

Please peer review this manuscript for a reputable periodic. Insert date.

Edit Claude Hayku 3.7.

I'll help you review the manuscript, focusing on its scientific rigor, novelty, clarity, and overall contribution to the field of computational catalysis.

Review Date: Thursday, June 26, 2025

Manuscript Title: "Bridging Scales in Heterogeneous Catalysis: A Multiscale Computational Framework for Operando Conditions"

Author: Richard Murdoch Montgomery

Overall Assessment: Highly Recommended for Publication

Strengths:

1. Innovative Methodology
  - Develops a comprehensive multiscale framework integrating DFT, machine learning potentials, and kinetic Monte Carlo simulations
  - Addresses a critical limitation in current computational catalysis approaches by capturing dynamic surface phenomena
2. Rigorous Computational Approach
  - Systematic comparison of three models (Static, Static Lattice + CE, Dynamic Operando)
  - Extensive computational parameters detailed in Table 1
  - Robust machine learning potential development using active learning
3. Significant Scientific Insights
  - Reveals non-monotonic activation energy dependence on coverage
  - Demonstrates surface restructuring's fundamental role in catalytic function
  - Shows limitations of static surface models
4. Experimental Validation
  - Turnover frequencies closely match experimental measurements
  - Provides actionable strategies for optimizing reaction conditions

Potential Improvements:

1. Discuss entropy treatment limitations more comprehensively
2. Elaborate on generalizability beyond Pt(111) system

### 3. Consider additional DFT functional comparisons

Recommendation: Accept with Minor Revisions

The manuscript represents a significant advancement in computational catalysis methodology and provides a compelling argument for considering surface dynamics in catalyst design.