# Supporting Information

# Unraveling the Adhesion Characteristics of Ruthenium as an Advanced Metal Interconnect Material Using Machine Learning Potential

Eunseog Cho[1,*], Won-Joon Son[1,*], Seungmin Lee[1], Hyeon-Seok Do[1], Kyoungmin Min[2,*], and Dae Sin Kim[1]

[1]CSE team, Semiconductor R&D center, Samsung Electronics, 1 Samsungjeonja-ro, Hwaseong-si, Gyeonggi-do, 18448, Republic of Korea

[2]School of Mechanical Engineering, Yonsei University, 50 Yonsei-ro, Seodaemun-gu, Seoul 03722, Republic of Korea

*Corresponding author.
eunseog.cho@samsung.com (E. Cho), wonjoon.son@samsung.com (W. Son), kmin.min@yonsei.ac.kr (K. Min)

## Calculation Details

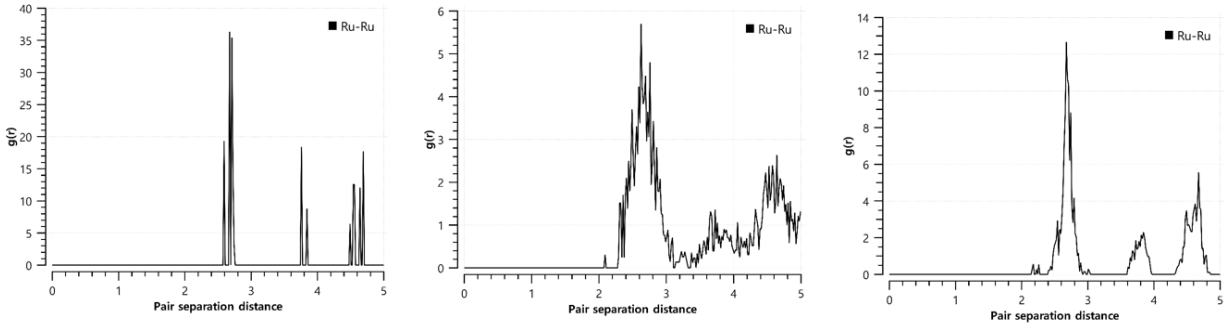
### I. Amorphous-like Ru

In this modeling, we constructed amorphous-like Ru structure and used it to build the interfacial Ru/$SiO_2$ structures. As mentioned in manuscript, previous studies of W/TiN reveal that W deposited on TiN does not exhibit the thermodynamically stable α-W structure but show an amorphous-like structures in X-ray diffraction.[1,2] In this regard, we assume that Ru on a-$SiO_2$ also has an amorphous-like phase; note that the amorphous-like structure does not represent a perfect amorphous structure but rather exhibits an imperfectly matched crystal structure. The amorphous-like Ru is constructed using the simulated annealing method. The MTP was developed with 2137 structures composed of various Ru phases like crystalline, molten, and quenched structures to accurately describe amorphous-like Ru structures. Bulk Ru structure was melted at 2500K temperature through MD simulation for 500 ps followed by gradual cooling down to room temperature of 300K. Then, another period of MD simulation was performed for an additional 500 ps using the quenching method resulting in the intermediate amorphous-like state between crystalline and molten states as shown in Figure S1.

### II. DFT calculations

The first-principles calculations were performed by the pseudopotential plane wave method, using the Vienna ab initio Simulation Package (VASP). We adopted the generalized gradient approximation (GGA) implemented by Perdew, Burke, and Ernzerhof (PBE) for the exchange-correlation energy functional with the D2 correction of Grimme to treat the van der Waals (vdW) interactions between Ru and a-$SiO_2$. The energy cutoff was selected to be 400 eV, and the atomic positions for bulk and slab structures were fully relaxed until the ionic force on each atom was below 0.02 eV/Å. The energy convergence criteria are set to be $10^{-5}$ eV. A dipole correction in the z-direction (perpendicular to the surface) is additionally used for an energy calculation of Ru/$SiO_2$ structures. A vacuum region of more than 15 Å is also selected along the z-direction to avoid spurious effects stemming from the periodic boundary condition. Ab initio MD simulations were performed ranging from 300K to 700K using a Nose-Hoover thermostat.

**Figure S1**. The RDF distribution of three different ruthenium structures; bulk Ru, molten Ru at 2500K, and quenched (amorphous-like) Ru at 300K.




[1] Kim, S.-H.; Kwak, N.; Kim, J.; Sohn, H. A Comparative Study of the Atomic-Layer-Deposited Tungsten Thin Films as Nucleation Layers for W-Plug Deposition. J. Electrochem. Soc. 2006, 153 (10), G887. https://doi.org/10.1149/1.2222966

[2] Kim, S.-H.; Yeom, S.-J.; Kwak, N.; Sohn, H. Phase and Microstructure of ALD-W Films Deposited Using B2H6 and WF6 and Their Effects on CVD-W Growth. J. Electrochem. Soc. 2007, 155 (2), D148. https://doi.org/10.1149/1.2815962

**Figure S2.** The variation of the total number of data as the active learning progresses.

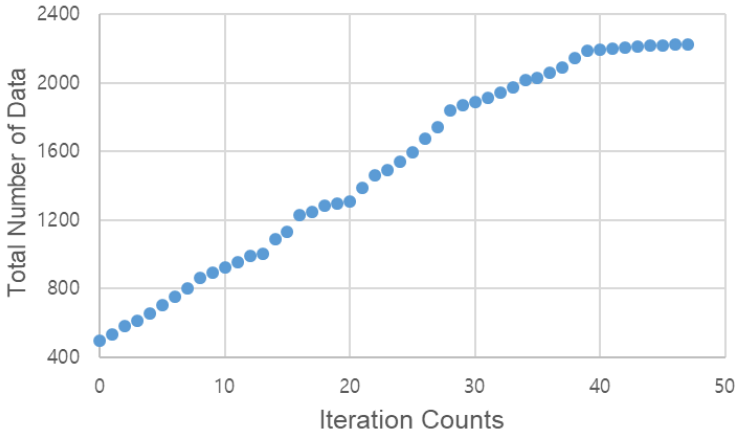
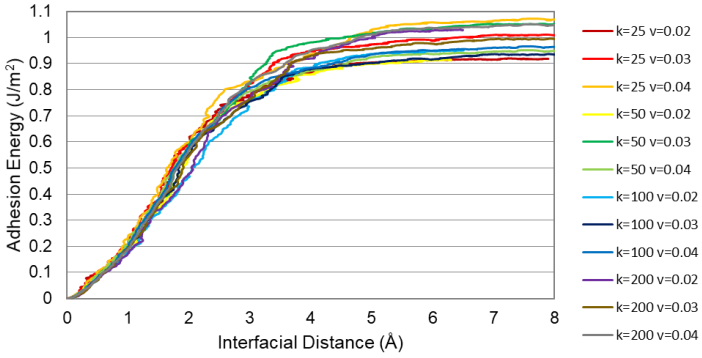


**Figure S3.** (a) Adhesion energy profile vs. interfacial distance and (b) adhesion energy depending on different SMD parameter sets. The k and v represents the spring force constant and the velocity, respectively.

(a)




(b)

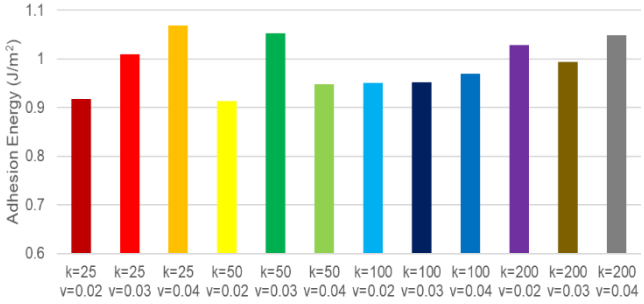


**Figure S4.** Adhesion force profile as the ratio of $O_{DB}$ ($O_{DB}$ ratio: 0.14, 0.29, 0.57, and 0.86) to the total surface oxygen atoms ($O_{tot}$) with $O_{tot}$ density of 7.9 $nm^{-2}$.

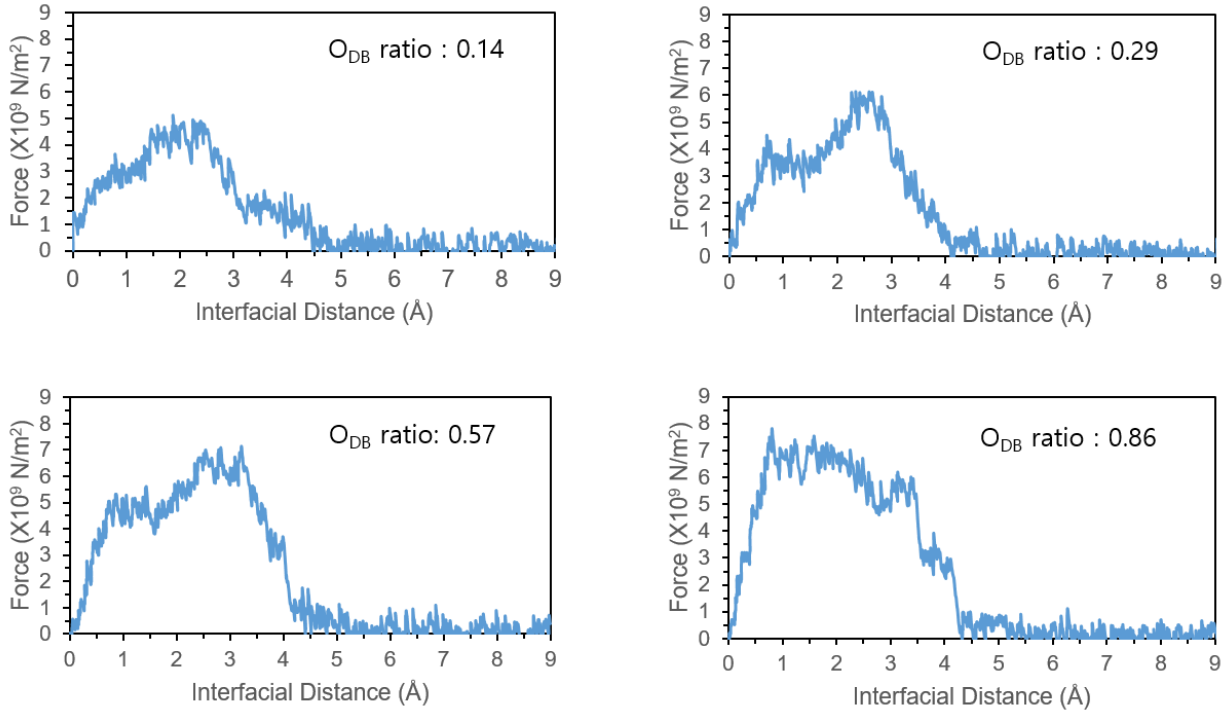


**Figure S5.** Adhesion energy profile as the ratio of $O_{DB}$ ($O_{DB}$ ratio: 0.14, 0.29, 0.57, and 0.86) to the total surface oxygen atoms ($O_{tot}$) with $O_{tot}$ density of 7.9 $nm^{-2}$.

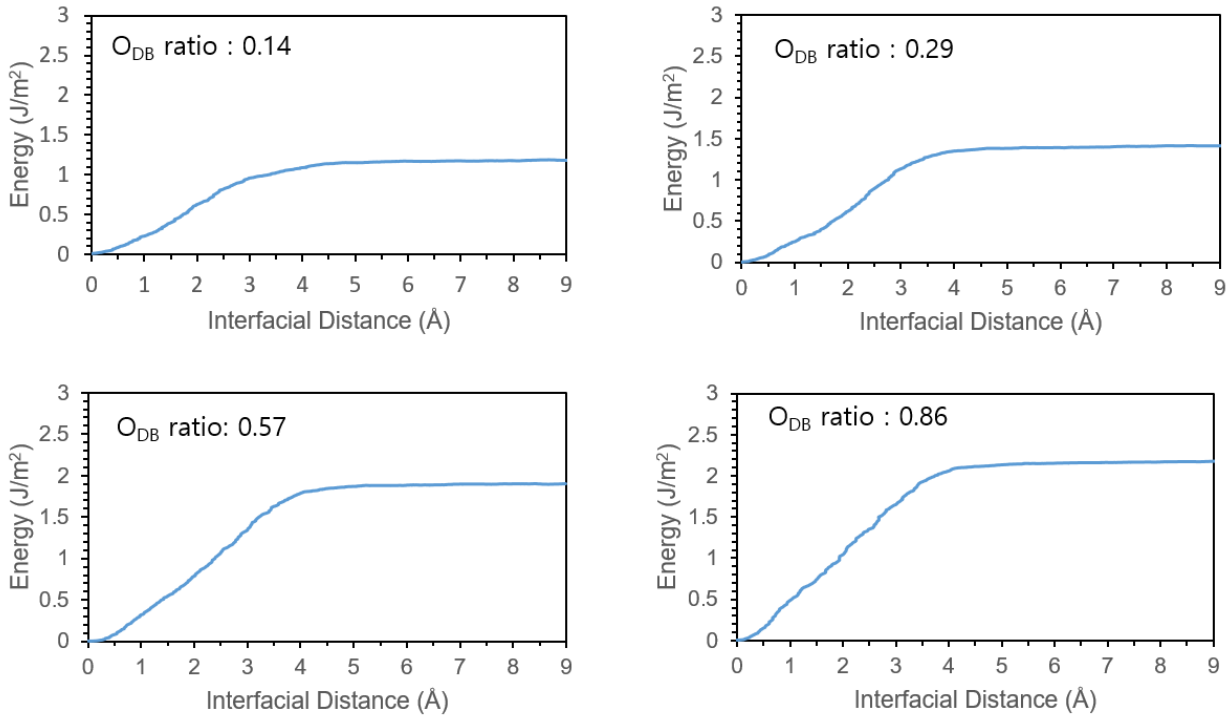


**Table S1.** The simulation cell size for interfacial area and number of atoms for different $Ru/SiO_2$ structures. The cell size of the perpendicular direction to the interfacial area is fixed to 80 Å for all structures.

| $O_{tot}$ ($O_{DB}$+OH) density (#/$nm^2$) | $O_{DB}$ ratio ($O_{DB}/O_{tot}$) | cell size | # of atoms |
|---|---|---|---|
| 7.2 | 0 | 13.71Å X 28.07Å | 684 |
| | 0.14 | 13.71Å X 28.07Å | 680 |
| | 0.29 | 13.71Å X 28.07Å | 676 |
| | 0.57 | 13.71Å X 28.07Å | 668 |
| | 0.86 | 13.71Å X 28.07Å | 660 |
| | 1.0 | 13.71Å X 28.07Å | 656 |
| 4.5 | 1.0 | 13.39Å X 26.57Å | 632 |
| 1.5 | 1.0 | 12.43Å X 26.24Å | 608 |