

REVIEWER REPORT

EVALUATION:

Reviewer's Responses to Questions

Please rate the importance compared to published work in this subject area

Reviewer #1: High (Top 20%)

Reviewer #2: High (Top 20%)

Please rate the novelty compared to published work in this subject area

Reviewer #1: High (Top 20%)

Reviewer #2: High (Top 20%)

Which aspects of scholarly presentation require improvement (if any)?

Reviewer #1: Clarity

Reviewer #2: (No Response)

Do the methods, data and analysis (including statistical analysis where applicable) adequately test the hypothesis and support the conclusions?

Reviewer #1: Yes

Reviewer #2: Yes

Are the methods, data and analysis described in sufficient detail to be reproduced?

Reviewer #1: Yes

Reviewer #2: Yes

Please indicate which other journal you consider more appropriate

Reviewer #1: (No Response)

Reviewer #2: (No Response)

Please indicate whether you have included attachments

Reviewer #1: No

Reviewer #2: No

Responses to referees' comments

Referee 1

The manuscript described the synthesis and reactivity of an imino-carbene supported low valent silicon species 3, which displayed the chemical behaviors of both mesoionic silylene and silylone. The low valent silicon compound has been studied by DFT calculations and a range of reactions with transition metal complexes and main group compounds S8, RSeSeR, RTeTeR et al. The title compound showed both silylone and silylene reactivity and might be very useful for the synthesis of new silicon heterocycles. All of the new compounds reported in the manuscript have been well characterized by NMR and X-ray single crystal analysis. The manuscript is well-organized and the SI is solid. However, as mentioned in the introduction part, the very similar Ge compound B

(Figure 1) by Kinjo was reported ten years ago, silicon compound 3 and germanium B have a very similar electronic structure. In addition the reactivities of the two compounds are also quite related. Thus, the authors should compare two compounds in the manuscript, and probably should extend the reactivity studies on 3, and delineate the novelty of the silicon species and differences of the two species. The detailed explanations on the differences between the structures and reactions of the two species should significantly improve the quality of the manuscript.

Referee 2

This work by Liu, Liand and coworkers describes the synthesis and reactivity of a mesoionic diazasilole. Although this compound class was described as a reaction product of a bisilylene with CO in 2022 by Jones, the revealed reactivity is much extended. Further, it differs from the previous example by containing only one low-valent silicon instead of two, which is expressed in different coordination chemistry. The manuscript is enjoyable to read, and the compounds are well characterized. Hence, given the broadness and novelties of the described reactions and the computational elucidation, it meets the high expectations for publication in ACIE. Before acceptance, I would suggest the following changes:

1. The references listed under [1] are not ideal, at least 1a and 1b. Rather include KutzIniggs ACIE paper from 1984 (for sp-hybridization defect) and some general Si review (for the general chemistry, hypercoordination).
2. Page 2, right, line 3: the sentence starting "More recently, Tobisu..." is not ideal.
3. I am unsure whether it is fitting and necessary to name the reaction with HNEt₃Cl as "skeletal rearrangement" (Figure 1). I think this is a buzzword that won't find too many friends addressed by this manuscript.
4. "In conclusion, our research has synthesized" ...it is rather "we have synthesized" or "as part of our research..."
5. Having an elemental analysis, at least for compound 3 would be ideal.