Supplementary Information

Revisiting Activity Tuning using Lattice Strain: CO Decomposition in Terrace Ru(0001) and Stepped Ru(1015) Surfaces

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Poisson Ratio Calculation

From the main text, Poisson ratio (μ) can be calculated with the equation below:

$$\mu = -\frac{\epsilon_{\mathrm{BEP}}}{\epsilon_{\mathrm{lateral}}} \quad \text{(S1)}$$

where $\epsilon_{\mathrm{transverse}}$ is the strain along the direction of applied force or constraint and $\epsilon_{\mathrm{lateral}}$ is that of along the perpendicular direction. While strain, ϵ, can be evaluated by

$$\epsilon = \frac{\Delta L}{L_{\mathrm{o}}} \quad \text{(S2)}$$

where ΔL and L_{o} is the change in length and the original length, respectively. Here, L_{o} is the optimized lattice vector length.

To induce expansion or compression, lattice contraint can be applied in a single direction while optimizing the the remaining lattice vector. Note that in slab calculations, the lattice vector normal to the plane surface is set large enough to suppress slab–slab interaction from the neighboring periodic image. This step then simply calls for a single lattice vector optimization alongside geometry optimization. ΔL can then be easily calculated as the difference of the strained and original lattice vector lengths. In this work, we calculated Poisson ratio to check the response of the slab to the strain with our calculation parameters.

Murnaghan Equation of State

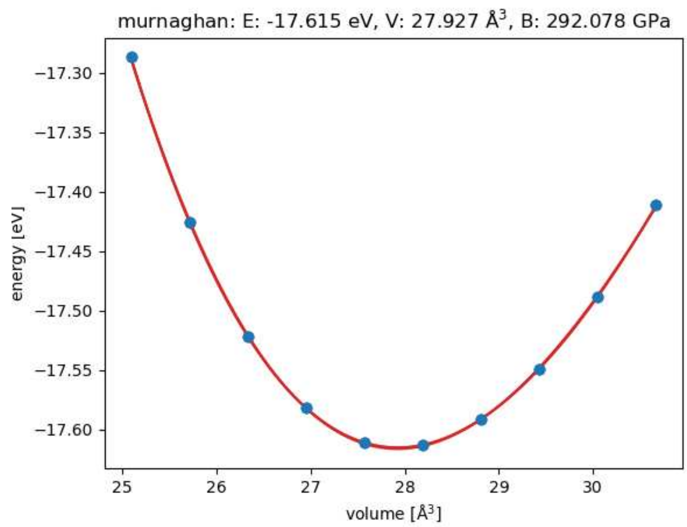
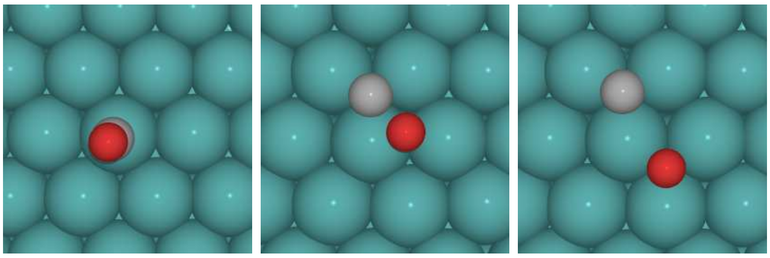


Fig. S1 Murnaghan Equation of State (EOS) plot for Bulk Ru using optimized calculation parameters described in main text. Murnaghan EOS fitting is done using the Atomic Simulation Environment.

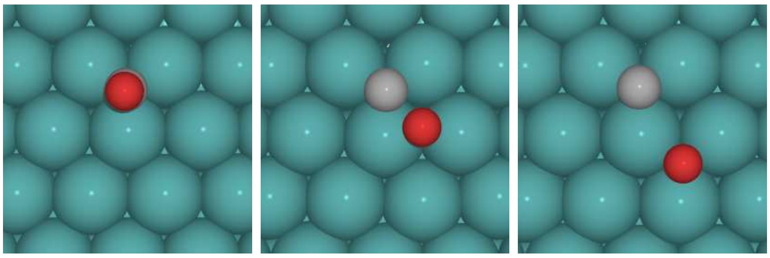
Optimized Structures CO dissociation

Relaxed Terrace Ru(0001) Surface



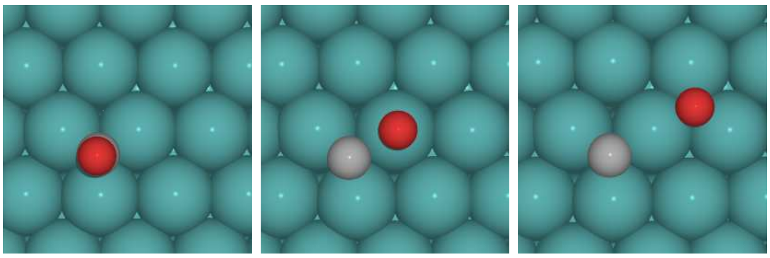
(a) (b) (c)

Fig. S2 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in relaxed terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



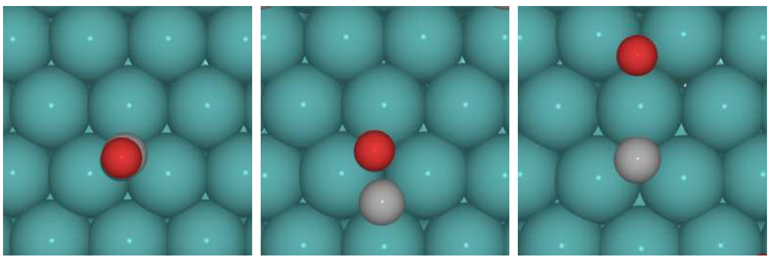
(a) (b) (c)

Fig. S3 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in relaxed terrace Ru(0001) surface for Path T2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

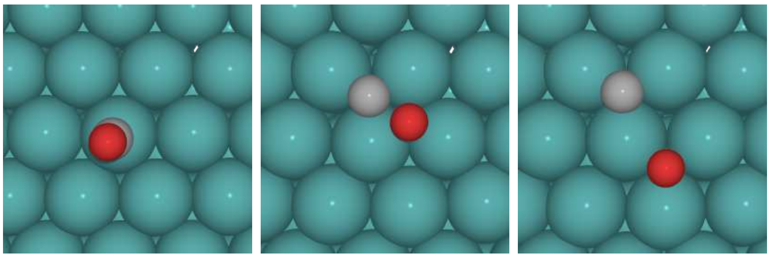
Fig. S4 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in relaxed terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

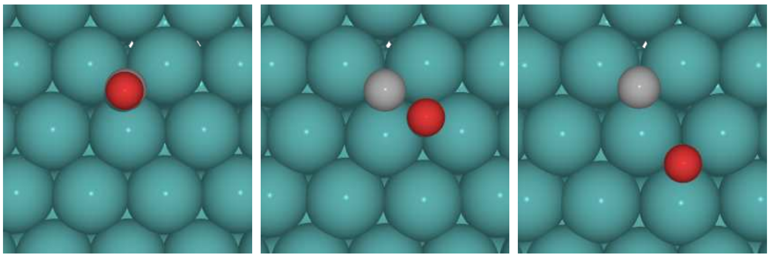
Fig. S5 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in relax terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Isotropic Expansion on Terrace Ru(0001) Surface



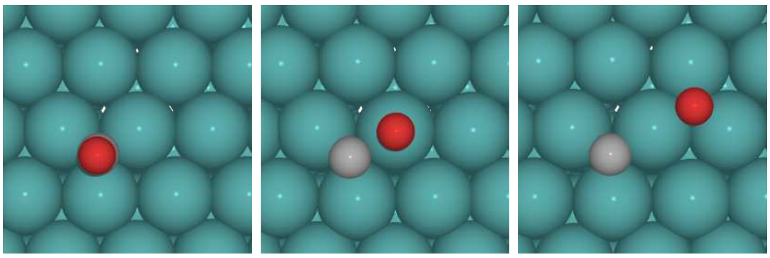
(a) (b) (c)

Fig. S6 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



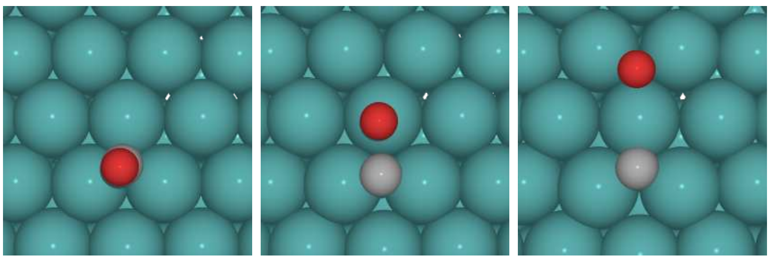
(a) (b) (c)

Fig. S7 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded terrace Ru(0001) surface for Path T2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

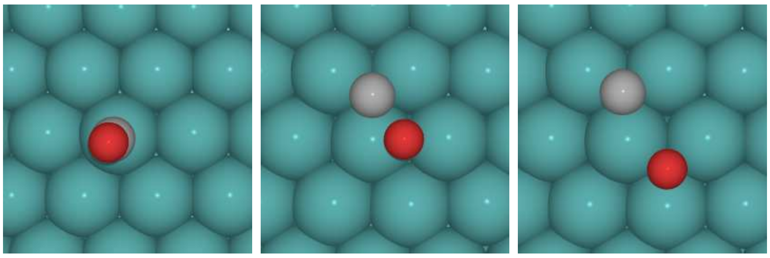
Fig. S8 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

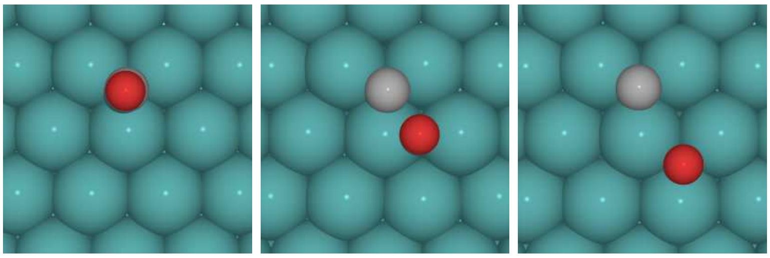
Fig. S9 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Isotropic Compression on Terrace Ru(0001) Surface



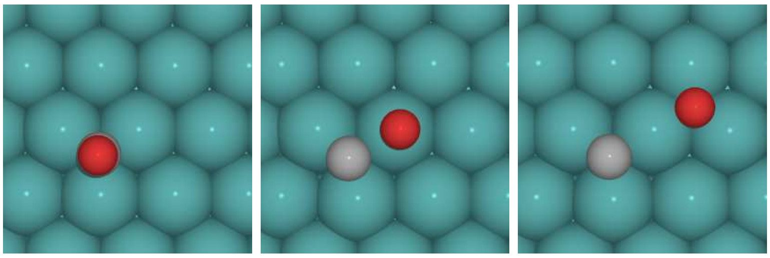
(a) (b) (c)

Fig. S10 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



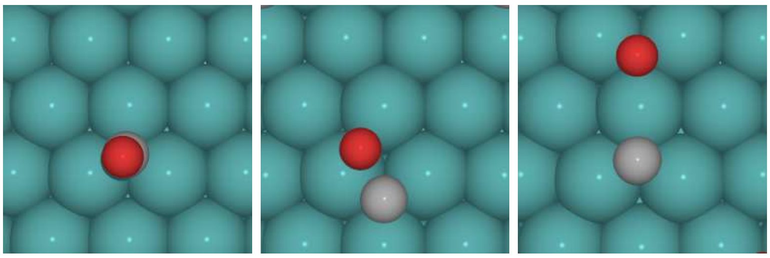
(a) (b) (c)

Fig. S11 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed terrace Ru(0001) surface for Path T2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

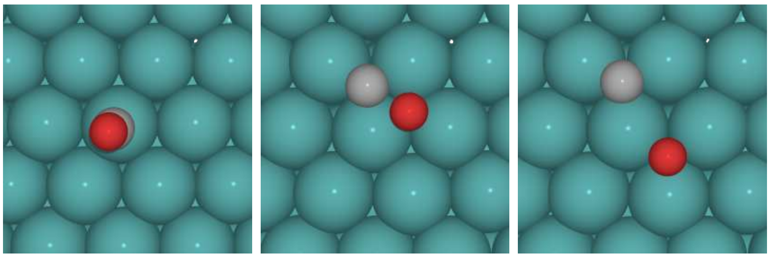
Fig. S12 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

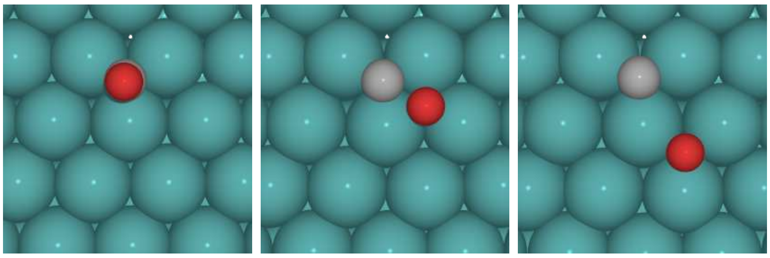
Fig. S13 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Anisotropic Expansion on Terrace Ru(0001) Surface



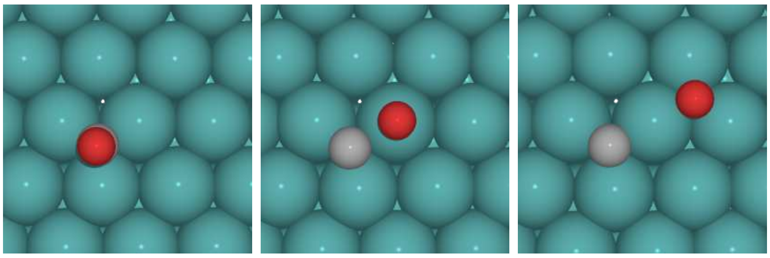
(a) (b) (c)

Fig. S14 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



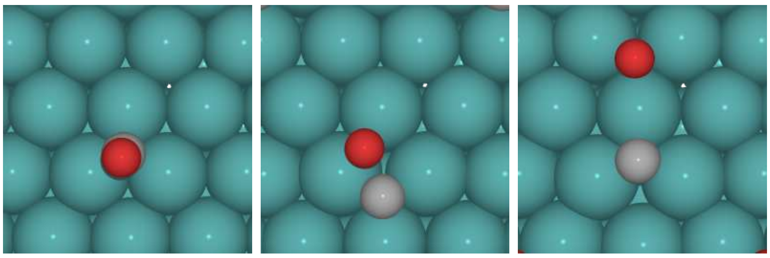
(a) (b) (c)

Fig. S15 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded terrace Ru(0001) surface for Path T2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

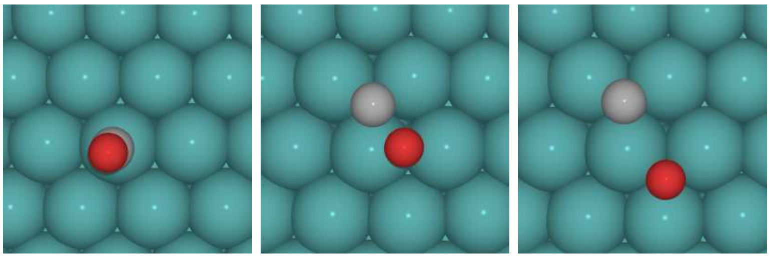
Fig. S16 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

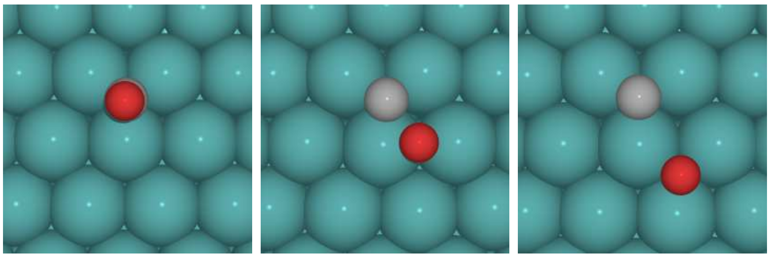
Fig. S17 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Anisotropic Compression on Ru(0001) Surface



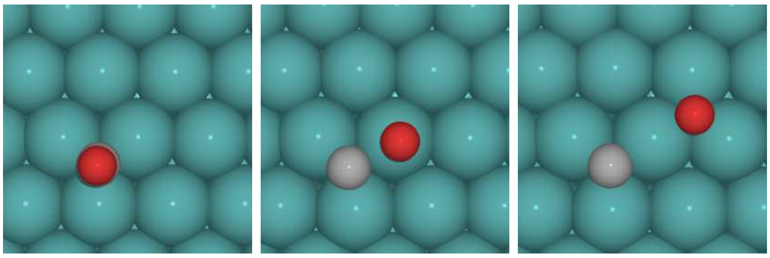
(a) (b) (c)

Fig. S18 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



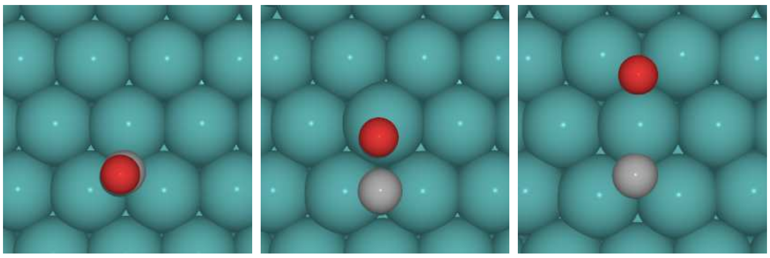
(a) (b) (c)

Fig. S19 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed terrace Ru(0001) surface for Path T2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

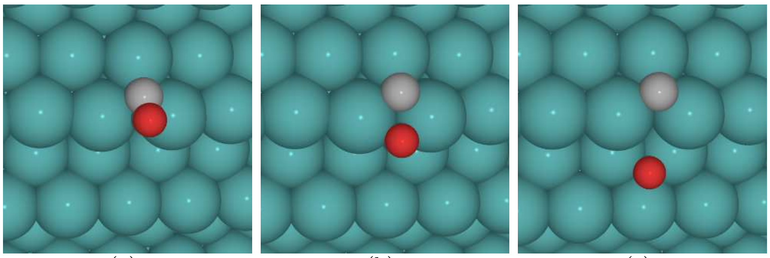
Fig. S20 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

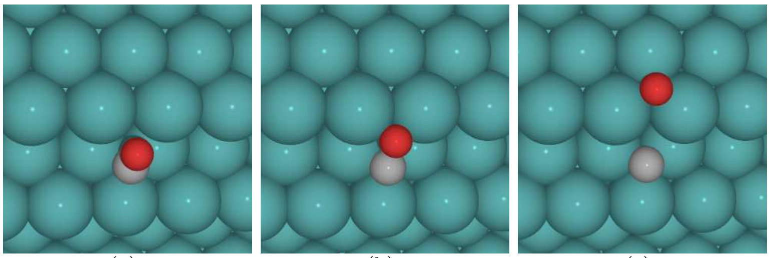
Fig. S21 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Relaxed Stepped Ru(1015) Surface



(a) (b) (c)

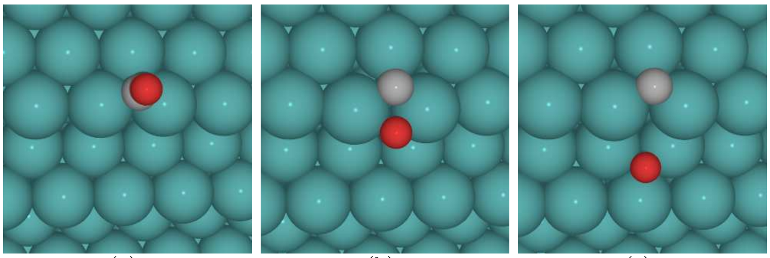
Fig. S22 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in relax stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

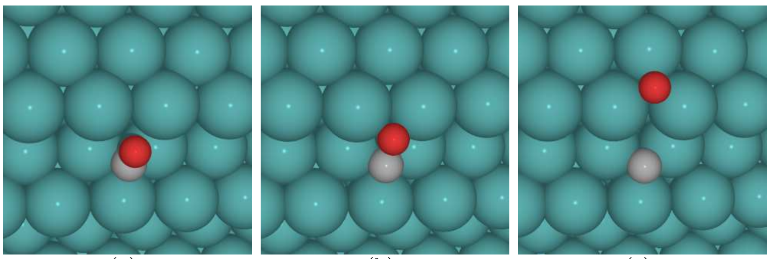
Fig. S23 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in relax stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Isotropic Expansion on Stepped Ru(1015) Surface



(a) (b) (c)

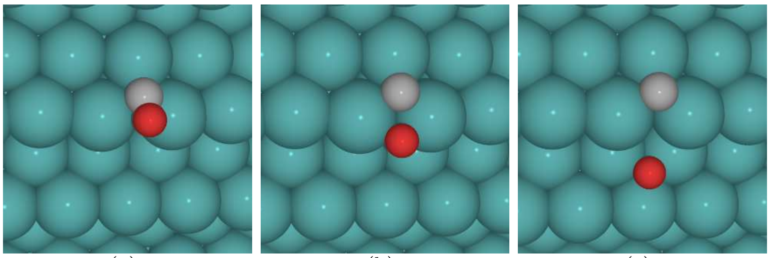
Fig. S24 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

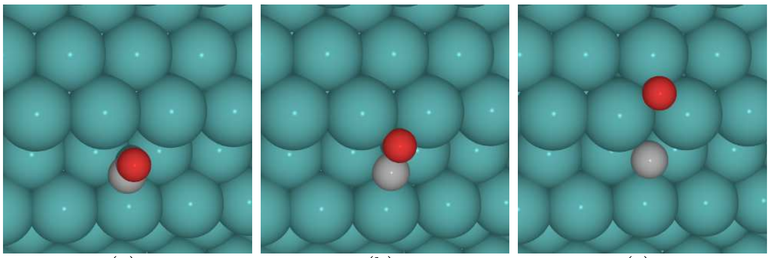
Fig. S25 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Isotropic Compression on Stepped Ru(1015) Surface



(a) (b) (c)

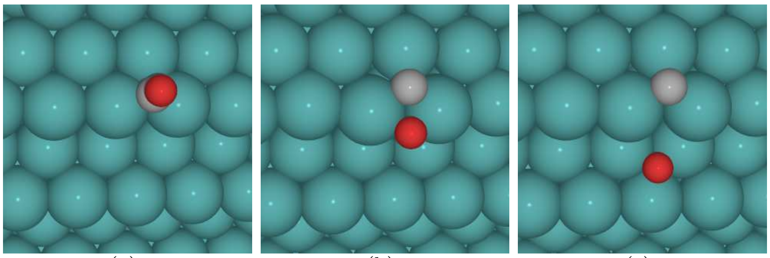
Fig. S26 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

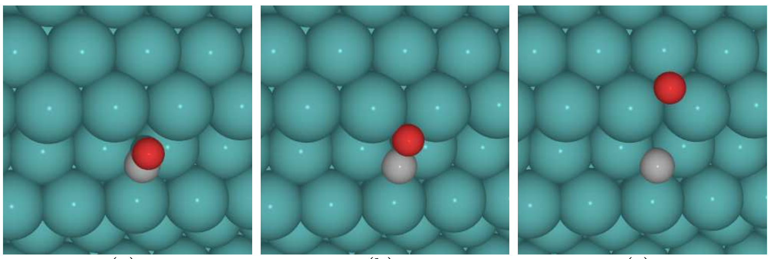
Fig. S27 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed stepped Ru(1015) surface for Path 2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Anisotropic Expansion in a of Stepped Ru(1015) Surface



(a) (b) (c)

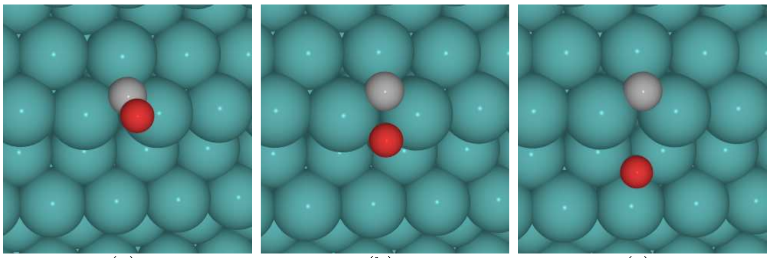
Fig. S28 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

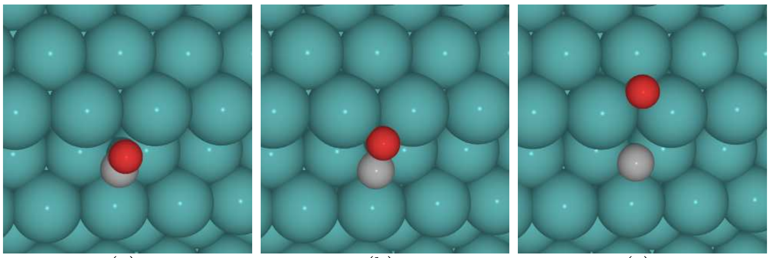
Fig. S29 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Anisotropic Compression in a of Stepped Ru(1015) Surface



(a) (b) (c)

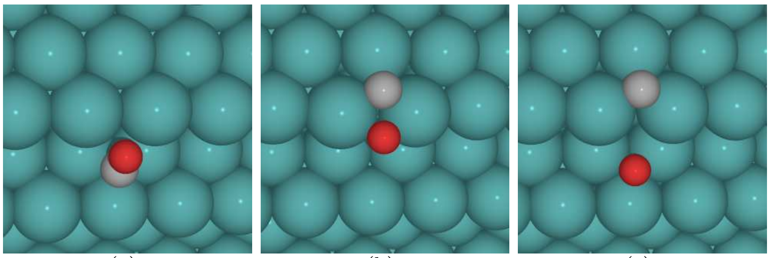
Fig. S30 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

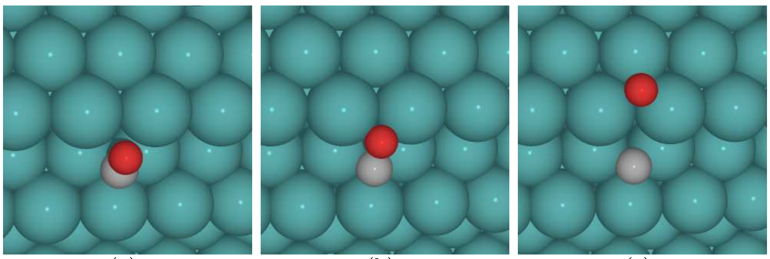
Fig. S31 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Anisotropic Expansion in b of Stepped Ru(1015) Surface



(a) (b) (c)

Fig. S32 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.



(a) (b) (c)

Fig. S33 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Anisotropic Compression in b of Stepped Ru(1015) Surface

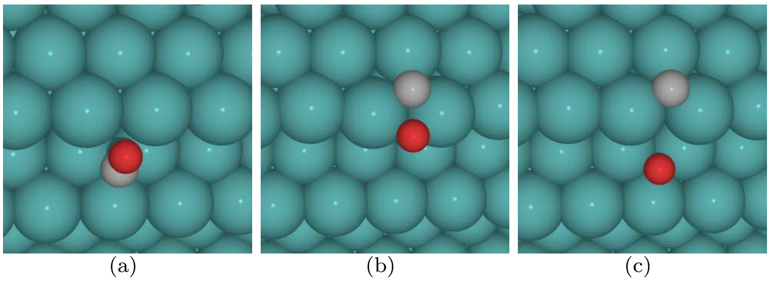


Fig. S34 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

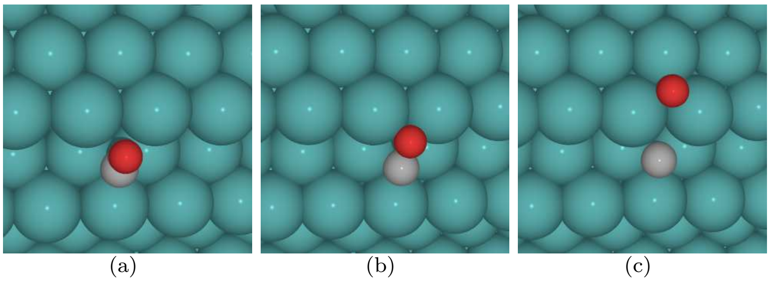


Fig. S35 Optimized structures of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively.

Projected Density of States (PDOS) and Integrated PDOS

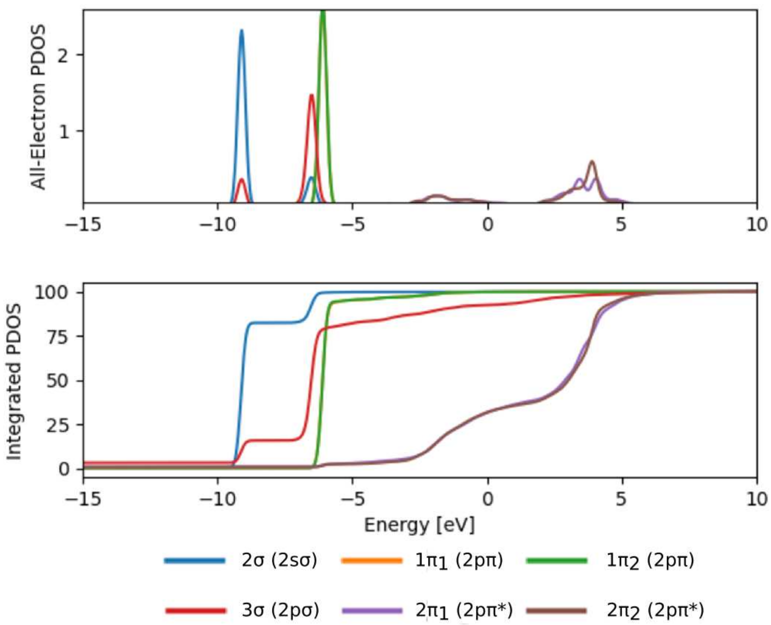


Fig. S36 Projected Density of States (PDOS) and Integrated PDOS of reactant absorbed in relax terrace Ru(0001) surface for Path T1

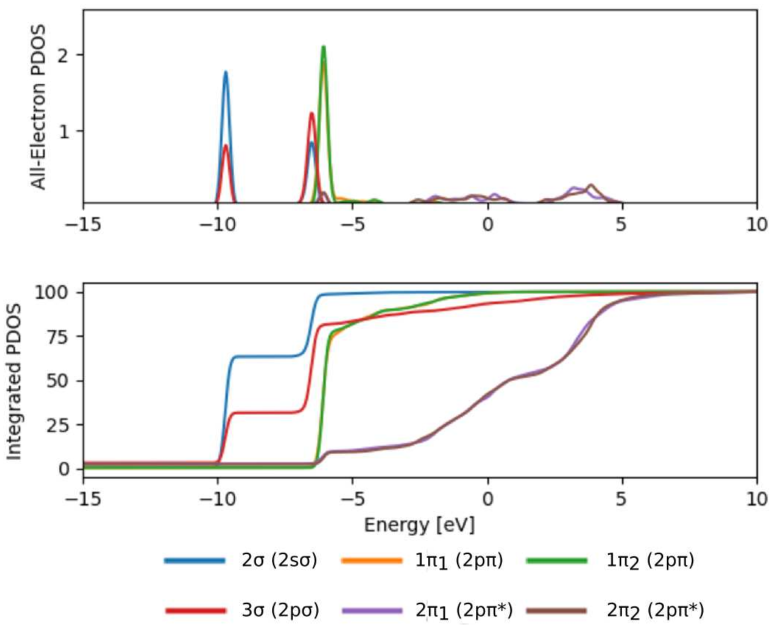
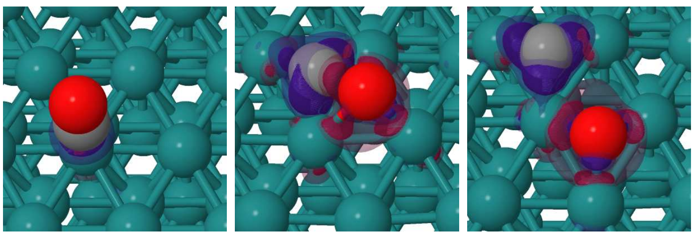


Fig. S37 Projected Density of States (PDOS) and Integrated PDOS of reactant absorbed in relax terrace Ru(0001) surface for Path T2

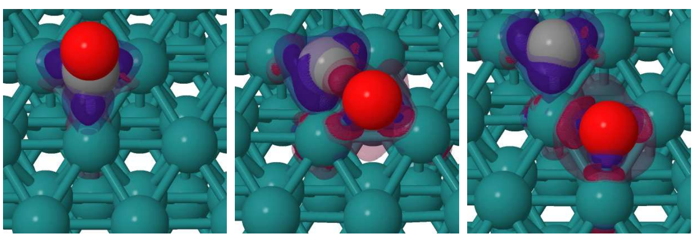
Charge Density Difference Maps

Relaxed Terrace Ru(0001) Surface



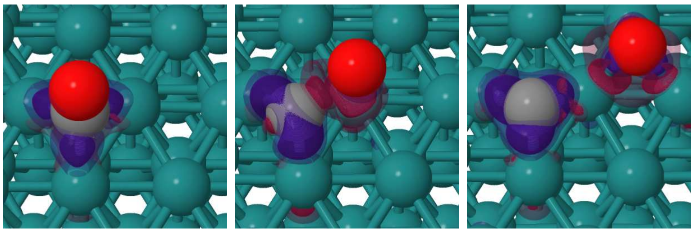
(a) (b) (c)

Fig. S38 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in relax terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



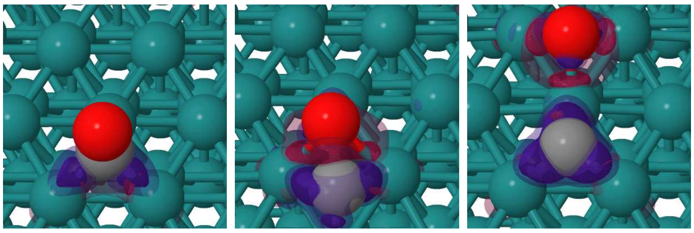
(a) (b) (c)

Fig. S39 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in relax terrace Ru(0001) surface for Path TT2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

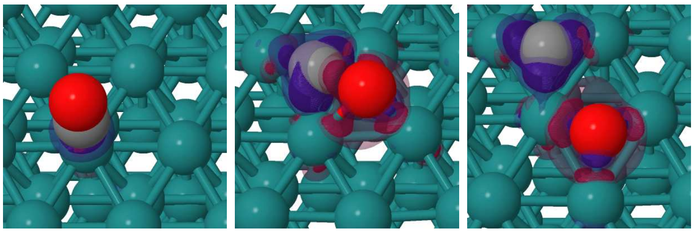
Fig. S40 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in relax terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

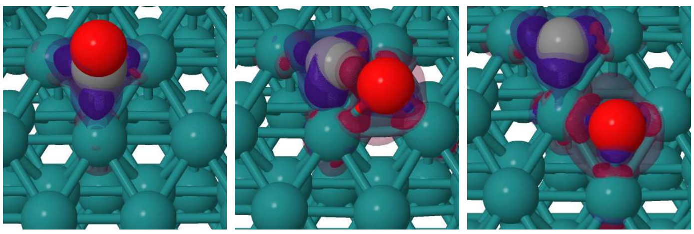
Fig. S41 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in relax terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Isotropic Expansion on Terrace Ru(0001) Surface



(a) (b) (c)

Fig. S42 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

Fig. S43 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded terrace Ru(0001) surface for Path T2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

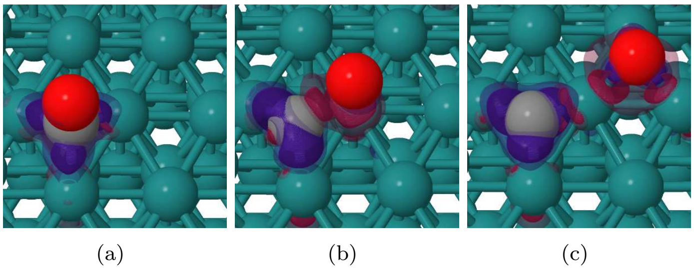


Fig. S44 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in relax terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

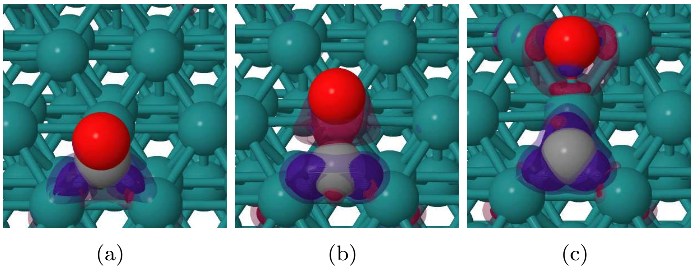


Fig. S45 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Isotropic Compression on Terrace Ru(0001) Surface

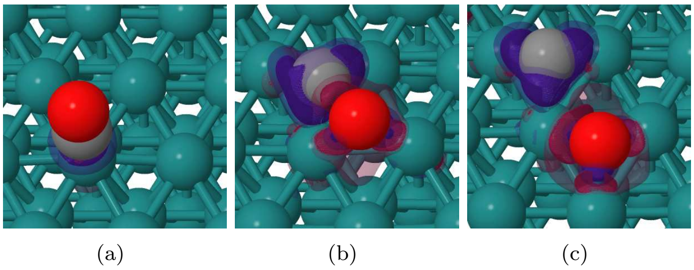


Fig. S46 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

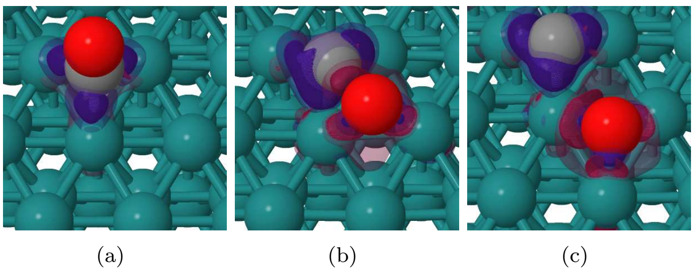
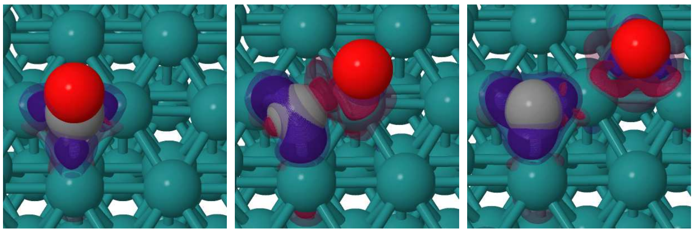
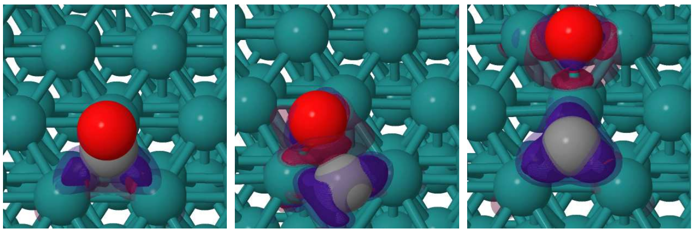


Fig. S47 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed terrace Ru(0001) surface for Path T2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

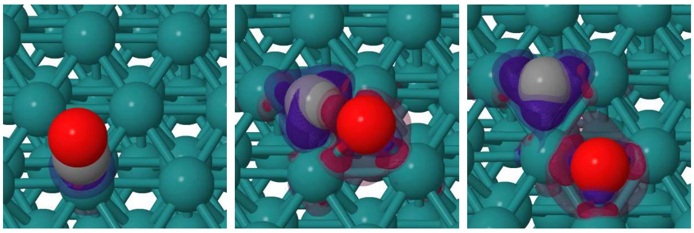
Fig. S48 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

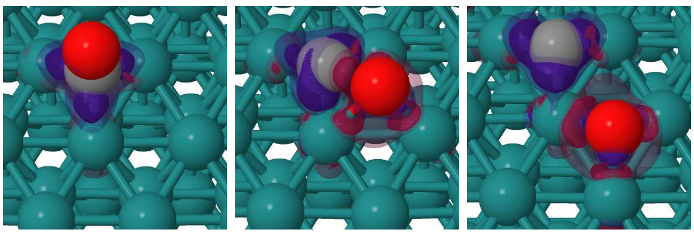
Fig. S49 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Anisotropic Expansion on Terrace Ru(0001) Surface



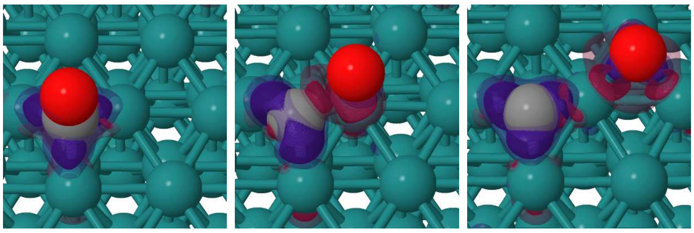
(a) (b) (c)

Fig. S50 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



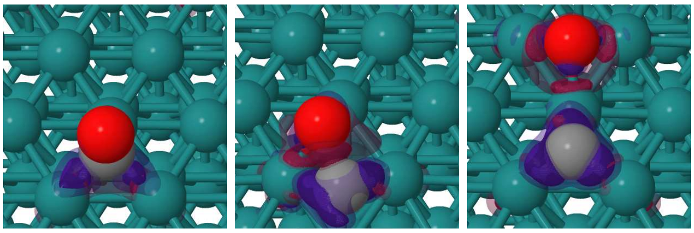
(a) (b) (c)

Fig. S51 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded terrace Ru(0001) surface for Path T2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

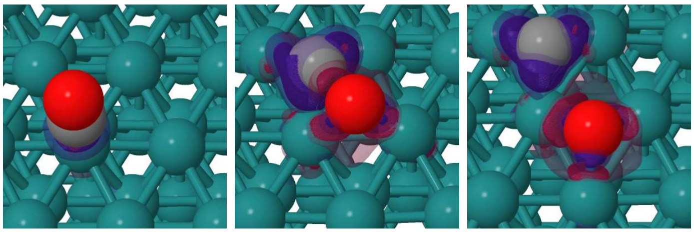
Fig. S52 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

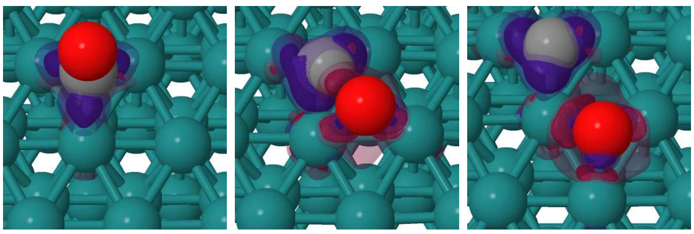
Fig. S53 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Anisotropic Compression on Ru(0001) Surface



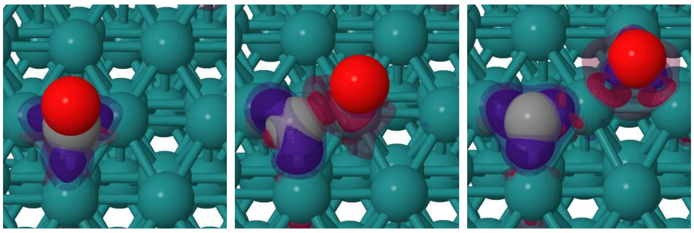
(a) (b) (c)

Fig. S54 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed terrace Ru(0001) surface for Path T1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



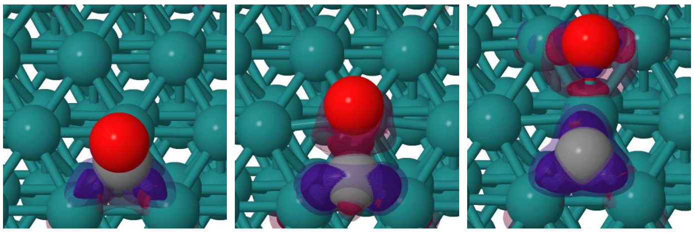
(a) (b) (c)

Fig. S55 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed terrace Ru(0001) surface for Path T2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

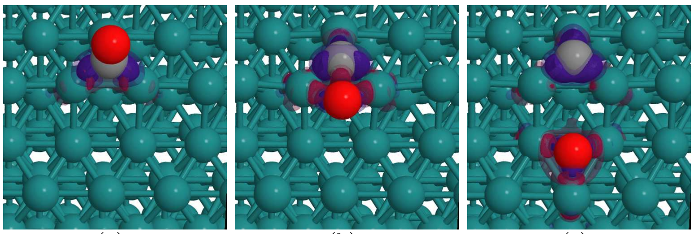
Fig. S56 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed terrace Ru(0001) surface for Path T3. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

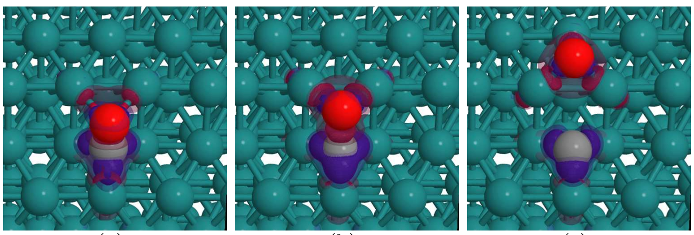
Fig. S57 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed terrace Ru(0001) surface for Path T4. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Relaxed Stepped Ru(1015) Surface



(a) (b) (c)

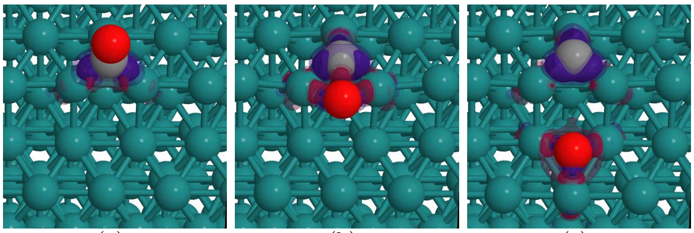
Fig. S58 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in relax stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

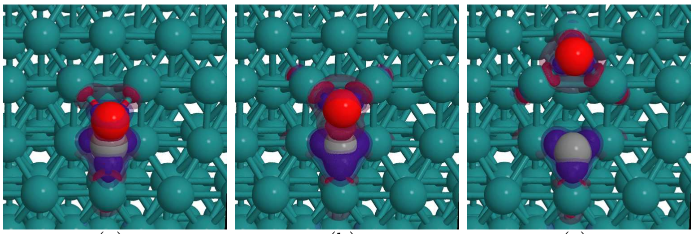
Fig. S59 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in relax stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Isotropic Expansion on Stepped Ru(1015) Surface



(a) (b) (c)

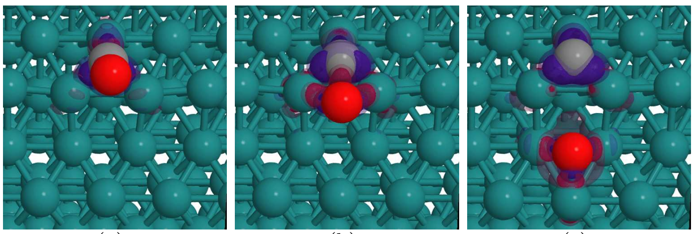
Fig. S60 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

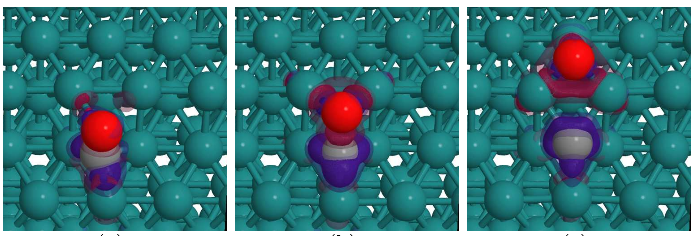
Fig. S61 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically expanded stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Isotropic Compression on Stepped Ru(1015) Surface



(a) (b) (c)

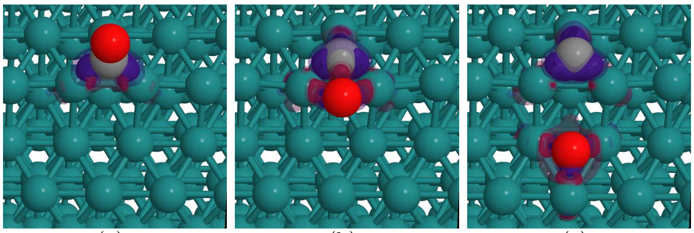
Fig. S62 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

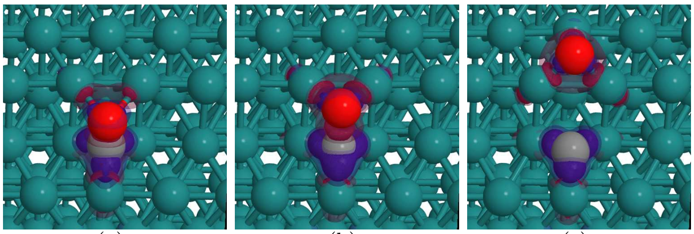
Fig. S63 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in isotropically compressed stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Anisotropic Expansion in a on Stepped Ru(1015) Surface



(a) (b) (c)

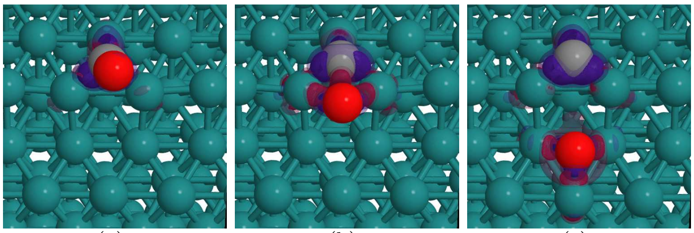
Fig. S64 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

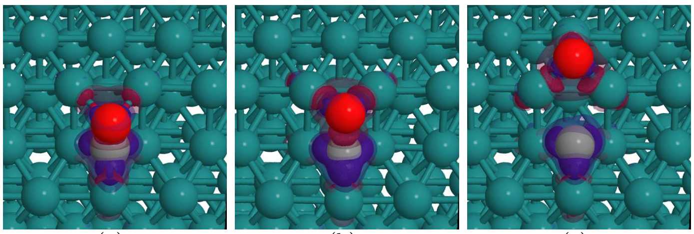
Fig. S65 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Anisotropic Compression in a Stepped Ru(1015) Surface



(a) (b) (c)

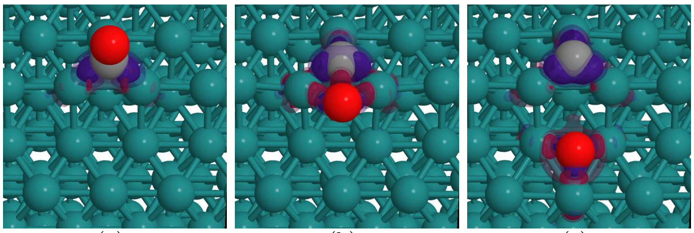
Fig. S66 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

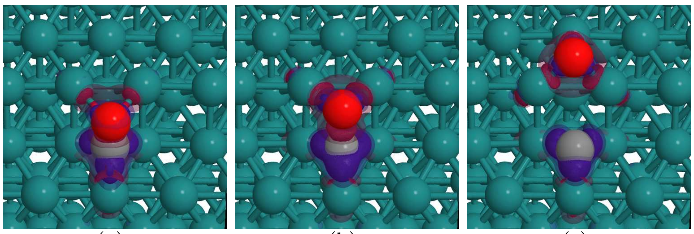
Fig. S67 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Anisotropic Expansion in b on Stepped Ru(1015) Surface



(a) (b) (c)

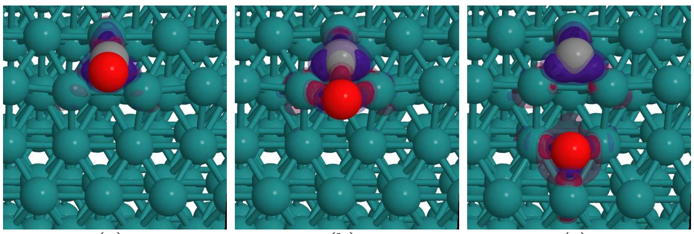
Fig. S68 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

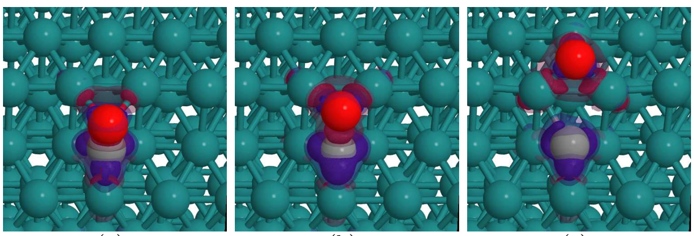
Fig. S69 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically expanded stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Anisotropic Compression in b Stepped Ru(1015) Surface



(a) (b) (c)

Fig. S70 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed stepped Ru(1015) surface for Path S1. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.



(a) (b) (c)

Fig. S71 Charge Density Difference Maps of (a) reactant, (b) transition state, and (c) product absorbed in anisotropically compressed stepped Ru(1015) surface for Path S2. Red, gray, and green atoms are O, C, and Ru atoms, respectively. Red and blue isosurfaces show increase and decrease of negative charge, respectively. Isosurfaces with isovalues of 0.01 and 0.005 are shown simultaneously with decreasing opacity.

Bader Charge Analysis

Terrace Ru(0001) Surface

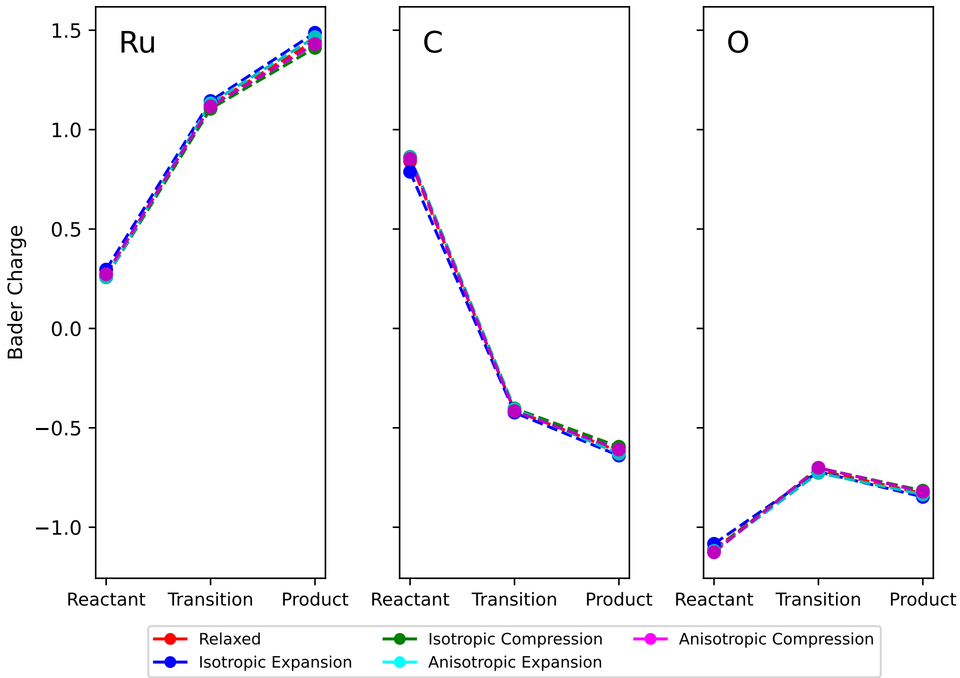


Fig. S72 Bader charge analysis along the reaction coordinates of Path T1 in terrace Ru(0001)

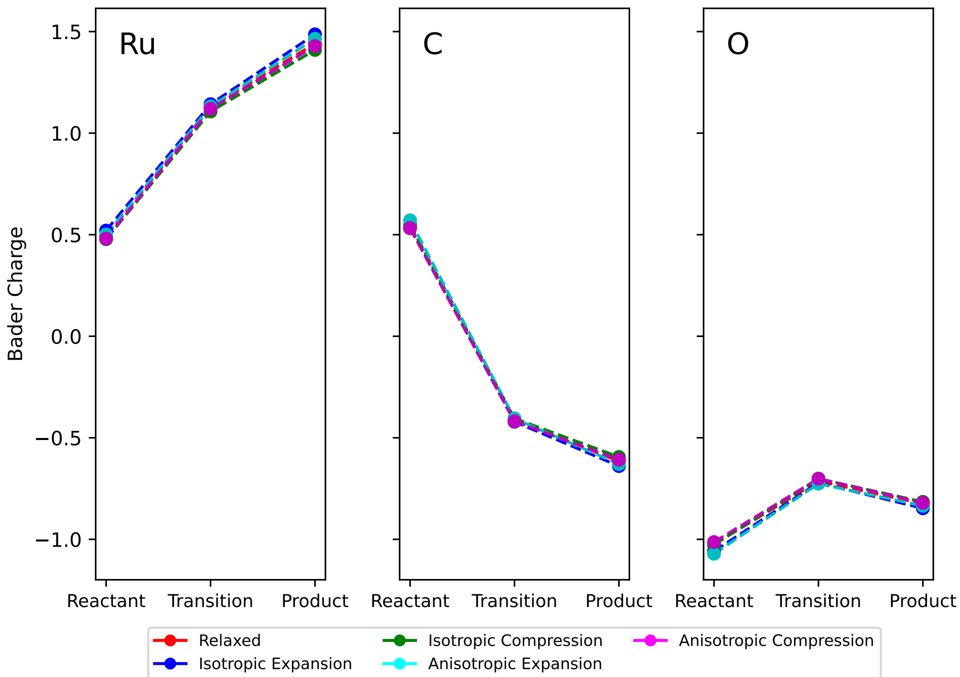


Fig. S73 Bader charge analysis along the reaction coordinates of Path T2 in terrace Ru(0001)

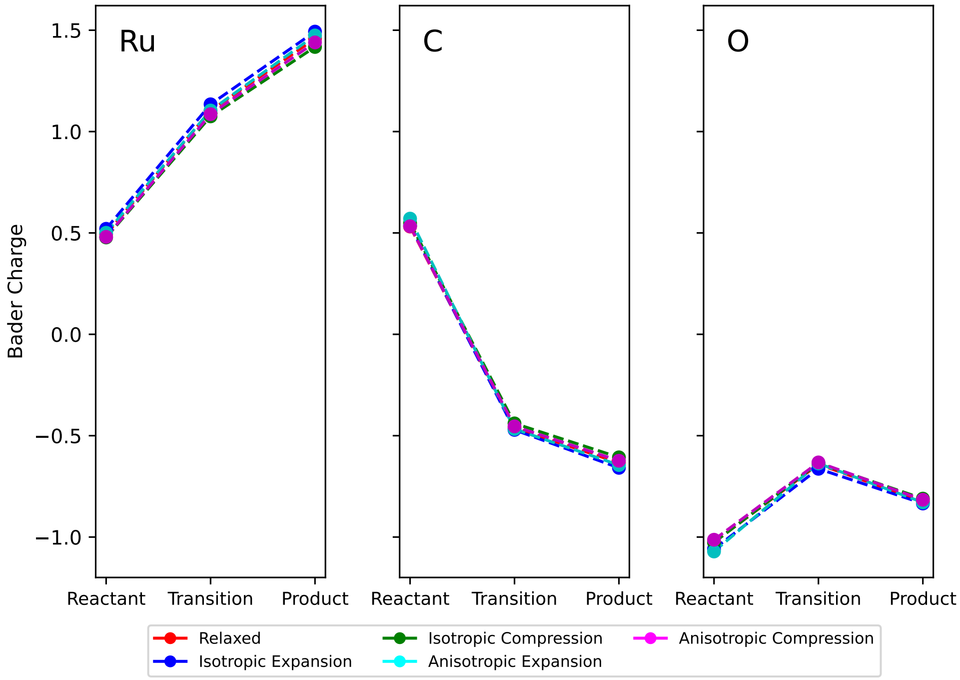


Fig. S74 Bader charge analysis along the reaction coordinates of Path T3 in terrace Ru(0001)

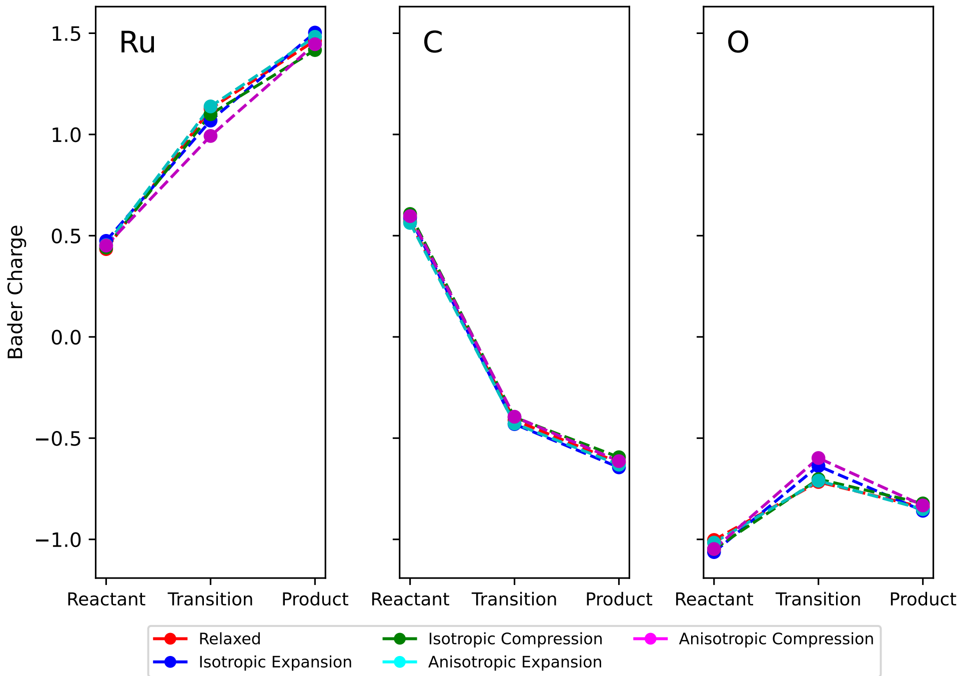


Fig. S75 Bader charge analysis along the reaction coordinates of Path T4 in terrace Ru(0001)

Stepped Ru(1015) Surface

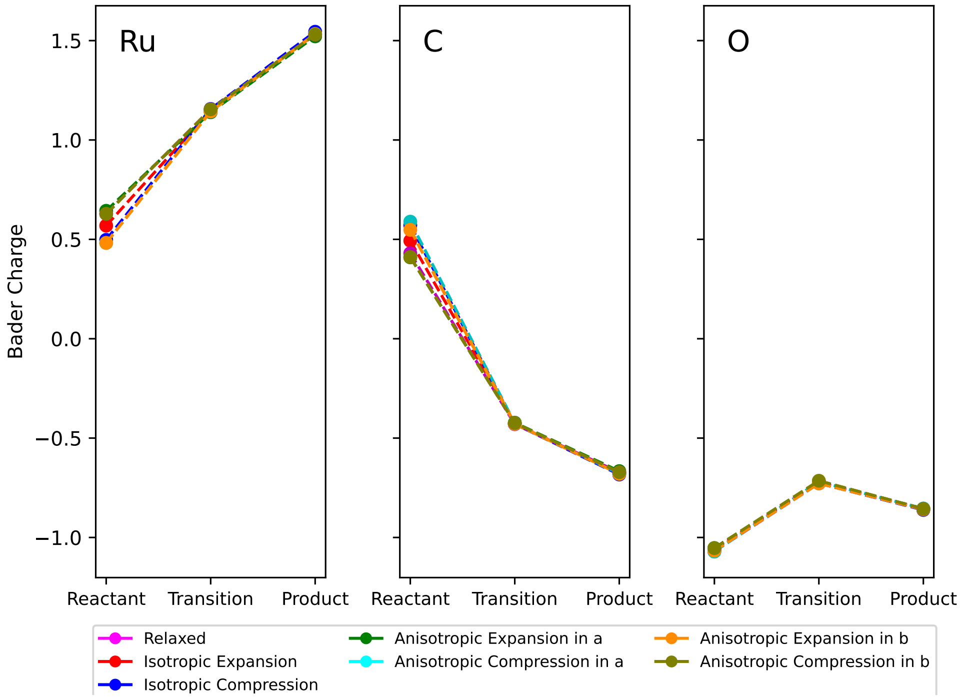


Fig. S76 Bader charge analysis along the reaction coordinates of Path S1 in stepped Ru(1015)

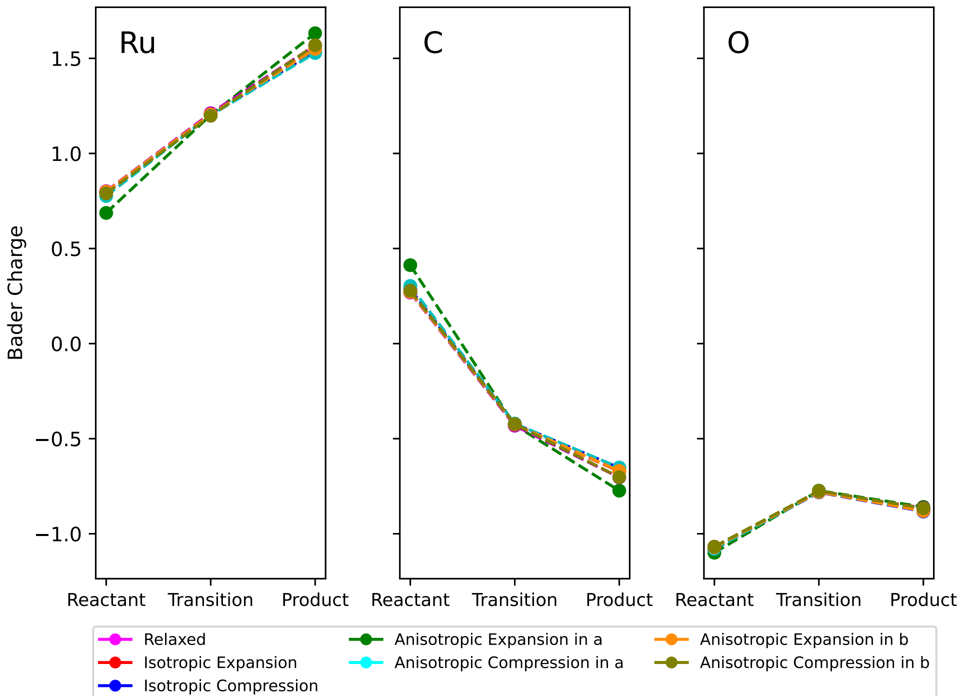


Fig. S77 Bader charge analysis along the reaction coordinates of Path S2 in stepped Ru(1015)

NEB Pathways

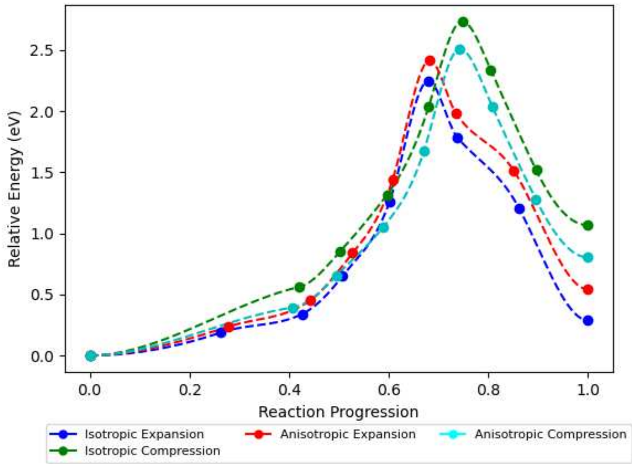


Fig. S78 NEB Pathways for Path T1 in Terrace Ru(0001) Surface

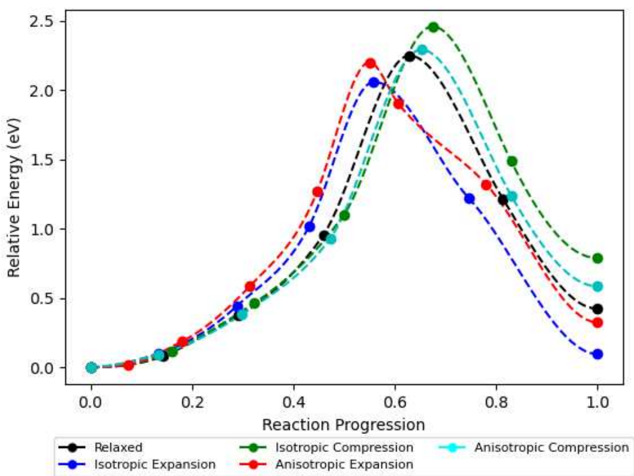


Fig. S79 NEB Pathways for Path T2 in Terrace Ru(0001) Surface

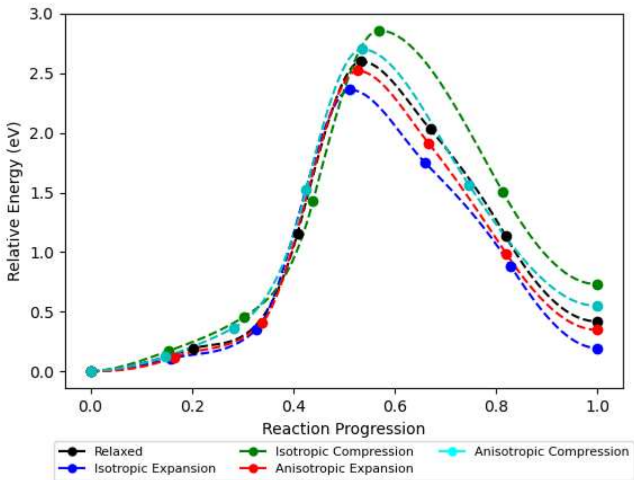


Fig. S80 NEB Pathways for Path T3 in Terrace Ru(0001) Surface

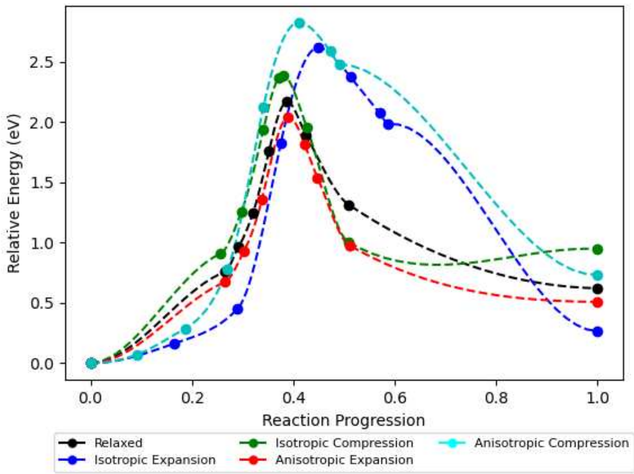


Fig. S81 NEB Pathways for Path T4 in Terrace Ru(0001) Surface

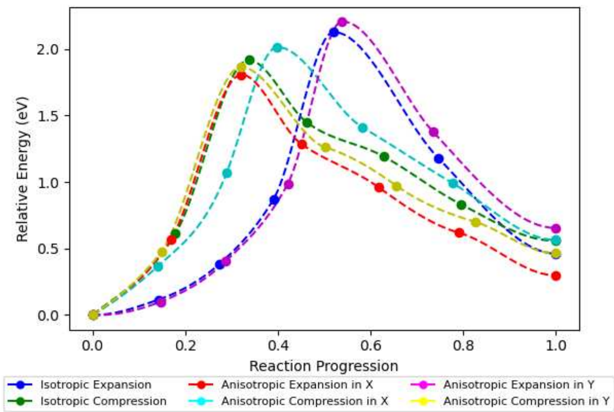


Fig. S82 NEB Pathways for Path S1 in Stepped Ru(1015) Surface

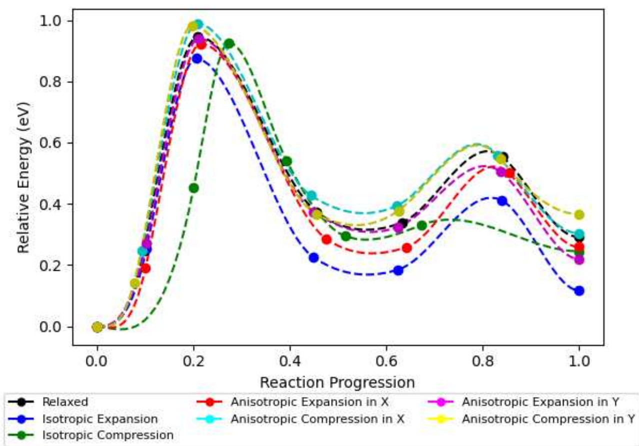


Fig. S83 NEB Pathways for Path S2 in Stepped Ru(1015) Surface