Nanodiamond synthesis mediated by electron irradiation through C-H couplings of adamantane

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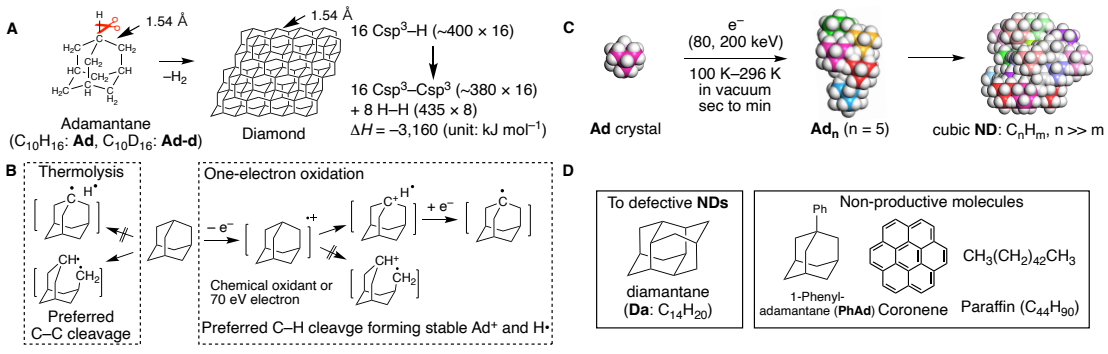
Nanoscale diamonds (NDs) are attracting significant attention due to their promising scientific and practical applications. However, the synthesis of NDs typically requires harsh conditions, such as explosions or high-temperature and high-pressure (HTHP) environments, which hinder controlled production on demand. Over nearly a century, little progress has been made in producing NDs under low-temperature and low-pressure conditions, largely due to a lack of suitable synthetic methods.

Here, we describe the electron-beam-mediated C–H coupling of adamantane (AD) molecules within solid AD to form NDs, as illustrated schematically in Fig. 1. Products, from oligomers to large single crystals, are ambiguously identified during reactions by transmission electron microscopy. This C–H coupling reaction proceeds at room temperature or cryogenic temperatures within seconds to minutes, even under vacuum, with high purity and yield. High-energy electrons selectively cleave the C–H bonds of the diamondoid molecules, forming AD radicals or AD radical cations while retaining the C–C framework. The reactive intermediates subsequently couple to yield NDs along with the hydrogen generations. The successful reactions using deuterated AD and diamantane demonstrate the general applicability of this electron-mediated C-H activation. We anticipate that the NDs produced via this method will be easy to functionalize, as their surfaces are predominantly terminated by hydrogen atoms. This feature suggests exciting prospects for a wide range of future applications.



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