species, a higher proportion of Mn^{4+} and good low-temperature redox properties.

From existing patents dealing with catalysts supported on ceramic monoliths with honeycomb structure for VOCs oxidation, the sequential adsorptive capture and catalytic oxidation defined in the US 6479022 B1 patent can be mentioned [188]. The inventors recommend grinding the active component to a powder with a particle size smaller than 20 microns, followed by a coating over the carrier such as cordierite. A method for the catalytic oxidation of VOC/CO in the presence of organosilicate compounds describes the patent EP 0993857 B1 [189]. As an example of suitable ceramic materials, it states cordierite, preferred for the treatment of gases containing halogenated organics. Cordierite-based monolithic catalyst without coating layer is the key idea of the CN 102989451 A patent [190]. The acidification treatment and the coating technology of the selected carrier are not required; the load capacity of the metal active component is controllable; the metal active component has good stability. The invention mainly solves the problems of complicated processes and the high cost of the preparation of the existing cordierite monolithic catalyst.

6. Au Monolithic Catalysts for VOC Oxidation

Gold catalysts for indoor air quality control already appeared as prototype products in the public domain [191]. Air-cleaning devices are required for removing CO, VOCs and ozone from ambient air in confined spaces such as submarines or space crafts on long missions and indoor office space [192]. The gold/titanium dioxide photocatalytic/thermocatalytic coating (where gold particles have a size lower than 3 nm) is used by Carrier Corporation (US) for the simultaneous oxidation of VOCs and carbon monoxide [193]. When photons of ultraviolet light are absorbed by Au/TiO₂ coating, reactive hydroxyl radicals are formed. They oxidize a contaminant, adsorbed on the coating, to H₂O, CO₂ and other substances. Gold is an oxidation catalyst that lowers the barrier energy of CO to be oxidized to carbon dioxide. It is particularly gainful that the activity of Au catalysts may be promoted by moisture. Oxidation of VOCs was studied using an inlet from the electrophotographic printing apparatus vapor stream [194]. The invention detailed the technology linking an apparatus and method for the oxidation of VOCs from a printer, particularly a combination of an unsupported and a supported oxidation catalyst. A method of dividing the supported oxidation catalyst into sections to obtain efficient oxidation of VOCs is included in the patent. Gas from within the printer body is conducted to a catalytic treatment system with separate contact regions for an unsupported oxidation catalyst (metal selected from the platinum group metals or from the first transition series) and a supported oxidation catalyst on different ceramic materials. Since 1992, Au/Fe₂O₃ supported on washcoated honeycomb has been used in Japanese toilets for the elimination of bad smells [195]. The commercial use of gold-based catalysts is rather limited for now, but this field is undergoing an exciting period of discovery" [196].

7. CeO₂-Based Cordierite Monolithic Catalysts for VOC Oxidation

Ce oxides have been known for many years as active and selective catalysts for VOC oxidation due to their oxygen storage capacity (OSC), redox properties and catalytic performances. The rheological properties of the suspension that contains the active elements are critical for obtaining a satisfactory washcoating [197]. The viscosity, concentration or pH of the slurry influences the adhesion ability of oxides towards cordierite honeycombs [198]. Table 3 displays several examples of successful depositions of ceria (possibly with other compounds) on cordierite monolith for VOC oxidation.

1st Author, Publ. Year	Ref.	Catalyst Supported on Cordierite Monolith Containing		VOCs to Oxidize
		Noble Metal	Ce and:	-
Sedjame, 2018	[199]	Pt	Al, Zr,	Acetic acid
Zhou, 2018	[187]	-	Mn, La, Al	Toluene
Azalim, 2013	[197]	-	Mn, Zr	N-butanol
Huang, 2013	[200]	-	Mn	Benzene
Jiang, 2013	[201]	Pt, Pd	Al, Zr	Toluene, benzene and styrene
Papavasiliou, 2009	[202]	Pt	Al, Zr, La	Propene
Liotta, 2009	[203]	Pt	Al, Zr	Propene
Zhi Min, 2007	[134]	Pt	Al, Zr, Mn, Y,	Toluene
González-Velasco, 2002	[204]	Pt	Zr	Propene
Agrafiotis, 2001	[205]	Pd, Rh	Al, Ca	Exhaust gases (hydrocarbons)

Table 3. Recently studied ceria containing VOC oxidation catalysts supported on cordierite monoliths.

Agrafiotis et al. [205] evaluated nanophase calcia-doped-ceria-based washcoats deposited on ceramic honeycombs (via sol-gel methods) with respect to catalytic activity for hydrocarbons conversion and thermal aging. The activity of the systems was found to depend strongly on the deposition method (directly from the sol or from suspensions of sol-gel derived powders), on the type of precursor used for preparing the starting sols (metallo-organic or inorganic) and on the method used to insert the noble metal(s) in/on the washcoat.

González-Velasco et al. studied 0.58Pt/Ce_{0.68}Zr_{0.32}O₂/ceramic monolith for simultaneous elimination of NO, C₃H₆ and CO [204]. Changes in activity have been related to the role of platinum and ceria–zirconia mixed oxide in the reaction milieu; the authors claim the advantages of using ceria–zirconia mixed oxides instead of CeO₂/Al₂O₃ to improve stability and oxygen storage capacity. Zhi Min et al. [134] demonstrated that the catalytic activity of Pt/ γ -Al₂O₃/Ce_{0.50}Zr_{0.50}O₂ monolithic catalyst could be greatly improved by doping Y and Mn into Ce_{0.50}Zr_{0.50}O₂.

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Remarkable stabilization of the ceria–zirconia solution towards CeAlO₃ formation operated by Pt under redox conditions managed to achieved Liotta et al. [203] by preparation of structured 1Pt/CeZr/Al catalyst, dip-coated over a cordierite monolithic support. Pt supported catalysts ceria–zirconia was present as a Ce_{0.6}Zr_{0.4}O₂ homogeneous solid solution, and the deposition over the cordierite does not produce any structural modification; moreover, no Pt sintering occurred.

Among the preparation procedures for Ce–Zr–La modified Pt/ γ -Al₂O₃ catalyst— (i) simultaneous coprecipitation of γ -alumina and ceria-based solid solution from metal nitrates, (ii) sequential coprecipitation of these components and (iii) wet impregnation with different γ -alumina powders—simultaneous coprecipitation led to the most thermally stable washcoat [202]. Papavasiliou et al. observed higher dispersion of the oxide modifiers in the alumina carrier, impeding the Pt sintering.

The monolithic Pt–Pd bimetallic catalysts supported on γ -Al₂O₃ using cordierite honeycomb ceramics as the first carrier were prepared by thermal adsorption method by Jiang et al. [201]. The Pt–Pd content of 0.1% was found to be suitable; the prepared catalyst shows high activity and stability. On the contrary, cerium-based catalyst supported on cordierite with no noble metal was used for catalytic combustion of benzene by Huang et al. [200]. Their results specified that the MnCeO_x/Cord catalyst with Mn/Ce molar ratio of 1:1, calcined for 7 h and Mⁿ⁺/(citric acid) molar ratio of 6 exhibited the highest catalytic activity. Recently, the direct deposition of Ce–Zr–Mn mixed oxide on the cordierite has been proposed as an alternative and promising way to the alumina-coated cordierite followed by noble metal impregnation for VOCs catalytic removal by Azalim et al. [197]. The retaining of the great textural and the redox properties of the Ce_{0.12}Zr_{0.40}Mn_{0.48}O₂ mixed oxides, already observed on the powder catalyst, was most likely the main reason for the excellent catalytic properties of the CeZrMn(0.48)/M monolith.

Several cordierite monolithic catalysts with ceria washcoat for VOC abatement were patented as well. An instance can be the US-8475755-B2 patent [206], which includes platinum group metals, one of which is either platinum or ruthenium.

8. Au/CeO₂ Cordierite Monolithic Catalysts for VOC Oxidation

Gold and ceria supported on a cordierite monolith are employed in several published patents. The US-7329359-B2 patent describes the use of Ce-Zr mixed oxides, together with a promoter from the transition metal oxides group, coated on a ceramic cordierite monolith for the destruction of VOCs in liquid media [207]. Exhaust gas (including VOCs) treatment catalyst is proposed in the WO 2013088091 A1 patent [208]. A catalytic system consisting of an oxide (preferably of fluorite crystalline structure) corresponds to the molar formula: $Ce_{1-u}M_vA_zD_vO_{2-x}$, where Ce is cerium, A represents at least one element having several degrees of oxidation, M represents at least one element chosen from Gd, Y, Se, Sm, Nd, Pr and Zn, D represents at least one element different from A and chosen from Pt, Pd, Rh, Ru, Cu, Fe, Ag and Au, u is between 0.05 and 0.45, y is between 0.01 and 0.4, z is greater than 0 and less than 0.4, v is between 0.001 and 0.4 and x is greater than 0. The VOC oxidation catalysts based on Ce-Zr mixed oxides, noble metals, and promoters are proposed in other patents [209,210]. For adequate exposure of the catalysts to the contaminated air stream, without producing excessive back pressure, the catalyst is deposited on structured support. Another catalyst compositions useful for destruction of VOCs in an oxygen-containing gas stream at low temperatures (the patent WO 2001045833 A1 [211]) comprises one or more first metals selected from the group consisting of: Ce and Zr; and at least one of: (a) one or more second metals selected from the group consisting of: Gd, La, Sr and Sc; (b) one or more third metals selected from the group consisting of: Ti, V, Mn, Fe, Co, Cr, Ni, Au, Ag and Cu; and (c) one or more fourth metals selected from the group consisting of Pt, Pd, Rh, Ru, Re, Os and Ir. Catalyst compositions provided may be single-phase, mixed-metal oxides, or multi-phase.

Monoliths coated with perovskites and ion exchange materials are described as highly efficient catalytic materials for VOCs abatement in the following patents: US 5882616 A [212] and US 6200483 B1 [213].

The method of coating a honeycomb monolith substrate with a catalyst component is proposed in the WO 2011080525 A1 patent [214]. Ceria is recommended as one of the suitable coatings for supporting the catalytically active metals, preferably from the following group: Pt, Pd, Au, Ag, Ir, Ru, Rh and Os. Methods of preparing highly dispersed monolithic gold catalysts are described in the WO2011106213 patent [215]. Nanosized gold catalysts are supported on the washcoated monoliths, stabilized by the addition of a third metal oxide, which may include a variety of transition metal oxides such as cerium oxide. The introduction of monolithic supported Au catalysts potentially can be beneficial for environmental catalyses, such as the low-temperature oxidation of VOCs and CO.

Recently, we proposed two methods of gold introduction during the preparation of $Au/Ce_{0.5}Zr_{0.5}O_2$ cordierite monolithic catalysts [137]. We have demonstrated that the order of gold and ceria introduction may influence gold particle size and thus the catalytic activity. The enhanced reducibility of the catalysts with higher Au dispersion led to an improved catalytic performance in ethanol oxidation.

9. Perspective

Except for further improvement of the preparation methods aimed to obtain required gold particle size and particle size distribution, the effect of gold nanoparticles on ceria or ceria-zirconia support and its catalytic activity would deserve further studies. For example, it is generally accepted that the optimal gold particle size is in the range of nanometers. However, we have shown that in the oxidation of chlorobenzene, even large gold particle agglomerates (~100 nm) may contribute to the catalytic performance of the $Au/Ce_{0.5}Zr_{0.5}O_2$ catalysts (cf. Figure 3) [138]. Furthermore, the role of the Au–CeO₂ interface is important and not yet fully understood. Depending on the type of VOC to be oxidized, the perimeter of Au nanoparticles may form the active site for the reaction. For example, for ethanol oxidation, it was reported that the presence of highly dispersed Au nanoparticles induced a new oxidation pathway [216]. Another interesting point might be the effect of ceria morphology (nanorods, nanocubes). It was already shown that the morphology of ceria might affect the interaction of the support with gold nanoparticles and thus the catalytic activity. For example, Bu et al. recently reported that ceria nanorods with exposed (110) and (100) planes possessed a large number of oxygen vacancies and promoted the creation of Au^{3+} near the surface [146].

10. Conclusions

Gold and ceria are well-known catalysts that already proved their performance in the total oxidation of VOCs. While ceria itself may be highly active, its ability to oxidize given VOC can be improved by depositing gold nanoparticles. Furthermore, gold nanoparticles can improve the selectivity of catalysts. This is crucial, as the oxidation by-products may be more detrimental than the original VOC. Choosing the proper preparation method is crucial to obtain catalysts that are able to totally oxidize VOCs at low temperatures and avoiding deactivation. Gold nanoparticles deposited on the partly reducible ceria is an especially suitable combination due to the oxygen storage/release capacity of such catalysts. The decisive role of oxygen vacancies and their mobility was confirmed in many studies. On the other hand, the catalytic activity and selectivity of gold/ceria catalysts may be substantially influenced by the nature of the VOC to be oxidized.

The kinetic behavior of the supported catalysts is usually affected mainly by their reducibility, acidobasic properties, morphology, and oxidation state of the active phase. The number and mobility of surface vacancies and lattice oxygens—involved in VOC oxidation through a Mars-van Krevelen reaction mechanism—is a key issue. The activity of the gold/ceria catalysts is influenced by gold particle size. It was revealed that the small particle size of gold, ~5 nm, has a positive effect on the catalyst performance; such Au/CeO₂

systems are preferably prepared by deposition–precipitation. New approaches, such as plasma deposition, allow to obtain the narrow size distribution of gold nanoparticles and achieve a better understanding of the effects of gold particle size. Attention should be paid to a proper determination of the gold particle size, especially when using an electron microscope; HAADF-STEM is preferable to distinguish gold nanoparticles on the ceria surface and should be confirmed by EDX. The limiting factors of the oxidation process using Au/CeO_2 based powder catalysts are poisoning phenomena (ceria is sensitive to halogen and sulfur-containing compounds), thermal stability (to prevent both gold and ceria agglomeration) and mass and heat transfer limitations.

Catalytic technology at an industrial scale uses mainly monolithic washcoated catalysts with precious metals (Pt, Pd, Rh) supported on oxygen storage components, such as ceria or ceria–zirconia mixed oxides. The application of monolithic catalysts to VOC removal is a crucial issue for practical applications in order to minimize the pressure drop in the reactor, allowing treatment of high flow rates frequently used in environmental applications. The factors influencing the activity of Au/CeO₂ based washcoats are similar to powder catalysts. However, the method used to introduce the gold in/on the washcoat (e.g., washcoating powder Au/CeO₂ catalyst or introducing gold to already washcoated CeO₂) may influence the gold particle size and thus the activity of the catalyst.

Author Contributions: J.G.: investigation, supervision, and original draft; P.T.: methodology, formal analysis, review and editing. Both authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Technology Agency of the Czech Republic, project number TN01000048, and by the Czech Science Foundation, grant number 19-08153Y.

Conflicts of Interest: The authors declare no conflict of interest.

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