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Simultaneous interaction of methyl radicals and atomic hydrogen with amorphous hydrogenated carbon films

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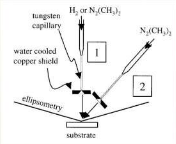
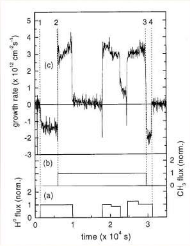




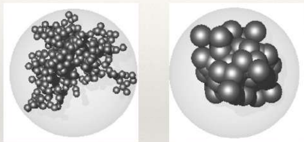
CH_3 Sticking = 10^{-4} w / o H
= 10^{-2} w H

H → Dangling bond; H_2; CH_3 → $sp^3 \to sp^2$ Erosion; CH_3 → $sp^2 \to sp^3$ Growth

How do these surface processes affect the particle shape?

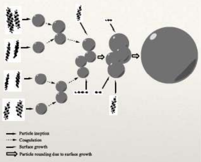
From combustion chemistry

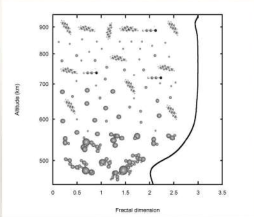
Only coagulation | Coagulation + surface chemistry



Mitchell & Frenklash (2003)

HETEROGENEOUS CHEMISTRY





Fractal aggregates

$$N_m = A\left(\frac{R_f}{r_m}\right)^{D_f}$$

Heterogeneous reactions affect the particle shape and the transition from spheres to aggregates

Main haze layer

(Pruppacher & Klett, Microphysics of clouds and precipitation, Kluwer Academic Publishers)

Pure microphysics

Continuity equation for particles

$$\frac{\partial n_p(\nu_p)}{\partial t} = -\frac{1}{r^2}\frac{\partial (r^2\Phi(\nu_p))}{\partial r} + \frac{1}{2}\int_0^{\nu_p} K(u,\nu_p-u)du - n(\nu_p)\int_0^{\infty} K(u,\nu_p)n(u)du + \frac{\partial n_p}{\partial t}\Big|_{prod}$$

Flux divergence | Growth from smaller particles | Loss to larger particles | Inception

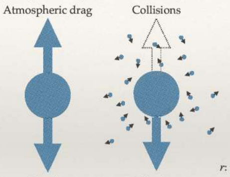
n_p: number density particles with volume ν_p
$\Phi(\nu_p)$: flux of particles
$K(u,u')$: coagulation Kernel of particles with volumes u and u'

$$\Phi(\nu_p) = -V_s(\nu_p)n(\nu_p) - K_{ZZ}n_a\frac{\partial (n(\nu_p)/n_a)}{\partial r}$$

$V_s(\nu_p)$: settling velocity
K_{ZZ}: atmospheric mixing
n_a: atmospheric density

Settling velocity

$K_n << 1$ Hydrodynamic regime, Atmospheric drag | $K_n > 1$ Molecular regime, Collisions

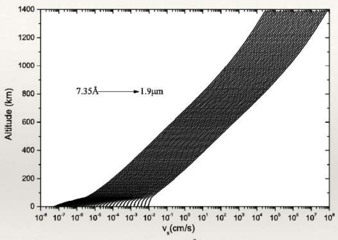


$K_n = \frac{\lambda_a}{r}$ Knusden number

For spheres $V_s(r) = \frac{2r^2\rho g}{9\eta_a}$ (Stokes limit) $\quad V_s(r) \sim \frac{r\rho}{\rho_a V_T}$

r: particle radius
ρ: particle density
ρ_a: gas density
g: gravity accel.
η_a: gas viscosity
V_T: thermal velocity

Settling velocity



$$V_s(r) = \frac{2r^2\rho g}{9\eta_a}f_{slip}$$

$f_{slip} = 1 + 1.257K_n + 0.4exp(-1.1/K_n)$
Cunningham slip-flow factor

Coagulation Kernels

Brownian: random collisions between particles

$$K_B(r,s) = \alpha_c 4\pi(D_r + D_s)(r+s)\beta \quad \text{(for spheres)}$$

$D_r = \frac{k_B T f_{slip}}{6\pi\eta_a r}$: Particle diffusivity

β, f: interpolation formulas between free-molecular (K_n>1) and continuum regimes (K_n<1)

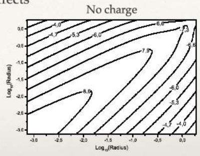
α_c: sticking efficiency $\quad K_n = \frac{\lambda_a}{r}$: Knusden number

Charge effects

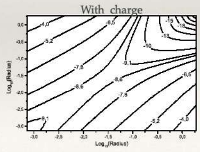
Reduction in sticking efficiency

$$\alpha \simeq \exp\left(-\frac{Z_1 Z_2}{k_B T(r_1+r_2)}\right) = \exp\left(-\frac{r_1 r_2 \chi^2}{k_B T(r_1+r_2)}\right)$$

No charge



With charge



Charge effects

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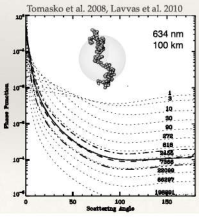
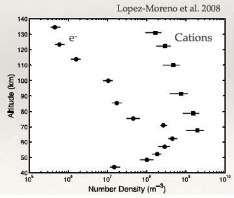
$\chi \sim 15\ e^-/\mu m$

Tomasko et al. 2008, Lavvas et al. 2010



Lopez-Moreno et al. 2008



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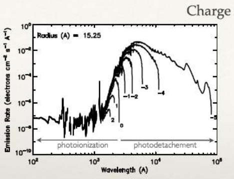
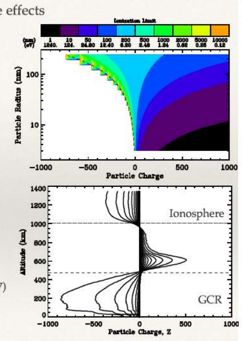
Charge sources:
- collisions with ions & electrons
- photoionization
- photodetachement

$$J^+ = J_{pi} + J_{h\nu}$$
$$J^- = J_{ni} + J_e$$

→ Charge distributions

Charge effects





$$J_{h\nu}(z,q,r) = \int_\lambda \Phi_\lambda(z)\sigma_{abs}(\lambda,r)Y_\lambda(q,r)d\lambda$$

$\Phi_\lambda(z)$: Solar flux
$\sigma_{abs}(\lambda,r)$: absorption cross section
$Y_\lambda(q,r)$: ionisation yield (Draine et al. 1987)

Particle Shape

Fractal aggregates

$$N_m = A\left(\frac{R_f}{r_m}\right)^{D_f}$$

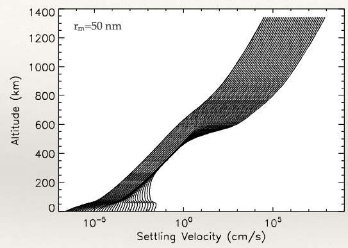
$D_f = 1$ Linear
$D_f = 2$ Fluffy
$D_f = 3$ Compact

Cabane et al. 1992

Growth of the cluster governed by:	$l_p \ll R_c$ (continuous regime)	$l_p \gg R_c$ (ballistic regime)
Particle–cluster aggregation (monomer–polymer)	$D_f \approx 2.5$	$D_f \approx 3$
Cluster–cluster aggregation (polymer–polymer)	$D_f \approx 1.75$	$D_f \approx 2$

$$r \to R_f = \frac{r^{3/D_f}}{r_m^{(3-D_f)/D_f}}$$ for formulas based on spheres

Settling velocity for aggregates with D_f=2



Particles settle with the velocity of the monomers!

Solution

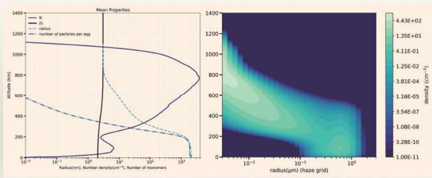
Continuity equation for particles

$$\frac{\partial n_p(\nu_p)}{\partial t} = -\frac{1}{r^2}\frac{\partial (r^2\Phi(\nu_p))}{\partial r} + \frac{1}{2}\int_0^{\nu_p} K(u,\nu_p-u)du - n(\nu_p)\int_0^{\infty} K(u,\nu_p)n(u)du + \frac{\partial n_p}{\partial t}\Big|_{prod}$$

Bin method: Solve for density of particles over a geometrically expanding volume grid.
Slow but more informative.

Moments method: Assume a distribution form (e.g. lognormal) and solve for the moments of the distribution.
Requires assumptions, but much faster and integrable to GCMs!
(de Batz de Trenquelléon et al. 2025)

Application to Titan



From A. Damiens, PhD

Haze mass flux ~ 3×10^{-14} g cm^{-2}s^{-1} corresponds to **30%** of N_2 & CH_4 photolysis
Haze formation yield

Cloud formation