JOURNAL OF SPACECRAFT AND ROCKETS
Vol. 35, No. 1, January–February 1998

Low-Earth-Orbit Atomic Oxygen Erosion of Polymer Surfaces

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Spacecraft in low Earth orbit (250–700 km altitude) encounter a residual atmosphere that is 90% atomic oxygen in its ground electronic state produced by the solar uv photodissociation of molecular oxygen. At a relative velocity of 8 km s^{-1}, an observer on the spacecraft at an altitude of 300 km experiences an average flux of 10^{15} oxygen atoms cm^{-2} s^{-1} approaching in the ram direction with a relative kinetic energy of 5 eV. When exposed to this flux, vitreous carbon, polymers, and many polymer-based composite materials suffer surface erosion. Upon microscopic examination, the roughened surface is seen to consist of troughs and points that have a shag-carpet appearance. We show that, when a target atom has weak bonds tangential to the surface, the impact energy of the incident O atom rapidly dissipates into the bulk of the solid, and the target atom cannot react. This produces a point. By contrast, when tangential bonds are strong, the reverse occurs, and a trough results. The anisotropic bonding on which this model depends can be found in both polymers and vitreous materials.

Nomenclature

E = energy of incident oxygen atom, J
E_A = reaction activation energy, J
E_1 = energy binding target atom to the surface of the solid, J
$F(t)$ = time-dependent force acting between oxygen atom and target atom, N
H = excitation energy in the bond between the target atom and the surface
I = collision impulse, Ns
$J_n(x)$ = ordinary Bessel function of order n; argument is x
k = spring constant, J/m^2
M = mass of an oxygen atom, kg
m = mass of an atom in the solid, kg
m_r = reduced mass, $mM/(M+m)$, kg
p = empirical parameter in Johnston estimate of E_A
Q = heat of reaction, J
R_j = displacement of the position of jth (where j is $1, 2, \ldots$) atom in the solid, m
R_o = position of the oxygen atom, m
t = time, s
v = initial speed of the incident oxygen atom
ΔE = energy transferred from oxygen atom to target atom, J
$\delta(t)$ = Dirac delta function, s^{-1}; argument is time t
λ = integration variable
ν = bond vibration frequency, s^{-1}
σ = Laplace transform variable, s^{-1}
τ = dimensionless time, $2\omega t$
ω = bond angular frequency of vibrations, s^{-1}

Introduction

SPACECRAFT in low Earth orbit (250–700 km altitude) encounter a residual atmosphere that is 90% atomic oxygen in its ^3P ground state produced by the solar uv photodissociation of O_2 (Refs. 1–4). The O-atom density depends on altitude, latitude, and time of day, as well as the sunspot cycle.[2,5] Under average conditions at an altitude of 300 km and a latitude of 30 deg, however, the atomic oxygen has a density of about 10^9 cm^{-3} (Refs. 1–3). The most probable Maxwell velocity of these atoms is 0.4–0.5 km s^{-1}, which is small when compared with the orbital velocity of 8 km s^{-1} of a spacecraft at this altitude.[3,6] Thus, to a good approximation, the relative velocity is 8 km s^{-1}, and an observer on the spacecraft experiences an average flux of 10^{15} O atoms cm^{-2} s^{-1} approaching in the ram direction with a relative kinetic energy of 5 eV (Refs. 1–3).

When exposed to this flux, graphite, vitreous carbon, polymers, and many polymer-based composite materials suffer erosion.[3,5,7–14] From vitreous carbon, for example, 0.02 C atoms are removed per incident O atom.[8] Kapton,[9,14] the polyimide polymer used in thermal control blankets, as well as polyethylene,[2] Mylar®,[14] and graphite,[8] all readily erode. In the case of polymers, the erosion rate is unaffected by deuteration.[2] The presence of aromatic rings and carbon-carbon double bonds, however, tends to reduce the reactivity.[15] Materials that exhibit similar erosion rates when exposed to 5-eV oxygen atoms sometimes display varying rates of erosion when exposed to thermal (0.04 eV) oxygen atoms.[15] A common and remarkable observation is that, when exposed to 5-eV atomic oxygen, polymers and vitreous carbon develop a highly roughened surface consisting of troughs and points oriented in the ram direction. This roughened surface is often described as having a shag-carpet appearance.[3,5,7,9–13] Because the bond energies are also of the order of 5 eV, the structure cannot be due to sputtering.[1,2]

We show that, when a 5-eV O atom collides with a surface, diffraction effects are unimportant. Whether a target atom in the surface becomes chemically activated[16] in a collision depends on the directional character of its bonds to other atoms. We suggest that this directional character is responsible for the shag-carpet appearance of the eroded surface of polymers.

Analysis

The atoms in a solid held at a temperature of less than 1000 K have kinetic energies of no more than a few hundred millielectron volts. Hence, in a collision with an oxygen atom moving with a relative kinetic energy of 5 eV, the atoms in the solid can be assumed to be at rest at their equilibrium positions. Moreover, quantum diffraction effects may be ignored in the scattering of the O atom because its DeBroglie wavelength (3 pm) at 5 eV is much less than the typical atomic spacing (100 pm) in a solid.[17] Hence, the O atom can be considered to collide with a single atom in the surface. Behind this atom we assume that there is an effectively infinite chain of other atoms extending into the bulk of the solid.[18] An O atom traveling with velocity v in the direction of such a chain is shown in Fig. 1. The first atom represents the surface of the solid, whereas atoms 2, 3, etc., represent the bulk. The coordinates R_o, R_1, and R_2 locate the atoms with respect to the various rest positions. We shall take the chain to be one dimensional and ignore any tangential bonding of an atom to any other atoms in the solid. We let M be the mass of the O atom and m be the mass of an atom in the solid, which has for convenience been taken to be monatomic. The displacement of the O atom from the surface of the solid is R_o, whereas the displacements of the atoms

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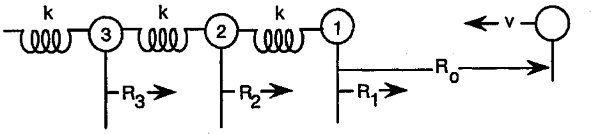

Fig. 1 O atom traveling with a speed *v* toward the first atom in a semi-infinite chain of atoms coupled by springs with spring constant *k*.

in the solid from their equilibrium positions are R_j ($j = 1, 2, \ldots$). The atom in the surface ($j = 1$) and the O atom exert a force F on one another. The Newtonian equations of motion for the system are

$$M\frac{\mathrm{d}^2R_o}{\mathrm{d}t^2} = F \tag{1}$$

$$m\frac{\mathrm{d}^2R_1}{\mathrm{d}t^2} = -F - k(R_1 - R_2) \tag{2}$$

$$m\frac{\mathrm{d}^2R_j}{\mathrm{d}t^2} = -k(R_j - R_{j-1}) - k(R_j - R_{j+1}), \qquad j = 2, 3, \ldots \tag{3}$$

where t is the time and k is the spring constant for each bond between atoms in the solid. Equations (1–3) are to be solved subject to the initial conditions

$$R_j(0^-) = 0, \qquad j = 0, 1, \ldots \tag{4}$$

$$\left(\frac{\mathrm{d}R_o}{\mathrm{d}t}\right)_{0^-} = -v \tag{5}$$

$$\left(\frac{\mathrm{d}R_j}{\mathrm{d}t}\right)_{0^-} = 0, \qquad j = 1, 2, \ldots \tag{6}$$

where $t = 0^-$ is the time an instant before collision. In general, F is derivable from an interatomic potential energy, which at long range is weakly attractive but which at short range is strongly repulsive. The strength of the attractive part, which serves to accelerate the O atom toward the surface, is usually much less than 5 eV and can be ignored.[19] The repulsive part, by contrast, increases rapidly as the separation between the two atoms diminishes.[19] For this reason, at 5-eV relative energy, the collision becomes similar to that occurring between hard spheres, where the force can be represented by

$$F = I\delta(t) \tag{7}$$

where I is the impulse delivered in the collision.

Upon substitution of Eq. (7) into Eq. (1), integration, and use of Eq. (5), one obtains for the O-atom postcollision velocity

$$\frac{\mathrm{d}R_o}{\mathrm{d}t} = -v + \frac{I}{M} \tag{8}$$

Equation (8) holds for $t \geq 0^+$, where $t = 0^+$ is a time an instant after collision. By use of a generating function method,[20] Zwanzig[18] showed how the remaining equations of motion, Eqs. (2) and (3), can be uncoupled and integrated. His result for the relative acceleration, $\mathrm{d}^2(R_o - R_1)/\mathrm{d}t^2$, can be written

$$\frac{\mathrm{d}^2(R_o - R_1)}{\mathrm{d}t^2} = \frac{F(t)}{m_r} - \frac{2}{m}\int_0^t \frac{J_2[2\omega(t-\lambda)]}{(t-\lambda)}F(\lambda)\,\mathrm{d}\lambda \tag{9}$$

where $\omega = (k/m)^{1/2}$ is the vibrational angular frequency of a bond,

$$m_r = mM/(m + M) \tag{10}$$

is the reduced mass, and $J_2(x)$ is the ordinary Bessel function of order two. After taking into account Eqs. (5) and (6), substituting Eq. (7) into the right-hand side of Eq. (9), and integrating, we find

$$\frac{\mathrm{d}(R_o - R_1)}{\mathrm{d}t} = -v + \frac{I}{M} + \left(\frac{2I}{m}\right)\frac{J_1(\tau)}{\tau} \tag{11}$$

where $\tau = 2\omega t$ and where we have used

$$\int_0^\tau \frac{J_2(\lambda)}{\lambda}\,\mathrm{d}\lambda = \frac{1}{2} - \frac{J_1(\tau)}{\tau} \tag{12}$$

which can be derived from a general Bessel function identity [see Eq. (11.33.11) of Ref. 21]. Combining Eqs. (8) and (11), we obtain

$$\frac{\mathrm{d}R_1}{\mathrm{d}t} = -\left(\frac{2I}{m}\right)\frac{J_1(\tau)}{\tau} \tag{13}$$

for the after-collision velocity of the first atom in the chain.

Letting σ be the Laplace transform variable, the transform of $J_n(\tau)/\tau$ is $\{n[\sigma + (\sigma^2 + 1)^{1/2}]^n\}^{-1}$ [see Eq. (10.16) of Ref. 22]. Using this transform and the initial value theorem,[22] one may show that $J_1(\tau)/\tau$ equals $\frac{1}{2}$ at $\tau = 0^+$, which corresponds to a time an instant after collision. In this limit, Eq. (13) becomes

$$\left(\frac{\mathrm{d}R_1}{\mathrm{d}t}\right)_{0^+} = -\frac{I}{m} \tag{14}$$

Because at $\tau = 0^+$, none of the springs are compressed and only the end atom ($j = 1$) in the chain has a nonzero velocity, the momentum transferred in the collision is $-I$. By substituting Eqs. (5), (8), and (14) into

$$\frac{M}{2}\left(\frac{\mathrm{d}R_o}{\mathrm{d}t}\right)^2_{0^-} = \frac{M}{2}\left(\frac{\mathrm{d}R_o}{\mathrm{d}t}\right)^2_{0^+} + \frac{m}{2}\left(\frac{\mathrm{d}R_1}{\mathrm{d}t}\right)^2_{0^+} \tag{15}$$

which is the law of conservation of energy, we find that

$$I = 2m_r v \tag{16}$$

After using Eqs. (10) and (16), Eq. (8) becomes

$$\frac{\mathrm{d}R_o}{\mathrm{d}t} = -\frac{(M - m)}{(M + m)}v \tag{17}$$

Equation (17) shows that the sign of the postcollision velocity of the O atom depends on the sizes of m and M. The energy transferred ΔE in the collision is the second term on the right in Eq. (15). After employing Eqs. (14) and (16), this term becomes

$$\Delta E = \frac{4Mm}{(M + m)^2}E \tag{18}$$

where $E = \frac{1}{2}Mv^2$ is the energy of the O atom before collision.

Using the initial conditions expressed by Eqs. (4) and (6), Zwánzig[18] showed that

$$R_1 - R_2 = -\frac{2}{k}\int_0^t \frac{J_2[2\omega(t-\lambda)]}{(t-\lambda)}F(\lambda)\,\mathrm{d}\lambda \tag{19}$$

After substitution of Eq. (7), Eq. (19) becomes

$$R_1 - R_2 = -\left(\frac{4I\omega}{k}\right)\frac{J_2(\tau)}{\tau} \tag{20}$$

The excitation energy H stored in the first bond in the solid is given by

$$H = \frac{m}{2}\left(\frac{\mathrm{d}R_1}{\mathrm{d}t}\right)^2 + \frac{k}{2}(R_1 - R_2)^2 \tag{21}$$

Upon substitution of Eqs. (13), (18), and (20), Eq. (21) takes the form

$$H = \left\{4\left[\frac{J_1(\tau)}{\tau}\right]^2 + 16\left[\frac{J_2(\tau)}{\tau}\right]^2\right\}\Delta E \tag{22}$$

According to Eq. (22), H is proportional to ΔE. The proportionality coefficient in brackets, however, is a decreasing function of time because energy is leaking out of the bond and propagating into the bulk of the solid.

Discussion

According to Eq. (18), the maximum energy transferred to the solid occurs when the mass M of the O atom is the same as the mass m of the target atom. Equation (17) shows that, when $M = m$, the velocity of the O atom after the collision is zero. By contrast, when $M < m$, the postcollision velocity of the O atom causes it to recoil in a direction away from the surface of the solid, whereas when $M > m$, the postcollision velocity of the O atom carries it on into the solid. By continuing into the solid, the O atom presumably becomes thermalized and is ultimately adsorbed.

Although initially at rest, the target atom ($j = 1$) recoils into the solid after the collision. Its speed is given by Eq. (13), which has been plotted in Fig. 2. The speed on the ordinate is in the units of $-I/m$. The abscissa is $\tau = 2\omega t$. In Fig. 3, we show a plot of $H/\Delta E$ as computed using Eq. (22). H has been divided by the energy transfer $\Delta E = 4MmE/(M + m)^2$, where E is the kinetic energy of the O atom before collision. At the instant $\tau = 0^+$, just after collision, the energy of the target atom is entirely kinetic. Subsequently, it becomes partly kinetic and partly potential and remains that way until $\tau = 3.832$, when, as shown in Fig. 2, the target atom comes to rest at its first turning point. By this time, as is apparent from Fig. 3, $H/\Delta E$ has decreased to less than 20% of its initial value. The missing energy has propagated into the bulk of the solid. During this compression phase, when the bulk of the energy brought to the collision gets dissipated, the harmonic force specified in Eqs. (2) and (3) is strongly repulsive and mimics in this regard the actual interatomic potential in the solid.

The actual timescale over which the missing energy propagates depends on the vibrational frequency of the chemical bond between the atoms in the solid, which in the Zwanzig[18] model is represented by a spring. The vibrational frequencies of the covalent chemical bonds in ploymers lie generally within the range from $\nu = 2.5 \times 10^{13}$ to 13×10^{13} s^{-1} (Ref. 23). Because $t = \tau/4\pi\nu$, then depending on ν, the dimensionless time $\tau = 3.832$ required to reach the first turning point corresponds to a dimensioned time lying between 2.3 and 12 fs. The short timescale for the dissipation of energy is a special property of the Zwanzig[18] model in which bonds linking atoms in the plane parallel to the surface are ignored.[24,25] When these tangential bonds are taken into account, the target atom retains more of the initial energy transferred to it in the collision.

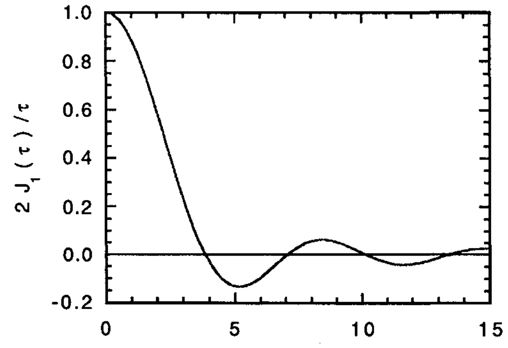

Fig. 2 Speed of the first target atom ($j = 1$) in the solid as a function of dimensionless time τ.

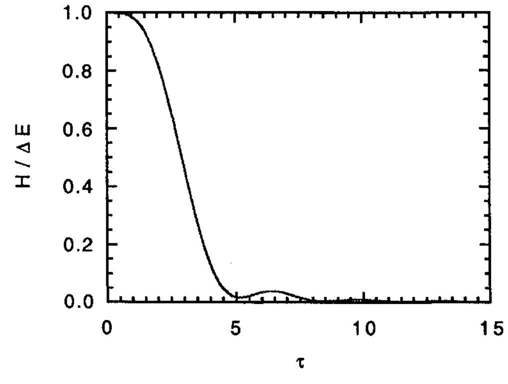

Fig. 3 Total energy H stored in the first target atom ($j = 1$) as a function of dimensionless time τ.

Surface erosion is due presumably to a chemical reaction between an O atom and an atom in the solid. If the target atom is to form a chemical bond with the O atom, it must surmount the saddle point of the potential energy barrier holding it to the solid. In the absence of the reaction, this saddle point would constitute a turning point in its classical motion.[26] Presumably, in the Zwanzig[18] model, this turning point occurs at $\tau = 7.02$ in Fig. 2, where the target atom is moving away from the surface. To react at this point, the target atom should have an energy at least equal to the reaction activation energy E_A. In the absence of an ab initio quantum mechanical calculation,[27] we can approximate E_A using Johnston's bond energy-bond order empirical relation[28]

$$E_A = E_1\left(1 - \left\{1 + [1 + Q/(E_1 - Q)]^{1/(p-1)}\right\}^{1-p}\right) \tag{23}$$

where $p > 1$ is an empirical parameter that is adjusted so as to fit a series of reactions of the same type, e.g., atomic abstractions. Evaluation of Eq. (23) shows that only in the case of a highly exothermic ($-Q \gg E_1$) reaction does E_A approach zero. This occurs when the bond broken is much weaker than the bond formed. According to Fig. 2, by $\tau = 7.02$, nearly all of the energy in the target atom has been dissipated, implying that only strongly exothermic reactions can be stimulated by direct collision between the O atom and an atom at the end of a linear chain.

The bonding between atoms in a real polymer chain is highly directional. When a number of chains are condensed to form a solid, there are regions of random chain orientation interspersed among regions of parallel fibrils and pleated sheets.[29] When an O atom strikes such a polymer surface, it may collide with a surface atom that is a part of any of these regions. Applying the Zwanzig[18] model, we suppose that, if the target atom is in a region that is only weakly bonded tangentially, its collisional activation will dissipate rapidly into the bulk of the solid, and the target atom will not react. If, however, the tangential bonding is significant, the reverse will occur. Thus, we suggest that a point on the shag-carpet eroded surface topology corresponds to an inert region characterized by weak tangential bonding, whereas a trough corresponds to an active region characterized by strong tangential bonding. This conclusion is supported by the observation that, in a solid polymer, it is possible to find bonds pointed in any direction desired,[29] as well as the fact that, regardless of the orientation of the surface, the shag-carpet points seen in O-atom eroded surfaces always point in the ram direction.[12] As a test of this concept, a careful microprobe analysis of the surface structure of a polymer could be correlated with the shag-carpet pattern produced by its subsequent exposure to atomic oxygen. In this respect, it would be interesting to compare low-density polyethylene with high-density polyethylene. The latter has a larger average molecular weight, fewer chain ends, and more crystallinity.

Quantum mechanical zero-point vibrational energy is present in chemical bonds no matter what the temperature. The zero-point energy of covalently bonded hydrogen atoms in polymers ranges from 80 meV (C–H bend) to 230 meV (O–H stretch).[23] Replacement of ^{1}H by ^{2}H reduces these to 57 and 163 meV, respectively. Because none of these energies is significant when compared with ΔE, which can be as large as 5 eV, it is clear why the polymer erosion observed in low Earth orbit is insensitive to deuteration.[15] By contrast, the rate of erosion caused by thermal (40 meV) O atoms depends for its existence on the small population of bonds that are already thermally excited to high vibrational states and that can be carried over the saddle point of the potential energy barrier by the addition of only 40 meV (Ref. 30). As these populations depend on the vibrational frequency, the reaction rate can be expected to be sensitive to deuteration.[15]

Finally, the dissociation energies of multiple bonds having both sigma and pi character, with or without resonance, are generally larger than those for single bonds. The dissociation energy presumably accounts for the reported relative insensitivity to 5-eV O-atom erosion of polymer regions containing aromatic rings and multiple bonds.[15]

The bond orientation mechanism for shag-carpet erosion is to be compared with the Monte Carlo erosion pit model proposed by de Groh and Banks.[31] They have pointed out that, once an erosion pit has been established, subsequent O atoms entering the pit can undergo multiple collisions with the walls and become trapped in the re-entrant geometry because of the small exit solid angle presented by the opening. Assuming some elastic scattering, an O atom undergoing more than one collision presumably has a greater opportunity to make a reactive collision with the solid. Consequently, the pit expands at the expense of the surrounding uneroded (point) material. Where the pit starts is treated by Monte Carlo methods, as are the subsequent O-atom collisions within the pit.

There is as yet no experimental evidence that favors our bond orientation mechanism over the Monte Carlo mechanism. Logically, it would seem that both could operate together. Indeed, it may be the presence of tangential bonding in an area, rather than pure chance itself, that inclines that area to erode as a pit. Subsequent erosion is enhanced by the re-entrant geometry.

Acknowledgments

This research was sponsored by the U.S. Air Force Office of Scientific Research through a contract with the U.S. Air Force Phillips Laboratory, Kirtland Air Force Base, New Mexico. The author would like to thank Joy Johnson for helpful conversations concerning classical collision dynamics and Roger Gerzeski for discussions of polymer structure.

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