

Role Of Catalysts In The Decomposition Of Volatile Organic Compounds (VOCs) In Air Stream

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ABSTRACT : This article reviews the role of catalysts for catalytic decomposition of VOC in air. Major classes of catalyst formulations studied can be divided into four main categories viz supported metal catalysts, metal exchanged zeolites, perovskites and structured catalyst. The most active metals are noble metals (Pt and Pd) and oxides of metals (Cr, Co, Cu, Fe, Mn and V) but activity varies with the type of VOC used. The high activity of a catalyst is commonly associated with high metal loading, high metal dispersion and the selection of suitable support. Chlorinated VOCs pose major challenge to the activity and viability of catalytic decomposition process. The decomposition of VOCs with low H/Cl ratio is relatively low and problems of catalyst deactivation and formation of higher chlorinated species still remain unsatisfactorily solved. Thermal and hydrothermal stability received much attention among researchers as the activity of the catalyst should be sustained in a prolonged high temperature operation in humid air stream. The decomposition of mixture of VOCs cannot be predicted from the behaviour of individual component as promotional or inhibition effect does occur. The presence of water in feed stream can retard the activity of a catalyst due to competitive adsorption but for low H/Cl ratio VOCs, it serves as H supplier to suppress the formation of higher chlorinated by-products in favour of HCl. Successful conversion of VOC should be evaluated with high selectivity toward deep oxidation products (H_2O , CO_2 or HCl).

Keywords : VOCs, catalysts, oxidative decomposition, deactivation, chlorinated VOC, multicomponent VOCs, humid feed stream, product of incomplete combustion.

INTRODUCTION

Volatile organic compounds (VOCs) are an important class of air pollutants, emitted from many industrial processes (Kim and Ihn, 1999) and transportation activities (Becker and Förster, 1998 ; Niu et al., 1999). Catalytic combustion is one of the most promising technology for the decomposition of VOCs due to its definitive character and save of energy (Zhang et al., 1997). Catalytic oxidative decomposition is similar to thermal oxidation in that the gaseous impurities to be destroyed are reacted with oxygen at elevated temperature in form primarily carbon dioxide and water vapour. It differs in that the reactions are made to occur at a relatively low temperature by the use of a solid catalyst (Kohl and Nielsen, 1997). Typically, the reaction temperature is in the range of 150° to 480°C (Rafson, 1998), and is determined on the basis of the material being oxidized.

THE CATALYST

Many catalyst chemical formulations and geometric shapes are used to promote oxidation-reduction reactions (Rafson, 1998). Chemical types used for VOC oxidation include platinum, platinum alloys, copper chromate, copper oxide, cobalt oxide, chromium oxide, manganese oxide and nickel. The catalysts are often categorized as platinum metal group (PMG) and base metal (or metal oxide) types (Schneider et al., 2000). The active catalysts are often supported on an inert carrier such as $\gamma-Al_2O_3$. Catalyst forms include metal ribbons, mesh and gauze, honeycomb monoliths and small

beads or particles that can be used in a fixed, fluidized or moving bed (Kohl and Nielson, 1997).

Noble metals (Pt and Pd) are among the most commonly used metal catalysts in investigations of VOC catalytic decomposition (Brink et al., 2000). Apart from the higher specific activity, they are favoured because of their resistance to sulfur poisoning (Niu et al., 1999) than oxide catalysts. In addition, highly dispersed Pt and Pd catalysts are easily prepared using a number of support materials (Zhang et al., 1997 ; Paulis et al., 2000).

The kinetics and mechanism of catalytic oxidation of hydrocarbons have been studied extensively. According to Zwinkels et al. (1993), there is correlation between the rate of hydrocarbon oxidation and metal-oxygen bond strength. Generally, this correlation can be presented as so-called volcano plot (Figure 1). A clear maximum in the plot is observed for platinum-group metals.

Metal (Group V_B to II_B) oxides are, in general, less active than the noble metals but are more resistant to poisoning (Sinquin et al., 2000). The most active oxides have a p-semiconductor nature and the most frequently used ones are the oxides of V, Cr, Mn, Fe, Co, Ni and Cu. Chromium oxide is claimed to be one of the most active catalysts in chlorinated VOC destruction (Padilla et al., 1999), but the authors reported the formation of highly toxic by-products such as chromium oxychloride ($Cr_2O_2Cl_2$) and phosgene ($COCl_2$).

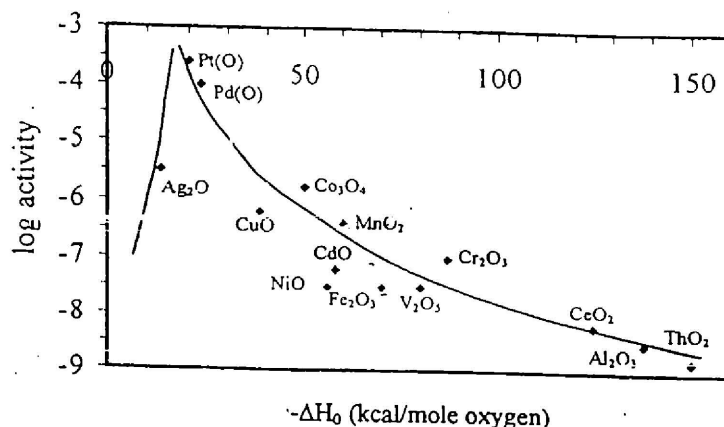


Figure 1 : Volcano-type plot indicating specific activity in hydrocarbon oxidation as a function of the metal-oxygen bond strength (Zwinkels et al., 1993).

Supported Metal Catalyst

γ - Al_2O_3 catalysts as well as Pt and Pd on various supports (TiO_2 , SiO_2 , ZrO_2 etc) have been extensively studied (Brink et al., 2000). However, it seems that these catalysts are deactivated more or less depending on the oxidation system. Sinquin et al. (2000) claimed that after a slight initial deactivation, the activity is normally stabilized over a long period. Several authors point to deactivation particularly at low temperature ($T < 350^\circ - 400^\circ\text{C}$) and on small metal particles (Tajima et al., 1997 ; Amores et al., 1999). The life time of these catalysts seems, therefore to depend on the reaction conditions and the preparation method.

Table 1 summarizes typical preparation method of support catalysts. As can be seen, most of the researchers employed either the incipient wetness or impregnation method to introduce the active metal catalysts into the support. Paulis et al. (2000) introduced a new method of impregnation via impregnation using citric acid solution. However, distinctive characteristics of the resulting final catalyst was demonstrated. In most of the recent researches, it is catalyst compositions and not the preparation method that received more emphasis.

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Table 1 : Supported catalysts composition for VOC decomposition.			
Reference	Catalyst	Support	Preparation/remarks
Tajima et al. (1997)	Pd, Fe, Co, Ni, Cr, Mn, Mo, V & W	TiO_2 - ZrO_2	TiO_2 - ZrO_2 support was prepared by sol-gel method. Metals were introduced through impregnation method. Aqueous solutions were added, stirred, dried, calcined (650°C , 2 hrs in air), grained and sieved (10-20 mesh).
Zhang et al. (1997)	Pt	Fluorinated carbon powder	Impregnation with H_2O - CH_3OH solution containing $\text{Pt}(\text{NH}_3)_4(\text{NO}_3)_2$, dried (105°C , 16 hrs), H_2 reduction (250°C , 16 hrs). Specific surface area= $150\text{m}^2/\text{g}$, Pt dispersion=9%.
Ng et al. (1998)	Cr, Ni, Mn & Co (chloride salts)	γ - Al_2O_3 & SiO_2 - Al_2O_3	Incipient wetness method. Chloride salts were dissolved in deionized H_2O , mixed and dried overnight at 120°C . Used for hydrolytic decomposition of CFC-12.
Amores et al. (1999)	Mn	TiO_2 (anatase)	Wet impregnation method. Adding salt precursor, stirring (100°C), drying (150°C , 3 hrs) & calcination (500°C , 3 hrs). Specific surface area= 72 - $85\text{m}^2/\text{g}$.
Patterson et al., (1999)	Rh	δ - Al_2O_3	Impregnation of δ - Al_2O_3 containing 22% wt. CeO_2 with Rh solution in HCl, calcination in air at 500°C . Specific surface area= $27\text{m}^2/\text{g}$.

Metal-Exchanged Zeolites

Zeolites can be formulated to carry active metals catalysts. The merit of zeolite-based catalysts is the active metal components are actually exchanged with the existing cations in the zeolites. Consequently, better metal dispersion down to nearly molecular level is expected. This property, coupled

with the size and shape features of the zeolites, allow the materials to be used as an effective catalyst in extremely selective reactions (Marcus and Cormier, 1999).

The presence of multivalent cations within the zeolite matrix has been found to increase its activity by creating highly acidic centres through hydrolysis of charged cations (Becker

and Förster, 1998). The following systems have been investigated: Cu-Y and Pd-Y/benzene (Becker and Förster, 1998), Pd-HFAU/*o*-xylene (Dégé et al., 2000) and Cr-ZSM-5/TCE (Chintawar and Greene, 1997) and the results are very promising. However, problems regarding catalyst deactivation due to poisoning by chlorinated VOCs still remain unresolved (Brink et al., 1998; Kim and Ihm, 1999).

Hydrophobicity and thermal resistance are found to increase with an increase in $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of zeolites (Marcus and Cormier, 1999). Hydrophobic zeolites have recently gained much attention due to their ability to selectively remove one or more organic pollutants from humid air streams (Dégé et al., 2000). More hydrophobic zeolites such as ZSM-5 types, dealuminated faujasites and silicalite are found to be effective in removing VOCs from dilute or humid air streams (Marcus and Cormier, 1999). This property is expected to eliminate some of the problems associated with catalyst deactivation in the presence of high humidity (Becker and Förster, 1998).

Perovskites

Perovskites can be used given their ability to stabilize the oxides on the catalytic system (Singuin et al., 2000). They are well known for their performances in total oxidation of hydrocarbons and/or CO as reviewed by Zwinkels et al. (1993) and Tofan et al. (2000). Among the mixed metal oxide catalysts, perovskites-type oxides with the general formula ABO_3 ('A' represents a lanthanoid and/or alkaline earth metal ion and 'B' a transition metal ion) have been found to be effective catalysts for the total oxidation of hydrocarbons and oxygenated compounds as well as chlorinated hydrocarbons (Schneider et al., 2000; Singuin et al., 2000).

New and efficient methods of preparation using low cost precursors assures production of fine high specific surface area perovskite powders. Some research groups have reported their success of developing perovskite compositions which can conserve high activity even after calcination at temperatures above 1300K as reviewed by Tofan et al. (2000). Most of the research works on perovskites development for VOC decomposition include manganese and cobalt (Schneider et al., 2000; Singuin et al., 2000) as an active element. Other compositions involve the use of strontium in addition to manganese and cobalt (Tofan et al., 2000). These materials were found to be equally promising and satisfactory for high temperature VOC catalytic combustion.

Structured Catalysts

Monolithic or honeycomb materials are unitary structures composed of inorganic oxides or metals in the structure of a honeycomb with equally sized and parallel channels which may be square, sinusoidal, triangular, hexagonal and round (Rafson, 1998). Monoliths are often washcoated with catalytically active high surface area materials such as γ -

Al_2O_3 (Barresi and Baldi, 1994), zeolite (Chatterjee and Greene, 1991) or perovskite (Schneider et al., 2000).

From the technical point of view, monolithic catalysts have several advantages compared with catalysts in pellet form e.g. the faster mass and heat transfer in the thin catalyst layer (Schneider et al., 2000) and the lower pressure drop in the reactor (Kohl and Nielsen, 1997). These materials can offer high thermal shock resistance, high mechanical strength and high melting point (Rafson, 1998). Other advantages are excellent attrition resistance, good mechanical properties, compact and freedom in reactor orientation as summarized by Heck and Farrauto (1995).

The washcoat influences the thermal shock resistance of the monolith (especially during rapid temperature changes) because it expands more than the monolith (Rafson, 1998). These differences must be factored into design of the finished catalysts to maintain good adherent coating without substantially weakening the monolith. There is also a limit to how much washcoat can be deposited on the monolith. Too much deposition results in a decrease of the effective channel diameter, thereby increasing the pressure drop to an unacceptable level (Kohl and Nielsen, 1997).

DECOMPOSITION OF CHLORINATED VOCs

Until recently, the use of catalytic oxidizers was restricted to nonhalogenated streams because halogen, especially chlorine, would poison the catalysts used. Brink et al. (2000) reported that noble metals were powerful oxidation catalysts, but can easily form inorganic chlorides with deactivation as a likely consequence. Reportedly the problem lies more with the catalyst carrier or washcoat rather than the catalyst (Kohl and Nielsen, 1997). Al_2O_3 support, for example, can react with chlorinated species to give AlCl_3 that subsequently can vaporize above 178°C (Velasco et al., 2000).

The quest for ideal catalysts for chlorinated VOC decomposition has received much attention among researchers. Chromia-alumina catalysts have been studied extensively as one of the most promising catalysts among commercially available materials (Kim and Ihm, 1999; Pradier et al., 2000). Brink et al. (2000) reported unsatisfactory result of their experiments on oxidative decomposition of chlorobenzene over Pt/ γ - Al_2O_3 . A study by Ng et al. (1998) concluded that metal chloride promoted γ - Al_2O_3 is an active catalyst for the hydrolytic decomposition of CFC-12 but the presence of water can depress the activity. Tajima et al. (1997) reported the excellent activity of $\text{WO}_3/\text{TiO}_2\text{-ZrO}_2$ supported catalysts for the decomposition of CFC113 but undesirable product CO in an amount equimolar to CO_2 was also detected.

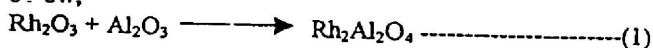
In the decomposition of high Cl/H ratio VOCs, the addition of water in the feed stream is actually needed to improve the conversion (Velasco et al., 2000). Typically, water as hydrogen-supplying reactant is added to generate HCl by suppressing the formation of Cl_2 because HCl could be readily scrubbed downstream of the catalytic oxidizer (Kim

and Ihm, 1999 ; Padilla et al., 1999). Beside water, hexane, toluene (Velasco et al., 2000) methane and propane (Schneider et al., 2000) have also been studied for the same positive role. Brink et al. (2000) have extended the scope of the study to the removal of chlorine from catalyst surface. They came to the conclusion that removal of Cl from the surface (mainly in the form of HCl) by hydrocarbons is responsible for reducing the formation of by-products.

THERMAL AND HYDROTHERMAL STABILITY

In order to develop various heat-resistant or steam resistant alumina materials, many research works have been done recently, such as modifying transition alumina with additives of rare earth metal oxides (Ng et al., 1998), alkaline earth metal oxides (Zwinkels et al., 1993), silica and other oxides such cerium oxide (Patterson et al., 1999). High temperature operation can cause conversion of catalyst supports to a new crystal structure. For example, when γ - Al_2O_3 convert to α - Al_2O_3 , there is significant stepwise decrease in the internal surface area from about 150 to < 50 m^2/g . Similarly, the higher surface area (60 m^2/g) TiO_2 anatase structure converts to rutile (< 10 m^2/g) at about 550°C. The conversion versus temperature curve will usually undergo a loss in activity (Heck and Farrauto, 1995).

Reaction of the active catalytic component with the carrier can be a source of deactivation if the product is less active than the initial dispersed species. Pradier et al. (2000) reported a formation of chromium silicate and chromium aluminate from interaction between chromia and alumina and silicate respectively. Rh_2O_3 is reported to react with a high surface area Al_2O_3 to form an inactive compound during high-temperature operation (Heck and Farrauto, 1995). The reaction is believed to be occurring is conceptually shown below;



Becker and Förster (1998) claimed that compared to alumina supporting Pd catalysts, zeolite exchanging Pd catalysts have higher and more durable activity in low-temperature catalytic combustion of benzene. Unfortunately, the catalysts are very unstable and lose their activity easily in steam because the

hydrothermal treatment results in sharp dealumination of zeolites framework and ultimately leads to the collapse of the structure. Luckily, an increase in $\text{SiO}_2/\text{Al}_2\text{O}_3$ of the zeolites has been demonstrated to significantly improve their hydrothermal stability (Chintawar and Greene, 1997).

DECOMPOSITION OF MULTICOMPONENT VOCs

The presence of mixtures of VOCs which is usual for 'end-of-pipe' air pollution control may affect the design criteria, operation control and performance characteristics of a catalytic incinerators (Dangi and Abraham, 1997). Sharma et al. (1995) and Barresi and Baldi (1994) reported that it is often not possible to predict what effect one component might have on the rate of oxidation of the other. Thus, it is very difficult to evaluate the performance characteristic of a given catalytic incinerator as applied to a specific VOC mixture, based on the behaviour of individual components.

Earlier studies have found that a mixture effect exists in the oxidation of hydrocarbons, mostly in the form of inhibition of oxidation of one hydrocarbon in the presence of others. Therefore, when inhibition effect is encountered, a higher temperature is usually needed to achieve a similar destruction level. It should also be noted that in some systems, promoting effect do occur (Table 2). However, no report has so far been made on inter-promoting effect between VOC components. Often, the promotional effect on the conversion of one component is at the expense of the others. Various reasons for this phenomenon can be put forward, ranging from simple competitive-reactions rationale (Barresi and Baldi, 1994) to additional heat production (Velasco et al., 2000) or the formation of specific intermediates in the conversion of the additive (Sharma et al., 1995) but none of them satisfactorily explains the result.

Dangi and Abraham (1997) used the Mars-van Krevelen model to obtain the kinetic parameters needed to predict the mixture conversion over platinum catalyst. They tried to extend pure-component model to the two-component mixture of benzene and MTBE. Padilla et al. (1999) suggested that for destruction of chlorinated VOCs, the reaction rate improves with an increase in H/Cl ratio brought about by addition of H supplying organic substances.

Table 2 : Summary of recent researches concerning VOC mixtures decomposition.

Reference	Experimental conditions			Finding/Conclusion
	VOC	Catalyst	Temperature	
Baresi and Baldi (1994)	Toluene, xylene, benzene & ethylbenzene	Pt/cordierite	420°-480°C	Toluene and xylene conversion are inhibited by benzene and ethylbenzene
Sharma et al. (1995)	Methanol, acetone & methylene chloride	Pt/ & Pd/fluorinated carbon	50°-400°C	Methanol : promoting destruction of acetone and methylene chloride. Acetone : promoting destruction of methylene chloride while inhibiting destruction of methanol. Methylene chloride : inhibiting destruction of methanol and acetone.

Becker and Förster (1998)	Benzene & toluene	Cu- & zeolite Y	Pd-	220°-350°C	Toluene promotes destruction of benzene only for Pd-Y catalyst.
Brink et al. (2000)	2-butene, toluene, heptane & chlorobenzene	Pt/ γ -Al ₂ O ₃		100°-500°C	Chlorobenzene conversion is promoted by 2-butene, toluene and heptane. Chlorobenzene retards toluene and heptane conversion.
Velasco et al. (2000)	TCE, hexane & toluene	Pd/ & Al ₂ O ₃	Pd/ γ -	200°-550°C	Hexane and toluene promote TCE conversion over both catalysts.

THE EFFECT OF HUMIDITY IN FEED STREAM

Most of the industrial waste gases are often saturated with water vapour (Ng et al., 1998). Moisture can cause considerable problems with the practical application of catalytic emission control in the sense that it generally inhibits or poisons the oxidation catalyst especially at low temperatures (Becker and Förster, 1998). Thus the development of low-temperature water-resistant catalysts is highly desirable (Zhang et al., 1997). The fact that the catalysts are more reactive in the absence of water shows that while water is a necessary reagent for the hydrolysis reaction (Sinquin et al., 2000), its presence

also has an inhibiting effect on the decomposition reaction (Ng et al., 1998).

Typically in a chlorinated VOC decomposition system, water is added to generate HCl by suppressing the formation of Cl₂ because HCl could be readily scrubbed downstream of the catalytic oxidizer before exiting the stack (Kim and Ihm, 1999). The presence of water in feed stream is known to affect components in the product gas. Table 3 summarizes some of the effect observed by some recent research.

Table 3 : Effect of water on product gas compositions.

Reference	Operating conditions		Temperature	Findings
	VOC/gas	Catalyst		
Ng et al. (1998)	CFC-12	LaO ₂ / & SiO ₂ / γ -Al ₂ O ₃	350°-400°C	H ₂ O increases CO ₂ yield but suppresses CCl ₃ F and CCl ₄ formation.
Kim and Ihm (1999)	TCE	CoCr ₂ O ₄ / and ZnCr ₂ O ₄ / γ -Al ₂ O ₃	330°C	H ₂ O suppresses Cl ₂ and tetrachloroethylene by products' formation.
Patterson et al. (1999)	Benzene/CO & toluene/CO	Rh/ & Rh/CeO ₂ /Al ₂ O ₃ with residual chlorine	100°-400°C	H ₂ O reduces formation of chlorobenzene from benzene/CO but increases yield of benzene from toluene/CO.
Holmgren et al. (1999)	CO, ¹⁶ O ₂ & ¹⁸ O ₂	Pt/CeO ₂	45° & 140°C	H ₂ O improves selectivity to C ¹⁶ O ₂ & C ¹⁶ O ¹⁸ O
Schneider et al. (2000)	dichloromethane	LaMnO ₃ coated monolith	500°-650°C	H ₂ O suppresses CCl ₄ , C ₂ HCl ₃ and C ₂ Cl ₄ formations but Cl ₂ formation is not influenced.

The weakly adsorbed water acts as an inhibitor of the reaction. However, without water replenishing the surface hydroxyl groups (Patterson et al., 1999), the hydrolytic activity is not sustainable. Water vapour can influence the activity of supported Pt catalysts in two ways. First, water molecules compete with reactant molecules for the surface Pt sites and therefore, affect the adsorption and activation of VOC and oxygen (Holmgren et al., 1999). Second, the accumulation of water on the support by adsorption and capillary condensation can affect the interaction between Pt and support, and can even deactivate some surface active sites (Zhang et al., 1997).

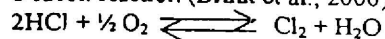
Hydrophobic catalysts have been intensively investigated for humid VOC decomposition applications (Chintawar and Greene, 1997; Niu et al., 2000). Hydrophobicity and thermal resistance are found to increase with an increase in SiO₂/Al₂O₃ ratio of zeolites (Chintawar and Greene, 1997; Marcus and Cormier, 1999). More hydrophobic zeolites such as ZSM-5 types, dealuminated faujasites and silicalite are

found to be especially effective in removing VOCs from dilute or humid air streams (Marcus and Cormier, 1999). This property is expected to eliminate some of the problem associated with catalyst poisoning in the presence of high humidity (Becker and Förster, 1998).

PRODUCTS OF INCOMPLETE COMBUSTION (PICS)

In catalytic combustion processes for hydrocarbons and oxygenates, partial oxidation reactions must be avoided and CO₂ and water being possibly the only products (Baldi et al., 1998; Zhang et al., 1997). According to the more strict limits for the emissions of some partially oxidized compounds, the presence of even small amounts of species e.g. aldehydes, can make the mixture resulting from catalytic combustion processes even more pollutant than the waste to be purified (Baldi et al., 1998).

Catalytic oxidation of chlorinated VOCs constitutes one of the most significant challenge as far as deep or complete oxidation is concerned. Ideally, there should be only HCl as the chlorine-containing species in the product gas. Chintawar and Greene (1997) have shown that among the first row transition metals, including Ni and Mn, only Cr has the ability to catalytically destroy TCE effectively. However, chromium residues are an environmental concern. Cr containing catalysts are also associated with the presence of elemental Cl₂ in the product gas due to well documented Deacon reaction (Brink et al., 2000);



Therefore, finding either an environmental friendly replacement or an active catalyst formulation containing very low levels of chromium would be worthwhile.

The addition of water as co-feed in the inlet gas has received considerable attention among researchers (Becker and Förster, 1998 ; Padilla et al., 1999 ; Velasco et al., 2000). Water can act as an H supplier to the decomposition of high Cl/H ratio substances to improve selectivity toward HCl (Velasco et al., 2000) and to suppress Deacon reaction. It can also suppress the formation of PICs in many systems as what have been summarized in Table 4. Decomposition of CFC113 in the presence of water was studied over WO₃, MoO₃ and V₂O₅ supported on TiO₂-ZrO₂ and satisfactory conversion was achieved but the product gas contained considerable amount of C₂Cl₄F₂ and C₂Cl₂F₄ by-products (Tajima et al., 1997). Normally, selectivity toward deep oxidation products deteriorates with time due to catalyst deactivation.

CONCLUSION

Supported metal catalysts, metal exchanged zeolites, perovskites and structured catalyst are main categories of catalyst systems currently being studied for VOC decomposition. The most active metals for this purpose are noble metals (Pt and Pd) and metal oxides (Cr, Co, Cu, Fe, Mn and V) but each metal has its own merits and weaknesses. Metal loading and dispersion as well as type of support are the main determinant in the activity of a catalyst. Problems of catalyst deactivation and formation of higher chlorinated species still remain unsatisfactory resolved in catalytic abatement of chlorinated VOCs. Thermal and hydrothermal stability are another key factors in successful application of catalytic process and the development of stable catalysts is under way. Promotion or inhibition effect might occur during the decomposition of mixture of VOCs and the behaviour of the decomposition cannot be based on individual component. Competitive adsorption might occur in the presence of water causing the retardation of the catalyst activity. Water can suppress the formation of chlorinated by-products during high H/Cl ratio VOCs decomposition and favour HCl formation. Successful conversion of VOC should be evaluated with selectivity toward deep oxidation products (H₂O, CO₂ or HCl).

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