

Evaporating grains in P/Halley's coma

M.K. Wallis, R. Rabilizirov, and N.C. Wickramasinghe

Department of Applied Mathematics and Astronomy, University College, P.O. Box 78, Cardiff CF1 1XL, Wales, UK

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Summary. Evidence is presented for the hypothesis that the organic grains streaming from a cometary nucleus lose substantial volatile constituents within 10^5 s. To develop the hypothesis, we study grain properties appropriate to both the comet-probe encounters at 0.8–0.9 AU and to comas observed at 4–5 AU. While submicron-sized grains of ice contaminated by 1% or more absorptive graphite can explain the strong OH coma far from the Sun, as in comet Bowell at 4.6 AU, the dynamical lifetime of the grain coma implies mm-sized and therefore cooler grains, with weaker H_2O or OH attachment than ice at about 10 kcal/mole. Grains in comet Halley, if composed of similar organic material must also contain fractions of more strongly bound components. In this study we adopt kerogen as a prototype organic complex having a range of bond energies. Temperatures and evaporation rates of kerogen grains are calculated using Arrhenius relations and Mie-Güttler radiative scattering theory with complex refractive index as measured for two cases: organic material of biological origin and UV-processed methane ice (“tholin”). A time-scale of 10^4 s at 0.9 AU corresponds to submicron grains with rather strongly-bound organics of ~ 40 kcal/mole, identified with bridges of ether, ketone, sulphide etc., bonds. The steeply increasing strength of H and O inside 1 AU ($\propto r^{-4}$), unlike OH, corresponds to increasing breakdown following rupture of the chemical bridges. The $10\ \mu\text{m}$ IR feature can be explained by grains of an organo-siliceous material or organic mantle on a siliceous core, a few μm in size and about 20 kcal/mole bond strength.

Key words: OH-coma – grain sublimation – organic composition – comet Bowell – comet P/Halley

1. Introduction

That cometary comas contain icy or ice-coated dust grains is a proposal of many years standing (Huebner and Weigert, 1966). In offering a larger surface area to the solar radiation, such grains on evaporation can even dominate over the nucleus as sources of gas and change the hydrodynamics of the coma. We now know from Vega and Giotto that small grains (femtogramme and smaller) are abundant, though it is unknown whether they arise directly from the nuclear material or condense out of the super-saturated gas near the nucleus (as in laboratory simulation of sublimating mixtures; see Dobrovolsky and Kaimakov, 1977). Optical studies indicate a new composition (Moroz, 1986), not studied hitherto. We know from in situ analysis (Kissel et al., 1986) and

spectroscopy of the 3–4 μm waveband (Wickramasinghe and Allen, 1986; Knacke et al., 1986) that much of the dust is organic rather than mineral in composition. High ratios of radiation pressure to solar gravity at $\beta \simeq 2.5$, inferred from tail structure (Lamy, 1986), are consistent with $\sim 0.1\ \mu\text{m}$ grains of graphite-like absorptive material being present in the inner coma and relatively abundant close to perihelion.

An organic composition for the grains was first proposed by Vanýsek and Wickramasinghe (1975). They hypothesised polyoxymethelene or polyformaldehyde compounds, whose radiation scattering properties are generally suitable. Such grains would release appropriate OH, CN, HCO, etc., under the solar radiation.

Evidence for the evaporation of grains as the gas and dust mixture expands from the comet comes from various directions. The comet-probes showed that the fraction of grains with Si:C exceeding 50% is lower close in to the nucleus (Kissel et al., 1986) while there are substantial amounts of C, CH, and C^+ in the gas and plasma phases (Balsiger et al., 1986; Krankowsky, 1986). Molecular CO increases outwards with a 10^4 km scale (Eberhardt et al., 1986) while CN increases both on this scale and a short (600 km) scale (Krasnopolsky et al., 1986). The report of radial structures in images covering the 388 nm CN band (A'Hearn et al., 1986) convinced many skeptics that grains are a source of some CN and other cometary molecules. The relative dominance of CN in comae at large heliocentric distance, compared with C_2 etc. (Wurm and Balazs, 1963) suggests that CN is more loosely bound and released at lower temperature.

The major H_2O component probably also comes in part from grains. An OH coma was marginally detected in Halley at 4.4 AU pre-perihelion (Festou et al., 1986), compatible with O I 630 nm observed six weeks earlier and of usual strength relative to CN; at such distance, sublimation from contaminated crystalline ice at ~ 130 K is too slow (cf. Sect. 2), so less strongly bound H_2O may be inferred. The H and O abundances, judged from H α and L α strengths (Craven et al., 1986) and from O I 630 nm intensities (Sherb et al., 1986), increased more strongly than OH (Feldman et al., 1986) with closeness to the Sun, especially inside 1 AU where the OH strength was roughly constant but with strong fluctuations. Suiséi's L α observations (Kaneda et al., 1986) imply a source of H, perhaps CH, with faster photo-production than from H_2O . The OH if it all came from evaporation of H_2O -ice from the nucleus surface, requires an emitting surface area of some $20\ \text{km}^2$ at the time of the Giotto encounter ($5 \cdot 10^{29}$ molecules/s, Feldman et al., 1986) from a surface normal to the solar radiation at 0.9 AU), which is 3–4 times the projected area of the “jet source”, that is itself structured (Keller et al., 1986; Huebner et al., 1986a). The R^{-2} variation of H_2O density, obeyed quite closely along the

Send offprint requests to: M.K. Wallis

probe trajectories even on the approach from behind the nucleus (Curtis et al., 1986), is understandable if much of the H_2O issues from the grains despite strongly anisotropic initial emission from the sunlit surface. Although the grains move sunward at $\lesssim 0.5 \text{ km s}^{-1}$, H_2O molecules evaporating at higher speed and isotropically from individual grains can fill the tail-side coma, resulting in much reduced anisotropy.

2. Grains at several AU from the Sun

2.1. What size icy grains in comet Bowell?

Comet Bowell 1980b had a well-developed particle coma more than a year prior to perihelion at 3.4 AU. In early 1981, it showed high OH strength over $r = 5.3\text{--}4.6$ AU (for $> 10^7$ s) but lower OH a year later around perihelion. If this arose from large ice grains composing the long-lived coma (the favoured explanation – A'Hearn et al., 1984; Sekanina, 1982; Jewitt, 1984), then the grains must be large enough for a long dynamical lifetime, which for a grain radius a in mm and $D = 50''$ diameter coma (Sekanina, 1982) under radiative acceleration g is

$$\sqrt{(D/g)} = 5 a [\text{mm}] 10^7 [\text{s}]. \quad (1)$$

Evidently this implies mm-sized grains in the 1981 coma. However, contaminated icy grains would be too cold to emit significant H_2O at 4.6 AU (Hanner and Campins 1986), being around the 130 K black-body temperature [their total surface area being inferred from the $20 \mu\text{m}$ IR flux (Hanner and Campins, 1986)]. If icy grains are to supply H_2O for the production of OH, they have to be warmer, as are micron-sized grains.

2.2. Analysis of micron-sized dirty-ice grains

Taking various fractions of contaminant absorbers embedded in the ice, we have calculated the grain temperature on the basis of Mie scattering theory for spherical grains. We could use the Maxwell-Garnett mixing rule for spherical inclusions (Bohren and Wickramasinghe, 1977) but such complexity makes a difference only for a large imaginary part to the refractive index near the solar spectrum peak. Mukai (1986) who does use the mixing rule also assumes a single size to the inclusions, but results are very sensitive to that. So we employed the simple linear combination

$$m = (1 - w_g) m_{\text{ice}} + w_g m_{\text{graphite}} \quad (2)$$

for fractional contamination (by volume) w_g , and adopted the tabulated values of refractive indices m for ice and graphite. We used a Planckian spectrum for the solar radiation as giving adequate accuracy. The temperature for slightly contaminated $5 \mu\text{m}$ grains agrees closely with Hanner (1981) for a real spectral distribution. We allowed for the energy lost in sublimation and calculated the grain lifetime against evaporation according to the half-size formula

$$\frac{1}{2} a / |\dot{a}| \quad \text{with} \quad |\dot{a}| = (p/q) (m/2\pi kT)^{0.5}. \quad (3)$$

Figures 1 depict results for various values of w_g , showing that the evaporation lifetime of $5 \mu\text{m}$ grains at 4.6 AU is 10^7 s if $w_g \simeq 1\%$ (more precisely, 0.25–1.5%), but longer for higher or lower contaminant fractions than this. Smaller grains have generally much shorter lifetimes (as the 0.3 and 0.1 μm grains in Fig. 1b, excepting the uncontaminated 0.3 μm example). We have compared results for the absorbing graphitic proportion confined to

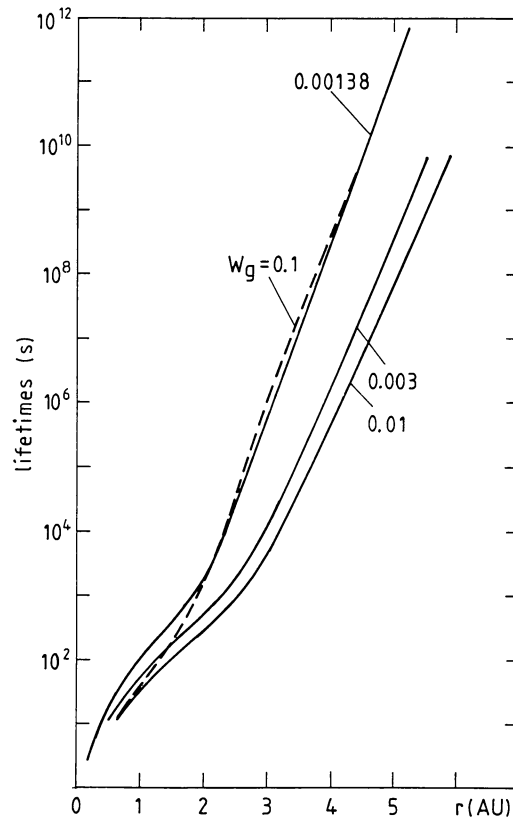


Fig. 1a. Lifetimes of $5 \mu\text{m}$ radius ice grains contaminated by small fractions of graphite w_g (by volume). The “lifetimes” given here are really half-lives, being the time to decrease to half the initial radius, by when the scattering properties and graphite fraction have changed drastically. The value $w_g = 0.00138$ is chosen to give the absorption coefficient $k = 0.002$ adopted by Hanner (1981). The fraction $w_g = 0.01$ gives hotter and faster sublimating grains than larger or smaller fractions for the $5 \mu\text{m}$ grains

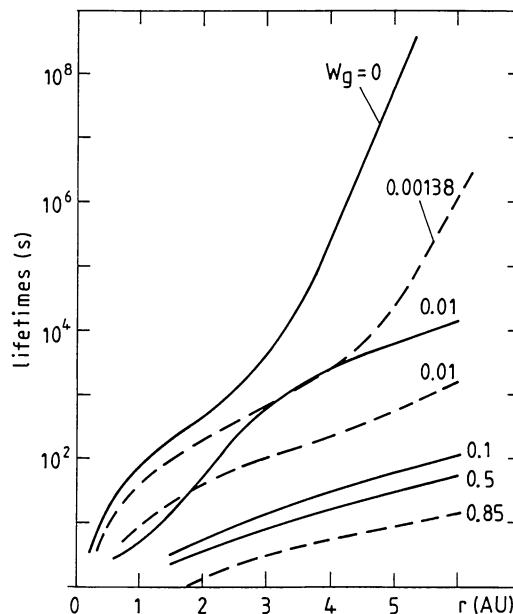


Fig. 1b. Lifetimes of submicron ice grains: solid lines – $0.3 \mu\text{m}$ radius; broken lines – $0.1 \mu\text{m}$ radius, with fractional contamination of dispersed graphite w_g as labelled

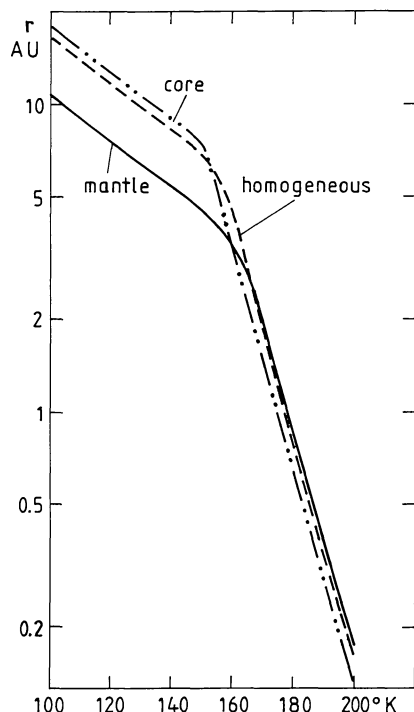


Fig. 2. Solutions of the sublimation equation for $0.1 \mu\text{m}$ ice grains containing 1% ($w_g = 0.01$) graphite, showing heliocentric distance where a given temperature is attained, for cases with the graphite in a mantle, in a central core, or homogeneously distributed as in Figs. 1. There is a marked change in slope where sublimation starts to become important in the energy balance (at 155 K). The 10 K spread in temperature at the 4.6 AU distance of comet Bowell is significant, showing that a contaminated grain that starts evaporating but develops a mantle of the non-volatile graphite will have its lifetime lengthened by several times

a central core, or to an outer mantle. Figures 2 give results for $w_g = 1\%$ graphite. The case with it concentrated in a core is close to the uniform distribution, T differing by under 5 K. A graphite-mantled grain, however, radiates efficiently and is substantially cooler beyond a few AU. The cooling is much stronger in the larger example shown ($0.4 \mu\text{m}$, Fig. 2b; cf. $0.1 \mu\text{m}$ of Fig. 2a). At 4.6 AU, mantled grains of $0.4 \mu\text{m}$ and larger are close to the 130 K black-body temperature and evaporate too slowly. Evidently grains of a few μm and initially uniform contamination could sublimate at 4.6 AU to produce the observed OH, but as the contaminating graphite accumulates at their surfaces they cool and sublimation is stopped. In any case, the residence time (1) of such grains in the coma is $< 10^6$ s, so continual emission of such grains in early 1981 is implied, but lower emission around perihelion one year later. The small grain hypothesis would not explain the relatively sharp coma boundary reported (Jewitt, 1984), nor would small grains alone give the $20 \mu\text{m}$ IR flux and the $10 \mu\text{m}:20 \mu\text{m}$ ratio observed (Hanner and Campins, 1986).

2.3. Alternative to ice grains

Our preferred explanation is for large grains of primarily organic composition (Hoyle et al., 1985) with temperature similar to a black body (130 K at 4.6 AU) that emit $\text{H}_2\text{O}/\text{OH}$ on the 10^7 s time scale. This requires slightly weaker binding energy than ice, at about $E = (130/155) \times L[\text{H}_2\text{O}] \simeq 10$ kcal/mole. On loss of

adsorbed H_2O , CO_2 and weakly bonded side-chains (e.g. CH_2 , NH , H , CN), these grains would shrivel and become more absorbant ($\text{C}=\text{C}$ bonds), giving in particular the dark grains observed postperihelion at 3.5 AU (1982 June) with temperature (Hanner and Campins, 1986) 15% above that of a black-body (i.e. IR absorptivity < 0.25).

3. Comet Halley's grains

The infra-red spectra of comet Halley imply that the grains probably consist of a complex mixture of organic compounds (Knacke et al., 1986). If of similar initial organic composition to those in comet Bowell, the submicron femtogram component would shed its loosely-bonded $\text{H}_2\text{O}/\text{OH}$, H , CN etc., within 100 s in free space at 0.9 AU (Fig. 1). Near the nucleus, however, IR radiation is strongly blanketed by gaseous H_2O , CO_2 , etc., (Bockelée-Morvan and Crovisier, 1986; Wallis, 1986) and also recondensation on the grains may occur (Kitamura and Yamamoto, 1986; Crifo, 1986). Still, the loosely-bonded (10 kcal/mole) fraction has surely left the grain surfaces within a few thousand km (Crifo, 1986). Rupture of $\text{C}=\text{C}$ bonds at 70–80 kcal/mole would not however proceed. Between these extreme bond strengths, various chemical components do come off.

3.1. Grains composed of kerogen

In the absence of specific composition data, we take as prototype the material classified as *kerogen* – the insoluble components of organic residues on Earth (Tissot and Welte, 1978) and thought to resemble that in carbonaceous chondrites and interstellar grains (Pelet, 1980). We assume the bonds of strength E_i are disrupted at rates given by the Arrhenius formula

$$d(\ln n/dt) = A_i \exp(-E_i/RT) \quad (4)$$

which simulates independent rupturing of bonds in a frozen lattice (Tissot and Welte, 1978 – Part V, Sect. 4.2). We adopt the bond strengths and proportions given for Type II terrestrial kerogen (reproduced in Table 1), but expect the weakly-bonded ($E \lesssim 30$) fractions to be higher in space organics. The radiative absorptivity of organic grains is uncertain, for the graphitic fraction found in terrestrial kerogens may not be prevalent in space. Organics are largely dielectric plus pigments, typified by chlorophyll with absorption peaks in the visible. Using measurements of terrestrial grass extracts and micro-organisms (Al-Mufti, 1985), we take the real part of the refractive index as 1.4 and imaginary part dominated by peaks around 433 and 660 nm and with significant value in the IR (details in Table 2). From polarization data, Mukai et al. (1986) derived the real refractive index as 1.4, while Krasnopolsky et al. (1986) give 1.35, but the difference is not significant for the present analysis.

The incident energy going to rupturing the bonds in the submicron grains is tiny, so the temperature given by ignoring that component can be calculated via Mie theory for spherical grains of various sizes. By setting the timescale for transformation at 10^4 s appropriate to submicron grains' residence time in the inner 10^4 km coma, we calculate from (3) the heliocentric distances where the various bonds E_i suffer rupture. The results for Type II kerogen are given in Table 1: the grain temperatures are numerically close to $14 E_i$ [in K for E in kcal/mole]. Evidently, bonds up to 50 kcal/mole rupture in Halley's grains near perihelion ($q = 0.6$ AU) where temperatures of 700 K are attained.

Table 1. Heliocentric distance r_{crit} for a time scale of 10^4 s for breaking bonds of strength E in three sizes of organic grains. The critical temperatures vary little: $T_{\text{crit}} \approx 14 E$. The E -data is a representation of experimental results on Type II kerogen (Tissot and Welte 1978, V-4.2); the constants A_i given there are not used as they apply to transformations in liquid rather than the solid lattice

E (kcal/mole)		10	30	50	60	70	80
f -fraction of bonds		0.022	0.034	0.251	0.152	0.116	0.120
$a = 0.05 \mu\text{m}$	T_{crit} (K)	140	420	670	820	965	1105
	r_{crit} AU	15	1.6	0.64	0.44	0.33	0.26
$a = 0.1 \mu\text{m}$	T_{crit} (K)	143	430	680	835	985	1125
	r_{crit} AU	16	1.6	0.64	0.44	0.33	0.26
$a = 0.3 \mu\text{m}$	T_{crit} (K)	148	445	710	860	1010	1160
	r_{crit} AU	13	1.25	0.50	0.34	0.25	0.20

Table 2. Complex refractive index of model organic grains. We employ $m = 1.4 - ik$ with $k(\lambda)$ based on experimental results for biological organic material (Hoover et al., 1986): a) in the UV over $\lambda = 0.2 - 0.3 \mu\text{m}$, Fig. 7.6 and Table 7.17 of Al-Mufti (1985) measured for a mixture of micro-organisms; b) at visual $\lambda = 0.4 - 0.9 \mu\text{m}$, values of absorbance for grass extracts; c) in the IR, $\lambda = 2.5 - 16 \mu\text{m}$, transmittance measured for *E. coli* bacteria (Fig. 4.7 of Al-Mufti). In the remaining wavebands, we have adopted constant values for the mass absorption coefficient K . At $\lambda < 0.2 \mu\text{m}$, $K = 25000$; at $\lambda > 20 \mu\text{m}$, $K = 500$ and $k = 1.2 \cdot 10^{-5} K [\text{cm}^2 \text{s}^{-1}]$. The value $\text{Re}(m) = 1.4$ corresponds to compact material and has been used to explain polarization of the scattered radiation (Mukai et al., 1986): lower values $\lesssim 1.2$ would better simulate porous or fluffy grains, but present results are not sensitive to it

$\lambda (\mu\text{m})$	0.21	0.23	0.25	0.35	0.45	0.55	0.65	0.75	0.85	0.95
k	0.08	0.077	0.375	0.075	0.19	0.01	0.156	0.0014	0.0015	0.0017
$\lambda (\mu\text{m})$	1.05	2.05	3.05	4.05	5.05	6.05	7.05	8.05	9.05	10.05
k	0.0019	0.0037	0.035	0.015	0.018	0.10	0.06	0.08	0.125	0.94

Figure 3 presents calculations of the evaporation lifetimes in fuller detail, including the energy absorbed in breaking the bonds (analogous to sublimation energy) and using the relative fractions of bonds f from Tissot and Welte (1978). We find, for example, that at 0.9 AU the 30 kcal/mole bonds have a very short 10–100 s lifetime (so diffusion of radicals through the grain matrix may in practice limit the escape rate), while 40 kcal/mole bonds last $4 \cdot 10^3$ s in $0.1 \mu\text{m}$ grains but $4 \cdot 10^5$ s in $0.3 \mu\text{m}$ grains (and ten times shorter at 0.8 AU).

3.2. Grains composed of synthesised organics

An alternative prototype for organic grain material is the “tholin” obtained by irradiating frozen methane and water (Khare et al., 1984). Mie calculations using their tabulated refractive index give lower temperatures (by ~ 100 K) and longer lifetimes, because the material lacks the absorption peaks in the visible of our biological organic. However, grains larger than $0.1 \mu\text{m}$ are still quite hot enough to evaporate and undergo chemical “cooking” (Hanner, 1986). The latter author shows the smallest grains of tholin ($0.01 \mu\text{m}$) to be very cold, but we find them to be hot (e.g. ~ 500 K at 0.9 AU) so still liable to rapid cooking. Figure 2 presents comparable calculations for $E = 30$ kcal/mole bonds in spherical grains of tholin – showing lifetimes up to twice as long but nowhere strongly different from the biological organics. Figure 2 shows also sample lifetimes for

ice grains, including the contamination fraction $w_g = 0.00138$ corresponding to absorptive $k = 0.002$ in the visual adopted by Hanner (1981). At 0.9 AU, life-times are under 100 s even for pure ice of $a = 0.3 \mu\text{m}$.

4. Discussion

The correspondence between bond strengths and particular chemical components is not very close, because of the complex chemical linkages and “cages”. Breaking one bond induces rearrangement of neighbouring ones, so modelling by independent breakages [Eq. (4)] may not be adequate. The 10 kcal/mole strength corresponds to physical/chemical absorption and some H/OH bonding. Carbonyl, carboxy and methoxy side-chains tend to be superficially attached with 20–30 kcal/mole. Ether (–O–), ketone (–CO–) and sulphide (–S– or –S–S–) bonds tend to serve as bridges with 40–50 kcal/mole, while carbon bonds are as strong as 60–80 kcal/mole.

Both the biological and the “tholin” organic prototypes (Sects. 3.1, 3.2) have significant absorption in the visible waveband, so grains of either substance are strongly heated in the outstreaming coma. If contaminant graphite were present as common in terrestrial kerogen, they would be still hotter.

Evidently the carbon bonds stay intact in Halley’s submicron grains but, with rupture of bonds up to 50 kcal/mole, substantial

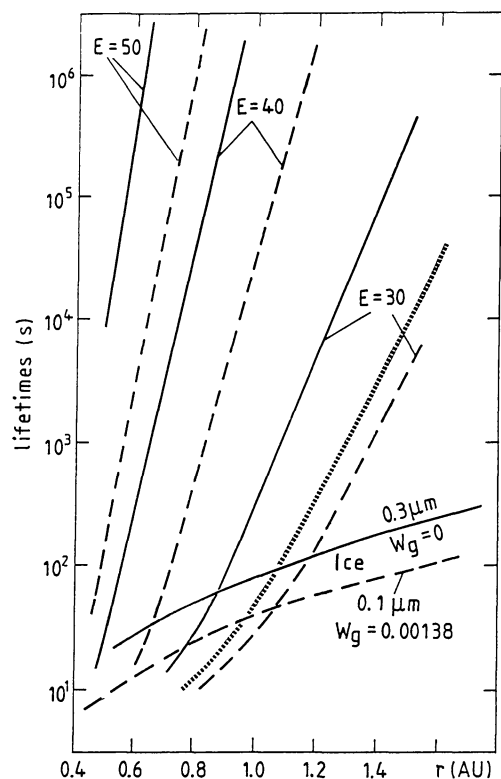


Fig. 3. Lifetimes of biological organic grains of $0.1\ \mu\text{m}$ (broken lines) and $0.3\ \mu\text{m}$ radius (solid lines) to evaporation or destruction of chemical bonds of various strengths E (kcal/mole) as indicated. Evidently for comet Halley at $0.8\text{--}0.9\ \text{AU}$, $0.1\ \mu\text{m}$ grains quickly lose their $E=40$ components, but $0.3\ \mu\text{m}$ grains retain them for $\sim 10^5\ \text{s}$. $E=50$ components are hardly affected within the $10^5\ \text{km}$ coma. Results for a laboratory organic "tholin" $0.1\ \mu\text{m}$ grain with $E=30$ are shown dotted, being some twice as long due to lower absorptivity. Sample ice-grain lifetimes are plotted for comparison, being under $100\ \text{s}$ for the Halley spacecraft encounters, so evaporation competes with condensation (Crifo, 1986) in the very inner coma

carbonisation may be achieved within $0.7\ \text{AU}$ (explaining the high radiative repulsion indicated by the $\beta \approx 2.5$ syndyne – Lamy, 1986 – see introduction). The time-scale at $0.9\ \text{AU}$ for breaking the chemical "bridges" is sensitive to grain size, but comparable to outstreaming times ($0.5\ \text{km s}^{-1}$ speeds). As bridge-breaking exposes interior CH_2 etc., this can explain the decreasing fraction of C with distance from the nucleus (Kissel et al., 1986 – correlation with grain size would check this). The bridge-breaking also allows increasingly efficient release of H (and O) at smaller heliocentric distance, unlike OH if it comes mainly from H_2O . Molecular H_2O and CO_2 do come off immature kerogen that is heated to $<600\ \text{K}$ (but at lower temperature in the presence of clay catalysts), so these would come off among the first components from fresh submicron grains released around $1\ \text{AU}$.

Why does comet Halley's $10\ \mu\text{m}$ "silicate" feature appear only inside $1.3\ \text{AU}$, becoming prominent at $1\ \text{AU}$? Such behaviour is known in other comets, and an evaporating organic coating has been proposed in explanation (Hanner, 1986). Do the grains have silicate cores (Mukai, 1986) or is $-\text{SiO}-$ a strongly bound structural component? As grains larger than a few microns are required, we have calculated the evaporation times for material with the refractive index of our organic mixture. Being cooler than

the submicron grains, time scales of $10^4\text{--}10^5\ \text{s}$ around $1\ \text{AU}$ are found for radii of $3\text{--}5\ \mu\text{m}$ if $E=20\ \text{kcal/mole}$. Since dielectric siliceous inclusions within a bulk of such weakly-bound organic material would not change significantly the absorption properties, such a composite would be suitable to explain the heliocentric distance dependence of the $10\ \mu\text{m}$ feature. An alternative would be the organo-siliceous mixture, used previously in grain modelling (Hoyle et al., 1985), which has a biological analogue in siliceous diatoms (Hoover et al., 1986).

5. Conclusion

The discovery that "CHON" grains (A'Hearn et al., 1986) composed of the light elements form an abundant component of comet Halley's dust (Kissel et al., 1986) has confirmed the earlier hypothesis that complex organics are a significant constituent, which partially evaporates from the grains and contributes to the gas coma. This is one part of the new cometary paradigm, that is superseding the icy conglomerate theory.

We have shown that micron and submicron grains composed of biological "kerogen" or synthetic "tholin" have appropriate radiation emission properties and suitable evaporation lifetimes. Specific studies of the gas emission properties of these classes of organics, and comparison with gas and dust distributions in the coma, to be detailed from the comet-probe and astronomical observations, should allow refinement of the new theory.

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