Air Pollution, SI Engine Emissions and Control

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Air Pollution,
SI Engine Emissions
and Control

Atmospheric Pollution

- **SMOG**

  O_3 – Ozone

  NO_2 Nitrogen dioxide

$$R\text{-}\overset{\overset{\displaystyle O}{\|}}{C}\text{-}OONO_2$$

PAN(Peroxyacyl Nitrate)

- **TOXICS**
  - CO, Benzene, 1-3 butadiene, POM (Polycyclic organic Matters), Aldehydes

Primary Pollutants: Direct emissions from vehicles

- ➢ CO, HC, NOx, PM(Particulate matters), SOx, aldehydes

Secondary Pollutants: From interaction of emissions with the atmosphere

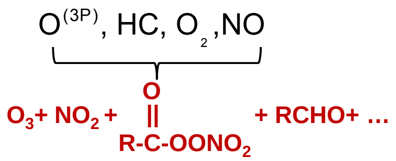
- ➢ O_3, PAN, NO_2, Aldehydes

Atmospheric Pollution

Smog formation:

$$NO + \frac{1}{2}O_2 \rightarrow NO_2$$

$$NO_2 + h\nu(\leq 415\ \text{nm}) \rightarrow NO + O^{(3P)}$$



$$\underbrace{O^{(3P)},\ HC,\ O_2,\ NO}\ \rightarrow\ O_3 + NO_2 + \text{R-}\overset{\displaystyle O \atop \displaystyle ||}{\text{C}}\text{-OONO}_2 + RCHO + \ldots$$

Acid rain:

$$SO_2 + \dot{O}H \rightarrow H\dot{O}SO_2$$

$$H\dot{O}SO_2 + O_2 \rightarrow H\dot{O}_2 + SO_3$$

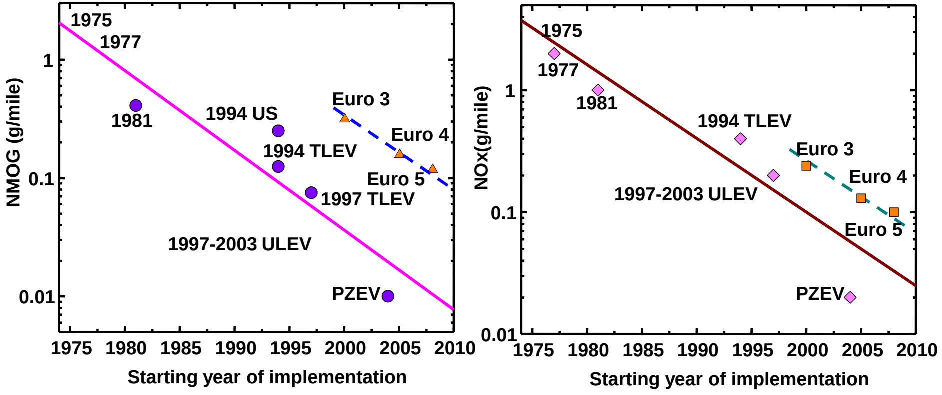
$$SO_3 + H_2O \rightarrow H_2SO_4$$

$$NO + \frac{1}{2}O_2 \rightarrow NO_2$$

$$NO_2 + \dot{O}H \rightarrow HNO_3$$

Emission requirements

(Gasoline engines)



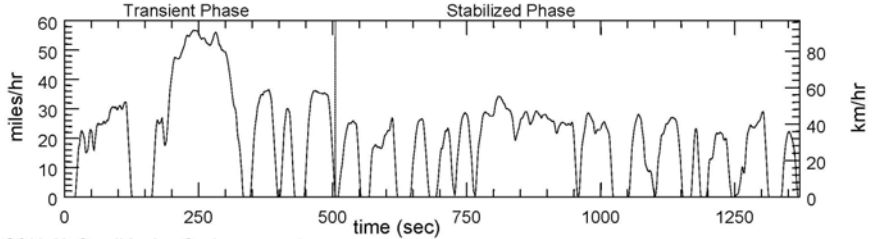
Historic trend: Factor of 10 reduction every 15 years

At 28.5 miles per gallon, 100 g of fuel is burned per mile.
Emission of 0.01 g/mile means 10^{-4} g/g-of-fuel

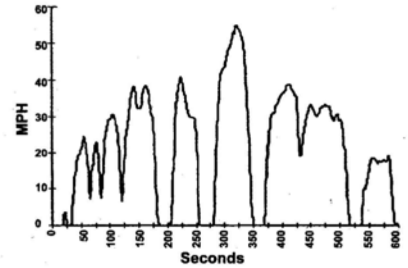
PZEV regulation
(120,000 miles guarantee):

NMOG	0.01 g/mile
CO	1.0 g/mile
NOx	0.02 g/mile

FTP 23 cycles – Each cycle consists of idle, accel., cruise, and decel. Three test phases: Transient phase (0-505s); stabilized phase (505 to 1376s); warm start (repeat of first 505 s test after 10 min. shut down)

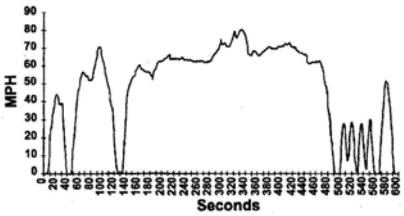


SC03 Air Conditioning Cycle



Total duration : 594 s Max. speed : 54.8 mph

US06 High speed/High load cycle



Total duration : 600 s Max. speed : 80.3 mph
Max. acceleration: 8 mph/second

Air Conditioning Cycle:
10 min. soak, 95°F ambient temperature, 40% relative humidity, 850W/m² solar load, AC max cooling

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EMISSIONS MECHANISMS

- CO emission
  - – Incomplete oxidation of fuel under fuel rich conditions
- NOx emisison
  - – Reaction of nitrogen and oxygen in the high temperature burned gas regions
- Particulate matter (PM) emission (most significant in diesel engines; there are significant PM emissions in SI engines in terms of number density, especially in direct injection engines)
  - – Particulates formed by pyrolysis of fuel molecules in the locally fuel rich region and incomplete oxidation of these particles
  - – Lubrication oil contribution
- Hydrocarbon emissions
  - – Fuel hydrocarbons escape oxidation (or only partially oxidized) via various pathways

Typical steady state SI engine-out emissions

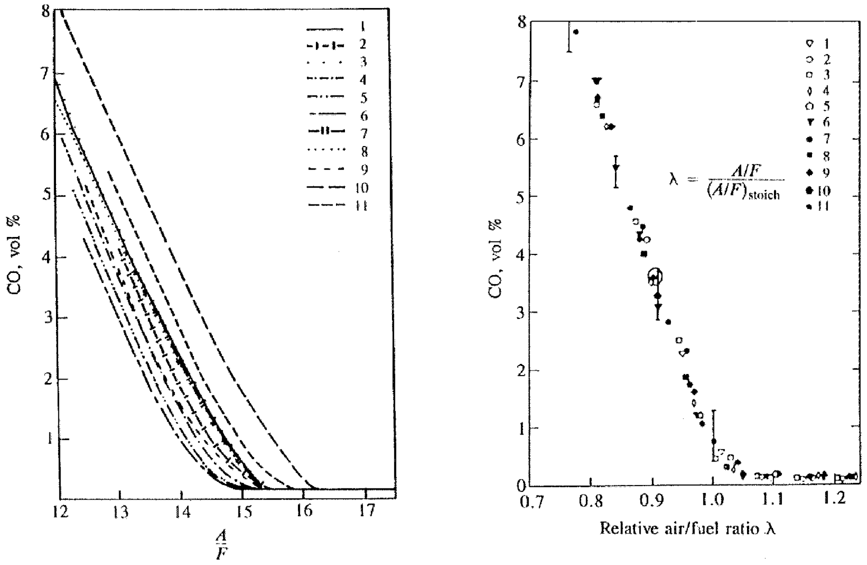
- NOx is a few thousand parts per million
- CO is around 0.5-1% for stoichiometric operation
- HC is 500-2000 ppm for fully warm up engine
- PM very small by mass

CO Emissions Mechanism

- CO is the incomplete oxidation product of the fuel carbon
- Significant amount in fuel rich condition
- Immediately following combustion, CO is in chemical equilibrium with the burned gas
- During expansion, as the burned gas temperature decreases, CO is ‘frozen’
  - Empirical correlation

$$\frac{[CO][H_2O]}{[CO_2][H_2]} \approx 3.7$$

CO is mostly an A/F equivalence ratio issue



NO FORMATION CHEMISTRY

- Zeldovich Mechanism

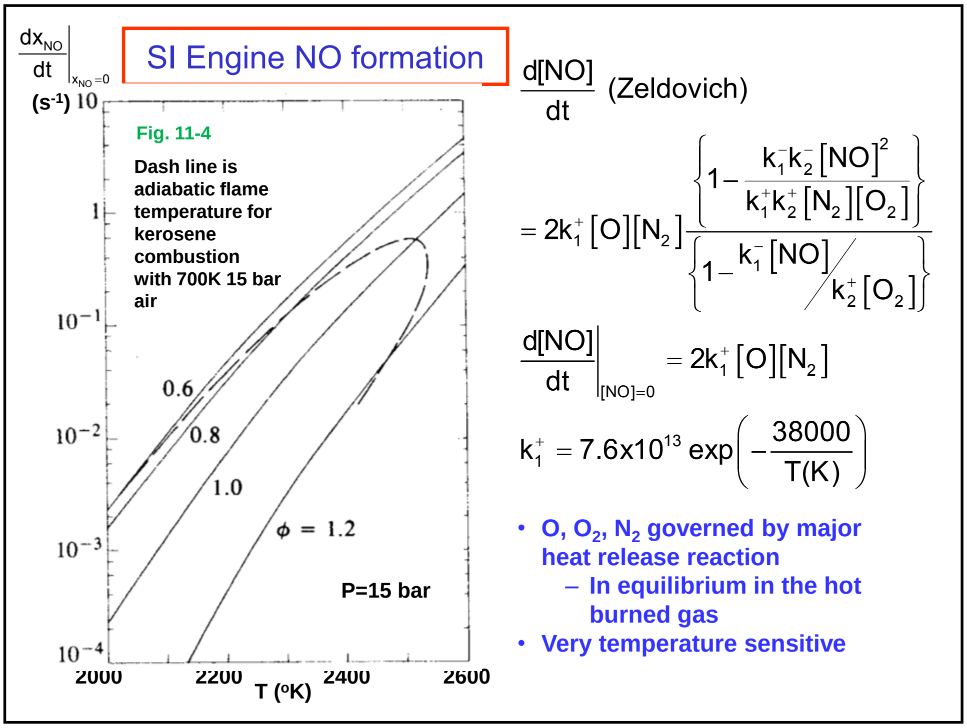
See table 11.1 for rates

$N_2 + O \underset{K^-_1}{\overset{K^+_1}{\leftrightarrow}} NO + N$ (1)

$N + O_2 \underset{K^-_2}{\overset{K^+_2}{\leftrightarrow}} NO + O$ (2)

$N + OH \underset{K^-_3}{\overset{K^+_3}{\leftrightarrow}} NO + H$ (3) **(extended Zeldovich Mechanism)**

  - ➢ NO formation is kinetically controlled
  - ➢ Reactions involving N is fast; N is in steady states ($d[N]/dt \approx 0$)
  - ➢ Very temperature sensitive
- At high temperature (≥ 1000K), equilibrium favors NO versus NO_2 formation
  - ➢ Engine-out $[NO_2]/[NO_x] \leq 2\%$



SI Engine NO formation

Fig. 11-4

Dash line is adiabatic flame temperature for kerosene combustion with 700K 15 bar air

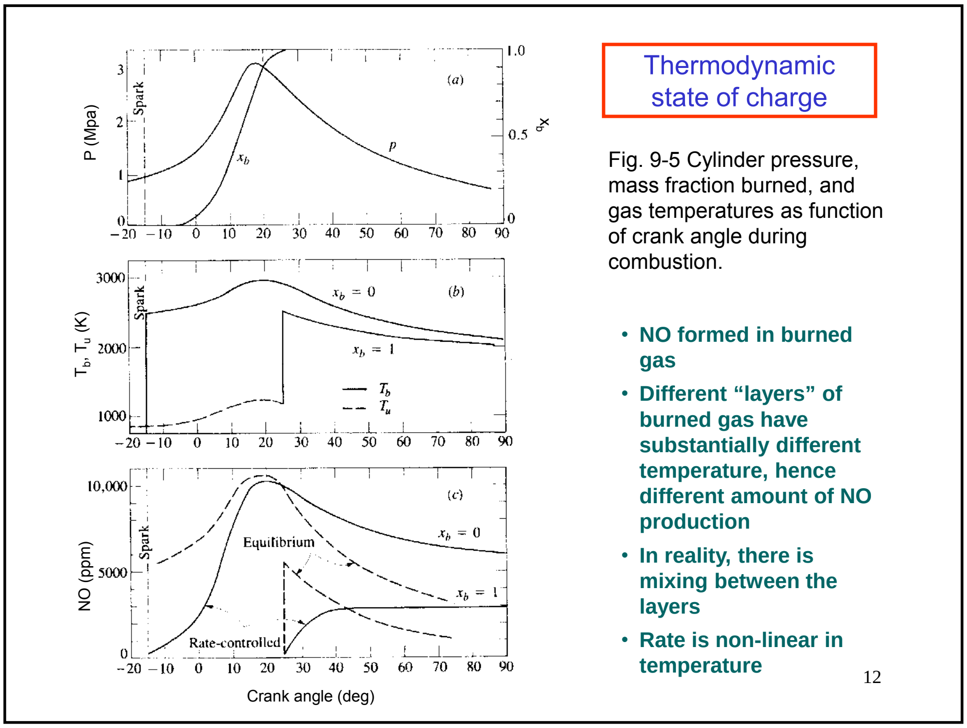
$$\frac{d[NO]}{dt} \quad \text{(Zeldovich)}$$

$$= 2k_1^+[O][N_2]\frac{\left\{1-\frac{k_1^- k_2^- [NO]^2}{k_1^+ k_2^+ [N_2][O_2]}\right\}}{\left\{1-\frac{k_1^-[NO]}{k_2^+[O_2]}\right\}}$$

$$\left.\frac{d[NO]}{dt}\right|_{[NO]=0} = 2k_1^+[O][N_2]$$

$$k_1^+ = 7.6\text{x}10^{13}\exp\left(-\frac{38000}{T(K)}\right)$$

- **O, O_2, N_2 governed by major heat release reaction**
  - **In equilibrium in the hot burned gas**
- **Very temperature sensitive**



Thermodynamic state of charge

Fig. 9-5 Cylinder pressure, mass fraction burned, and gas temperatures as function of crank angle during combustion.

- **NO formed in burned gas**
- **Different "layers" of burned gas have substantially different temperature, hence different amount of NO production**
- **In reality, there is mixing between the layers**
- **Rate is non-linear in temperature**

Engine-out NO emission as function of Φ

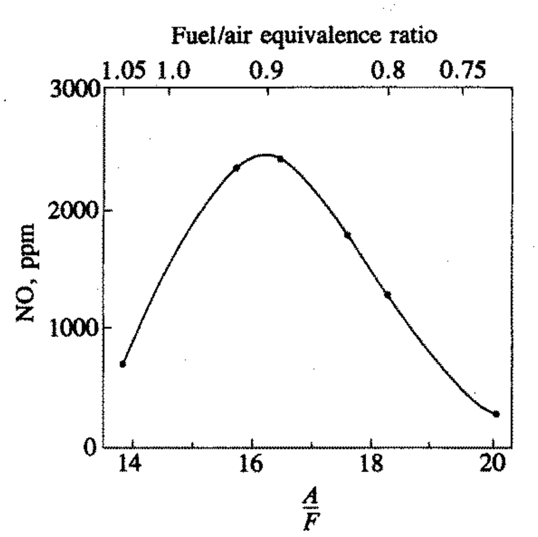


Fig. 11-9

SI engine, 1600 rpm, MBT timing, η_v=50%

In-cylinder NO control

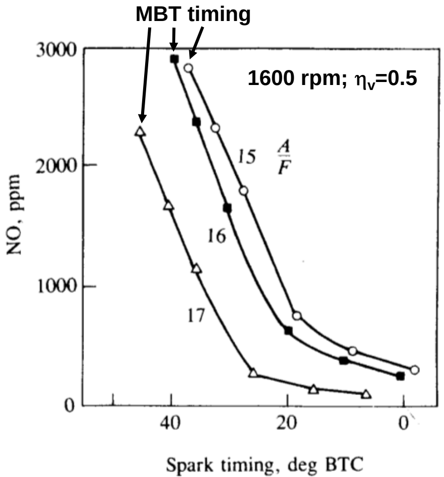
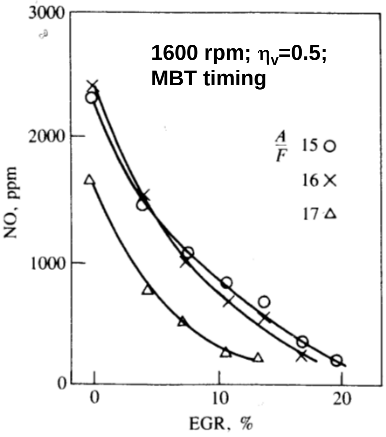


Fig. 11-13

- **Temperature is the key**
  - Spark retard
  - EGR (Exhaust Gas Recirculation)

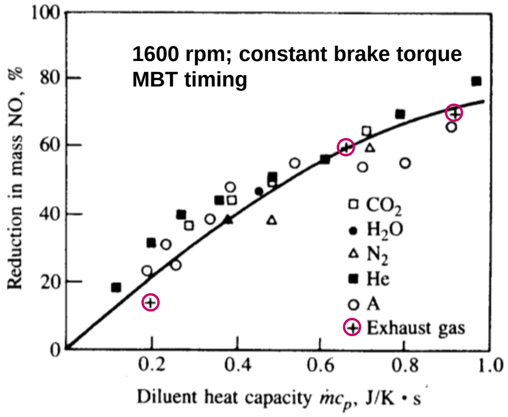
NO control by EGR

- EGR is a dilution effect
  - Reduce burned gas temperature via increase in thermal inertia



1600 rpm; η_v=0.5; MBT timing

Fig. 11-10



1600 rpm; constant brake torque MBT timing

Fig. 11-11

HC emissions

- Importance
  - Photochemical smog (irritant; health effects)
  - Significant loss of fuel energy
- Measurement
  - Flame Ionization Detector (FID)
    - ➢ Chemi-ionization process
    - ➢ Signal proportional to C atom concentration
- Emissions regulation: NMOG as g/mile
  - EPA definition of HC
    - ➢ Normal gasoline $CH_{1.85}$
    - ➢ Reformulated gasoline $CH_{1.92}$
    - ➢ Compressed natural gas $CH_{3.78}$
  - Need speciation to detect CH_4

HC Impact on smog formation

- Species dependent
  - Assessed as MIR of individual VOC
- VOC = volatile organic compounds

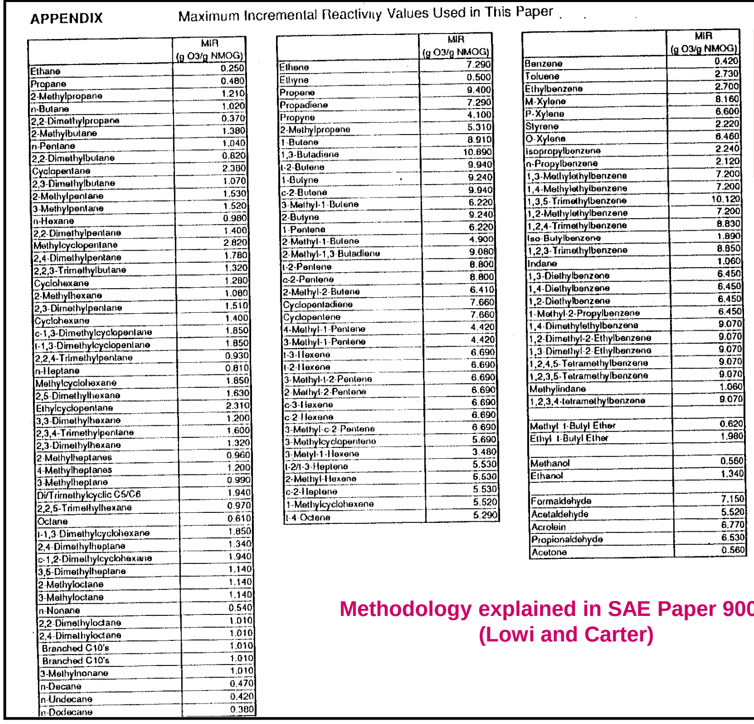
$$\text{Kinetic reactivity} = \frac{\text{VOC reacted}}{\text{VOC input}}$$

$$\text{Mechanistic reactivity} = \frac{\text{Ozone formed}}{\text{VOC input}}$$

Maximum Incremental Reactivity (MIR)

$$\text{MIR} = \frac{m_{\text{ozone,test case; max}} - m_{\text{ozone,base case; max}}}{\text{VOC increment to base case}}$$

EKMA (Empirical Kinetic Modeling Approach) methodology: follow air column (Lagrangian) from 0800 using O3 as indicator. Maximum O3 formation occurs at about 1500-1700 hr.



APPENDIX Maximum Incremental Reactivity Values Used in This Paper

	MIR (g O3/g NMOG)
Ethane	0.250
Propane	0.480
2-Methylpropane	1.210
n-Butane	1.020
2,2-Dimethylpropane	0.370
2-Methylbutane	1.380
n-Pentane	1.040
2,2-Dimethylbutane	0.820
Cyclopentane	2.380
2,3-Dimethylbutane	1.070
2-Methylpentane	1.530
3-Methylpentane	1.520
n-Hexane	0.980
2,2-Dimethylpentane	1.400
Methylcyclopentane	2.820
2,4-Dimethylpentane	1.780
2,2,3-Trimethylbutane	1.320
Cyclohexane	1.280
2-Methylhexane	1.080
2,3-Dimethylpentane	1.510
Cyclohexane	1.400
c-1,3-Dimethylcyclopentane	1.850
t-1,3-Dimethylcyclopentane	1.850
2,2,4-Trimethylpentane	0.930
n-Heptane	0.810
Methylcyclohexane	1.850
2,5-Dimethylhexane	1.630
Ethylcyclopentane	2.310
3,3-Dimethylhexane	1.200
2,3,4-Trimethylpentane	1.600
2,3-Dimethylhexane	1.320
2-Methylheptanes	0.960
4-Methylheptanes	1.200
3-Methylheptane	0.990
Di/Trimethylcyclic C5/C6	1.940
2,2,5-Trimethylhexane	0.970
Octane	0.610
t-1,3-Dimethylcyclohexane	1.850
2,4-Dimethylheptane	1.340
c-1,2-Dimethylcyclohexane	1.940
3,5-Dimethylheptane	1.140
2-Methyloctane	1.140
3-Methyloctane	1.140
n-Nonane	0.540
2,2-Dimethyloctane	1.010
2,4-Dimethyloctane	1.010
Branched C10's	1.010
Branched C10's	1.010
3-Methylnonane	1.010
n-Decane	0.470
n-Undecane	0.420
n-Dodecane	0.380

	MIR (g O3/g NMOG)
Ethene	7.290
Ethyne	0.500
Propene	9.400
Propadiene	7.290
Propyne	4.100
2-Methylpropene	5.310
1-Butene	8.910
1,3-Butadiene	10.890
t-2-Butene	9.940
1-Butyne	9.240
c-2-Butene	9.940
3-Methyl-1-Butene	6.220
2-Butyne	9.240
1-Pentene	6.220
2-Methyl-1-Butene	4.900
2-Methyl-1,3-Butadiene	9.080
t-2-Pentene	8.800
c-2-Pentene	8.800
2-Methyl-2-Butene	6.410
Cyclopentadiene	7.660
Cyclopentene	7.660
4-Methyl-1-Pentene	4.420
3-Methyl-1-Pentene	4.420
t-3-Hexene	6.690
t-2-Hexene	6.690
3-Methyl-t-2-Pentene	6.690
2-Methyl-2-Pentene	6.690
c-3-Hexene	6.690
c-2-Hexene	6.690
3-Methyl-c-2-Pentene	6.690
3-Methylcyclopentene	5.690
3-Metyl-1-Hexene	3.480
t-2/t-3-Heptene	5.530
2-Methyl-Hexene	5.530
c-2-Heptene	5.530
1-Methylcyclohexene	5.520
t-4-Octene	5.290

	MIR (g O3/g NMOG)
Benzene	0.420
Toluene	2.730
Ethylbenzene	2.700
M-Xylene	8.160
P-Xylene	6.600
Styrene	2.220
O-Xylene	6.460
Isopropylbenzene	2.240
n-Propylbenzene	2.120
1,3-Methylethylbenzene	7.200
1,4-Methylethylbenzene	7.200
1,3,5-Trimethylbenzene	10.120
1,2-Methylethylbenzene	7.200
1,2,4-Trimethylbenzene	8.830
Iso-Butylbenzene	1.890
1,2,3-Trimethylbenzene	8.850
Indane	1.060
1,3-Diethylbenzene	6.450
1,4-Diethylbenzene	6.450
1,2-Diethylbenzene	6.450
1-Methyl-2-Propylbenzene	6.450
1,4-Dimethylethylbenzene	9.070
1,2-Dimethyl-2-Ethylbenzene	9.070
1,3-Dimethyl-2-Ethylbenzene	9.070
1,2,4,5-Tetramethylbenzene	9.070
1,2,3,5-Tetramethylbenzene	9.070
Methylindane	1.060
1,2,3,4-tetramethylbenzene	9.070
Methyl t-Butyl Ether	0.620
Ethyl t-Butyl Ether	1.980
Methanol	0.560
Ethanol	1.340
Formaldehyde	7.150
Acetaldehyde	5.520
Acrolein	6.770
Propionaldehyde	6.530
Acetone	0.560

Carter Index for Ozone Forming Potential
(CARB July, 1992)

Table from SAE Paper 932718 (Tauchida et.al)

Methodology explained in SAE Paper 900710 (Lowi and Carter)

HC sources

- Non-combustion sources
  - Fueling loss
  - Diurnal emissions
  - Running loss
  - Hot soak
  - Blow by
    - A few L/min; depends on load and RPM
    - At light load, 1500 rpm, blow by ~ 4L / min

HC sources (cont.)

- Combustion sources
  - 300 to 3000 ppmC1 typical
    - Stoichiometric mixture is ~120,000 ppmC1
  - Main combustion: very little HC except for very lean/ dilute or very late combustion (misfires/ partial burns)
    - Various mechanisms for HC to escape from main combustion
  - Cold start emissions (wall film) especially important

SOURCES OF UNBURNED HC IN SI ENGINE

a) Crevices

b) Absorption and desorption in oil layers

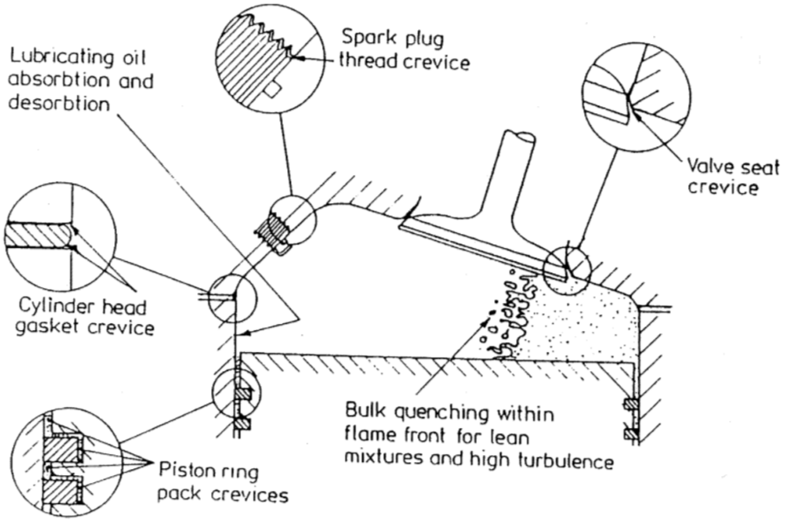
c) Absorption and desorption in deposit

d) Quenching (bulk and wall layer)

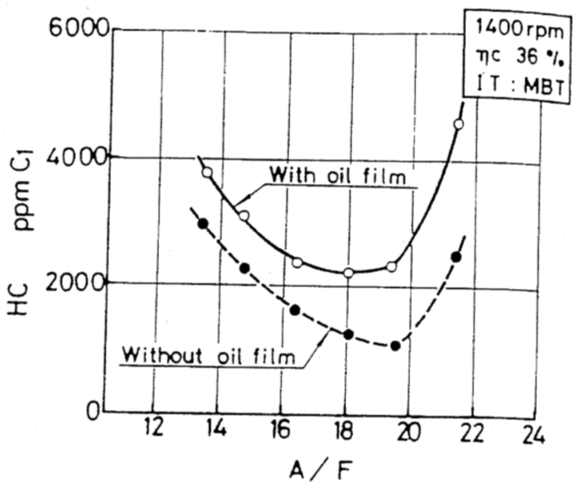
e) Liquid fuel effects

f) Exhaust valve leakage

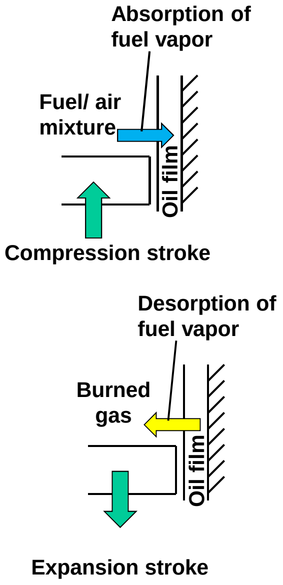
Crevice HC mechanism



Absorption and desorption of fuel vapor



Ishizawa and Takagi (Nissan)
JSME Int. Jnl. 1987 Vol. 30 No. 260 pp. 310-317



Absorption of fuel vapor

Fuel/ air mixture

Oil film

Compression stroke

Desorption of fuel vapor

Burned gas

Oil film

Expansion stroke

HC pathway

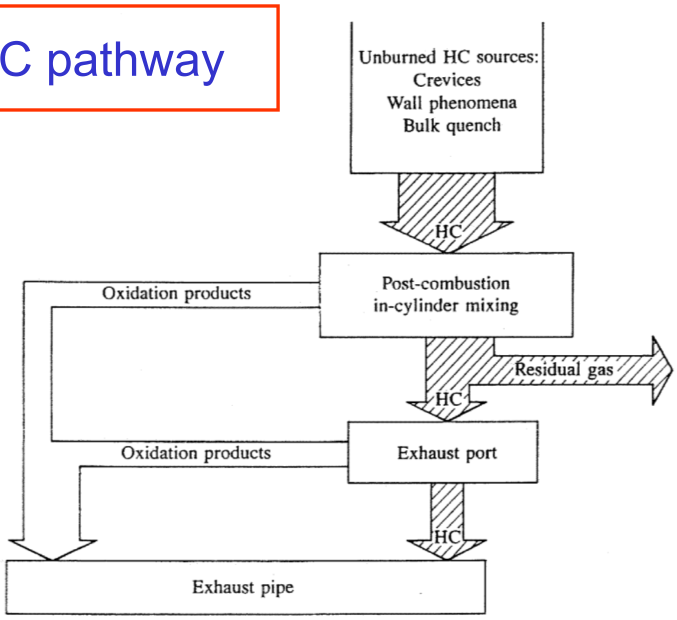
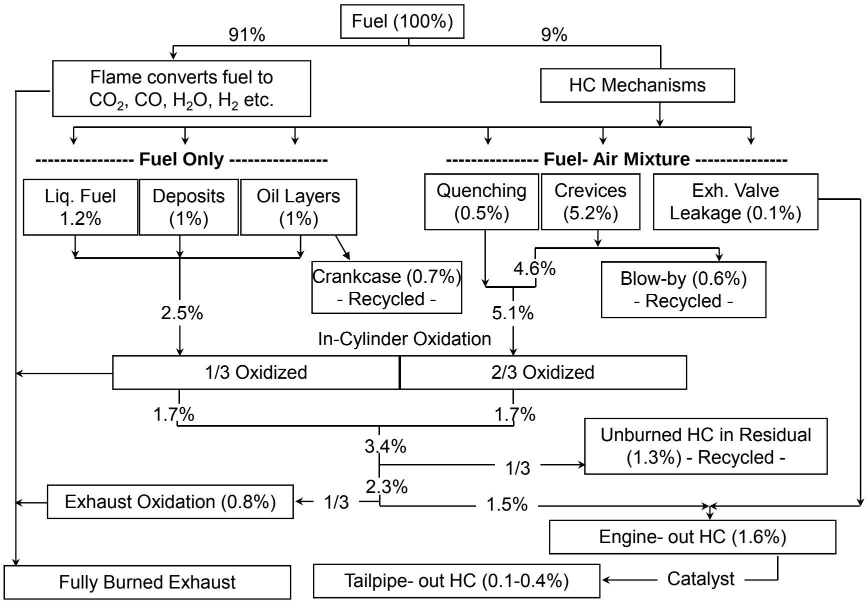


Fig. 11-31

Hydrocarbon Pathway - Steady State, cruise condition



Fuel (100%)

91%

9%

Flame converts fuel to CO_2, CO, H_2O, H_2 etc.

HC Mechanisms

Fuel Only

Fuel- Air Mixture

Liq. Fuel 1.2%

Deposits (1%)

Oil Layers (1%)

Quenching (0.5%)

Crevices (5.2%)

Exh. Valve Leakage (0.1%)

Crankcase (0.7%) - Recycled -

4.6%

Blow-by (0.6%) - Recycled -

2.5%

5.1%

In-Cylinder Oxidation

1/3 Oxidized

2/3 Oxidized

1.7%

1.7%

Unburned HC in Residual (1.3%) - Recycled -

3.4%

1/3

2.3%

Exhaust Oxidation (0.8%)

1/3

1.5%

Engine- out HC (1.6%)

Fully Burned Exhaust

Tailpipe- out HC (0.1-0.4%)

Catalyst

HC Sources: Magnitudes and Percent of Total Engine-out Emissions*

(SAE Paper 932708)

Source	% Fuel Escaping Normal Combustion	Fraction Emitted as EOHC	% Fuel as HC Emissions	% of Total EOHC Emissions
Crevices	5.2	0.15*	0.682*	42.6
Quench	0.5	0.15	0.074	4.6
Oil Layers	1.0	0.09**	0.090**	5.6
Deposits	1.0	0.30	0.300	18.7
Liquid Fuel	1.2	0.30	0.356	22.2
Valve Leakage	0.1	1.00	0.100	6.3
Total	9.0		1.60	100

* Blowby (0.6%) subtracted

** Amount to crank case (0.7%) subtracted

*steady state cruise condition (1500 rpm, 2.8 bar NIMEP)

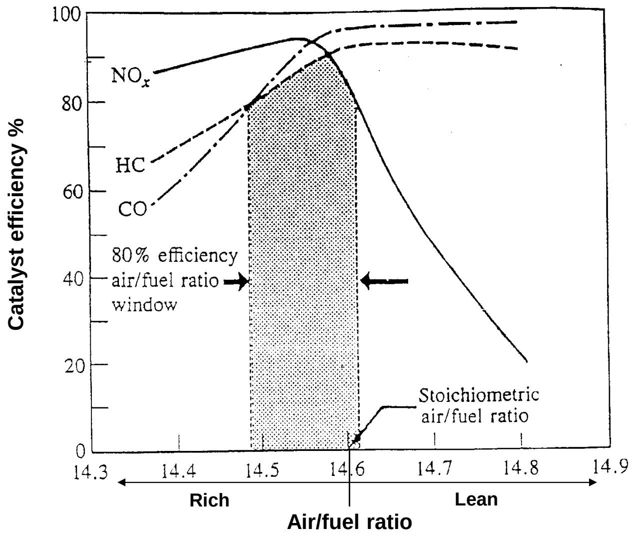
HC control

- Reduce crevice volume
- Keep liner hot
- Spark retard
  - Higher burned gas temperature in the later part of expansion stroke and higher exhaust temperature
- Comprehensive cold start strategy
  - Retard timing, fuel rich followed by exhaust air injection

2.61 Internal Combustion Engines
Spring 2017

SI Engine Catalyst

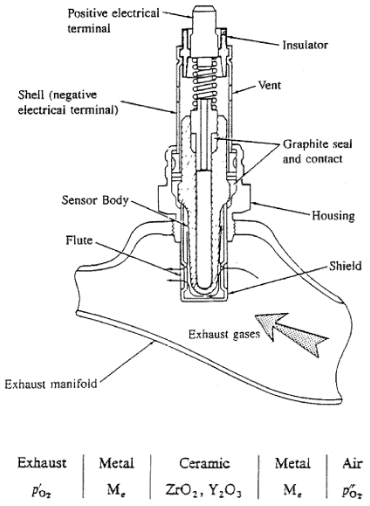
Requirement for the 3-way catalyst

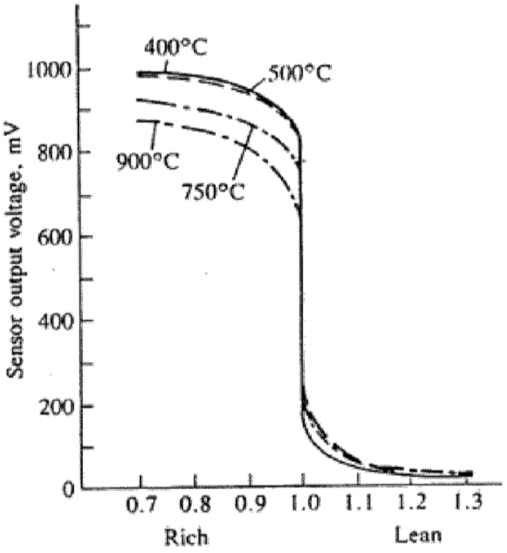


Modern catalyst peak efficiency is better than 97%

Fig 11-57

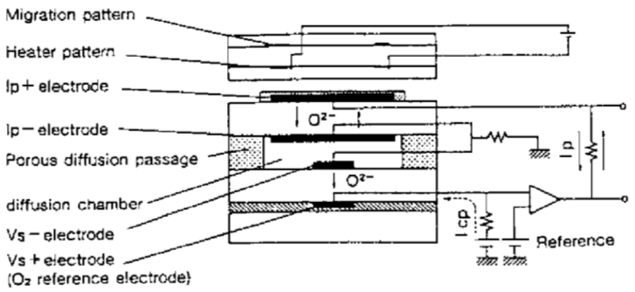
EGO (exhaust gas oxygen) sensor

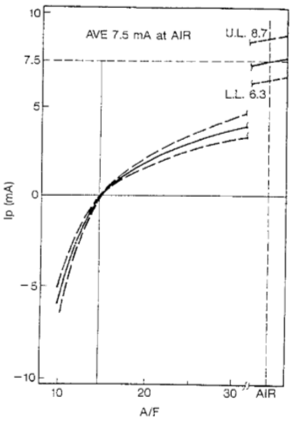




Nerst Eq.: $V_o=(RT/4F)\ell n(P''_{O2}/P'_{O2})$

UEGO sensor

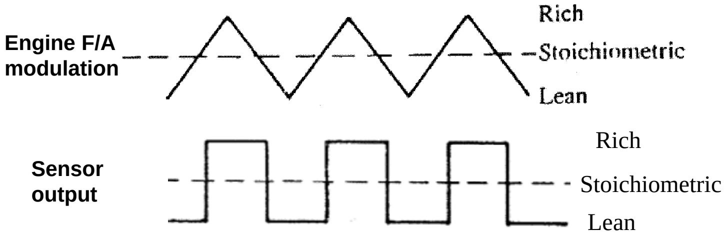




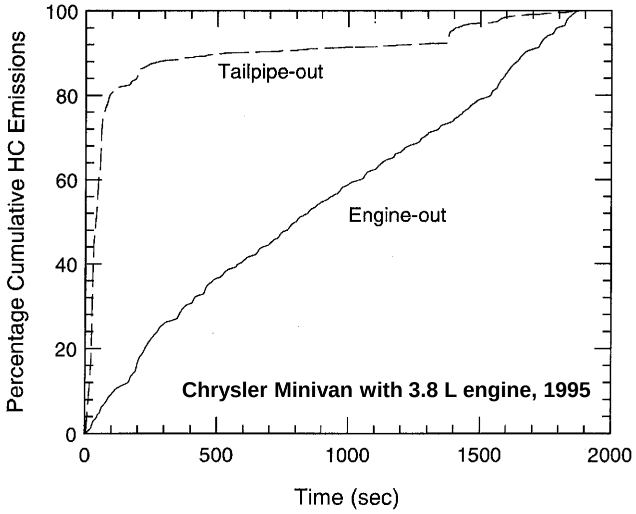
SAE Paper 920234

λ control strategy

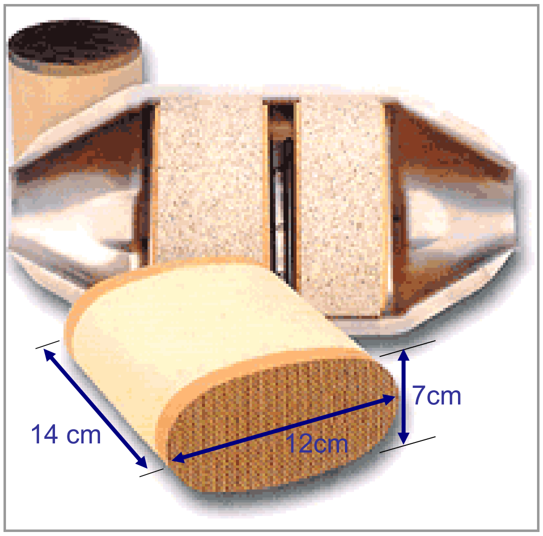
- Modulate A/F ratio around stoichiometric (typically by +/- 2% at around 1 Hz)
  - Enable EGO sensor to read average λ value
  - Make use of O2 storage capability of catalyst so that only average λ = 1 is needed



Engine out and Tailpipe out Cumulative HC emissions in FTP cycle

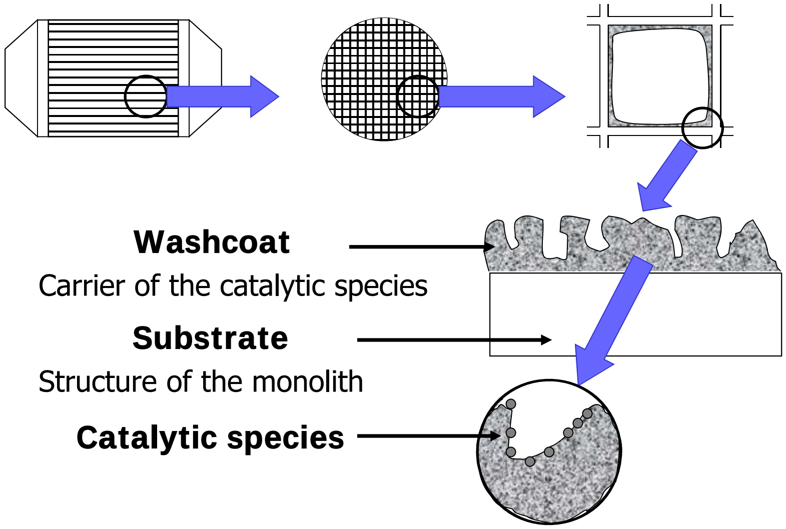


Monolithic reactors



(Typical
dimensions
for a 2.4 L
engine)

Monolithic catalysts’ elements



Washcoat
Carrier of the catalytic species

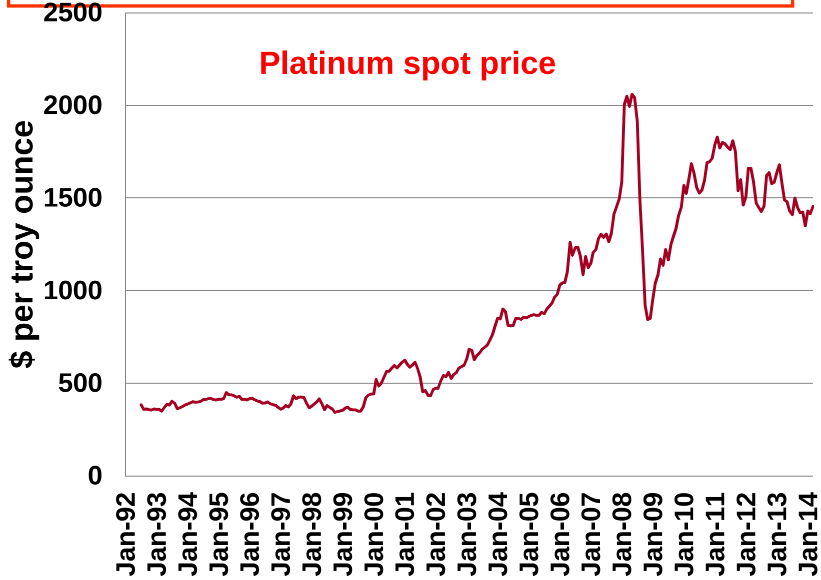
Substrate
Structure of the monolith

Catalytic species

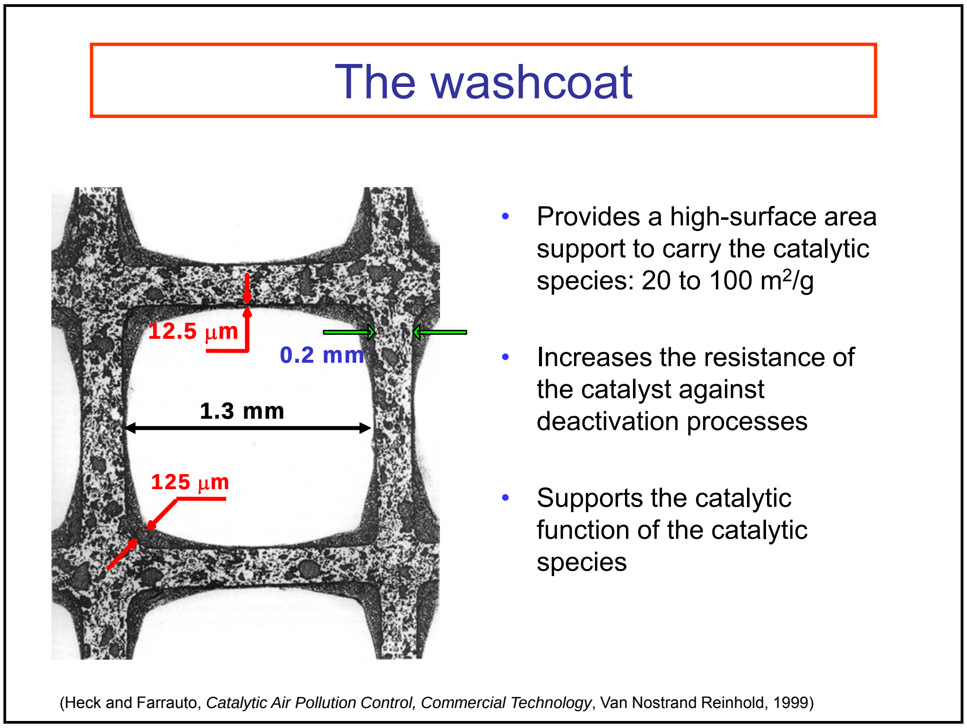
Materials

- Substrate
  - Synthetic cordierite ($2MgO \bullet 2Al_2O_3 \bullet 5SiO_2$)
- Washcoat
  - γ-alumina (γ- Al_2O_3)
- Active materials
  - Platinum (~ 1-2 g/L)
  - Palladium (~0.5-1 g/L; usually in front brick)
  - Rhodium (~0.2 g/L; for NOx and HC reduction)
  - Ceria (for oxygen storage)
    - ➢ $Ce_2O_3 + 1/2\ O_2 \leftrightarrow 2CeO_2$

Cost of catalyst active material



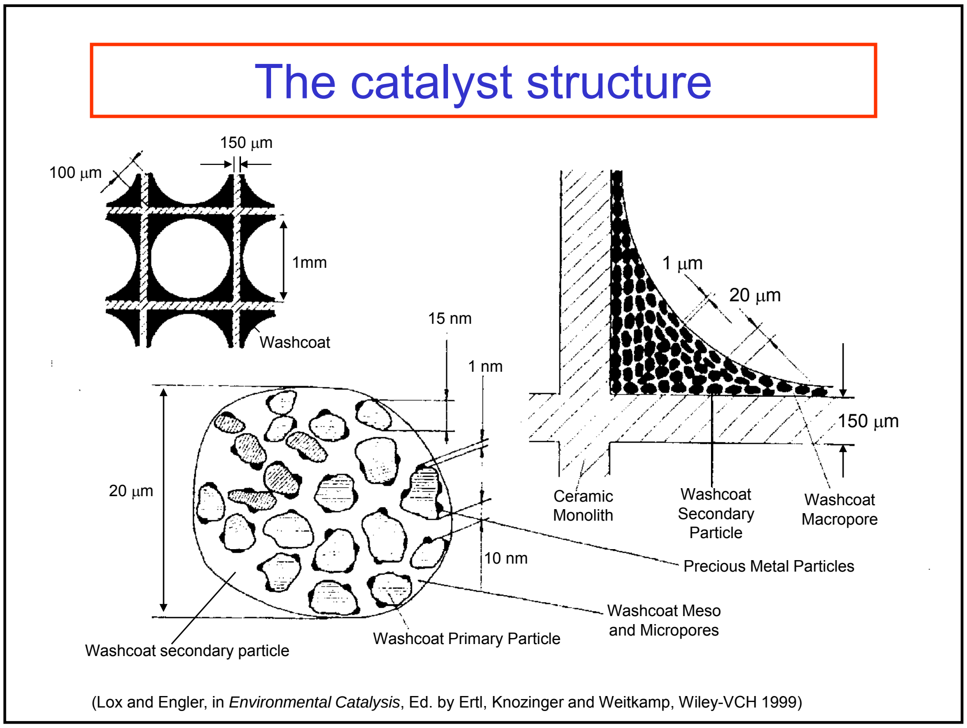
1 troy ounce = 31.1 g



The washcoat

- Provides a high-surface area support to carry the catalytic species: 20 to 100 m^2/g
- Increases the resistance of the catalyst against deactivation processes
- Supports the catalytic function of the catalytic species

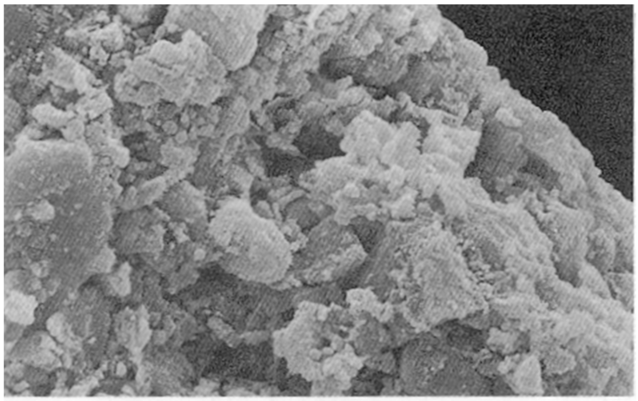
(Heck and Farrauto, *Catalytic Air Pollution Control, Commercial Technology*, Van Nostrand Reinhold, 1999)



The catalyst structure

(Lox and Engler, in *Environmental Catalysis*, Ed. by Ertl, Knozinger and Weitkamp, Wiley-VCH 1999)

The washcoat secondary particles



- **Secondary particle size** ~ 2 to 30 μm
- **Macro-pore dimensions** ~ microns

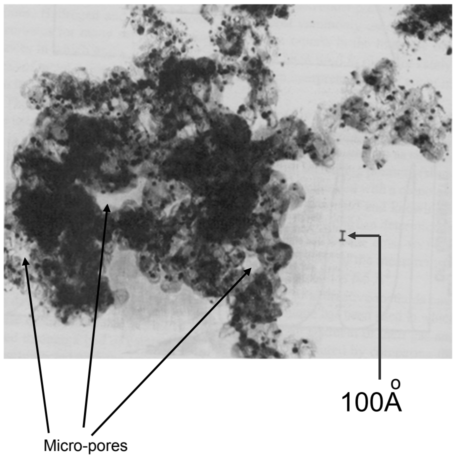
Scanning electron microscope view of washcoat
(Lox and Engler, in *Environmental Catalysis*, Ed. by Ertl, Knozinger and Weitkamp, Wiley-VCH 1999

The catalytic species on the washcoat primary particles

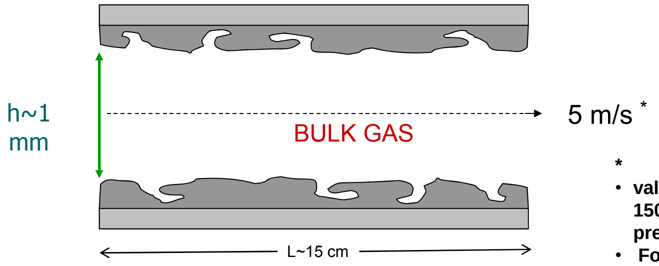
- **Primary washcoat particle size** ~ 10-20 nm
- **Typical size of active material (e.g. Pt) on fresh catalysts**

  less than 50 angstroms,
  (30 angstroms on the figure)
- **Atomic spacing of Pt atom** 2.8 angstroms
- **Average distance between two particles**

  65 angstroms
- **Micro-pore dimensions** ~ 10 to 100 nm



Transport time scale



*
- value for 2.0 L Engine at 1500 rpm, 0.4 bar intake pressure.
- For V_{cat} = 1L space velocity is 1×10^5/hr

T=900°K, p=1bar

Mass diffusivity = 4×10^{-5} m²/s; mean free path = 200 nm; molecular speed c = 450 m/s

External diffusion time $\tau_{ext} = (h/2)^2/D$ = 6 ms

Internal diffusion time:

Macro-pore (size ℓ = 10 μm; continuum limit)

$\tau_{int,\ macro} = \ell^2/D$ = 2.5 μs

Micro-pore (size ℓ' = 100 nm; Knudson limit)

$\tau_{int,\ micro} = \ell'/c$ = 0.2 ns

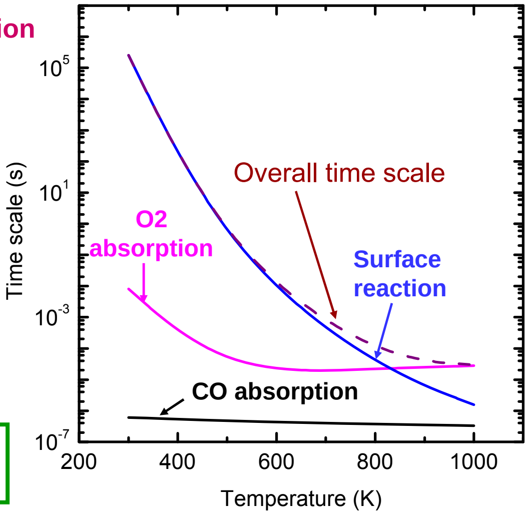
Residence time L/U = 30 ms

Transport time dominated by external transport

Chemical time

Example: Catalytic CO oxidation

- O_2 absorption
  - $O_2 + 2\,S \leftrightarrow 2\,O^*$
- CO absorption
  - $CO + S \leftrightarrow CO^*$
- Surface oxidation and CO_2 release
  - $CO^* + O^* \leftrightarrow CO_2$

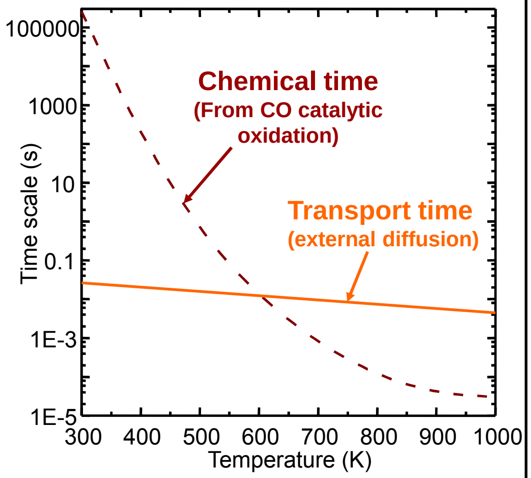


Overall time scale dominated by surface reaction

Limiting time scale

Conclusions:

- For fully warm-up catalyst, overall reaction is rate limited by external diffusion
- At low temperatures, surface chemistry is rate limiting



Catalyst deterioration

- Poisoning
  - Lead
  - Phosphorus (from oil additives)
  - Sulfur (fuel S from 300 to 30 ppm)
    - ➢effect reversible to a large extend
- Thermal degradation
  - Sintering (T>1000°K)
    - ➢Active ingredients: loss of reactive surface
    - ➢γ-alumina: occluding the active ingredients
    - ➢Oxidation of Rh
- Glazing — lubrication oil covering catalyst
- Erosion

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