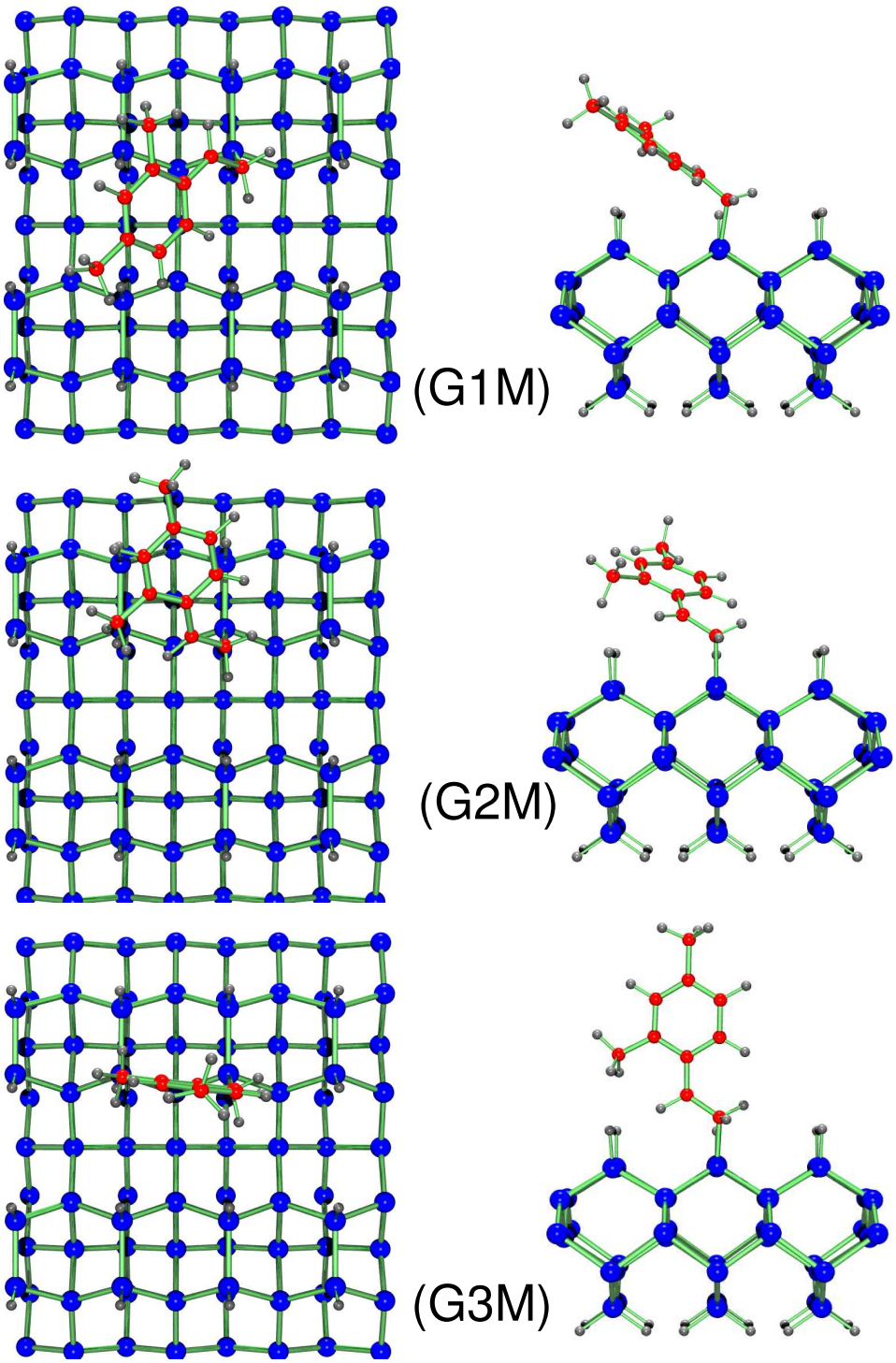

Figure S1 Initital (C-radical containing) adsorption structures of a styrene molecule on the H-Si(001)-(2×1) surface. Energies are –0.83, -0.73, -0.66 eV for G1M**,** G2M, G3M, respectively, relative to the non-interacting molecule + surface system. Big blue circles correspond to Si atoms, small gray circles represent H atoms, and red circles are C atoms.

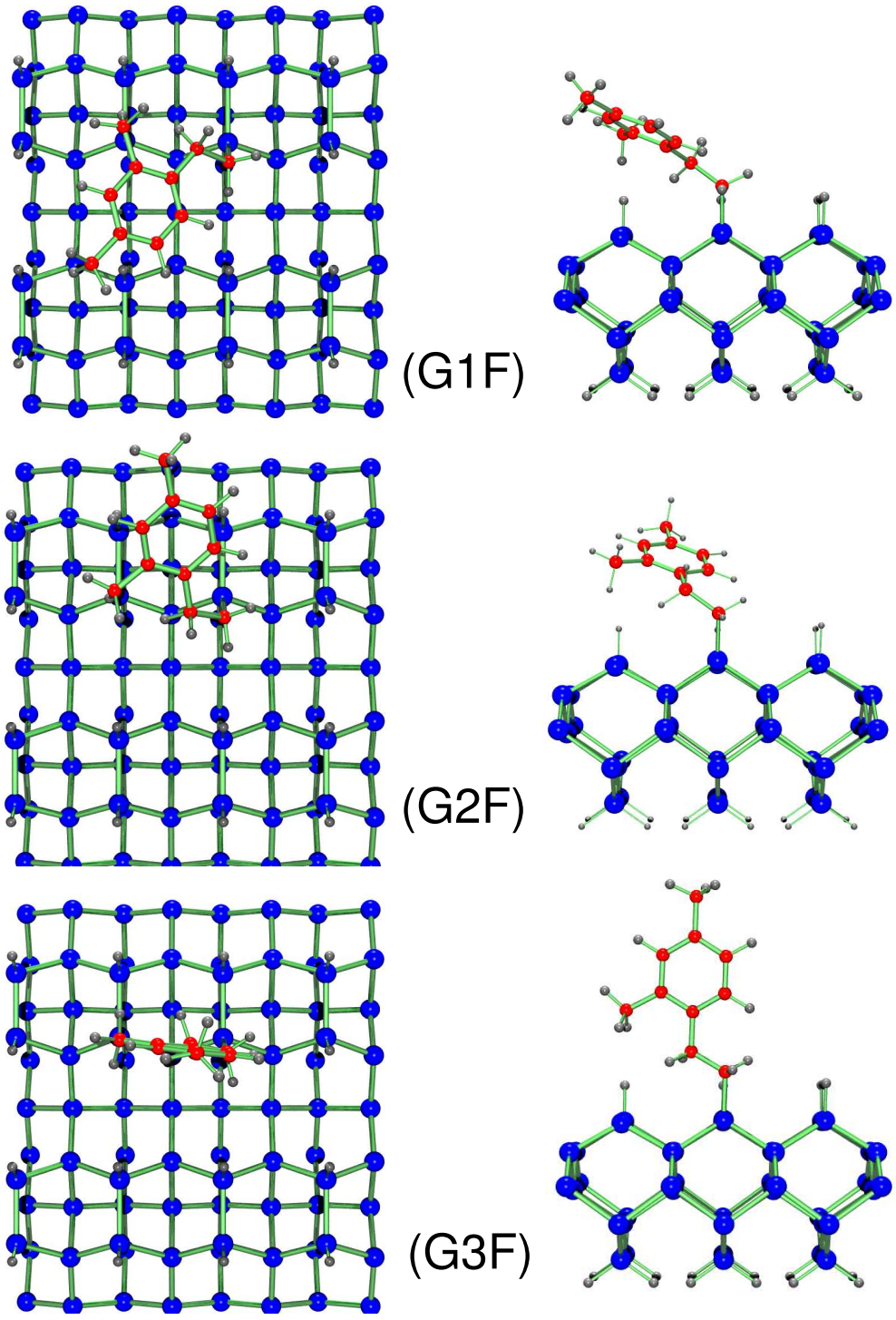

Figure S2 Final adsorption structures of a styrene molecule on the H-Si(001)-(2×1) surface, after hydrogen abstraction. Energies are –0.92, -0.99, -1.03 eV for G1F, G2F, G3F**,** respectively, relative to the non-interacting molecule + surface system. Big blue circles correspond to Si atoms, small gray circles represent H atoms, and red circles are C atoms.

2,4 Dimethyl-styrene molecule, structures after hydrogen abstraction.

After the 2,4 dimethyl-styrene molecule attaches to the molecule and form the configurations of figure S1, the created highly reactive carbon radical abstracts a hydrogen atom from a neighboring Si-H group. Figure S2 shows such adsorption geometries. Note the creation of a new dangling bond on the neighboring site. It is found that for each initial structure in figure S1, there is a corresponding structure with a stable molecule.

Beside the H atom that is abstracted, atomic positions change little. In Geometry G1F, the length of the Cα-Cβ bond is close to 1.55 Å**,** slightly larger than the length of a single carbon-carbon bond, and it is tilted by θ ~ 36º with respect to the surface plane, while the C-Si bond length is 1.92 Å, indicating a stronger bond than in the initial state. The βC-1C forms a 22° angle with the surface. As shown in Table II, this state is more stable than the system formed by the non-interacting molecule with the surface by 0.92 eV. The structural parameter for Geometry G2F are as following: the Cα-Cβ bond has a length $d_{C\text{-}C}$ ~ 1.54 Å, indicating a single bond, and is tilted by θ ~ 36º with respect to the surface plane, while the C-Si bond length is 1.93 Å; the Cβ-C1 bond forms a 27° angle with the surface. The adsorption energy of this configuration is 0.99 eV, larger than the energy of geometry G1F.

In structure G3F, the length of the Cα-Cβ bond is close to 1.53 Å. As shown in Table I, this state is more stable than the non-interacting molecule and surface by 1.03 eV, and it is more stable than the two previous configurations, indicating that it is the ground state.