

APPLICATIONS ARE PREFERABLY DRAWN UP IN ENGLISH. AN ENGLISH TRANSLATION HAS TO BE ENCLOSED WITH APPLICATIONS SUBMITTED IN DUTCH.

The application form is available in English on the website <https://vscentrum.be/>.

Title of the application:

[Modeling aldol condensations in metal-organic frameworks with hybrid functional calculations](#)

Name and first name of the applicant:

[Hajek Julianna](#)

Institution:

[Ghent University](#)

Research group / department:

[EA17, Center for Molecular Modeling](#)

Title / position:

[PhD fellow](#)

e-mail address:

Julianna.Hajek@ugent.be

Total computing time that is needed, in node days:

[4608](#)

Total disk storage that is applied for:

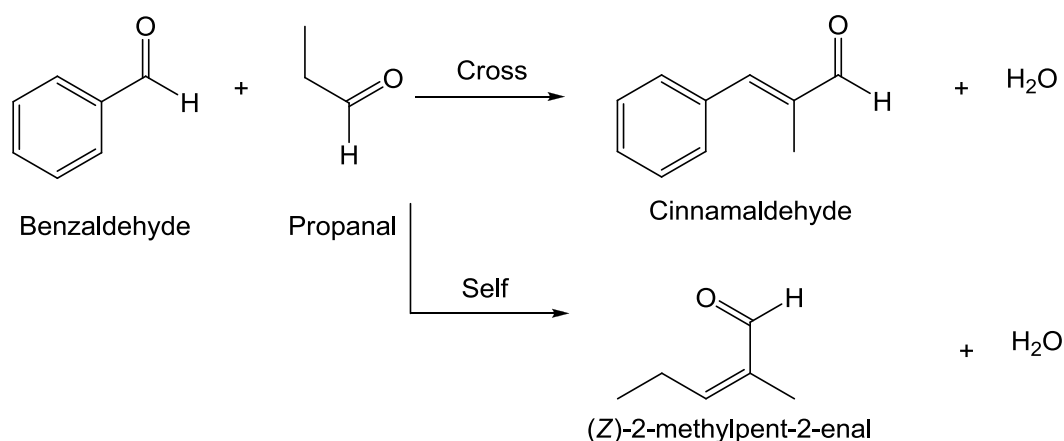
[At maximum 0.5 TB will be needed on the gpilot scratch](#)

1. Title of the research project (with IWETO link if available) within the framework of which computing time is applied for

Combined experimental and computational study of electronically modulated metal-organic framework (MOF) catalysts, FWO project 3G048612 between COK (Prof. Dirk De Vos) and CMM (Prof. Veronique Van Speybroeck)

2. Short description of the research project within the framework of which computing time is applied for (max. 1 A4 in Arial 12):

In this project, we want to study of the Metal-Organic Frameworks (MOFs) UiO-66 and UiO-66-NH₂ for aldol condensation reactions. We chose to investigate the cinnamaldehyde condensation between benzaldehyde and propanal (Scheme 1).



Scheme 1: Aldol condensation between benzaldehyde and propanal (cfr. cross and self aldol condensation).

The catalytically active sites of such Zr-terephthalate UiO-66 materials were identified as accessible Zr sites that arise during the synthesis of these materials (*Angewandte Chemie Int. Ed.*, 51(20), 4887-4890, **2012**). On these active sites, we have already computed a catalytic cycle for the aldol condensation reactions on both periodic and cluster models of the UiO-66. An extension of this research to the amino-functionalized counterpart is warranted, as this material was found to be more active and selective (*Chem. Commun.*, 47, 1521-1523, **2011**).

This will enable us to unravel the role of the amino group during the catalytic cycle.

To achieve a good comparison between the UiO-66 and the amino-UiO-66 for their selectivity and activity, we need accurate hybrid functional calculations using the periodic code VASP. Why? First, this brings the results of periodic models on a comparable level of theory with the cluster models. Second, as the environment is better accounted for in periodic simulations, it makes it possible to estimate the effect of extra stabilization on the activation barriers compared to cluster calculations. These two issues are of particular importance, since accurate reaction barrier prediction is necessary if we want to compare experimental and theoretical results.

3. Financing institution or channel, financing the research project in full or in part (FWO, BOF, IWT, EU, ...): Please attach the confirmation letter as enclosure (see instructions in enclosure 3).

- FWO project 3G048612, confirmation letter can be found at the end of the file.
- Personal FWO postdoc fellowship of Matthias Vandichel, confirmation letter can be found at the end of the file.

4. Promoter of the research project:

Prof. Dr. ir. Veronique Van Speybroeck

Prof. Dr. Dirk De Vos

5. Persons mandated by the Applicant to compute on the TIER1 within the framework of the present project: Please provide for every person:

- name and first name
- institution
- research group / department
- title / position
- experience with TIER1/TIER2 infrastructure in Belgium and abroad

Julianna Hajek
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Experience with Tier1 and Tier2 infrastructure of UGent

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Veronique Van Speybroeck
Ghent University
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Professor
vsc40021
Experience with Tier2 infrastructure of UGent

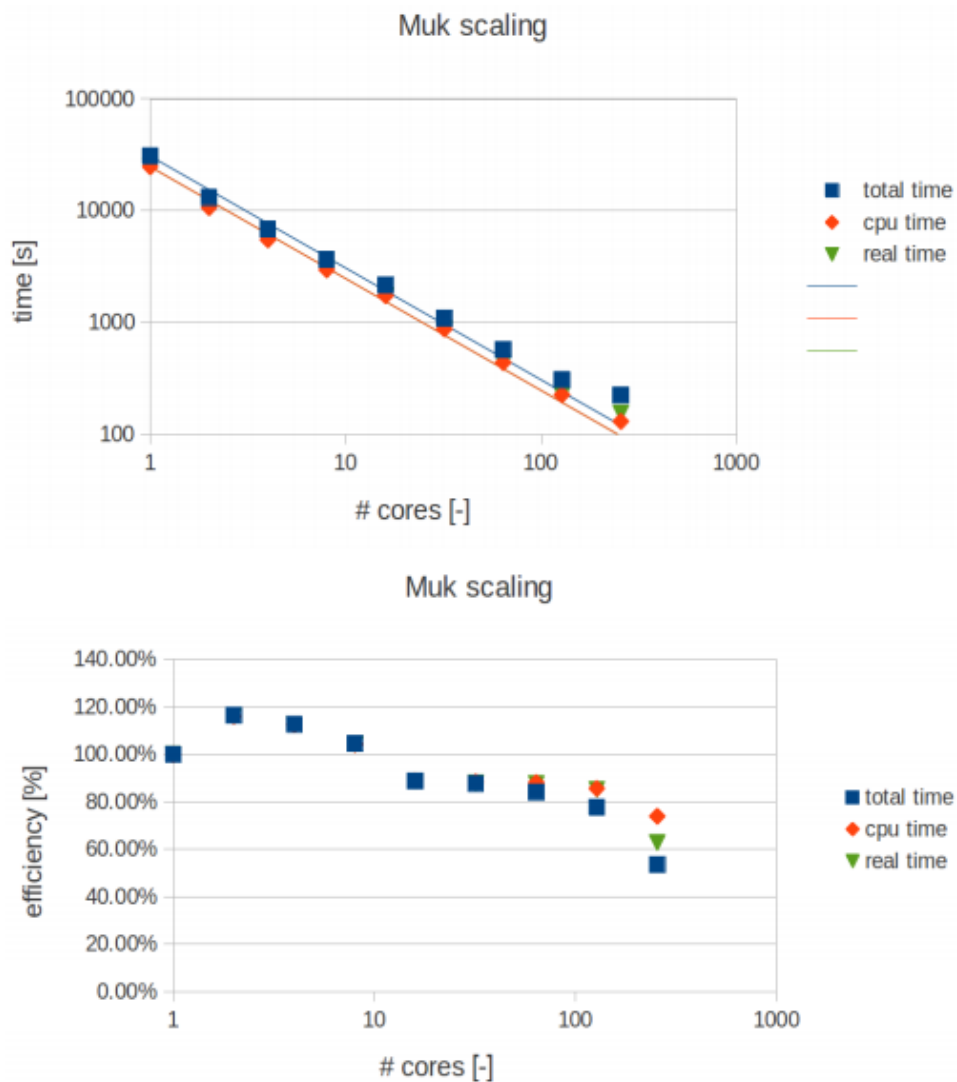
6. Description of the computing task, justification for the computing time, disk storage and memory that are applied for, and description of the software tools required (max. 3 A4 in Arial 12). Please clearly provide the following in this regard:
 - the number of nodes/cores that are applied for per computing task, with a subdivision of the computing time in sub-tasks indicating the sequence of the sub-tasks
 - whether these tasks use diversification (OpenMP, MPI, hybrid OpenMP/MPI ...)
 - the estimated memory use of a computing task (maximum 64GiB/node)
 - whether a vSMP system will be used
 - the requirements for disk storage (estimated volume in GiB and the total number of files), more specifically for:

- required input files (data set, parameter files, etc.)
- SCRATCH volume used during the performing of the computing tasks
- result files

Gavin J. Pringle provided details of the scaling behaviour of the hybrid functional code in VASP;

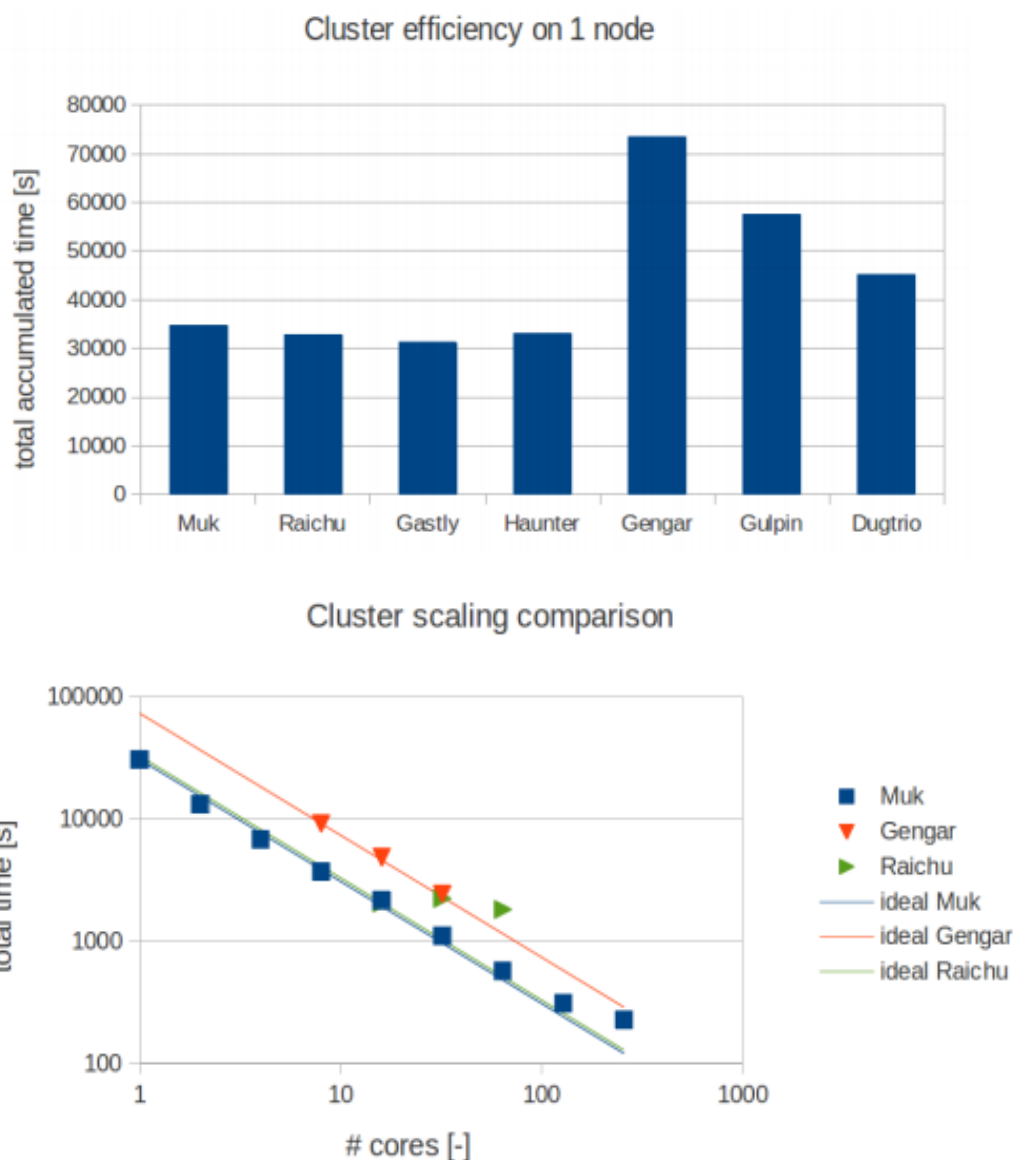
<http://www.hector.ac.uk/cse/distributedcse/reports/vasp02/vasp02.pdf>.

Recently Kurt Lejaeghere has done the Muk-specific test to assess the parallelization of the VASP-code. The graphs with the results are shown below.



Muk displays a very good scaling behaviour up to 100 cores.

Cluster comparison:



For our particular systems, we did small scaling test at the PBE0 level of theory. The scaling efficiency from going from 1 node to 4 nodes on the Tier-2 cluster Delcatty is still good (we obtained efficiency values higher than 85%), so we will use parallelization up to 4 nodes (64 cores) for the calculations.

Based on this info and initial VASP-benchmark calculations from our previous TIER1 project, we could make a fair estimate of the computational time needed (Table 1) and the memory use (Table 2).

Table 1: Estimated node and core days for the project.

Estimated computational time for one of the 18 intermediates in the catalytic cycle.					
Catalytic Cycle	Optimization PBE0 (#nodes x node-days)	Frequency PBE0 (#nodes x node-days)	Single point SCF B3LYP (#nodes x node-days)	Total (node days)	Total (core days)
Normal UiO-66 self aldol condensation (234 atoms)	4 x 4	4 x 1.5	4 x 2.5	18x32	18x32x16
Normal UiO-66 cross aldol condensation (230 atoms)	4 x 4	4 x 1.5	4 x 2.5	18x32	18x32x16
Amino-UiO-66 self aldol condensation (252 atoms)	4 x 4	4 x 1.5	4 x 2.5	18x32	18 x32x16
Amino-UiO-66 cross aldol condensation (248 atoms)	4 x 4	4 x 1.5	4 x 2.5	18x32	18x32x16
Estimate of total computational time needed				4608	36864

Table 2: Estimated memory use

	Maximum Memory/core (MB)	SCRATCH (GB)	Permanent storage (GB)
Optimization PBE0	1088	1-1.5	0.3
Frequency PBE0	1088	1-1.5	0.3
Single point SCF B3LYP	1088	1-1.5	0.3
Total (18 x 4)**		216-324	65

** 4 catalytic cycles containing each 18 structures

As one job requires approximately 1-1.5 GB of storage, we estimate to need at maximum around 0.5 T of temporary storage on the pilot scratch.

We will not use a vSMP scheme.

7. Please indicate why the TIER1 is the appropriate machine to perform the computing task (max. 1/2 A4 in Arial 12):

In this work, hybrid functional calculations on periodic models are needed (1) to truthfully compare the periodic and cluster calculations; (2) to more accurately describe environmental effects. Due to the size of the systems and the amount of calculations that need to be performed, it is clear that these calculations go beyond what can comfortably be done (within a reasonable timeframe) on a TIER2 cluster.

8. Summary of the software required to perform the computing task, and possible installation and compilation instructions (max. 2 A4 in Arial 12). Please clearly provide the following per item in this regard:
- a reference to the software's web page
 - the software licence system (open source, GPL, etc.)
 - if there is no free academic use of the software, state which licence makes the installation and the use valid on the TIER1 by the Applicant (+ add a copy of the signed licence)
 - if need be, which licence server will be used (name + IP address)
 - whether the software is already available on the TIER1 (see <https://vscentrum.be/nl/tier1-rekenen>) and, if this is not the case, compilation and installation instructions (possibly with reference to existing TIER2 installation)

VASP 5.3.3

- <http://www.vasp.at>
- License: see at the end of the file
- The software is already available on the TIER1 infrastructure.

9. Period during which the task is to be performed:

Optimally the calculations would start in July and end in January (~ 6 months). Our intension is to submit the research papers during this period, so that we can address the questions from the reviewers before the end of computing period.

10. Describe the results that were obtained within the framework of computing time that was attributed during the past two years on the TIER1 or on other TIER1 or TIER0 supercomputers (max. 2 A4 in Arial 12):

In our earlier TIER1 project, we successfully identified the right collective variables to model the citronellal cyclization within the metadynamics simulations on different UiO-67-X materials (NPT ensemble). We obtained good estimates for the reaction barriers, accounting for both framework and linker flexibility. This type of computations goes beyond the state of the art. We are finalizing the data-mining and writing a research paper. Our intention is to publish this paper in JACS (Journal of the American Chemical Society).

Should you have any questions or encounter any difficulties during the electronic submission of an Application, please contact by e-mail:

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