# How To Make Your Computational Paper Interesting – and Have It Published

In 1987, the German chemist Peter Hofmann wrote: "*We still have a long way to go until a computation will be able to compete with or to substitute a lab experiment [...] one could then conclude that theoretical work in this field - not being quantitatively reliable anyhow - is rather useless, except for the purpose of keeping theorists busy.*" However, he proceeded "*If applied properly and with their limitations in mind, methods of various levels of sophistication can all contribute their part to a basic understanding of organometallic systems*".[1]

Without doubt, the last 30 years have shown the usefulness of computational methods to provide information about the properties of molecules, including the activities and selectivities of organometallic systems.[2,3,4] Since the late 1990's, the preferred method of computational chemists has been Density Functional Theory (DFT).[4,5] A search of papers containing DFT in *Organometallics* shows an increase from 113 papers in 2000 to 379 in 2012 (Fig. 1).[6]

*Organometallics* is now receiving an ever increasing number of manuscripts that report DFT calculations in whole or in part. Unfortunately, computational manuscripts sometimes are written in a way that is not very appealing to experimental readers (or reviewers and editors). Further, computed results often are not sufficiently correlated with experimental data. **The consequence may be a rejection of the manuscript.** How can you increase the likelihood that your computational study is publishable in *Organometallics*? 3 things are essential:

- The scientific problem should be intriguing
- The presentation should be appealing
- The computed results should be validated

The following provides (personal) advice on how to make a computational paper intriguing, appealing, and validated. The main focus is on mechanistic papers.

**The Scientific Problem:** A paper whose scientific hypothesis is restricted to: "*We computed the mechanism that the experimentalist proposed, and it seems to work okay*" is not very exciting to read. Use computations to study **an interesting problem**, and state it clearly in the text. For example, "*Why does catalyst 1 convert ketones, but not aldehydes, which should be more reactive?*".[7] Choose problems where theory can provide answers that experimentalists would love to have, but cannot access easily.

**The Presentation:** An abstract or discussion should be easy to follow. "*Our calculations show that Int1 becomes Int2 via TSab_II, whereas the pathways through Int7 and*
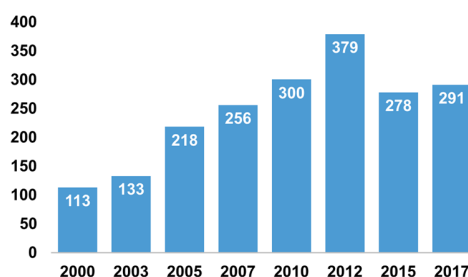


**Figure 1.** Approximate number of articles containing DFT in *Organometallics.*[6]

*TS19 are clearly disfavored*" is everything but clear. Try instead: "*The preferred pathway proceeds via a carbene intermediate*". Give the exact experimental conditions in your discussion: solvent, temperature, mol% catalyst. The reader wants to know what you are trying to model. Refer to those that proposed the mechanism, also in figure captions. **Talking about figures:** Show clear pictures of relevant transition states (TSs), but instead of Ball-and-Stick, consider Chemdraw figures, which are easier accessible to experimentalists (Fig. 2). Avoid overloading energy profiles with 4-5 different pathways. A mechanistic figure should not require half an hour of contemplation (Fig. 3), so let an experimental colleague evaluate your figures and text before submission. Give the interested reader the possibility to visualize structures by providing them in a separate Supporting Information (SI) file that can be opened with e.g. Mercury (see ref. 8). **Most importantly, keep it short.** Probably, you would not be excited to read 15 pages about experiments that failed. Do not expect that an experimentalist wants to read 15 pages about computations that are not relevant. For non-preferred mechanisms, refer briefly to the results, but place the details in the SI.
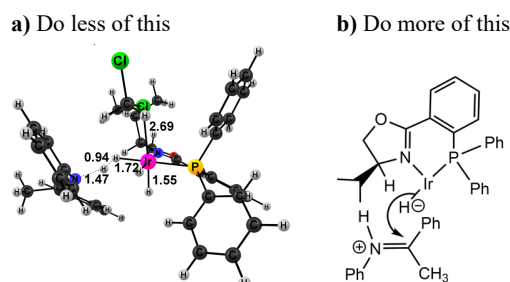


**a)** Do less of this   **b)** Do more of this

**Figure 2**. **a)** A Ball-and-Stick versus **b)** a Chemdraw figure of the same TS (axial ligands omitted, from the author's own work[9]).

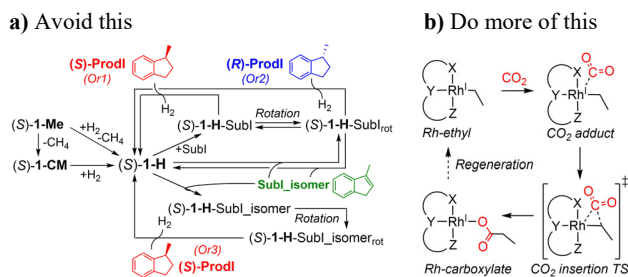**a)** Avoid this    **b)** Do more of this

**Figure 3**. **a)** An incomprehensible figure by the author that she is not very proud of today.[10] **b)** A comprehensible figure.[11]

**The Methods and Models:** The model should be as close as possible to the real system. If in experiments, 4-chlorostyrene was converted, but styrene did not react, do not base your mechanistic investigation on styrene. Choose an appropriate DFT method. Today, this normally means including dispersion corrections.[12,13] **Do not use DFT for mechanisms it cannot handle.** Be careful if many ligands enter or leave; this is difficult for conventional DFT to describe accurately (therefore, use a large basis set and correct for basis set super position errors[12,13]). Avoid using static DFT for a reaction with many loosely interacting components (e.g. a catalyst, substrate, additive, and solvent molecule), as these may form *tenth* of different supramolecular complexes; this requires dynamics to describe properly. As a general rule, do not include explicit solvent, unless it is strongly bound to a complex.

**The Results: Be critical** to your results! Is the computed barrier meaningful? At 298K, a barrier should be below 25 kcal/mol.[14] Also if it is below, make sure to test mechanistic alternatives. Always discuss in the manuscript if the results really are in agreement with experiment. If an additive speeds up the reaction by a factor of 4, this corresponds to a lowering of the barrier by 0.8 kcal/mol (298K). If your computations predict that the additive lowers the barrier by 9 kcal/mol, the trend may look right, but this is a factor of 4 million and hence not in agreement with experiment. If you locate an intermediate that has a low energy, do not ignore it. If it is easy to form, it needs to be considered as an on- or off-cycle species. If this makes your barriers too high, your mechanism may be incorrect.

Most importantly: **always validate the final mechanism.** How? *i)* Compare computed intermediates to experimentally observed intermediates. *ii)* Show that the proposed mechanism can reproduce substrate preferences and selectivities. If in experiments, ketones are better substrates than aldehydes, your mechanism needs to reproduce that. When you compute enantiomeric excesses (*ee*'s), estimate the error in kcal/mol. If you wanted 33% *ee* (*R*), but got 10% *ee* (*S*), you may be worried, but the error is only 0.5 kcal/mol (298K). If you wanted 99.5% *ee* (*S*) and got 90% *ee* (*S*), you may not be worried, but maybe you should be, as the error is 1.8 kcal/mol. This may indicate your TS or

mechanism is incorrect. Some computational papers make predictions that have not yet been tested experimentally. This should be done in combination with studying something known. If your computed results about the known ligand **A** match experiment well, your predictions about the novel ligand **B** are more credible. The most compelling papers may be those where computations and experiments are employed together.[2,15] This provides the opportunity to use theory to immediately rationalize interesting experimental results, and to use experiment to immediately validate interesting computational predictions.

**The Proposed Reviewers:** A computational study is often based on a specific experimental paper. Surprisingly, computational authors sometimes indicate the experimentalist as a *non-preferred* reviewer. Note that editors can disregard this request. If you instead **indicate the experimentalist as a preferred reviewer**, it shows that you are not worried to have the results scrutinized by an expert on this system. And if he/she finds the work sound and convincing, this is a quality stamp - and increases the likelihood that your results will impact future experimental setups. Isn't that a main goal of any computational study?

*Organometallics* **is looking forward to receive your intriguing, appealing, and validated computational study.**



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**Notes**
Views expressed in this editorial are those of the author and not necessarily the views of the ACS

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(6) Search performed at https://pubs.acs.org/search/advanced with search string *"Density Functional Theory" OR DFT*. Reviews, editorials and corrections were excluded from the results. Analysis of the search results for 2000 and 2003 reveals that circa 10% of returned articles referred to DFT results by others, but did not report own computations. Numbers shown in Figure 1 are therefore approximate.

(7) This and all text examples given here are invented by the author and do not originate from real manuscripts. Any similarity is purely accidental.

(8) This is explained in the author guidelines of *Organometallics*: https://pubs.acs.org/paragonplus/submission/orgnd7/orgnd7_authguid e.pdf

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