FORMATION OF INTERSTELLAR ICES BEHIND SHOCK WAVES

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ABSTRACT

We have used a coupled dynamical and chemical model to examine the chemical changes induced by the passage of an interstellar shock in well-shielded regions. Using this model, we demonstrate that the formation of H2O in a shock will be followed in the postshock phase by depletion of the water molecules onto the grain surfaces. To attempt to discriminate between the creation of ices behind shocks and their production by means of grain surface chemistry, we examine the deuterium chemistry of water before, during, and after a shock. We show that chemical evolution in the postshock gas can account for both the deuterium fractionation and the abundance of CO2 relative to H2O observed in interstellar and cometary ices. Given the pervasiveness of shocks and turbulent motions within molecular clouds, the model presented here offers an alternative theory to grain surface chemistry for the creation of ices in the interstellar medium, ices that may ultimately be incorporated into comets.

Subject headings: comets: general — ISM: abundances — ISM: clouds — ISM: molecules — molecular processes — shock waves

1. INTRODUCTION

There is mounting evidence that some interstellar material survives inside objects in the solar system. Such evidence is found in the compositional similarity between cometary ices and interplanetary grains with those found in the interstellar medium (Mumma 1997). That cometary ices and organics may have supplied the molecular seeds for prebiotic life on the earth, or even the oceans, leads to the intriguing possibility that the molecules essential for life might have their origin in interstellar space (e.g., Chyba & Sagan 1997). As such, it is important to gain an understanding of the processes that lead to the formation of ices in the interstellar medium (ISM).

By far, the most abundant constituent of ices in the interstellar medium, and also in comets, is H2O. Because the low-temperature ion-molecule chemistry that is active in dense regions of the ISM is incapable of reproducing the observed H2O ice observations (Jones & Williams 1984), it has been proposed that water ice is formed through the hydrogenation of oxygen atoms on the surfaces of cold (K;~ 200 K for shock velocities v; between 10 and 30 km s−1, respectively (Kaufman & Neufeld 1996). The high-temperature chemistry in the shock is allowed to evolve for a cooling timescale (t~100 yr for a 30 km s−1 shock; see BMN), whereupon the third stage commences with a return to quiescent temperature conditions.

We use the three-stage model described in BMN to examine the evolution of chemical abundances in preshocked (stage 1), shocked (stage 2), and postshock gas (stage 3). The chemical evolution in each stage is treated independently, except that the final composition of the preceding stage is used as the initial composition of the next. The first stage is assumed to be initially atomic in composition and has physical conditions appropriate for quiescent gas in molecular cores (T = 10–30 K; nH2 = 1015–1016 cm−3). The stage 1 preshock chemistry evolves until t~106 yr, when the gas is assumed to be shocked. The stage 2 shock dynamics are modeled as an increase in the gas temperature, which scales with the shock velocity—peak gas temperatures range between 400 and ~2000 K for shock velocities v; between 10 and 30 km s−1, respectively (Kaufman & Neufeld 1996). The high-temperature chemistry in the shock is allowed to evolve for a cooling timescale (~100 yr for a 30 km s−1 shock; see BMN), whereupon the third stage commences with a return to quiescent temperature conditions.

We use the University of Manchester Institute of Science and Technology (UMIST) RATE95 database (Millar, Farquhar, & Willacy 1997) for all computations of nondeuterated molecules. For the deuterium chemistry, we have created a smaller network containing all of the important species and reactions that lead to the formation and destruction of HDO and OD. The database was created using reactions taken from the literature (Croswell & Dalgarno 1985; Millar, Bennet, & Herbst 1989; Pineau des Forêts, Roueff, & Flower 1989; Rodgers & Millar 1996). The principle reaction in the deuterium chemistry is H2 + HD H2D+ + H, for which we have used the rate coefficients in Caselli et al. (1998). We have adopted the H3+ electron recombination rate and branching ratios given in Sundstrom et al. (1994) and Datz et al. (1995), respectively. The H2D+ recombination rate and branching ratios are from Larsson et al. (1996). The deuterium chemical network was tested against the larger network of Millar et al. (1989), and the degree of deuterium fractionation in HD, OD, and HDO was found to be in excellent agreement for the temperature ranges considered here. As a corollary to the high-temperature reactions which form H2O in the shocked gas (see Kaufman & Neufeld...
TABLE 1

| Reaction                  | \( A \) (cm\(^{-3}\) s\(^{-1}\)) | \( B \) (K) | References |
|---------------------------|----------------------------------|-------------|------------|
| \( O + H D \rightarrow OD + H \) | \( 1.57 \times 10^{-11} \) | 1.7         | 4639       | 1          |
| \( O + H D \rightarrow OH + D \)   | \( 9.01 \times 10^{-11} \)     | 1.9         | 3730       | 1          |
| \( OH + H \rightarrow H_{2}O + D \) | \( 2.12 \times 10^{-11} \)     | 2.7         | 1258       | 2          |
| \( OH + HD \rightarrow HD + H \)    | \( 0.60 \times 10^{-11} \)     | 1.9         | 1258       | 2          |
| \( OD + H \rightarrow OD + H \)     | \( 1.55 \times 10^{-11} \)     | 1.6         | 1663       | 3          |

* Rate coefficient \( k = A/T^{3}e^{-B/T} \).

References.—(1) Fit to theoretical predictions of Joseph, Truhlar, & Garret 1988; (2) from Talukdar et al. 1996; (3) rate same as OH + H; Talukdar et al. 1996.

1996), we have included similar reactions that form HDO. These reactions and rate coefficients are listed in Table 1.

We have used the gas-grain adaptation of the UMIST network discussed in Bergin & Langer (1997). For the first and second stages we assume that molecules are depleting onto bare silicate grains, while in the third stage the molecules deplete onto a grain mantle dominated by solid \( H_{2}O \). This will increase the binding potential for all species by a factor of 1.47.

In the shock stage, we assume \( v_{s} = 20 \) km s\(^{-1}\), which will heat the gas to \( \sim 1000 \) K; similar results would be found for any shock velocity between \( \sim 10 \) and 40 km s\(^{-1}\). In the preshock quiescent stage, we find significant levels of D-fractionation at: at \( t = 10^{6} \) yr we find \( [HDO]/[H_{2}O] \sim 10^{-3} \) in both the gas and solid phases. Thus, \( [HDO]/[H_{2}O] > [HD]/[H_{2}] = 2.8 \times 10^{-3} \), which is the result of low temperatures and low electron abundances favoring production of \( H_{2}D^{+} \) relative to \( H_{2}^{+} \). These enhancements are mirrored in the daughter products of \( H_{2}D^{+} \), such as HDO. In the shock itself, HDO is released from the grain surface, leading to an increase in its gas-phase abundance. Thus, in stage 2 the HDO abundance shows little change because the fractionation via the ion-molecule chemistry is halted at higher temperatures. There is some production via the high-temperature reactions listed in Table 1; however, these reactions do not increase the fractionation significantly. Therefore, the rapid hydrogenation of oxygen is not followed by a similar increase in the HDO abundance in the shock [HDO]/[H_{2}O] = 5 \times 10^{-7}. Pineau des Forêts et al. (1989) examined the D-chemistry in shocks and found little change in the water D/H ratio. As noted by the authors, their work did not examine the high temperatures required to produce abundant HDO and therefore did not probe this change.

In the postshock gas (Fig. 1, stage 3) the abundance of gaseous HDO is approximately constant with a small, but increasing, abundance frozen on the grain surfaces. At later times \( (t \sim 4 \times 10^{4} \) yr), HDO begins to deplete onto the grain surfaces, removing the primary destruction pathway for \( H_{2}^{+} \) and \( H_{2}D^{+} \). As the abundance of \( H_{2}D^{+} \) ions increase, they react directly with water, forming HDO (along with OD and O) and further increasing the fractionation.

As shown in the top panels of Figure 1, the abundance of DCO\(^{+} \) is quite high with [DCO\(^{+}/[HCO]^{+} = 0.002, but in the shock the abundances of both DCO\(^{+} \) and HCO\(^{+} \) decline due to reactions with HDO) and fractionation is reversed such that [DCO\(^{+}/[HCO]^{+} \sim 2 \times 10^{-3} \). Thus, DCO\(^{+} \) should be an excellent tracer of quiescent gas in star-forming regions, since chemical theory predicts that it will be destroyed in an outflow.

In the first two stages, active CO sublimation at \( T_{g} = 30 \) K suppresses the net CO depletion onto bare silicate grains. In stage 3, at \( \sim 10^{4} \) yr, the gaseous CO abundance declines because we have assumed that in the third stage the grains are a priori coated by water ice which results in a greater binding energy, resulting in an exponentially decreasing sublimation rate compared to bare silicates (see § 2). This leads to a solid CO abundance given by [CO]/[\( H_{2}O \) = \( 8 \times 10^{-3} \) at \( t = 10^{7} \) yr. Second, on similar timescales the abundance of OH is enhanced due to the destruction of water molecules. The reaction of OH with CO will form CO\(_{2}\), which readily depletes onto grain surfaces.

3. RESULTS

Figure 1 presents the time evolution of abundances for the three-stage gas-grain chemical model. The peak gas-phase H\(_{2}O \) abundance is low ([H\(_{2}O]/[H_{2}] = 3 \times 10^{-7}) in the preshock gas, rises to \( \sim 10^{-4} \) following the passage of a shock, and depletes onto the grain surface at \( t = 10^{6} \) yr (for \( n_{H_{2}} = 10^{4} \) cm\(^{-3}\)) in the postshock gas. In the shock stage, we assume \( v_{s} = 20 \) km s\(^{-1}\), which will heat the gas to \( \sim 1000 \) K; similar results would be found for any shock velocity between \( \sim 10 \) and 40 km s\(^{-1}\). In the preshock quiescent stage, we find significant levels of D-fractionation: at \( t = 10^{6} \) yr we find [HDO]/[H\(_{2}O \sim 10^{-3} \) in both the gas and solid phases. Thus, [HDO]/[H\(_{2}O \sim [HD]/[H_{2}] = 2.8 \times 10^{-3} \), which is the result of low temperatures and low electron abundances favoring production of \( H_{2}D^{+} \) relative to \( H_{2}^{+} \). These enhancements are mirrored in the daughter products of \( H_{2}D^{+} \), such as HDO. In the shock itself, HDO is released from the grain surface, leading to an increase in its gas-phase abundance. Thus, in stage 2 the HDO abundance shows little change because the fractionation via the ion-molecule chemistry is halted at higher temperatures. There is some production via the high-temperature reactions listed in Table 1; however, these reactions do not increase the fractionation significantly. Therefore, the rapid hydrogenation of oxygen is not followed by a similar increase in the HDO abundance and in the shock [HDO]/[H\(_{2}O \sim 5 \times 10^{-7} \). Pineau des Forêts et al. (1989) examined the D-chemistry in shocks and found little change in the water D/H ratio. As noted by the authors, their work did not examine the high temperatures required to produce abundant HDO and therefore did not probe this change.

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4. DISCUSSION

4.1. Comparison with Observations

Observations of ices in the ISM and in comets have yielded limits on the level of deuterium fractionation in water along with the quantity of CO\(_{2}\) frozen in the water matrix (see Whittet et al. 1998). One question is whether our model can simultaneously reproduce both the observed [CO\(_{2}\)]/[\( H_{2}O \) and [HDO]/...
[H₂O] ratios. Equilibrium models of diffusion-limited grain surface chemistry (i.e., chemistry that is limited by the depletion rate onto the surface) can reproduce high D-fractionation of water (Tielens 1983) and produce [CO₂]/[H₂O] ~ 0.01%–10% (Tielens & Hagen 1982; Shalabiea, Caselli, & Herbst 1998).

In Figure 2 we present the comparison of observations with our model predictions as a function of time. To illustrate the sensitivity of these results to our assumed gas and dust temperature of 30 K, results for higher (40 K) and lower (25 K) gas and dust temperatures are also shown. The small circles (with error bars) depict HDÖ and CO₂ measurements for comets Halley, Hyakutake, and Hale-Bopp, the only comets for which all of these quantities have been determined. In the ISM there are no similar complementary measurements; HDÖ and H₂O have been observed in the gas phase in molecular hot cores, while CO₂ ices have only been observed along single lines of sight toward stars behind molecular clouds or embedded within them. We have therefore represented these two independent measurements for the ISM as a dotted box in Figure 2. In hot cores, the gas-phase D-fractionation is believed to trace the solid (D/H) ratio because the levels of deuterium fractionation are larger than expected for pure gas-phase chemistry evolving at the observed temperatures (T ~ 100–200 K). Thus, the observed HDÖ and H₂O are proposed to be “fossilized” remnants from a previous low-temperature phase, remnants that were frozen on the grains and evaporate in the hot core (e.g., Gensheimer, Mauersberger, & Wilson 1996).

The contours of time-dependent abundance ratios for the range of dust temperatures provided in Figure 2 nicely encompass the observations. For this model to be applicable to interstellar ices, the initial gas clouds that collapsed and ultimately formed the molecular hot cores must have evolved at T ~ 25–40 K to account for the observed fractionation. For both CO₂ and HDÖ production and where CO₂ ices have been observed, our model also suggests that the quiescent clouds that formed hot cores would have undergone at least one shock within the past 10⁶–10⁷ yr. This timescale is consistent with the range of inferred shock timescales in molecular clouds (Reipurth, Bally, & Devine 1998; BMN).

Over the past 20 years or more, there has been considerable debate about whether comets are composed of pristine interstellar material or whether significant processing has taken place within the proto-solar nebula (or indeed whether comets represent a mixture of pristine and processed material). The striking similarity between the composition of comets and that of interstellar ices, and in particular the very similar level of deuterium fractionation, might suggest that the degree of processing in the proto-solar nebula is relatively small. That the composition of interstellar ices was preserved during their passage into the outer parts of the proto-solar nebula is perhaps unsurprising given the relatively benign physical conditions; in particular, the typical shock velocity during accretion onto

3 Note that while the three comets have similar [HDO]/[H₂O] ratios, comet Hale-Bopp shows a larger [CO₂]/[H₂O] ratio than either Halley or Hyakutake. These differences are significant relative to the observational uncertainties but may be an artifact of the various Sun-comet distances at which the measurements were made: ISO observations of CO₂ in Hale-Bopp were obtained when the comet was 2.9 AU from the Sun, while the other cometary measurements were made at a Sun-comet distance of ~1 AU. The lower binding energy of CO₂, compared to H₂O, could therefore lead to differences in the outgassing of these species at larger Sun-comet distances. Interestingly, ISO observations of the short-period comet 103P/Hartley 2 at 1 AU from the Sun also indicate high CO₂ abundances, but this comet does not have a measured D/H ratio (J. Crovisier 1998, private communication).

![Fig. 2.—Plot of the grain surface [HDO]/[H₂O] against [CO₂]/[H₂O] shown as a function of time [log (t)] and gas and dust temperature (T). The calculated abundance ratios are shown only in the postshock phase (stage 3). The points are the observed ratios in comets Halley, Hyakutake, and Hale-Bopp. For Halley, Hyakutake, and Hale-Bopp, the HDÖ data are from Eberhardt et al. (1995), Balsiger, Altwegg, & Gies (1995), Bockelée-Morvan et al. (1998), and Meier et al. (1998). The CO₂ measurements are from Kranowsky (1991), McPhate et al. (1996), and Crovisier et al. (1997). The box shows the range of representative values observed in the interstellar medium and [HDO]/[H₂O] ratios are from Gensheimer et al. (1996), and the [CO₂]/[H₂O] ratios in polar ices are from Schuttle (1998) and Whittet al. (1998).

4.2. Sensitivity to Physical Conditions

Although our model assumes n_H₂ = 10⁵ cm⁻³, the results are similar at both higher and lower densities because most chemical processes show the same dependence on density. We adopt A_v = 10 mag in our models, which are therefore only applicable in regions in which the ultraviolet field is heavily attenuated. Variations in the dust temperature can affect our results, and our model can only create ice mantles at dust temperatures greater than 25 K. Below 25 K, O₂ remains frozen on the grain surface in the preshock quiescent stage, leaving little free gas-phase O for shock chemistry to form H₂O in abundance. Under these conditions, a problem arises in accounting for the high abundance of water ice (~10⁻³, Schuttle 1998) observed in the cold foreground gas toward Elias 16. However, this limitation can be mitigated by adopting a lower density (n_H₂ ≤ 10⁹ cm⁻³) since the reduced depletion rate results in a smaller and less constraining O₂ abundance on grains.

One unresolved issue concerns the large abundance of CO ice predicted in our model, which is quite high (~70% relative to H₂O) and is larger than observed in the ISM or in comets in which abundances are generally less than 20% (e.g., Mumma 1997). However, if we consider a single CO molecule approaching a surface with nearly equal amounts of CO and H₂O, it is difficult to imagine that CO will always be sharing a (stronger) physisorbed bond with H₂O rather than a (weaker) bond to a CO molecule. In laboratory experiments in which CO and H₂O (with CO/H₂O = 0.5) are co-deposited onto metal

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substrate at 10 K, the evaporation rate of amorphous CO is dominated by the vapor pressure of pure CO and is not affected by the presence of H$_2$O (Kouchi 1990). Although the sublimation of pure CO is not included in our model (we include sublimation of CO embedded in a water mantle), in reality some co-deposited CO will evaporate off the grain mantle, provided that the dust temperature exceeds 17 K, the sublimation temperature of pure CO ice. The large abundance of frozen CO predicted by our model (under the assumption that CO sublimation is negligible) may therefore be unrealistic and should be regarded as an upper limit.

5. CONCLUSION

In this Letter, we have demonstrated that in well-shielded regions, the gas-phase chemistry after the passage of a shock is capable of reproducing the observed abundance of frozen water, the HDO/H$_2$O ratio, and the abundance of CO$_2$ ice. Thus, the creation of grain mantles behind shock waves is a viable alternative theory to the production of ices through grain surface chemistry. We note that this mechanism does not preclude grain surface chemistry, and indeed it may act as a supplement. A future extension of our model will be to determine whether shock models can account for the enhanced abundances of methanol observed in shocked regions relative to surrounding gas (e.g., Bachiller et al. 1995). Such an extension of our model might explain the formation of methanol ices—an observed constituent of interstellar grain mantles—as arising from the “freezeout” of CH$_3$OH gas produced by gas-phase chemistry in the warm shocked gas. Other tests could involve an examination of other proposed outflow chemistry tracers (e.g., SO) and an expanded examination of the deuterium chemistry, including species such as DCN, HDCO, and D$_2$CO.

It has previously been suggested that the high degree of deuterium fractionation observed in cold cloud cores can explain the observed deuterium fractionation in cometary ices (e.g., Geiss & Reeves 1981). However, our results indicate that shock waves are a critical ingredient in the production of ices from gas-phase chemistry. Ion-molecule chemistry in quiescent gas can reproduce the degree of deuterium fractionation observed in molecular hot cores; however, it cannot reproduce the observed abundance of water ice (Jones & Williams 1984; Bergin, Langer, & Goldsmith 1995). In our model, the abundance of water ice in the preshock gas at 10$^4$ yr is less than 10$^{-5}$, much less than the value estimated along the line of sight toward Elias 16 (∼10$^{-4}$; Schutte 1998). At $n_{H_2} = 10^8$ cm$^{-3}$, the gas-phase chemistry could, with time, eventually approach the measured abundance. However, the molecular ice absorption observations are taken along lines of sight that likely have lower densities, which makes reproducing the observed abundances more difficult. Thus, either grain surface chemistry, creation of ice mantles behind shock waves, or other unknown theories are necessary to account for the presence of molecular ices.

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