Simulation of plasma-assisted catalytic reduction of NO\textsubscript{x}, CO, and HC from diesel engine exhaust with COMSOL

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**Highlights**
- This study was conducted to simulate removal of diesel engine exhaust gas pollutants.
- Proposed process consists of plasmacatalytic after treatment.
- Nitrogen oxides, propane, and radical produced dependence of the electric field modeled.
- Nitrogen oxide, carbon monoxide, methane, and propane reduction in different velocities and temperatures achieved.

**Abstract**
In the present paper, computational fluid dynamic (CFD) model was applied to simulate for the first time a synergetic application of a hybrid plasma and catalyst comprising DOC (Diesel Oxidation Catalyst) with Pt/\text{CeO}\textsubscript{2}-\text{Al}\textsubscript{2}O\textsubscript{3} and SCR (Selective Catalytic Reaction) with V\textsubscript{2}O\textsubscript{5}/TiO\textsubscript{2}. Plasma-catalytic can be used in cold start diesel engine exhaust to reduce pollutants when catalyst is not hot enough. Plasma system was a pulsed corona consisting of the coaxial wire-cylinder reactor. NO\textsubscript{x} and C\textsubscript{3}H\textsubscript{6} reduction, NO\textsubscript{2}, NO, and radical produced concentration dependence of the electric field intensity were achieved. Transient temperature and CO distribution at catalyst channel, catalyst back pressure, and NO distribution all over the CC (catalytic converter) concentration were investigated to determine the steady state reduction efficiency of NO, CO, CH\textsubscript{4}, and C\textsubscript{3}H\textsubscript{6} with respect to inlet velocities and temperatures. In velocity 3 m/s, temperature 600 K, highest electric field before sparkover at 17 kV/cm, and 80 s engine cold start time, the reduction efficiencies of NO and C\textsubscript{3}H\textsubscript{6} got about 9% and 99% respectively. In steady state condition CO, NO, and C\textsubscript{3}H\textsubscript{6} reduction are about 98%, 29%, and 95% in order.

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**1. Introduction**

Diesel engine converts diesel fuel chemical energy into mechanical power (Nett Technologies Inc., 2017). Diesel fuel is mainly used in trucks, buses, and off-road engines (ToxProbe Inc., 2017). In comparison with gasoline, diesel is more durable, has more particulate matter exhaust and thermal efficiency, and less fuel cost. Carbon monoxide (CO), nitric oxides (NO\textsubscript{x}), and hydrocarbons (HCS) are three of the most important diesel engine exhaust gases. CO, NO\textsubscript{x}, and HCs have an important role in ozone and smog formation in the lower atmosphere. NO\textsubscript{x} can cause acid rain. HCs can be carcinogenic and contribute to the greenhouse effect (Steffens, 2003). Britain, France, and China have decided to stop diesel cars production due to their combustion pollutants. Madrid and Athens are two major metropolises that ban using these cars. Volvo Corporation moves toward manufacturing electric engines. However, German car producers in July 2017 decided to survive the diesel car industry because of hundreds of thousands of people employed...
combined with an activated Ag/Al₂O₃ catalyst for NOx removal. Non-thermal plasma for diesel exhaust treatment modeling (Mok et al., 2012; Francke et al., 2000). In recent publications, the idea of this research was created to use plasma assisted catalytic converter in a diesel engine to reduce gas pollutants like NOx and hydrocarbons in engine cold start, which exhaust gas temperature isn't warm to react well, and take time to reduce pollutants efficiently. The focus of this research is to find out the coupling between non-thermal plasma and catalytic converter effects on NOx and HCs in diesel engine exhausts. This includes consideration of the complete device, including discharge electrode (a thin wire) stretched along the axis of a grounded cylindrical electrode as plasma part and multi-channel catalytic converter. Computational fluid dynamic (CFD) is used to simulate the plasma assisted catalyst in this paper. Plus, we consider oxidation of carbon monoxide and hydrocarbons, and nitrogen oxide over V₂O₅/TiO₂ and Pt/CeO₂-Al₂O₃ catalysts. Density and viscosity of exhaust gas are not constant and depend on temperature and pressure.

### 2. System description

3D CFD modeling is resource intensive, thus two-dimensional structure is considered in simulations. This calculates efficiently and retains the important physics. The geometry is similar to the one used by previous work (Agrawal et al., 2012). The main difference with the earlier works is that the flow equation in the entire device is solved with plasma and catalytic reactions, fluid convection and diffusion, and heat transfer effects. This let us to understand how those affect flow distribution. The geometry has 55 channels. The modeled catalytic monolith consists of 27.5 channels.

### Nomenclature

**Greek letters**
- \( \delta_w \) wall offset (m)
- \( \eta \) the eddy viscosity (m²/s)
- \( \rho \) gas density (kg/m³)
- \( \varepsilon \) porosity
- \( \eta \) gas dynamic viscosity (Pa s)

**Dimensionless numbers**
- \( X \) total number of mole fraction of any radical

**Abbreviation**
- CC catalytic converter
- CFD computational fluid dynamic
- DC direct current
- DOC diesel oxidation catalyst
- EHD electro hydrodynamic
- eV electron volt
- HC hydrocarbon
- SCR selective catalytic reduction
- VOC volatile organic compounds

**Dimensionless numbers**
- \( X \) total number of mole fraction of any radical

| \( |N_i|_d \) | the total radical concentration produced |
|-----------|
| \( Q_m \) | mass source (kg m⁻³ s⁻¹) |
| \( Q_T \) | heat source (W m⁻³) |
| \( D_i \) | diffusion coefficient of reactant i (m² s⁻¹) |
| \( R_i \) | reaction rate of reactant i (mol m⁻³ s⁻¹) |
| \( c_i \) | concentration of reactant i (mol m⁻³) |
| \( k_{pre} \) | permeability (m²) |
| \( \Gamma_c \) | radical concentrations |
| \( a \) | constant factor |
| \( A \) | frequency factor |
| \( b \) | constant factor |
| \( c \) | constant factor |
| \( \sigma_p \) | specific heat capacity (J kg⁻¹ K⁻¹) |
| \( d \) | constant factor |
| \( D \) | dipole moment (D) |
| \( E \) | activation energy (J/mol) |
| \( h \) | mesh element diameter |
| \( h_r \) | hour |
| \( l_T \) | turbulent intensity |
| \( k_d \) | dissociation rate constant (cm³ molecules⁻¹ s⁻¹) |
| \( k_e \) | depletion reaction constant rate of O(1D) (cm⁻³ molecules⁻¹ s⁻¹) |
| \( K_\Gamma \) | constant rate of reactions (cm³ molecules⁻¹ s⁻¹) |
| \( k_T \) | thermal conductivity (W m⁻¹ K⁻¹) |
| \( l_T \) | turbulent length scale (m) |
| \( M \) | third body reaction |
| \( n \) | temperature power |
| \( O(1D) \) | the excited atomic oxygen (molecules cm⁻¹) |
| \( \Omega \) | radical |
| \( OH \) | radical |
| \( H \) | radical |

**Dimensionless numbers**
- \( X \) total number of mole fraction of any radical

| \( P \) | gas pressure (pa) |
| \( r \) | reaction rate (cm³ molecules⁻¹ s⁻¹) |
| \( \bar{c} \) | reactants concentrations (mol/m³) |
| \( \bar{s} \) | species molecular weight (kg/mol) |
| \( T \) | absolute temperature (K) |
| \( u \) | gas velocity (m/s) |
| \( U \) | the velocity field of the gas (m/s) |

### Abbreviation
- CFD computational fluid dynamic
- DOC diesel oxidation catalyst
- EHD electro hydrodynamic
- HC hydrocarbon
- SCR selective catalytic reduction
- VOC volatile organic compounds

(Eddy and Ewing, 2017). Many efforts have been made on studying non-thermal plasma for diesel exhaust treatment modeling (Mok and Nam, 2002; Penetrante et al., 1997; Chae, 2003; Mok and Ham, 1998). The experimental study of CC (catalytic converter) behavior in a diesel engine is discussed in many resources (Athrashalil Phaily et al., 2014). CO, NO, and HC conversion efficiency, velocity, and temperature distribution inside the monolith are described (Braun et al., 2000). Pressure drop at the CC, transient 2D temperature profiles inside the monolith and velocity vectors inside the monolith channels are modeled in many sources (Tsinoglou, 2004). Synergetic plasma-assisted CCs have been widely used in diesel engine industry to decompose contaminant gases. Structure of set-ups, different parts, and materials have been explained in many papers (Penetrante et al., 2001). These combinations are made to remove volatile organic compounds (VOCs) and NOx (Miessner et al., 2002; Francke et al., 2000). In a recent publication, the interaction between carbon dioxide and ethane in a plasma reactor with a vanadium/alumina catalyst dispersed on the BaTiO₃ was investigated (Gómez-Ramírez et al., 2013). Plasma combined with an activated Ag/Al₂O₃ catalyst for NOx removal with using hydrocarbon SCR (Selective Catalytic Reaction) was studied in some references (Stere et al., 2014). In all researches about catalytic reduction, one thing is common that catalyst can’t start properly in cold start time, because in this period, catalyst isn’t warm to react well, and take time to reduce pollutants efficiently. Engine cold start is the time that most of the harmful exhausts are emitted before catalytic converter has warmed up in all time engine running (Liu et al., 2001; Guojiang and Song, 2005). The main questions are that: can plasma be helpful in cold start in automobile? How much time does plasma need to work in lack of high catalytic reduction? What is the reduction efficiency of the pollutants? Which pollutants are affected by plasma and why? The geometry (will be explained in the next part) is close to the one used by previous research (Agrawal et al., 2012). From all previous works, the idea of this research was created to use plasma assisted catalyst in a diesel engine to reduce gas pollutants like NOx and hydrocarbons in engine cold start, which exhaust gas temperature is not high enough that catalyst can’t reduce them. The focus of this research is to find out the coupling between non-thermal plasma and catalytic converter effects on NOx and HCs in diesel engine exhaust. This includes consideration of the complete device, including discharge electrode (a thin wire) stretched along the axis of a grounded cylindrical electrode as plasma part and multi-channel catalytic converter. Computational fluid dynamic (CFD) is used to simulate the plasma assisted catalyst in this paper. Plus, we consider oxidation of carbon monoxide and hydrocarbons, and nitrogen oxide over V₂O₅/TiO₂ and Pt/CeO₂-Al₂O₃ catalysts. Density and viscosity of exhaust gas are not constant and depend on temperature and pressure.
due to axial symmetry. In the present work, a two-dimensional CFD model in COMSOL Multiphysics 3.5a and COMSOL Reaction Engineering Lab was applied to calculate the concentrations of CO, NO, and HC in a plasma-assisted CC in a diesel engine exhaust. The results show pollutant reduction efficiency with respect to inlet velocities and temperatures. Furthermore, transient catalyst temperature and back pressure were analyzed using this simulation. Fig. 1 presents 3D schematic of the proposed plasma reactor assisted CC in a car. The filter is placed horizontally in the car exhaust system. The catalyst was arranged in a separate downstream to the plasma reactor. Engine exhaust enters the plasma reactor to convert HC and NOx gases. Plasma reactor is shown in Fig. 1. In addition, to reduce CO, the reactor is connected to a CC, such that the filtered clean gas exits after two stages filtration. Fig. 1 indicates different areas of CC in simulation. Three areas are defined for this purpose: Area (1) is washcoat, area (2) is a wall, and area (3) is free space that flow crosses the catalyst channel. Fig. 1 demonstrates all the areas of catalytic converter. The plasma reactor in this research contains a discharge electrode (a thin wire) stretched along the axis of a grounded cylindrical electrode. In this study, we analyze the pulsed corona that is a system consisting of a metallic tube (aluminum or copper) as the ground electrode with a 400 mm length, 30 mm diameter, and a stainless steel rod as the high-voltage electrode with a 0.5 mm diameter. The size of the monolith is 200 mm long and 88 mm in diameter. Substrate wall thickness is 400 μm, and channel diameter is 1 mm. The geometrical data is got from previous experiments in plasma and catalyst fields (Mok and Nam, 2002; Agrawal et al., 2012).

3. CFD modeling

The commercial CFD package, COMSOL is used for simulations in this work. The flow inside the catalytic convertor is assumed to be two-dimensional. The gas is assumed to be non-ideal and compressible. The typical range of operating velocity inside monolithic channels is from 1 to 8 m/s. The inlet temperature is in range of 400 K to 600 K. The flow inside the monolith channel is laminar. Since the flow upstream and downstream of the monolith is turbulent, the standard k-ε model is used. Parameters, like turbulent length scale were default values in COMSOL. The specific heat and other thermodynamic properties are calculated from the COMSOL database. The solid properties are constant. Modeling consists two parts plasma and catalyst, which are explaining in sequence.
3.1. Plasma simulation

When plasma reactor connects to high voltage, gas discharge happens, so the following reactions take place in an electron-molecule collision.

\[ e + H_2O \rightarrow OH + H + e k_{d1} = 3.3 \times 10^{-10} \]  
(1)

\[ e + O_2 \rightarrow O + O + e k_{d2} = 1.0 \times 10^{-9} \]  
(2)

\[ e + O_2 \rightarrow O + O^{(1D)} + e k_{d3} = 5.2 \times 10^{-9} \]  
(3)

\[ e + N_2 \rightarrow N + N + e k_{d4} = 2.0 \times 10^{-11} \]  
(4)

Where \( k_{d1}, k_{d2}, k_{d3}, k_{d4} \) are dissociation constant rate that are expressed in cm\(^3\) molecules\(^{-1}\) s\(^{-1}\). In an electron-oxygen collision, O radical and the excited atomic oxygen \( O^{(1D)} \) are produced. These atoms with high level of energy collide with molecules, and make more O and OH radicals.

\[ O^{(1D)} + H_2O \rightarrow OH + OH k_{e1} = 2.3 \times 10^{-10} \]  
(5)

\[ O^{(1D)} + N_2 \rightarrow O + N_2 k_{e2} = 2.6 \times 10^{-11} \]  
(6)

\[ O^{(1D)} + O_2 \rightarrow O + O_2 k_{e3} = 4.0 \times 10^{-11} \]  
(7)

\[ O^{(1D)} + H_2O \rightarrow O + H_2O k_{e4} = 1.2 \times 10^{-11} \]  
(8)

Where \( k_{e1}, k_{e2}, k_{e3}, k_{e4} \) are the depletion reaction constant rate of O \((1D)\). The bond dissociation energy of \( N_2, H_2O, \) and \( O_2 \) are 9.8, 5.2, and 5.1 eV, respectively. Because of the high chemical bond energy of nitrogen, electrons affect the \( H_2O \) and \( O_2 \) more. Thus O, OH, and H radicals play an important role in pollutant reduction (Mok and Ham, 1998).

\[ X_{OH} = X_H = \frac{k_{d1}[H_2O]}{2[k_{d1}[H_2O] + (k_{d2} + k_{d3})[O_2] + k_{d4}[N_2]]} \]  
(9)

\[ X_O = \frac{(2k_{d2} + k_{d3})[O_2]}{2[k_{d1}[H_2O] + (k_{d2} + k_{d3})[O_2] + k_{d4}[N_2]]} \]  
(10)

\[ X_{OH}(1D) = \frac{k_{d3}[O_2]}{2[k_{d1}[H_2O] + (k_{d2} + k_{d3})[O_2] + k_{d4}[N_2]]} \]  
(11)
\[ X_N = \frac{k_{d4}[N_2]}{[k_{a1}[H_2O] + (k_{a2} + k_{a4})[O_2] + k_{d4}[N_2]]} \]  

(12)

Where the total radical concentration produced by direct electron dissociation impact \([N_i]_d\) is (Mok and Ham, 1998):

\[
[N_i]_d = (a + b\varepsilon + cE^2 + dE^3) \times 10^3
\]

(13)

\(a, b, c, d\) are constant factors.

\(a = 111.60, b = 2.4975, c = -18.59, d = 0.46\)

H, N, OH, and O radical concentrations are:

\[ [H]_c = X_{Ni}[N_i]_d \]  

(14)

\[ [N]_c = X_{Ni}[N_i]_d \]  

(15)

\[
[OH]_c = \frac{k_{d4}[H_2O][N_i]_d \exp[-(k_{d4} + k_{a4})[H_2O] + k_{a2}[N_2] + k_{a4}[O_2]]t_c - 1}{-[k_{d4} + k_{a2}[H_2O] + k_{a4}[O_2] + k_{a4}[N_2]]} \times X_{Ni}[N_i]_d
\]

(16)

\[ [O]_c = (k_{a2}[N_2] + k_{a4}[O_2]) + k_{d4}[H_2O] + k_{a2}[N_2] + k_{a4}[O_2]]t_c - 1}{-[k_{d4} + k_{a2}[H_2O] + k_{a4}[O_2] + k_{a4}[N_2]]} + X_{Ni}[N_i]_d
\]

(17)

Radicals have a high tendency to react with contaminants and convert them. The most important reactions are (Chae, 2003):

\[ H + O_2 + M \rightarrow HO_2 + M \]  

(18)

\[ NO + O + M \rightarrow NO_2 + M \]  

(19)

\[ N + NO \rightarrow N_2 + O \]  

(20)

\[ HO_2 + NO \rightarrow OH + NO_2 \]  

(21)

\[ NO_2 + O \rightarrow NO + O_2 \]  

(22)

\[ H + NO_2 \rightarrow OH + NO \]  

(23)

\[ OH + N \rightarrow H + NO \]  

(24)

\[ N + O_2 \rightarrow NO + O \]  

(25)

\[ O + C_2H_6 \rightarrow CH_2CO + CH_3 + H \]  

(26)

\[ CH_2 + O_2 \rightarrow O + CH_3O \]  

(27)

\[ O + O_2 + M \rightarrow O_3 + M \]  

(28)

\[ NO + O_3 \rightarrow NO_2 + O_2 \]  

(29)

\[ NO + OH + M \rightarrow HNO_2 + M \]  

(30)

Where M is a very important parameter and is considered as the third body reaction. It participates in reactions and is dependent on the pressure, but is not consumed. It also participates in engine combustion-compression reactions. M rises reaction rate and equals the sum of all reactant concentrations, but each gas has special factor (COSMOL, 2008). \(N_2\) is a solvent, so it has a constant concentration. Furthermore, space velocity is another factor in pulse electric field intensity that is effective on pollutant reduction. Its unit is h\(^{-1}\).

\[
\text{Space velocity} = \frac{\text{reactor inlet velocity}}{\text{reactor length}}
\]

(31)

Space velocity residence time in the reactor should be long enough, so electric discharge has enough opportunity to act on pollutants. Space velocity range that does not affect efficiency is 9200–29,500 h\(^{-1}\) (Mourimine, 2005). To simulate chemical reaction on filter geometry, two equations mass transfer and momentum transfer are solved at the same time.

\[
\frac{\partial c_i}{\partial t} + \nabla \cdot (-D_1 \nabla c_i) = R_i - u \nabla c_i
\]

(32)

\[
\nabla \cdot U = 0
\]

(33)

\[
\rho (U \cdot \nabla) U = -\nabla P + \nabla \cdot \left( \eta \left( \nabla u + (\nabla u)^T \right) \right) + \rho s \nabla V
\]

(34)

Where \(c_i\) is concentration of reactant \(i\) (mol m\(^{-3}\)), \(D_1\) is diffusion coefficient of reactant \(i\) (m\(^2\) s\(^{-1}\)), and \(u\) is gas velocity (m/s).

### 3.2. Catalyst simulation

In oxidation catalyst DOC (Diesel Oxidation Catalyst), the most important reactions are (Tsinoglou, 2004):

\[ CO + 0.5O_2 \rightarrow CO_2 \]  

(35)

\[ C_2H_6 + 4.5O_2 \rightarrow 3CO_2 + 3H_2O \]  

(36)

\[ CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O \]  

(37)

\[ H_2 + 0.5O_2 \rightarrow H_2O \]  

(38)

SCR, which is used for NO reduction in diesel engines, needs an NH\(_3\) injection to react with NO and O\(_2\) (CHEMKIN-II. Version3.6., 1994).

\[ 4NO + 4NH_3 + O_2 \rightarrow 4N_2 + 6H_2O \]  

(39)

\[ 4NH_3 + 3O_2 \rightarrow 2N_2 + 6H_2O \]  

(40)

Reactions occurring in washcoat are similar to those in the other two areas (COSMOL, 2008). Mass transfer is the same as Eq. (32). Other equations are:

\[
\rho C_p \frac{\partial T}{\partial t} + \nabla \cdot (-k_1 \nabla T) = Q_T - \rho C_p u \cdot \nabla T
\]

(41)

\[
\frac{\partial}{\partial t} (\rho u) + \nabla \cdot (\rho u u) = 0
\]

(42)

\[
\frac{\rho u}{\varepsilon} \frac{\partial}{\partial t} (\rho u) + \left( \frac{\eta}{k_{per}} + Qu \right)u = \nabla \cdot \left[ -p I + \frac{1}{\varepsilon} \left( \eta (\nabla u) + \eta (\nabla u)^T - \frac{2}{3} \eta \right) \nabla u \right] + F
\]

(43)

Eq. (41) is heat transfer, where \(c_p\) is specific heat capacity (J kg\(^{-1}\)K\(^{-1}\)), \(T\) is an absolute temperature (K), \(k_1\) is thermal conductivity (W m\(^{-1}\) K\(^{-1}\)), and \(Q_T\) is heat source (W m\(^{-3}\)). In washcoat areas, heat transfer is conduction (area 1) and in other areas exists both conduction and convection. To make a more effective catalyst, flow in channel has to be laminar (area 3). Momentum transfer occurs according to Eqs. (33) and (34); however, in washcoat (area 1), these equations are turned into Eqs. (42) and (43). Where \(Q_{in}\) is mass source (kg m\(^{-3}\) s\(^{-1}\)), \(k_{per}\) is permeability (m\(^2\)), and \(\varepsilon\) is porosity. Washcoat specifications and physical properties, such as both SCR and DOC are incorporated in the modeling. Washcoat characteristics in two types of catalysts are listed in Table 1 (COSMOL, 2008).
3.3. The computational grids

Since flow is turbulent, meshes near the wire and wall are finer than middle of plasma reactor to show differences well. Boundary layer meshes are in the inlet and outlet. Number of edge elements are 45 and element ratio is 7. Distribution method is linear and symmetric. On the wire and wall, number of edge elements are 35, element ratio is 1, because in Z direction, differential changes are not as important as r direction and distribution method is linear. Predefined mesh sizes are fine. All the meshes are quadrilateral. Mesh number of degrees of freedom, points, and elements are 55,000, 1692, and 1610 respectively. In calculation domain for 2D mesh, element ratio is 1, because in Z direction, differential changes are less than middle of plasma reactor to show differences well. Boundary conditions are mentioned in Table 4 (ToxProbe Inc., 2017).

3.4. Solver parameters and details

In developing the kinetic model, following assumptions are applied:

- It is assumed that the radicals are produced immediately after discharging, because this time is less than nanoseconds.
- There are no concentration differences in the radial direction, so gas flow can be considered as plug flow.
- Since the plasma region cannot be defined, assumed that plasma can happen in whole of the reactor.
- The reactor is isothermal, because increase in temperature is negligible.

The number of mole fraction of any radical is the ratio of the concentration of any radicals produced by discharge to total radical concentration produced by direct electron dissociation impact at the initial stage of (O/D) depletion reactions. It is modeled with MATLAB. Produced radical concentrations were imported to COMSOL Reaction Engineering Lab. All the chemical reactions are simulated in COMSOL Reaction Engineering Lab 3.5. Since plasma reactions are non-thermal, so temperature is assumed to be constant and energy equation is neglected; however, transport parameters are incorporated in the reaction. Reactor type is defined as plug-flow and corona discharge occurring throughout the reactor (Mok and Nam, 2002). To solve the reactions, parameters like constant rate of reactions $k$, activation energy $E$ (J/mol), frequency factor $A$, reaction rate $r$, species molecular weight (kg/mol), potential characteristic length ($10^{-10}$ m), potential energy minimum (K), Dipole moment (D), and initial reactants concentrations (mol/m$^3$) are imported to the software (ToxProbe Inc., 2017; CHEMKIN-II, Version3.6., 1994; NASA Polynomial, 2015). Both density and dynamic viscosity are dependent on pressure and temperature. After solving chemical reaction, to simulate them on filter geometry, reactions are executed to COMSOL Multiphysics 3.5 and two equations mass transfer and momentum transfer are solved at the same time. In an isotherm case, $D_i$ depends on pressure (Santos and Costa, 2008). The flow field is affected by the corona discharge, so there is electro hydrodynamic (EHD). Corona discharge takes place in an adiabatic heating process. EHD flow is under the assumption of incompressible, steady-state, and isothermal flow. The governing equations are the continuity equation and the Navier-Stokes equation. The dimension of filter, fluid characteristics, and inlet velocity make the flow turbulent (COSMOL, 2008). Gas pressure is $P$ (pa), $p$ is gas density, $\eta$ is gas dynamic viscosity (Pa s), $U$ is the velocity field of the gas (m/s), $\eta_1$ is the eddy viscosity, and $\rho_V$ Coulomb force (N/m$^3$). There is a coupling factor between flow field model and corona discharge model. Geometry is solved in an axial symmetry case. All the boundary conditions are mentioned in Table 4.

$I_T$ is turbulent intensity, $L_T$ is turbulent length scale, $\delta_w$ is wall offset, and $h$ is mesh element diameter. All the chemical reactions are simulated in COMSOL Reaction Engineering Lab 3.5. Temperature changes and transport parameters are incorporated of reactor modeling. To model the reactions, in addition to the parameters mentioned in a plasma reactor, other ones including species Lower, Midpoint, and Upper temperature (K) and species Low and High polynomial coefficients are imported to the software (CHEMKIN-
II. Version3.6., NASA Polynomial, 2015). After solving the chemical reactions, to simulate them on catalyst geometry, reactions are exported to COMSOL Multiphysics 3.5 and three equations mass, momentum, and heat transfer are solved at the same time. Boundary conditions in CC simulation are presented in Table 5. The walls of the CC are non-catalytic, no-slip, and zero flux boundary conditions. The reaction takes place only on the surface of washcoat. In case of non-isothermal simulations, the heat of reaction is also included in the model.

The COMSOL CFD code solves simultaneously the mass, momentum, species and energy balance equations. The equations are expressed in the standard conservative form and solved using the finite element method (COSMOL, 2008).

4. Results and discussion

The results from CFD simulations are divided into two parts in this section. The first one is plasma impact analyzing and the second one is about catalyst effects on pollutants.

4.1. Plasma effect on pollutants

Radical produced concentrations according to Eqs. (1)–(8) at different electric field intensities are presented in Table 6. As electric field is increased, radical concentrations are also raised. It is obvious in Eq. (13) that the total radical concentration [Ni]d is related to electric field, thus it can affect H, N, OH, and O radical based on Eqs. (14)–(17). It means that incline in applied voltage will ascend the electric field; consequently, those will improve radical production. In stronger electric field, more forces are applied on free electrons; hence they collide with molecule with more intensity. Finally, more radicals are gained. According to the Table 6, O and OH have the highest radical concentration. Therefore, these two radicals have more important role than N and H in contaminant descend. Based on the defined range velocity according to the space velocity in Eq. (31), maximum inlet velocity can be 3.2 m/s. If higher velocity is demanded, the reactor length should be increased.

NOx concentration distribution in electric field intensity 17 kV/cm, temperature 600 K, steady-state conditions, and axial symmetry are presented in Fig. 2. According to Eqs. (18)–(30) and (32)–(34), in Fig. 2a, NO2 concentration is increased, while in Fig. 2b, NO is reduced. It describes that in plasma, radical reactions with pollutants, tend to increase NO2, which is not favor for us. However, plasma effect on NO is acceptable and is in interested direction for environment. Totally, NOx is declined. In the other words, NO reduction is higher than NO2 production. Since plasma is non-thermal, temperature along the reactor is not changing, and there is no need to solve energy equation. It is clear that plasma has affected on pollutants at the beginning of reactors, unlike the catalytic converter that can make impact along the catalyst. All the concentration units are in mol cm⁻³.

4.2. Catalyst impact on pollutants

Transient temperature distribution according to the Eqs. (35)–(43) at catalytic channel in inlet velocity 3 m/s and temperature 600 K in the first 90 s of the engine power-on, where catalyst is still too cold (cold start), is presented in Fig. 3. At the end of 10 s, the beginning of channel's temperature numbers is near the inlet temperature. With time passing (20 s, 30 s, and 50 s), channel's temperature is increased along the reactor and at the end of the 90 s, almost all of the channel has a temperature near the inlet gas. Hence, 90 s is the time lasts that catalyst to get to its highest performance. Plasma, which described in previous part can show its role inside first 90 s. In channel, heat transfer consists conduction and convection, while in washcoat surface there is only conduction. Consequently, in this CC, about 90 s is needed to have a warm catalyst to affect pollutants well. The shape of flow is parabolic that shows it is a laminar flow. It proves that simulation is correct.

The time dependency of the CO concentration distribution at catalyst channel at inlet velocity 3 m/s and temperature 600 K is given in Fig. 4. Based on previous part, with the passing of time, washcoat temperature rises, thus Arrhenius factor is improved, and CO reduction efficiency is enhanced according to Eq. (35). At the end of 90 s, CO in all the channel is converted. That’s because, by increasing the catalyst temperature, the energy needed to overcome activation energy of chemical reactions is reduced. Therefore, with time passing, engine exhausts and catalyst can react easier, and pollutants are converted more. At 10 s, the catalyst is still cold; hence CO reduction efficiency is low. When time increases, e.g. reaches 20 s, 30 s, and 50 s, the catalyst becomes hotter, so more efficient reactions may occur. Most of these reactions take place in 90 s, which leads to the highest CO reduction efficiency. In higher temperature, reaction rate goes up, while activation energy sets down. From this, it is clear that about 90 s takes to CC gets warm to get more efficient in reduction, and agrees well with part (a) in timing. The highest reduction rate happened in the spaces, where washcoat is placed, because velocity is in lowest level there in catalyst. Conversely, thefewest conversion happens in the center of the catalyst that flow is in top speed. Flow inside the channel is laminar and parabolic to have high reduction efficiency through the channel. Concentration units are in mol cm⁻³.

<table>
<thead>
<tr>
<th>Table 5</th>
<th>Boundary conditions in catalyst chemical reaction simulation.</th>
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</thead>
<tbody>
<tr>
<td>Equations</td>
<td>Inlet</td>
</tr>
<tr>
<td>M Mass ass</td>
<td>$c_m$</td>
</tr>
<tr>
<td>Heat Transfer</td>
<td>$T_{in}$</td>
</tr>
<tr>
<td>Momentum</td>
<td>$U_{in}$</td>
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<table>
<thead>
<tr>
<th>Table 6</th>
<th>Produced radical concentrations in different electric field intensity.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electric Field (kV/cm)</td>
<td>15.5</td>
</tr>
<tr>
<td>H (mol/m³)</td>
<td>$1.35 \times 10^{-7}$</td>
</tr>
<tr>
<td>N (mol/m³)</td>
<td>$2.37 \times 10^{-7}$</td>
</tr>
<tr>
<td>OH (mol/m³)</td>
<td>$0.47 \times 10^{-5}$</td>
</tr>
<tr>
<td>O (mol/m³)</td>
<td>$0.27 \times 10^{-4}$</td>
</tr>
</tbody>
</table>
Fig. 5 illustrates temperature variation along the washcoat at different times at the inlet velocity and temperature 3 m/s and 600 K for two periods: a) 0–10 s and b) 80–90 s. In Fig. 8a, with the passing of time, the washcoat temperature in contact with exhaust gas is increased. That's why each curve is coming upper than the previous period along the channels. Since the beginning of channel is in contact with exhaust gas more than the middle and the end of the channel, its temperature is upper than other areas. In Fig. 8b, washcoat reaches a steady-state condition and its temperature does not vary considerably. It is like a horizontal line till reaches end of catalyst near the ambient temperature. All the areas, such as beginning, middle, and the end of the channel have the same temperature. Thus, at maximum 80 s, CO will be reduced.

Fig. 6 illustrates CO concentration variation along the washcoat at different times at the inlet velocity and temperature 3 m/s and 600 K for a) 0–10 s and b) 60–70 s. In Fig. 6a, with the passing of time, CO is decreased, because according to Fig. 5, washcoat temperature is increased. Thus, reaction (34) happens more efficiently. For this reason, each curve is going down with respect to time. According to the previous part, the beginning of channel has the higher temperature in comparison to other areas; Thus CO reduction is higher than middle and the end of the channels. In part b, CO reaches steady-state conditions and its concentration reaches a constant state, since washcoat temperature no longer changes with time. Therefore, for CO, 60 s is needed from engine start to reach the highest reduction rate that is less than 80 s, which in part (a) was mentioned. The most reduction happens at the end of the channel, which contaminants have enough interaction with washcoat to get decreased.

NO concentration distribution in catalyst and channels at the inlet velocity 1 m/s and temperature 600 K in steady-state condi-
tions based on Eqs. (39)–(43) is seen in Fig. 7. NO concentration is reduced through the catalyst channels in contact with washcoat. At the CC inlet, NO concentration is high, but at the outlet of the converter, NO concentration is reduced considerably. Flow in each channel is laminar. That’s why flow streamline is parabolic. Reduction in areas near the axial symmetry boundary is lower than wall of CC. That’s because the velocity near the wall is lower than center of CC. Also, in channels, the flow is slower in washcoat compare to center of channel; hence much more reaction can happen in porous spaces. Finally, reduction is much higher in washcoat rather than other areas, thus it can be concluded that velocity reduction and channel’s length increase can go up the efficiency. Other pollutants steady state conversion for different velocities and temperatures are collected in next parts.

NO reduction efficiency according to the Eqs. (39) and (40) with respect to inlet velocities and temperatures is shown in Fig. 8 clearly. At inlet 550 K and 3 m/s, reduction efficiency is 12%. As can be seen, at low temperatures, reduction efficiency is not high until 500 K and 1 m/s that reduction is near 10%. However, at 600 K efficiencies are improved. As a result, the lowest and highest efficiencies are achieved in 7 m/s (14%) and 1 m/s (68%). For nitro-
gen oxide is clear that with rising temperature and decreasing velocity, reduction is increased. That’s because high temperature ascends reaction rate. Plus, in upper temperature, the energy, which is needed to overcome reaction activation energy is descended. As a result, reaction can happen efficiently. Likewise, in slow velocities, pollutants can stay in catalyst more, hence there will be more reduction reaction in CC. As it is considerable, NO reduction is low for different range of velocities and temperature, but in 600 K and lower than 3 m/s, minimum efficiency achieves 30%. In conclusion, 600 K and 3 m/s are important boundaries in NO decrease. Therefore, in cold start, which CC is not hot enough, catalyst does not affect NO very much. Generally, SCR can’t reduce NO very well.

CO reduction efficiency in different inlet velocities and temperatures is presented in Fig. 9. For all of the contaminants, with increasing inlet temperature and reducing the velocity, the pollutant reduction efficiency is also enhanced. According to the Eq. (35), CO reduction efficiency in 400 K is not high, but in 450 K and the velocity lower than 3 m/s, efficiency exceeds 80%. At 500 K and the highest inlet velocity (7 m/s), reduction efficiency becomes 77%. Carbon monoxide reduction in higher temperatures and less velocities is improved, because raised temperature inclines reaction rate and declines activation energy. Consequently, reaction can happen easier. Furthermore, in low velocities, pollutants resident time in catalyst is increased, so there will be more reduction reaction in CC. Remember for CO, reduction under 450 K for variety of velocities is low. For above 450 K, in maximum 5 m/s, minimum efficiency gets 61%. Consequently, 450 K and 5 m/s are boundary conditions in CO decrease.

Fig. 10 demonstrates CH4 reduction efficiency with respect to inlet velocities and temperatures. Methane reduction efficiency according to the Eq. (37), up to 450 K is not high. At temperature 500 K and in velocities lower than 3 m/s reduction efficiency exceeds 70%. At 550 K and velocity of 5 m/s, efficiency reaches 73%, while in 600 K efficiencies are higher than 92% for all velocities. For methane is clear that higher temperatures and less velocity can make reduction better because of reaction rate and activation energy. Hence, reaction can happen more efficiently.
addition, in slow movement, pollutants resident time in catalyst is increased, so there will be more reduction reaction in CC. Remember for CH₄, reduction under 500 K for variety of velocities is low. For above 500 K, in maximum 5 m/s, minimum efficiency gets 50%. Consequently, 500 K and 5 m/s are critical boundaries in methane conversion.

C₃H₆ reduction efficiency in different inlet velocities and temperatures are analyzed in Fig. 11. For propene in 400 K and 1 m/s, the reduction efficiency according to the Eq. (36) is 82%; however, at higher velocities, the efficiency is not good enough. At the same temperature and 7 m/s, the efficiency plunges to about 10%, while in higher temperatures for example, at 600 K and fixed velocity, it becomes 78%. As a result, this trend is clear that higher temperatures and less velocity can make reduction better. That’s because, ascended temperature raises reaction rate, so reaction can happen efficiently. Besides, lower velocities give opportunity to pollutants to have more contact with catalyst; finally, can reduce more and more. For C₃H₆, reduction under 450 K for variety of velocities is low. For above 450 K, in maximum 5 m/s, minimum efficiency gets 62%. As a result, 450 K and 5 m/s are important boundary temperature and velocity in propene reduction.

Fig. 12 illustrates that at inlet velocity 10 m/s and temperature 650 K, with increasing NH₃ to NO ratio, NO reduction efficiency is enhanced. Efficiency in the lowest ratio (0.5) is 11%, while in the highest ratio (3.5) is 23%. That’s because according to the Eq. (39), reaction rate is proportion to fourth power of NO multiplied by NH₃. With increasing reaction rate, more reactants are reduced and there will be more products, so for going up NO reduction, more NH₃ injection can help to achieve this goal.

Fig. 13 according to the Eqs. (41)–(43), presents the catalyst back pressure variation in DOC, SCR, and DOC + SCR for various inlet velocities and temperatures. It is observable that with increasing velocity or temperature, also catalyst back pressure rises, because contact between gas and CC is increased. More back pressure can increase the fuel consumption that it is a negative point.

4.3. Hybrid plasma and catalyst simulation impact

In conclusion, it was calculated that engine cold start lasts 80 s in 3 m/s and 600 K. In these conditions and electric field 17 kV/cm, in cold start, NOₓ and C₃H₆ reduction efficiency are gained 9% and 99% that are presented in Table 7. As was mentioned, plasma has no effect on CO because of high level of energy between its bonds. After cold start, when catalyst reaches its steady state temperature,
Fig. 11. C\textsubscript{3}H\textsubscript{8} reduction efficiency in different inlet velocities and temperatures.

Fig. 12. Influence of NH\textsubscript{3} to NO ratio on NO reduction efficiency.

Fig. 13. Catalyst back pressure variation in different inlet velocity and temperature.
NO, CO, and C\textsubscript{3}H\textsubscript{6} reduction efficiencies are 29, 98, and 95% in order. CC’s backpressure reaches 105 Pa in this situation.

5. Model validation

5.1. Plasma part validation

Fig. 14 presents the effect of electric field intensity on NO\textsubscript{x} reduction efficiency obtained from experimental and simulated results in 400 K according to Eqs. (18)–(30). In the experiment, the plasma reactor is a dielectric-barrier discharge and comprises concentric cylinders. A high voltage AC power was applied between the electrodes. Center electrode diameter varied from 0.1 mm to 4 mm. The inner diameter of the outer electrode tube ranged from 22 mm to 25 mm. The length of the plasma reactor tube was 30.5 cm (McLarnon and Penetrante, 1998). All results are in qualitative agreement with each other. Below 16.5 kV/cm, the NO and NO\textsubscript{2} concentration does not change considerably, because based on table 6, total radical produced are not high enough in this range, but exceeding that electric field, more ions are produced. As it was stated in previous parts, NO is reduced and NO\textsubscript{2} is increased much more. Consequently, plasma is undesirable in NO\textsubscript{2} production, while it is in favor on NO reduction. Although plasma is not effective for NO\textsubscript{2} reduction, it can totally reduce NO plus NO\textsubscript{2} concentrations. Up to 16.5 kV/cm, the efficiency does not change much. In 17 kV/cm, the efficiency is 9%. Exceeding 17 kV/cm, sparkover initiates. As a result, plasma does not affect NO\textsubscript{X} perfectly, but it can be helpful in cold start engine, which catalyst temperature is not high enough. Results between modeling and experiments are closer to each other till 16.5 kV/cm. From 16.5 to 17 kV/cm, range of efficiency difference between modeling and experiments are closer to each other till 16.5 kV/cm.

Fig. 15 presents the effect of electric field intensity on NO\textsubscript{x} reduction efficiency. In experiment, a tubular furnace was used. The flow rate of the gas injected was adjusted to 2 L.min\textsuperscript{-1} with the temperature of the plasma/HC-SCR reactor at 230 °C. Hydrocarbon reduction efficiency. In experiment, a tubular furnace was used. The flow rate of the gas injected was adjusted to 2 L.min\textsuperscript{-1} with the temperature of the plasma/HC-SCR reactor at 230 °C. Hydrocarbon reduction efficiency. In experiment, a tubular furnace was used. The flow rate of the gas injected was adjusted to 2 L.min\textsuperscript{-1} with the temperature of the plasma/HC-SCR reactor at 230 °C. Hydrocarbon reduction efficiency. In experiment, a tubular furnace was used. The flow rate of the gas injected was adjusted to 2 L.min\textsuperscript{-1} with the temperature of the plasma/HC-SCR reactor at 230 °C. Hydrocarbon reduction efficiency.

Effect of C\textsubscript{3}H\textsubscript{6} on NO\textsubscript{x} reduction efficiency presented in Fig. 15. Experimental set-up consists of a wire anode inside a tube electrode. Test was done with 500 p.p.m. NO, at 300 °C and 1000 p.p.m C\textsubscript{3}H\textsubscript{6}. According to Eqs. (26)–(28), propene leads to form ozone, so it causes NO\textsubscript{x} reduction based on Eq. (29). At 17 kV/cm and 400 K, NO\textsubscript{x} reduction efficiency is 1.5%, but with C\textsubscript{3}H\textsubscript{6} addition, efficiency reaches 9% that is 6 times higher. Although hydrocarbons are pollutants themself, it is important to have them in exhaust gases in plasma to have a significant decrease in NO\textsubscript{x}. This effect is mentioned in some references (Rezaei et al.). At engine exhaust temperatures, the efficiency for conversion of NO to NO\textsubscript{2} is very low in the absence of hydrocarbons in the gas stream. Back conversion of NO\textsubscript{2} to NO by the O radical is responsible for the low oxidation efficiency. The O radicals that normally would react with NO or NO\textsubscript{2} are now consumed in reactions with C\textsubscript{3}H\textsubscript{6}. The H would be turned to the powerful oxidizing radical HO\textsubscript{2} upon reaction with O\textsubscript{2} and then oxidize NO fast. Likewise, the CH\textsubscript{3} will react with O\textsubscript{2}, followed by a series of reactions that effectively oxidize NO to the OH radical would in turn break up more C\textsubscript{3}H\textsubscript{6} molecules and lead to more hydrocarbon radicals that could oxidize more NO to NO\textsubscript{2}. Hence, in the presence of hydrocarbons, one O radical could form the oxidation of many NO molecules. Additionally, the O radical will be prevented from back converting NO\textsubscript{2} to NO. The hydrocarbon decreases the energy requirement by production of HO\textsubscript{2} radicals that then become the main radical for conversion of NO to NO\textsubscript{2} (Penetrante, 1999). There is no significant deviation between modeling and test results.

Fig. 16 presents the influence of electric field intensity on hydrocarbon reduction efficiency. In experiment, a tubular furnace with the temperature of the plasma/HC-SCR reactor at 230 °C was used. The flow rate of the gas injected was adjusted to 2 L.min\textsuperscript{-1} (space velocity: 8300 h\textsuperscript{-1}) and n-heptane was used as pollutant. Analyses of the reactor components were performed by a gas chro-

<table>
<thead>
<tr>
<th>Contaminants</th>
<th>Cold start efficiency (%)</th>
<th>Steady state efficiency (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO</td>
<td>0</td>
<td>98</td>
</tr>
<tr>
<td>NO</td>
<td>9</td>
<td>29</td>
</tr>
<tr>
<td>C\textsubscript{3}H\textsubscript{6}</td>
<td>99</td>
<td>95</td>
</tr>
</tbody>
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Table 7

Contaminants reduction in transient and steady states conditions.
matograph. According to Eq. (26), O is the most important radical in n-heptane decomposition. Because of lower chemical bond energy between hydrocarbon molecules, electric field intensity’s effect on n-heptane is higher than NOx. With increasing electric field, C3H6 reduction efficiency is also improved. At low electric field such as, 15.5 kV/cm, the deviations between experimental and simulation is about 15%, while in higher electric field, the results are converged to each other. The reason behind some deviations between simulation and experimental results are the difference between two reactors’ geometry. Plus, the hydrocarbon analyzed in experiment is heptane not propene (Jo and Mok, 2017). From this, it is clear that plasma is completely effective on hydrocarbons reduction.

5.2. Validation of CO, NO, and C3H6 reduction in catalyst

NO, C3H6, and CO reduction efficiency according to the Eqs. (35)–(40) for simulations and reference results are illustrated in Figs. 17–19 at initial conditions 600 K and 7 m/s. In Fig. 17, NO reduction from 400 to 575 K and 7 m/s for experiments and simulations are agreed well (Braun et al., 2000). Experiment catalyst is Pt-Rh/CeO2-Al2O3 with 22 mm in diameter, 29 mm in length, 1 mm in channel diameter, porosity (micropores) 27.9%, and washcoat thickness 100 μm. It is obvious that in low temperatures, efficiency is not high. In more than 500 K, with increasing temperature, efficiency is rised up. It is clear that experiment and modeling results are so closed to each other.

In Fig. 18, C3H6 reduction efficiency simulation results are close to experimental results, especially in higher temperature from 550 to 600 K. That’s because heat transfer between catalytic converter and surrounding is not considered in modeling and the walls are assumed insulation. Thus, temperature inside the CC is higher than reality, while in real there are some heat losses from walls (Braun et al., 2000). In conclusion, SCR is not very useful in NOX decrease and the efficiency is not as high as CO and hydrocarbons.

Fig. 19 illustrates the temperature effects on CO reduction according to the experimental and computed results. Catalyst Pt/CeO2-Al2O3 was used in simulation; however, experiment catalyst was Pt/Al2O3 with total flow rate = 100 mLmin⁻¹. In low temperatures between 400 and 425 K, results are close to each other. Deviations between experiments and simulations from 450 to 500 K are high. With increase in temperature, efficiency should rise up, but in experimental results until 500 K, it is not as high as simulation. The reason behind low temperature differences is the same as C3H6 reduction efficiency differences in simulation and experiment in previous part. In higher temperatures from 525 to 600, two types of results are more converged, because temperature inside the CC in experiment is increased significantly (Royer and Duprez, 2011).

5.3. Catalyst back pressure validation

In Fig. 20, there is a dilemma between back pressure among simulated and previous experimental results in SCR, DOC, and SCR + DOC in 650 K, 100 mm CC length and 7 g/s flow velocity (Tsinoglou, 2004). The results between modeling and reference agree well.

Overall, the simulation results are almost in coordinate with the experimental data, which indicates the reliability of the model. Some deviations in plasma reactor may be caused by the differences in reactor geometry, which lead to differences of NOx reduction efficiency. In catalyst part, computed and experimental results are agreed well.
6. Conclusion

In the present research, NOx, CO, and HC reduction from diesel engine exhaust with plasma assisted catalyst were modeled using COMSOL software. In a plasma reactor, in cold start conditions, in highest electric field before sparkover, NOx reduction efficiency was low, but C3H6 efficiency was almost high. Hence, in sedan size, plasma is not a good solution for NOx reduction. In larger cars with more spaces for enlarging plasma reactor and with using stronger battery with more capacity, this problem would be solved. Moreover, when there was C3H6 in the exhaust, NOx reduction is improved. As a result, plasma is very effective in hydrocarbons reduction. In catalyst section, reactions in SCR and DOC catalysts were simulated in COMSOL Reaction Engineering Lab 3.5. Cold start time was calculated 80 s. That’s important, because it defines the plasma working time. Transient temperature and CO concentration distribution at catalyst channel were modeled. An increase in velocity and temperature resulted in a higher backpressure NH3 to NO ratio influence on NO reduction was modeled and found that an incline in this ratio could increase NO conversion efficiency. Finally, one specific condition introduced, and plasma effects on pollutants reduction calculated. These results provide a promising base for future development of vehicle exhaust after treatment during cold start, which remains a serious problem in terms of pollutants emissions.

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