Catalytic oxidation of carbon aerosols: Influence of the Pt-C interparticle contact on the kinetic parameters (E_a and k_0)

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Combustion Generated Nanoparticles

Regeneration of diesel soot particle filters:

Reducing the temperature of thermal soot oxidation from the range of 550-650°C to lower temperatures → *Application of catalyst*

Under some circumstances no catalytic activity of platinum was observed → ?? → *What do we know about catalyst-Carbon particle contact?*

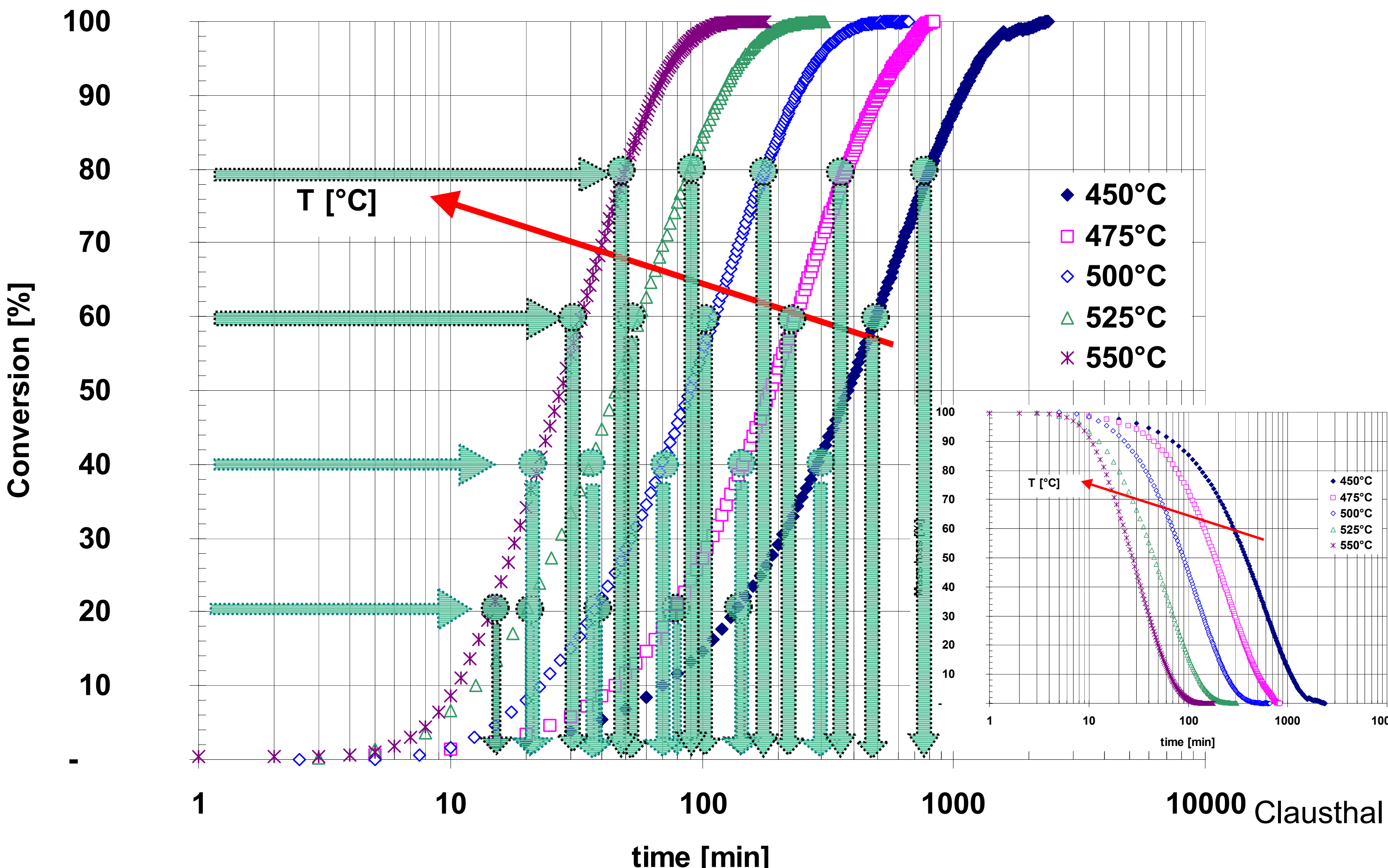
↓

Investigation on influence of *contact intensity* on increase of the oxidation rate

Oxidation set-up for bulk powder

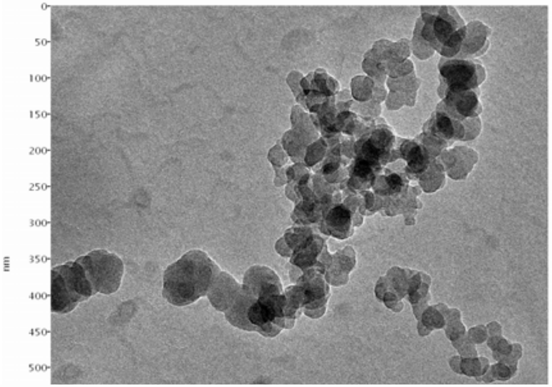
Oxidation in the form of bulk powder
TGA (thermogravimetric analysis)

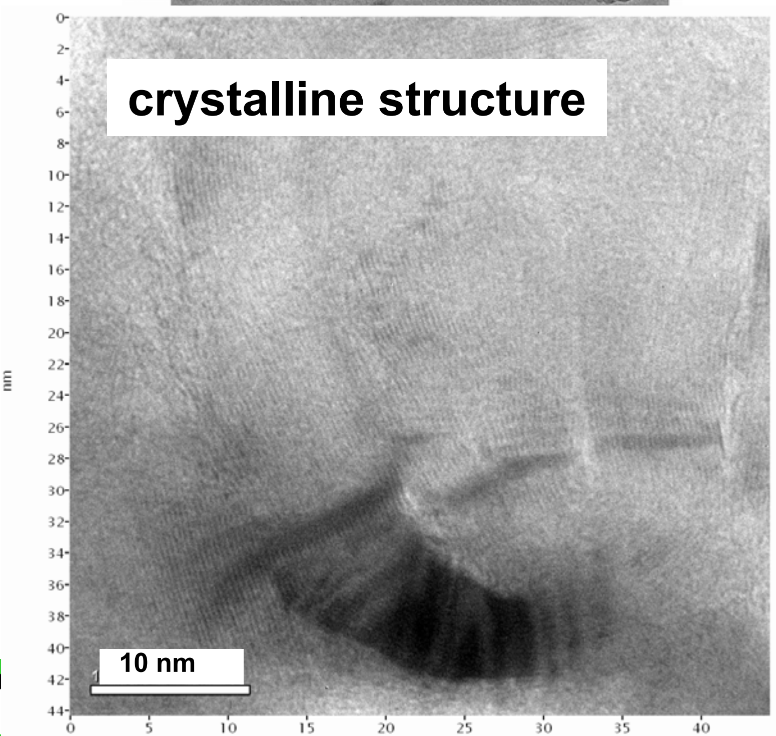
Generating carbon particles → sampling → Oxidation of powder with TGA



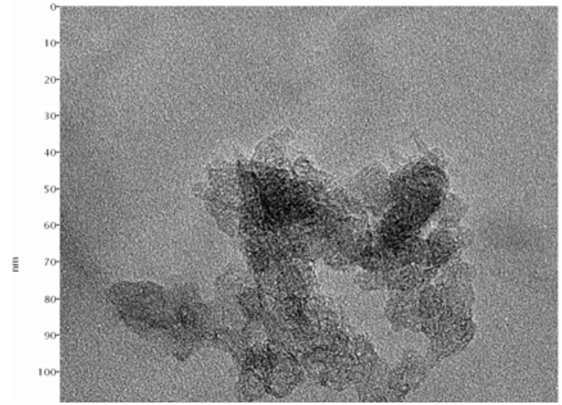
Carbon

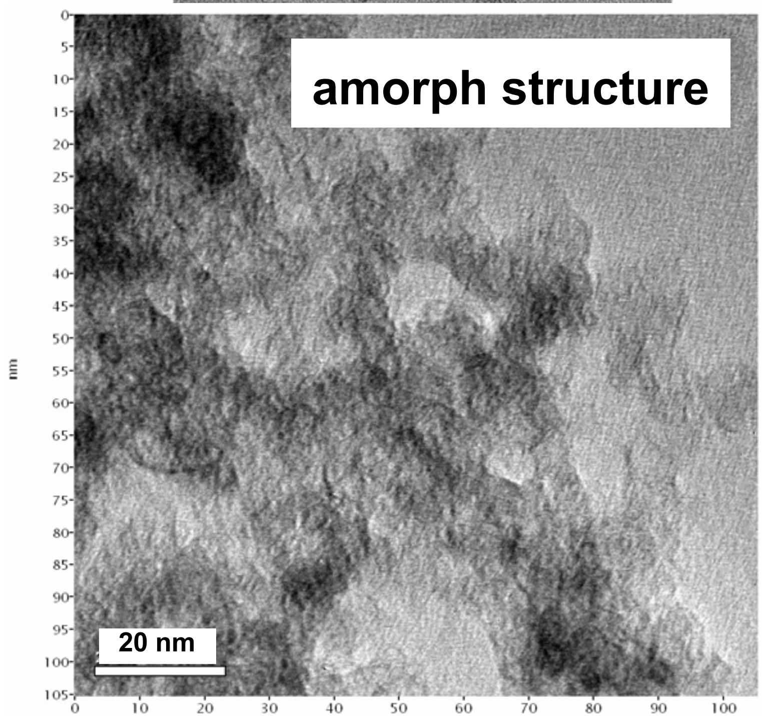
Printex U: industry soot generated by flame process





SD-Carbon: generated by spark erosion





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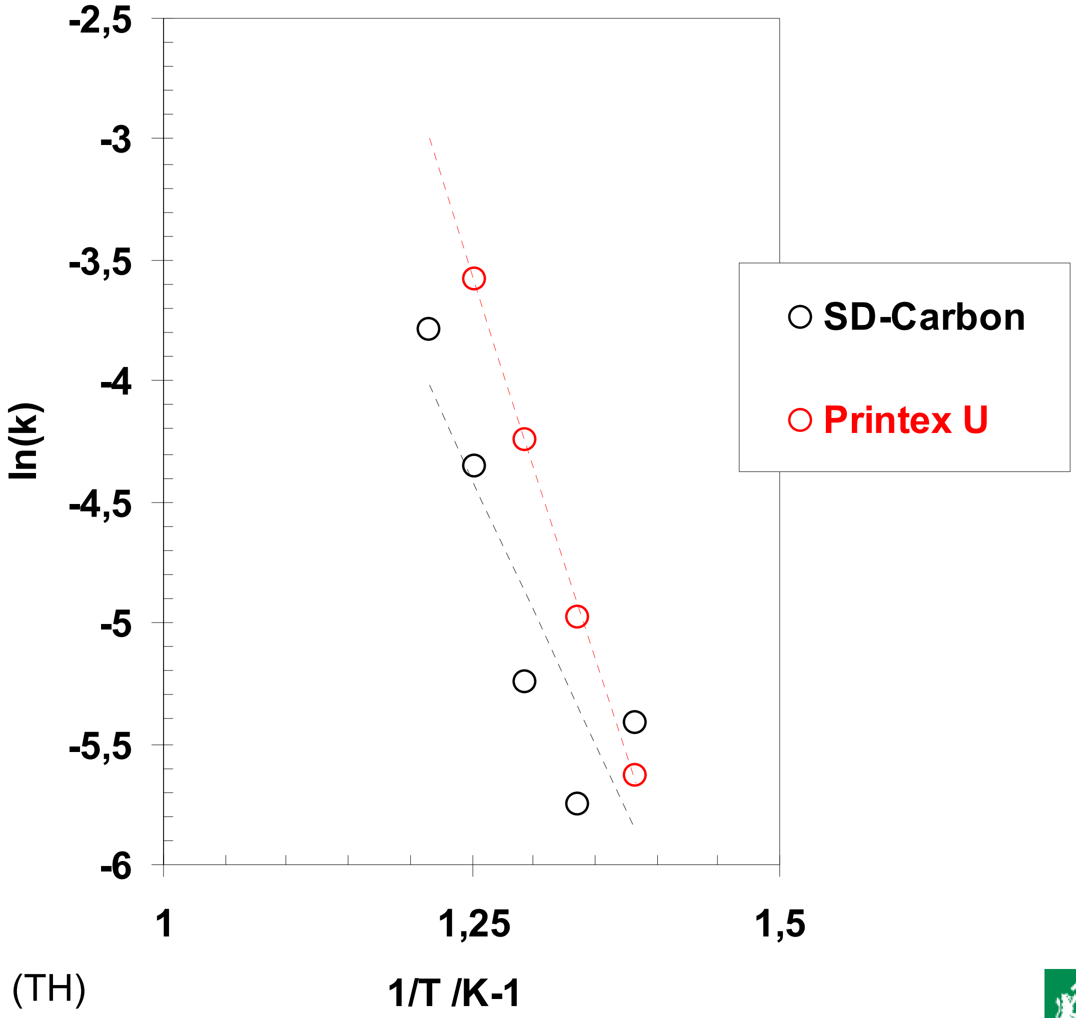
Arrhenius diagram

Thermal oxidation measured by TGA

E_a= 135± 5 kJ/mol

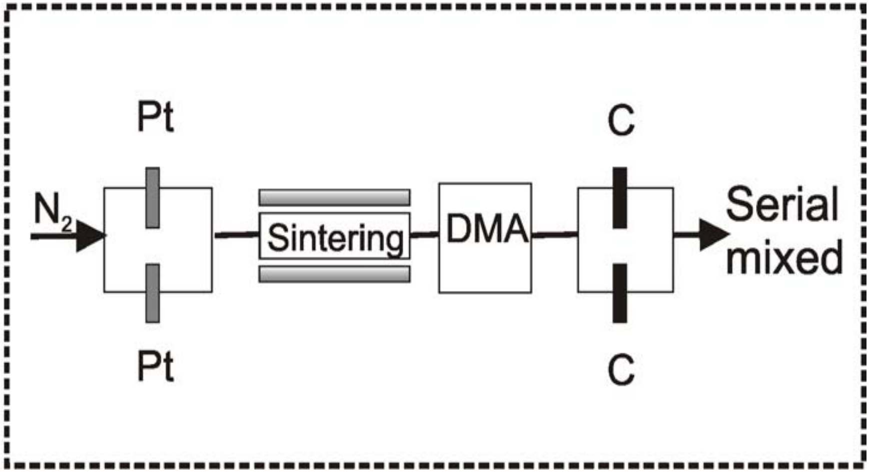
k_0 = 2,05 x 10^7

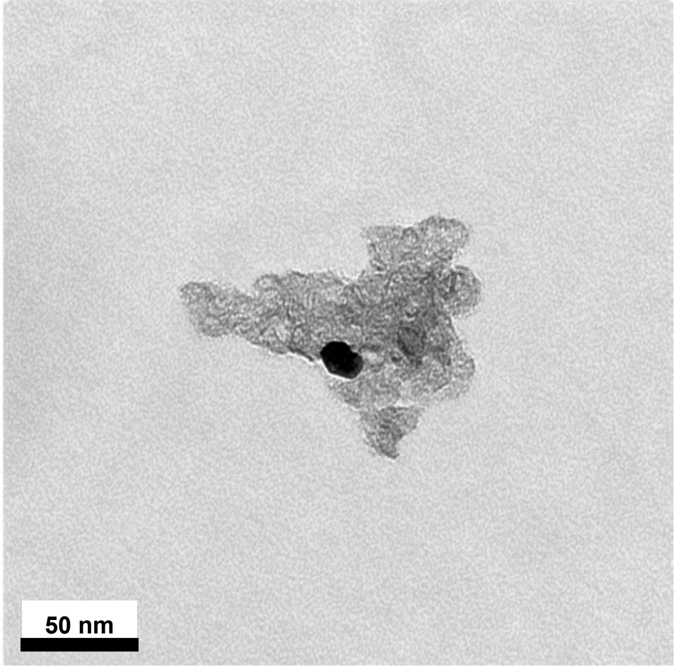


Types of Pt/C contact

Co-condensation contact

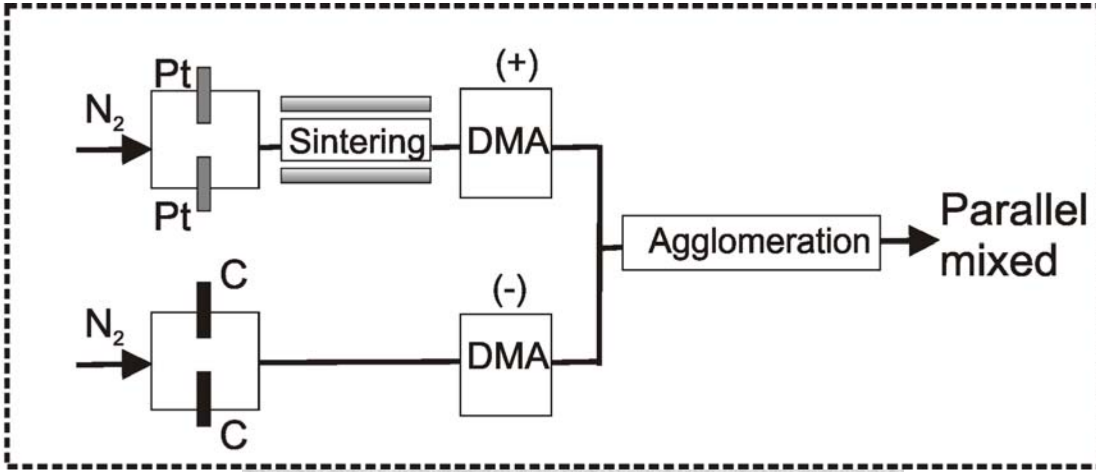
Pt nano sphere in a random diffusional contact with a carbon nano agglomerate (*Co-condensation contact*)

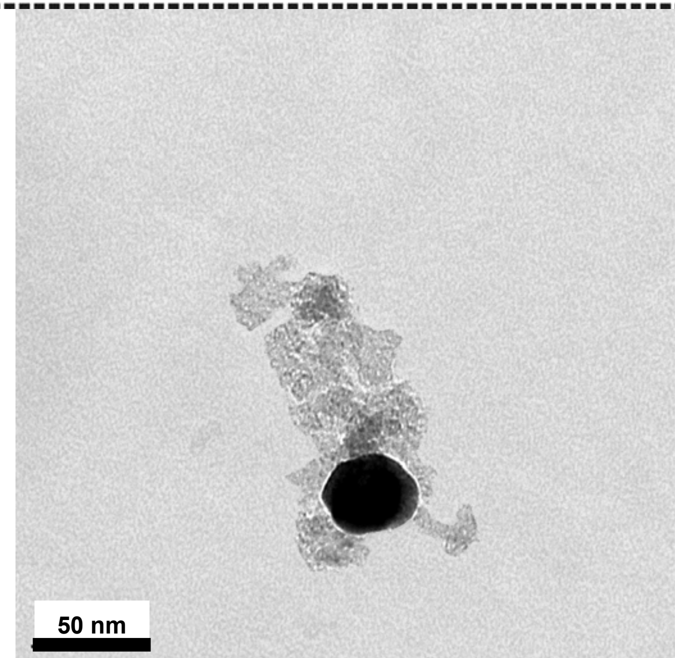




Co-agglomeration contact

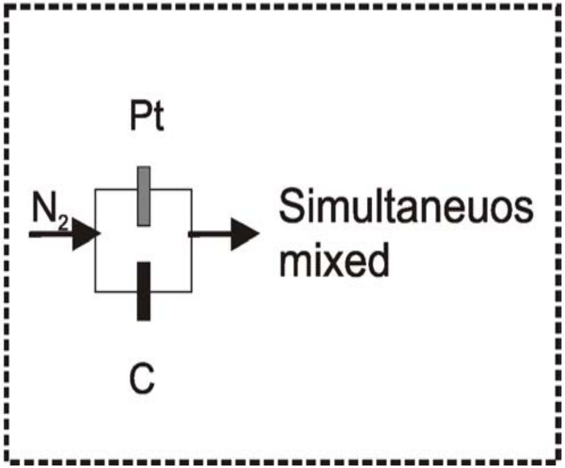
Pt nano sphere in a random diffusional contact with a carbon nano agglomerate (*Co-agglomeration contact*)

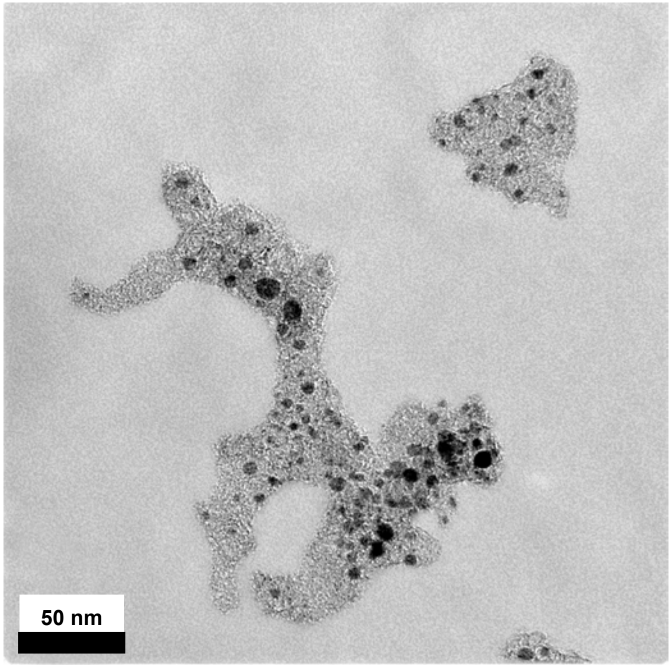




Physical Vapor Deposition (PVD) contact

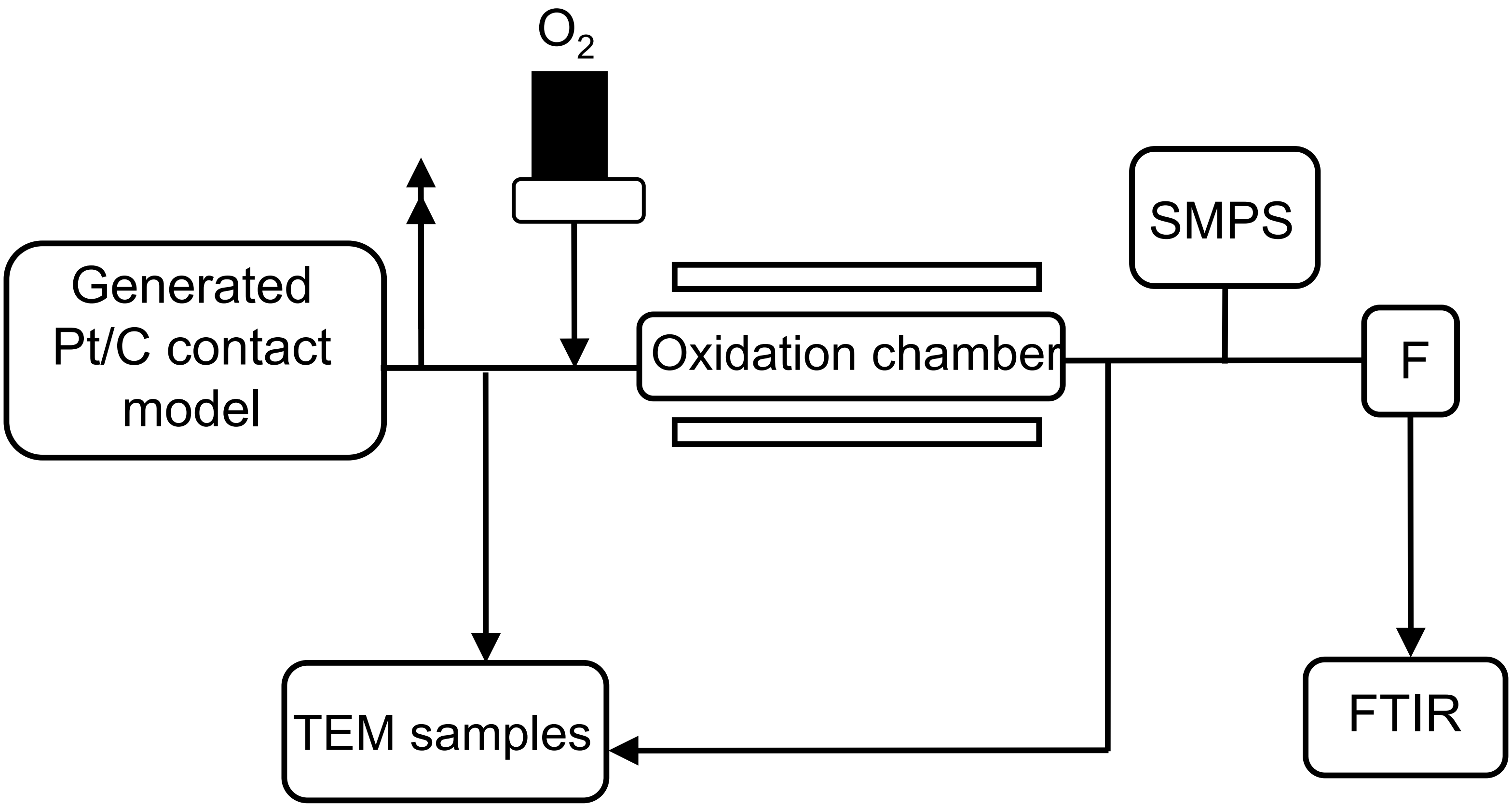
Pt nano sphere partially embedded in a carbon nano agglomerate (*PVD contact*)





Oxidation set-up in aerosol state

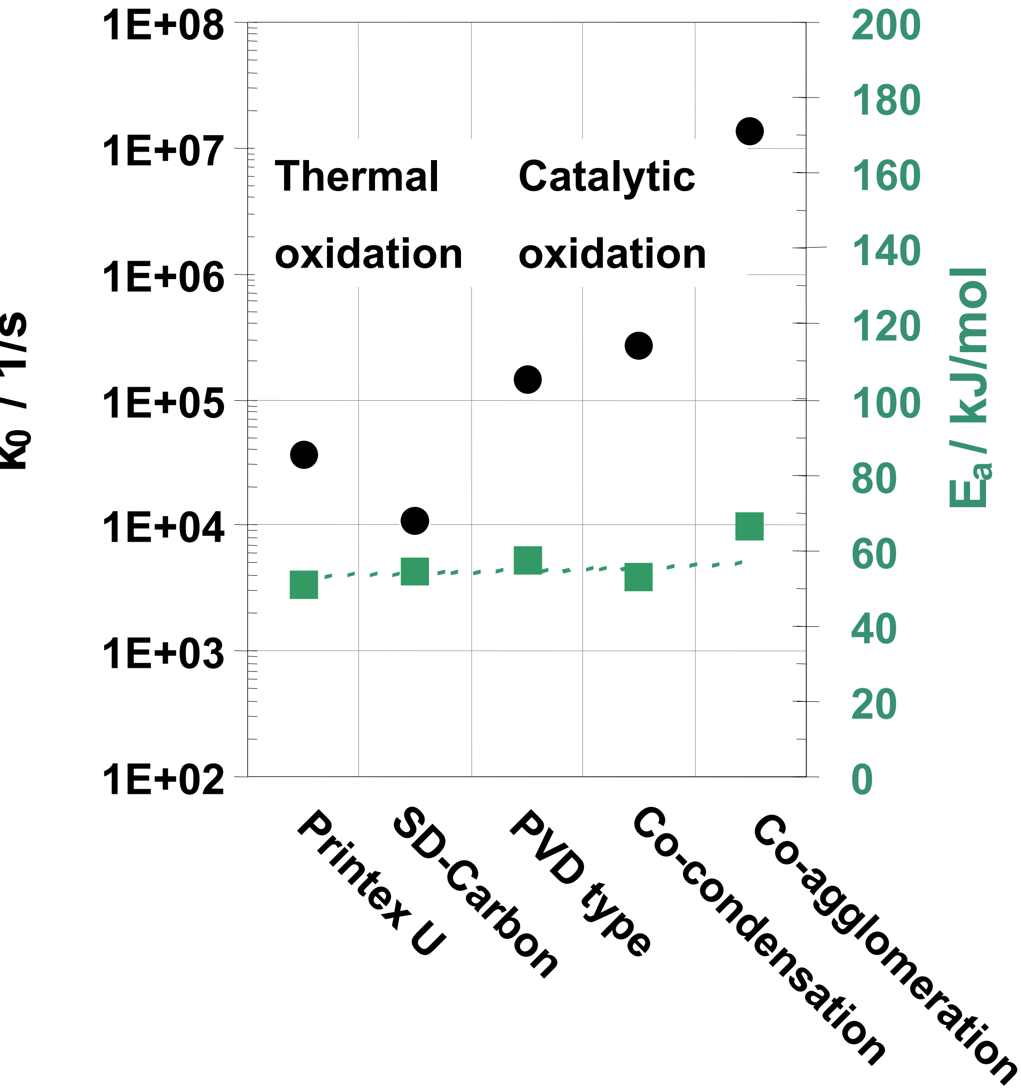
Oxidation in aerosol state
On-line measurement of gas concentration using FTIR

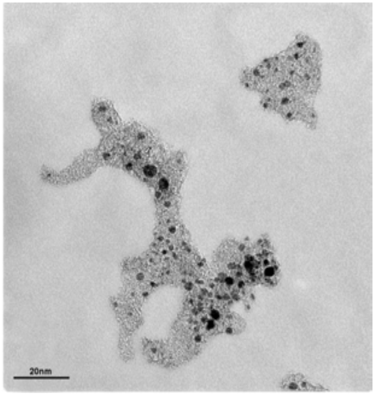


Determination of CO_2 concentration

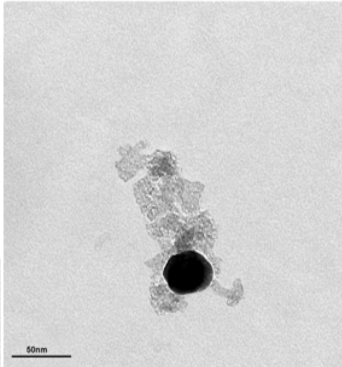
Results

E_a and k_0
(results of the oxidation in the aerosol state)

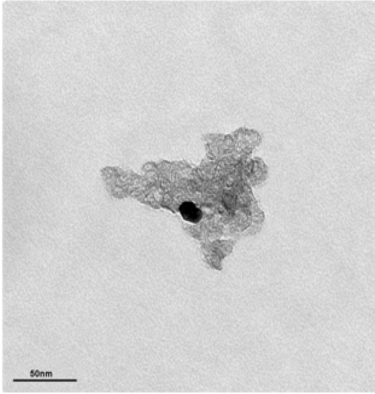




PVD contact

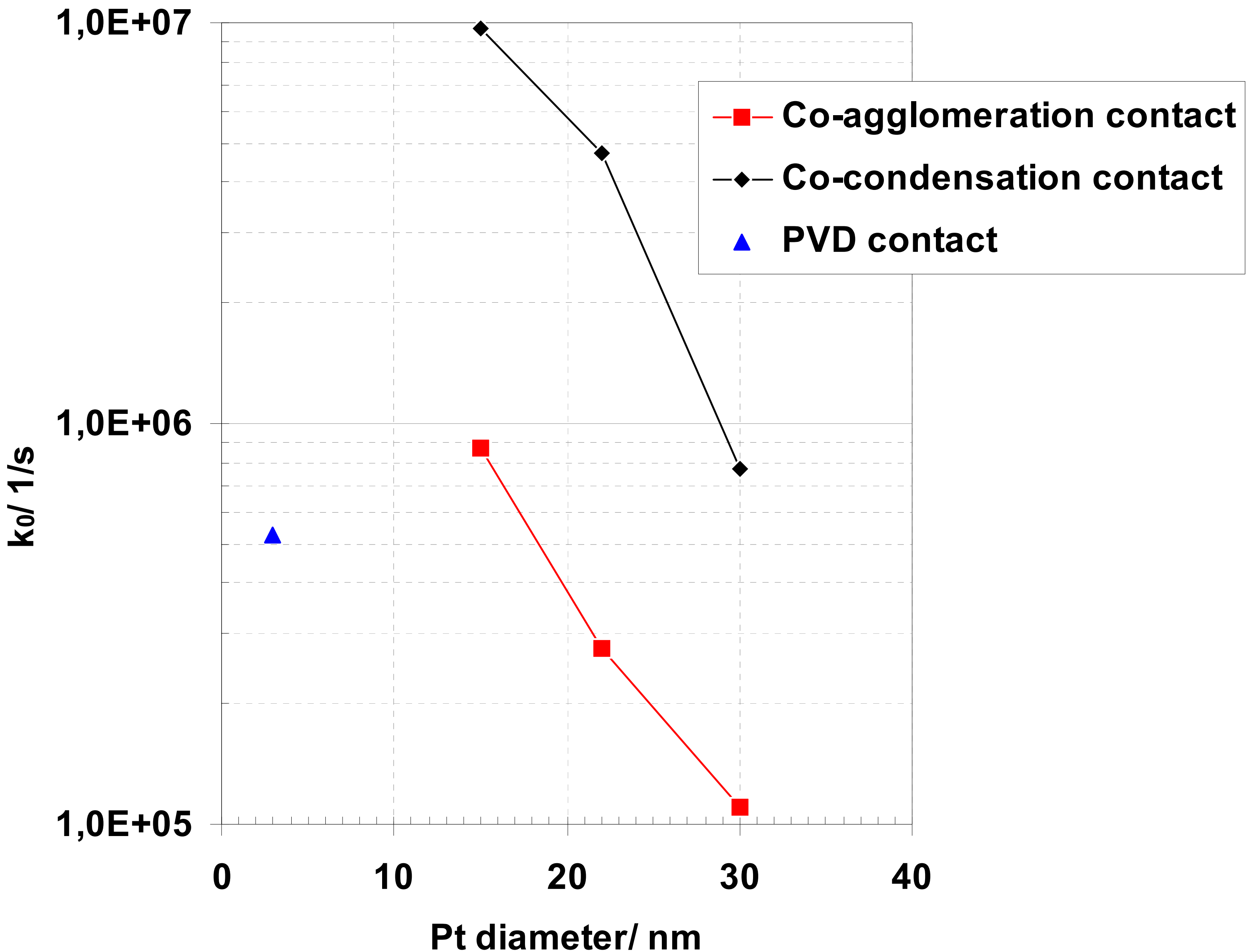


Co-agglomearation contact



Co-condensation contact

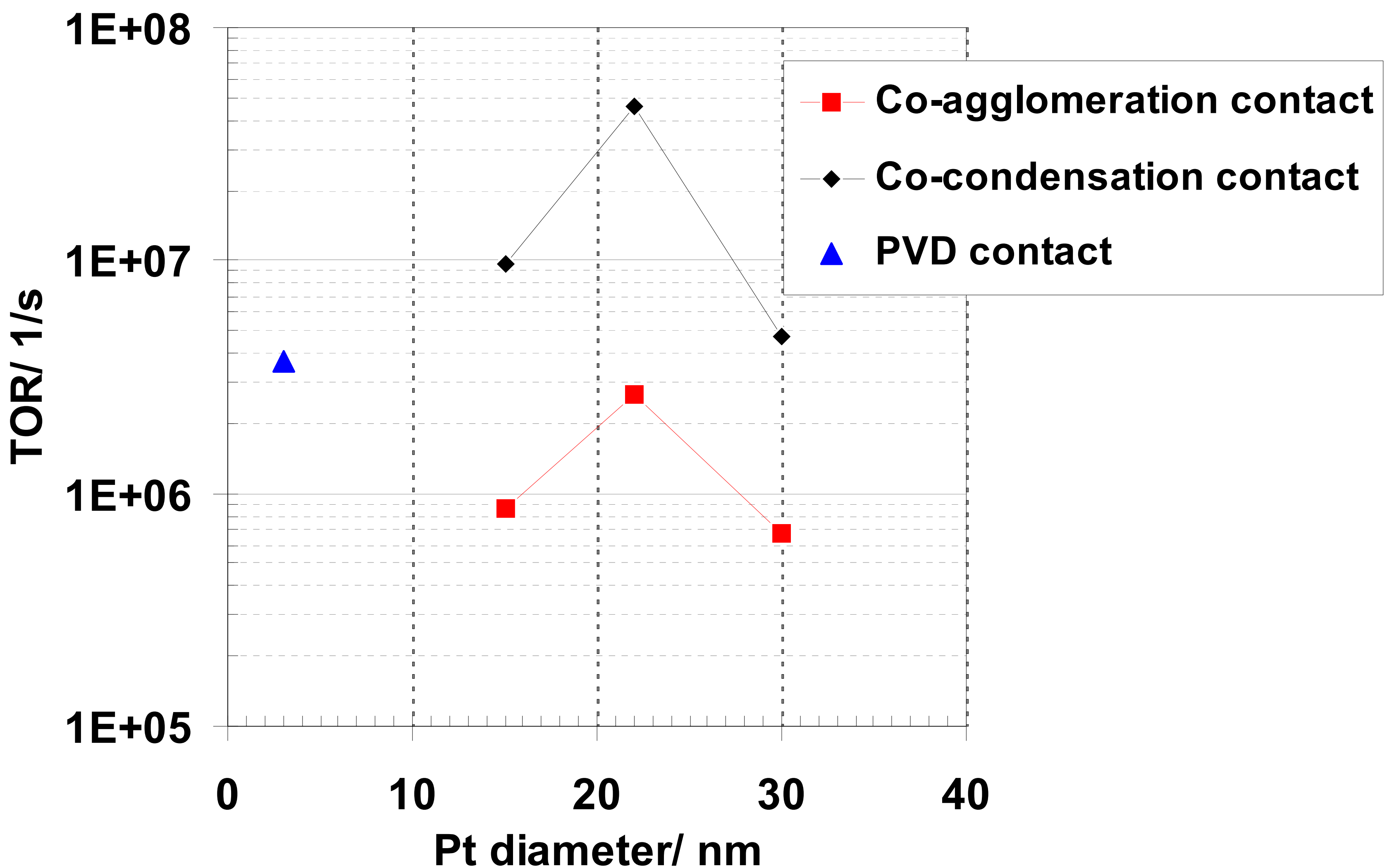
Velocity coefficient vs. Pt particle size



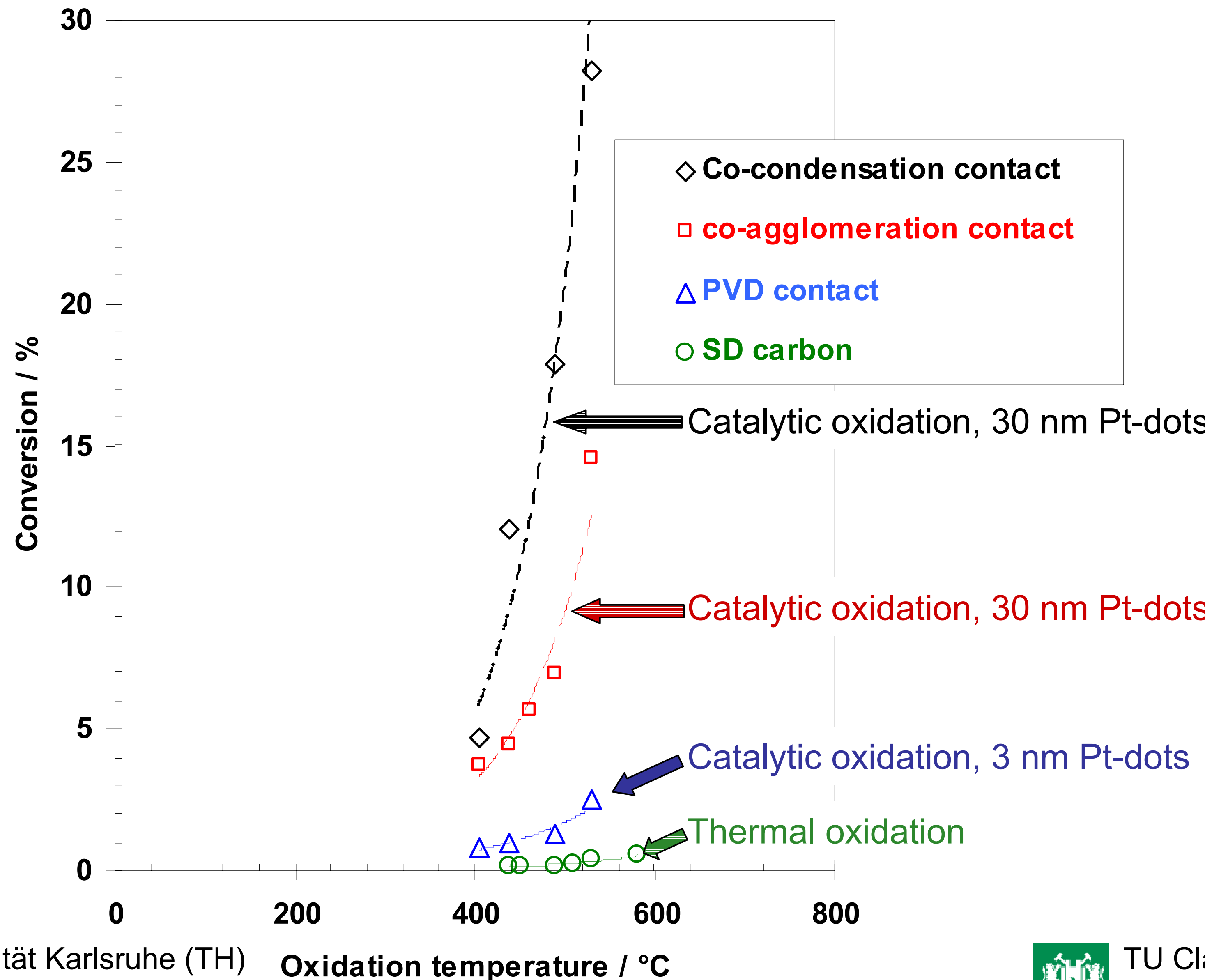
Univer

TOR vs. Pt particle size

Turn Over Rate (TOR) = reaction rate based on the surface of catalyst



Conversion – Reaction Temperature



Comparison of the kinetic parameter for Co-condensation and Co-agglomeration contact

T /°C	time s	Size of Pt nm	$k_{0,co\text{-}aggl.}$ 1/s	$k_{0,co\text{-}cond.}$ 1/s	Conversion Co-agglomeration	Conversion Co-condensation	$k_{0,co\text{-}cond.}/k_{0,co\text{-}aggl.}$
405-530	1,52	15	4,8E+05	9,7E+06	17%	30%	20
405-530	1,74	22	2,9E+05	4,7E+06	14%	28%	16,53
405-530	1,72	30	1,1E+05	7,7E+05	9%	14%	7,3

1)d_P smaller→ higher specific surface area → increase of conversion

2) Co-condensation contact →

Higher reaction rate
intensive contact than Co-agglomeration contact

Conclusion

Oxidation of carbon aerosol (thermal and catalytic):

E_a = 50 ± 10 kJ/mol

Different contact type→ different k_0

An optimum size of approx. 22 nm of Pt spheres is observed.

The best Pt/C contact model → Co-condensation type

Acknowledgement:

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