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Repeated addition of atomic hydrogen to fullerene cations, dications and trications

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Abstract

The reactivity of the fullerene cations C_{56}^{n+}, C_{58}^{n+}, and C_{60}^{n+} (n = 1, 2, 3) with atomic hydrogen has been studied in a selected-ion flow tube apparatus at a temperature of 294 ± 2 K and a helium buffer-gas pressure of 0.36 ± 0.01 Torr. Repeated addition of hydrogen atoms was inferred in the reactions of all the fullerene cations. Rate coefficients for each hydrogen-atom addition step could not be determined separately. However, a "mean addition rate coefficient" for the observed series of sequential hydrogen-atom additions could be determined, yielding values of 3.0×10^{-10}–4.5×10^{-10} cm^3 $molecule^{-1}$ s^{-1} for singly and doubly charged fullerene ions, and 8.0×10^{-10}–10×10^{-10} cm^3 $molecule^{-1}$ s^{-1} for fullerene trications. The lower limits obtained for the effective bimolecular rate coefficient for the primary addition step were 1.0×10^{-10} cm^3 $molecule^{-1}$ s^{-1}, 3.0×10^{-10} cm^3 $molecule^{-1}$ s^{-1}, and 1.0×10^{-9} cm^3 $molecule^{-1}$ s^{-1} for singly, doubly and triply charged cations, respectively. In contrast with previous studies which have established a substantial enhancement in the efficiency of addition to the adjacent-pentagon fullerene ions C_{56}^{n+} and C_{58}^{n+} over addition to the isolated-pentagon fullerene ions C_{60}^{n+}, little or no difference in the rate of addition or the overall extent of hydrogenation was apparent between C_{56}^{n+}, C_{58}^{n+}, and C_{60}^{n+} ions possessing the same degree of positive charge. None of the fullerene cations reacted measurably with H_2 under the experimental conditions.

Keywords: Fullerene cations; Hydrogen atoms; Ion/molecule reactions; SIFT technique

1. Introduction

One of the many aspects of the emerging fullerene chemistry [1,2] to have received considerable attention in the last few years is that of the hydrogenation of fullerenes. Several syntheses have recently been reported for hydrogenated fullerenes ("fulleranes") [3–6]: for Buckminsterfullerene, C_{60}, levels of hydrogenation ranging from $C_{60}H_2$ to $C_{60}H_{44}$ [3–5] have been achieved, although Banks et al. [6] noted that the Birch reduction of C_{60} appears to yield a range of fullerane products dominated by $C_{60}H_{32}$, rather than $C_{60}H_{36}$ as

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