Laboratory-based sticking coefficients for ices on a variety of small-grain analogues

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Abundances and the partitioning between ices and gases in gas–grain chemistry are governed by adsorption and desorption on grains. Understanding of astrophysical observations relies on laboratory measurements of adsorption and desorption rates on dust grains analogues. On flat surfaces, gas adsorption probabilities (or sticking coefficients) have been found to be close to unity for most gases. Here we report a strong decrease in the sticking coefficients of H$_2$O and CO$_2$ on substrates more akin to cosmic dust, such as submicrometre-sized particles of carbon and olivine, bare or covered with ice. This effect results from the local curvature of the grains, and then extends to larger grains made of aggregated small particles, such as fluffy or porous dust in more evolved media (for example, circumstellar disks). The main astrophysical implication is that accretion rates of gases are reduced accordingly, slowing the growth of cosmic ices. Furthermore, volatile species that are not adsorbed on a grain at their freeze-out temperature will persist in the gas phase, which will impact gas–ice partitions. We also found that thermal desorption of H$_2$O is not modified by grain size, and thus the temperature of snowlines should be independent of the dust size distribution.

Grain size distributions, from a few nanometres to a micrometre, and compositions can be derived from the radiative properties of grains. There may be small grains in diffuse clouds; more evolved, aggregated grains in dense clouds; and bigger, porous and complex grains of micrometre- to centimetre-size in protoplanetary disks. They include various components: polycyclic aromatic hydrocarbons (PAH), particles of carbon (graphite, amorphous carbon, organic refractory material), silicate (olivine) and/or a mixture of both. Laboratory experiments have explored many aspects of gas–grain interactions, from the initial stage of surface atom additions to the more advanced stages of ice photochemistry. Yet in the few studies carried out on substrates close to dust in the interstellar medium (ISM), such as carbon nanoparticles, nano-structured silicates, bare or mixed with ice, effects on the sticking coefficient of gases were not investigated.

To this end, we have performed a set of X-ray photoelectron spectroscopy (XPS) experiments to study the adsorption of H$_2$O and CO$_2$—which are abundant constituents of astrophysical ices—on submicrometre-sized particles of carbon and olivine, the main minerals that make up cosmic dust. We also used other materials such as TiO$_2$ or Al$_2$O$_3$ to help unravel the role of chemical composition from size and morphology. Evolving with the gas exposure, the XPS signals of both the substrate and the adsorbate provide the coverage versus pressure; that is, the value of the sticking coefficient. We present here the sticking coefficients of H$_2$O and CO$_2$ on these grain analogues, bare or pre-covered with ice, to model the icy mantle of cosmic dust.

As references for adsorption on plane surfaces, we used a gold foil and crystals of highly oriented pyrolytic graphite (HOPG), olivine, TiO$_2$ and Al$_2$O$_3$. As dust grain analogues, we used both a graphite powder with a particle size of ∼2.5 µm (graphite microparticles) and powders of submicrometre-sized particles: olivine microparticles (∼0.30 µm), TiO$_2$ microparticles (∼0.24 µm), Al$_2$O$_3$ microparticles (∼0.24 µm) and an organic carbon soot Cl (∼0.115 µm) produced by a soot laboratory generator. To mimic the smallest dust grains, we used nanoparticles consisting either of a graphitic carbon soot C2 (∼0.025 µm) produced by the same generator, or a carbon soot produced by a candle CS (∼0.025 µm), and the same soot after an oxygen plasma oxidation, called Ox-CS. The grain size corresponds to the mode of the particle size distribution; that is, the most commonly found diameter. Figure 1 presents a selection of electron microscopy images of the substrates; more details of the samples are given in the Methods. The substrates are cooled but the gases are not, which raises the question of whether our results can be extrapolated to the ISM, which is discussed in Methods. Figure 2 presents the mean S of H$_2$O at 20 K on all substrates (Fig. 2a), and for CO$_2$ at 20 K on fewer substrates (Fig. 2b). Further experiments on H$_2$O at 80 K led to values close to those measured at 20 K (Supplementary Information 5). The sticking coefficients are graphically presented as a function of the substrate size (note the logarithmic scale). Values of S for increasing exposures are also listed in Table 1 (the mean values are plotted in Fig. 2). In Table 1, changes in S with exposure are related to the evolution of the growth mode with the coverage (Supplementary Information 2). In Fig. 2, the horizontal bar indicates the diameter range of ISM grains in dust models consistent with astronomical observations. Decreasing the grain size causes a decrease in S for H$_2$O and CO$_2$. The highest values of S are obtained for adsorption on plane surfaces. These values are relatively dispersed (0.40 < S < 1 for H$_2$O and 0.30 < S < 1 for CO$_2$), owing to different chemical structures leading to different densities of adsorption sites. It is worth mentioning that S differs from 1 on surfaces commonly used as grain analogues in laboratory astrochemistry: S≈0.65 on olivine and 0.40 on graphite HOPG, in agreement with refs. 17,18. The values of S are substantially lower on submicrometre-sized grains (C1, olivine microparticles, Al$_2$O$_3$ microparticles, TiO$_2$ microparticles) and micrometre-sized grains (graphite microparticles): 0.13 < S < 0.21 for H$_2$O and 0.06 < S < 0.25 for CO$_2$. There is also a certain dispersion of the values of S, showing that adsorption is still influenced by chemical composition, as on plane surfaces. Finally, the values of S are the lowest for adsorption onto CS, C2 and Ox-CS: 0.06 < S < 0.09 for H$_2$O, and 0.04 for CO$_2$. There is no difference after a strong oxidation of CS (S≈0.09 for CS and Ox-CS) whereas oxidized carbon functions should favour water adsorption by hydrogen bonding. This indicates that the chemical composition of such nano-sized particles has little or no influence on adsorption. Furthermore, it is important
to note that thermal desorption experiments of H2O indicate that the desorption temperature is not dependent on the substrate’s size (Supplementary Information 4).

To model condensation on the icy mantle of cosmic dust, we have adsorbed CO₂ on grains pre-covered with a thin layer of ice, preserving the morphology of the underlying grain (Methods). As the degree of porosity of cosmic ices is unknown, two kinds of ice were deposited: a porous amorphous ice (p-ASW, for porous amorphous solid water) and a non-porous amorphous ice (ASW). It can thus be determined whether adsorption depends on the ice porosity. Figure 3 presents S for CO₂ at 20 K on a selection of bare and icy substrates; for the sake of clarity, only the TiO₂ plane surface is presented (further substrates are listed in Table 1). Ice substantially increases the sticking of CO₂, from S = 0.53 on bare TiO₂ to S = 0.74 on p-ASW/TiO₂, and S = 0.93 on ASW/TiO₂, a value in good agreement with He et al.15. This is also consistent with the work of Noble et al.19, who reported that CO₂ sticks better to non-porous ice. Adsorption of CO₂ is also enhanced by ice on submicrometre particles, from S = 0.25 and 0.15 (TiO₂ microparticles and Al₂O₃ microparticles) to 0.34 and 0.28 (p-ASW/TiO₂ microparticles, p-ASW/Al₂O₃ microparticles) and to 0.49 and 0.45 (ASW/TiO₂ microparticles, ASW/Al₂O₃ microparticles). However, the values of S are not as high as those measured on ice surfaces deposited on flat TiO₂, indicating that the size of the icy grain still influences adsorption. Finally, S values for CO₂ on bare and ice-covered carbon nanoparticles (CS) are extremely low and identical (S = 0.04 in both cases). This adds to the evidence that surface chemistry plays very little role in the case of nanoparticles.

In the Earth’s atmosphere, it is well established that the ability of solid aerosols to condense water depends on their chemistry16, but the size of the particle also matters. Increasing curvature weakens the attractive forces between the adsorbed water molecules, and the equilibrium vapour pressure of a gas is always higher at convex surfaces than at flat surfaces; this reduces or hinders the condensation of water onto small aerosol particles. This effect, classically described by Kelvin’s equation, becomes crucial at nanometre scales21. It causes the poor cloud-nucleating ability of small atmospheric aerosols, and is independent of the composition for micrometre-sized particles22. Regarding ice, molecular simulations indicate that heterogeneous nucleation is indeed less efficient at surfaces whose convex curvature is not negligible at the molecular scale23. For instance, CO₂ interacts more strongly in surface valleys than on hills of the amorphous ice surface24. Simulations also show that on rough ice nanoparticles, molecules physisorb in crevices and indentations rather than on bumps and protrusions25. For an amorphous ice cluster, quantum-mechanical simulations show that the low-lying potential minima are located in cavities26. This is the microscopic rendering of Kelvin’s equation, which also states that concave surfaces facilitate nucleation by increasing the number of van der Waals interactions with the wall of the confined space, a phenomenon called capillary condensation. On nanoparticles such as soot, morphology is dominated by highly convex surfaces where gases will then condense less efficiently. On such small particles, a modification of the surface chemistry by oxidation or ice coating does not make any difference because the grain morphology remains unchanged. For larger, submicrometre-sized particles (TiO₂ microparticles, Al₂O₃ microparticles, olivine microparticles), strongly convex areas are necessarily fewer; their morphology is mostly made up of slightly curved surfaces, and adsorption is

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**Fig. 1 | Selected electron microscopy images of the substrates.** a-e, Scanning electron microscopy images of the dust grain analogues: graphite microparticles (a); olivine microparticles (b); TiO₂ microparticles (c); Al₂O₃ microparticles (d); and candle soot nanoparticles (e). f-j, Transmission electron microscopy images of TiO₂ microparticles (f); Al₂O₃ microparticles (g); candle soot (h); carbon soot C1 (i); and carbon soot C2 (j).
mainly controlled by chemistry. Finally, as $S$ is governed by the local features of the grain, it is not determined at the grain's overall scale, but rather at the scale of the particles that comprise it. Our results therefore apply to various cosmic environments, whether dust consists of single grains (diffuse ISM) or small grains coagulated into larger aggregates (dense clouds, protoplanetary disks).

One main general astrophysical implication is that accretion rates of gases on grains will be reduced, and cosmic ices will grow at slower rates. Furthermore, as not all species adsorb when hitting a grain at their freezing temperature, they can remain in the gas phase instead of forming the expected ices. This must be considered to model the partition between gases and ices in cold regions, along with possible non-thermal desorption processes that can reallocate condensed molecules into the gas phase. Until experimental values are determined, some recommendations for the values of $S$ can be provided to reassess astrochemical models of interstellar and circumstellar environments. For highly curved, small grains ($\leq 0.025\mu$m in diameter), or bigger aggregates made of such grains,
For the pre-stellar core L1689B, this halves the CO freeze-out rate and leads to timescales closer to the nominal free-fall time, in better agreement with the observed abundance\textsuperscript{29}. Finally, with water playing a central role in planet-forming regions (disk composition, coagulation)\textsuperscript{30}, assessing realistic conditions for direct condensation and sublimation of H\textsubscript{2}O on small grains is important for interpreting water abundances. Our study shows that although grain size impacts the condensation process, it has no effect on the desorption temperature (Supplementary Information 4). An important consequence is that the isotherm of the water snowline in planetary disks will not be dependent on the size distribution of the grains.

**Methods**

**Samples.** As plane, centimetre-sized substrates, we used (1) a gold foil of 10 × 10 × 0.5 mm\textsuperscript{3} (99.95%, Alfa Aesar); (2) a highly oriented pyrolytic graphite (HOPG, Alfa Aesar); (3) a crystal of TiO\textsubscript{2} (rutile) of 10 × 10 × 2 mm\textsuperscript{3}, (4) a crystal of Al\textsubscript{2}O\textsubscript{3} (corindon) of 10 × 10 × 5 mm\textsuperscript{3} and (5) a crystal of period of 10 × 10 × 1 mm\textsuperscript{3}. Peridot is a gem variety of a natural Mg-rich San Carlos olivine of composition (Mg\textsubscript{95}Fe\textsubscript{4}Si\textsubscript{24})\textsubscript{24}O\textsubscript{36} as determined by XPS. Samples (3)–(5) were provided by the collection of mineralogy of Aix-Marseille University, and cut and polished mechanically. Powders with particles of various sizes were used as grain analogues: (6) graphite microparticles made graphite grains of ~5.0 ± 2.4 μm in diameter, sometimes aggregated in bigger grains of 20–30 μm; (7) an olivine microparticle powder obtained after fine grinding of an olivine crystal, leading to grains of 0.30 ± 0.14 μm in diameter, mixed with some bigger grains of a few micrometres; (8) TiO\textsubscript{2} microparticles (anatase) made of slightly faceted round grains of 0.24 ± 0.05 μm in diameter, obtained after drying of an ultrapure alumina suspension (99.98%, Pesti). We also used carbon soot: (10) a candle soot (CS) emitted from a pure paraffin candle, made of fractal aggregates of graphitic carbon nanoparticles (~90%), the rest being aliphatic and aromatic organic hydrocarbons) with a diameter of 0.025 μm (that is 25 ± 15 nm), exhibiting an imperfect graphitic structure (turbostatic disorder); (11, 12) two carbon soot samples (C1 and C2) generated by a MiniCAST generator under two different combustion conditions\textsuperscript{31}. C1 is an organic carbon soot (called CAST3 in ref. \textsuperscript{31}), containing 38% aliphatic and aromatic hydrocarbons and 62% graphic carbon), consisting of almost indistinct carbon nanoparticles forming aggregates of 0.115 ± 0.02 μm in diameter. C2 is a graphitic carbon soot (94% graphic carbon) made of fractal aggregates of distinct carbon nanoparticles of 0.025 μm (that is 25 ± 15 nm) with a turbostratic disordered structure (called CAST1 in ref. \textsuperscript{31}). XPS allowed surface oxidation ratios [O]/[C] of 9%, 4% and 3%
to be determined for C1, C2 and Cs, respectively. No other atomic species, such as nitrogen, were detected. The crystal structures of all samples were obtained by X-ray diffraction carried out with a PANalytical X’Pert diffractometer (not presented). The grains were also studied with electron microscopy, using a Jeol JSM-6340F microscope in scanning mode (for scanning electron microscopy) and a Jeol JEM-2010 microscope in transmission mode (for transmission electron microscopy). The particle size distributions were determined from electron microscopy images using the open source image processing software Imagej v1.53 (https://imagej.nih.gov/ij). Scanning or transmission electron microscopy images displaying a large number of particles were loaded in the software. The spatial scale was then calibrated using the scale provided by the microscopes (horizontal bars on the images). The area selection tool of the software allows areas for several hundred particles to be manually defined. For each selected area, the software provides the area, the perimeter and the Feret’s equivalent diameter, taken as the particle sizes. Statistics including the mean size and standard deviation were then calculated.

The substrates were also pre-covered with two kinds of ice. In a first set of experiments, ice was condensed at 20 K on to form a microporous amorphous ice film (p-ASW). In a second set of experiments, ice was condensed at 20 K, then annealed at 110 K to form a non-porous amorphous ice film (ASW)32,33. The ice layers and the condensation method did not lead to any substantial increase in the particle’s size. We found that the XPS signal from the substrates becomes negligible, indicating that they are fully covered by the ice layers.

XPS. Ethanol solutions of the powders were drop-cast on the XPS sample holders and dried to form a thin film. The MiniCAST samples were deposited by thermomigration onto a silicon wafer (LQG optics), like the flat references (graphite, TiO2, Al2O3, and olivine), were glued to the sample holders using a thermally conductive silver paint. Candle soot was directly deposited on a sample holder by passing it through the flame. Another sample of candle soot (Ox-CS) was oxidized under ultrahigh vacuum with an O3 plasma from a microwave source (Tectora GmbH). XPS showed that the oxygen concentration at the surface increased seven-fold, from 3% to 21%, mainly owing to ketone and carbonyl functionalities. Graphite HOPG was cleaved before its introduction under ultrahigh vacuum, whereas TiO2, Al2O3, and olivine crystals were cleaned by sputtering Ar+ at 3 kV for 5 min under ultrahigh vacuum at room temperature. The TiO2 microparticle, Al2O3 microparticle and olivine microparticle samples were sputtered at the same time. All samples submitted to sputtering should be amorphous over a depth of few nanometres from the surface. The substrates were cooled down overnight to 20 ± 1 K, and the proper thermalization of their surface was systematically verified by measuring the desorption temperature of an ice layer with XPS (Supplementary Information 4). The bare substrates were exposed to H2O or CO2 via background dosing (Supplementary Information 1).

As noted previously, the gases are not cooled, which raises the question of whether our results can be extrapolated to the ISM. For H2, it is known that the gas temperature substantially impacts the sticking coefficients, to a lesser extent for D2, as surface thermalization is more efficient for higher masses34. Gas temperature is then required to obtain realistic experimental values for the sticking coefficients of H2, D2, H and D. For heavier species such as H2O or CO2, the ISM temperatures are in practice impossible to obtain. However, their high mass should favour their thermalization35. As it is extremely difficult to experimentally investigate this effect it has rarely been studied, but was confirmed by an experiment that showed that the thermal energy of CO from 30 K to 600 K is totally absorbed by a tungsten surface maintained at 20 K (ref. 36). This also agrees with calculations predicting the full thermalization of CO at 300 K landing on water ice at 40 K (ref. 37). For thermal CO as well as for Ar+38, the number of collisions is two to three before sticking, slightly increasing with the incident energy of the gas. For hydrogen at 300 K on crystalline ice at 10 K, sticking occurs a few picoseconds after the collision, that is ~0.5–0.6 nm (ref. 39), in agreement with the calculations of Buch et al. for hydrogen at 200 K on 96 K ice cluster40. Thus, gas species, even the lightest, do not travel large distances after collision on a surface held at low temperature. Further experimental evidence is the fact that background deposition of H2O at 300 K on cold substrates held below 90 K results in the growth of p-ASW films at the atomic scale, a consequence of the limited molecular mobility. Therefore, if the grain size is larger than the few interatomic distances necessary for a complete thermalization of the gas during the sticking trajectory, there is no impact of the size on the sticking probability due to this dynamic aspect, provided the surface is cold and the gas is at room temperature and not too hot. Then, we assume that the sticking coefficients of heavy species at ambient gas temperature might not be very different from those at the colder gas temperatures of the ISM.

The XPS data were recorded using a Resolve 120 hemispherical electron analyser (PSP Vacuum), and an unmonochromatized X-ray source (Mg Kα at 1.253.6 eV, PSP Vacuum) operated at 100 W at an angle of 15° with respect to the analyser axis, and a detection angle of 55° between the surface normal and the analyser axis. At this angle, the magic number effect, the angle influence of roughness on the signal intensity is null or very limited (Supplementary Information 2). During the experiments, neither the substrates nor the condensed H2O or CO2 layers showed any detectable radiation damage, such as chemical changes. We observed no temperature increase of the substrates due to the proximity of the X-ray tube owing to the low power used, the large source–sample distance (2 cm) and the adsorption by the crystal of the thermal radiation emitted by the X-ray tube. The S value on each substrate was graphically determined from the XPS intensities (Supplementary Information 2) by finding the exposure necessary to obtain the same XPS intensity as on gold, where S = 1 by calibration. Uncertainty in the values of S was estimated to be between ±5% and ±10% of the standard deviation, considering the accuracy of the exposure, error due to roughness and the accuracy of the XPS data analysis (±5%) (Supplementary Information 3).

### Data availability
A text version of Table 1 is available at https://figshare.com/articles/dataset/112724840. All of the datasets generated and analysed during the current study are available from the corresponding author on reasonable request.

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Author contributions

C.L. and P.P. conceived, performed, analysed and interpreted the XPS experiments. O.G. provided and prepared most of the samples, and carried out the X-ray diffraction experiments. D.F. and O.G. performed and analysed the scanning and transmission electron microscopy images. P.P. wrote the manuscript. All authors contributed ideas to this Letter.

Competing interests

The authors declare no competing interests.

Additional information

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