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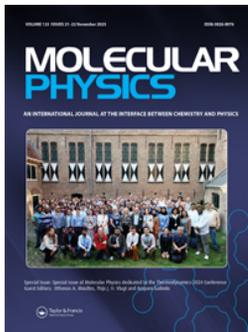
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## The RASPA2024 workshop in Delft, The Netherlands

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### ABSTRACT

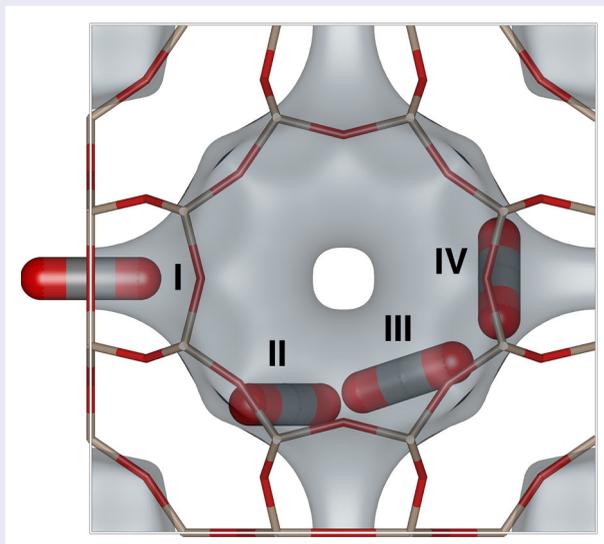
In the days prior to the Thermodynamics2024 conference in Delft (The Netherlands), the annual RASPA workshop/school took place at Delft University of Technology with 55 participants (both industry and academics) from all over the world. RASPA is a popular open-source molecular simulation software package for studying adsorption and diffusion in fluids and nanoporous materials, and it is especially popular in the metal-organic frameworks and zeolite communities. The main contributors to RASPA (and organisers of the RASPA workshop/school in Delft) are David Dubbeldam, Sofia Calero, Randall Q. Snurr, and Thijs J.H. Vlugt. In this short paper, we briefly explain the history of RASPA and the RASPA workshops, as well as our strategy to teach the workshop participants how to use RASPA for their specific research projects.

### ARTICLE HISTORY

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### KEYWORDS

Molecular simulation; adsorption; porous materials; zeolites; metal-organic frameworks



## 1. Introduction

Microporous and nanoporous materials like zeolites and metal-organic frameworks (MOFs) are important in applications such as separation technology and catalysis. In recent decades, molecular simulation has developed into a popular research tool to investigate the adsorption and diffusion properties of guest molecules inside these porous materials for the following reasons: (1) One can predict properties of adsorbed guest

molecules without having to synthesise the nanoporous host material first; (2) One can obtain a direct atomistic or molecular insight into adsorption and diffusion of guest molecules; (3) Molecular simulations are usually faster than experiments, thereby allowing for a fast screening of materials for a particular application. Due to the length- and time scales involved, these simulations are usually classical force field-based, which is usually justified unless hydrogen at very low temperatures

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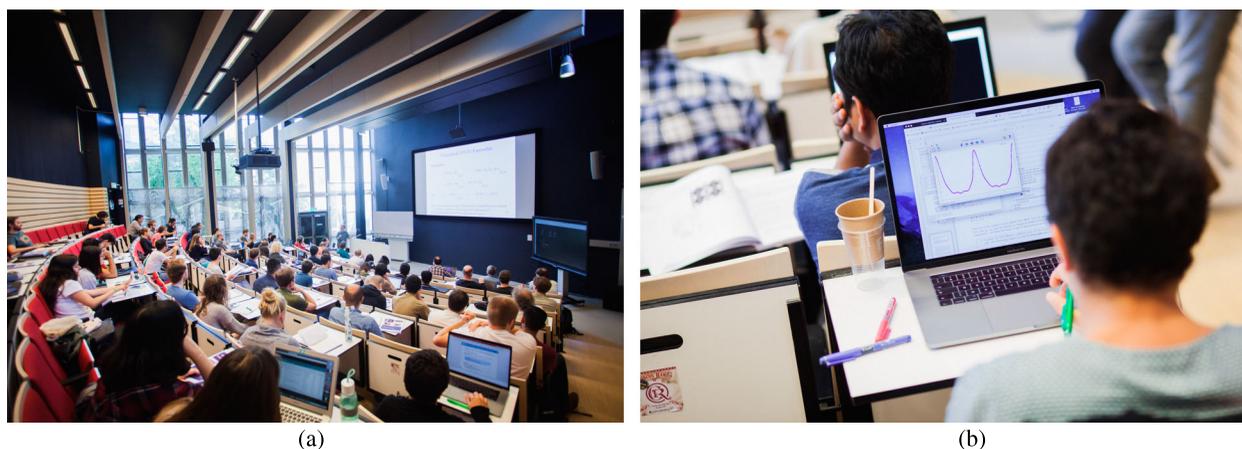
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is considered or when chemical bonds are broken or created [1]. The RASPA software [2, 3], first introduced in 2007 by David Dubbeldam and co-workers, is especially designed and tailored for classical Monte Carlo and Molecular Dynamics simulations of adsorbed guest molecules in these nanoporous materials. The software can perform Monte Carlo and Molecular Dynamics simulations in the most popular ensembles [1]: NVT-ensemble, NPT-ensemble, grand-canonical ( $\mu$ VT) ensemble, the Gibbs ensemble, and the reaction ensemble [4, 5]. Both rigid and flexible frameworks can be simulated, with or without non-framework molecules or cations. In Monte Carlo simulations with RASPA, the internal degrees of flexible (linear or branched) molecules are sampled with the Configurational-Bias Monte Carlo (CBMC) algorithm [1, 6]. In ensembles that critically rely on insertions and deletions of molecules, those molecule exchanges are facilitated by CBMC [1, 6] and the Continuous Fractional Component Monte Carlo (CFCMC) algorithm [7, 8], as well as by combinations thereof [9]. Interactions between atoms are described via the usual intramolecular bonded interactions (bond-stretching, bond-bending, torsion, as well as higher-order interactions [10]) and non-bonded interactions via Lennard-Jones and electrostatic interactions (either via the Ewald summation or the Wolff method). Polarisation between the adsorbed guest molecules and the framework can also be described by neglecting back-polarisation [11]. Typical properties of single- and multi-component systems that can be computed are: equations of state, vapor-liquid equilibria, free energies, adsorption isotherms, Henry coefficients, heats of adsorption, and transport properties such as self- and Maxwell-Stefan diffusivities and viscosities. There is a direct integration with iRASPA [12] for visualisation and RUP-TURA [13] for the computation of breakthrough curves.

Recent development are a GPU-accelerated version of RASPA2 [14] and a complete re-build of RASPA3 in C++ [15], which is ca. 4 times faster than RASPA2. In the near future, RASPA3 will also incorporate all features of the Brick-CFCMC Monte Carlo software package [16, 17]. For an overview of the full functionality of RASPA, we refer the readers to Refs. [2, 3, 10, 15] and the RASPA manual at <http://www.iraspa.org>.

## 2. History of the RASPA workshops

In 2017, the authors of this manuscript took the initiative to organise an annual RASPA workshop/school to teach the participants how to use the RASPA software. The first RASPA workshop took place at DIFFER in Eindhoven, The Netherlands, on May 30, 2017, with ca. 50 participants. The workshop was turned into a 4-day event in 2018 at Delft University of Technology, The Netherlands (June 25–28, 2018) and in Evanston, United States from (July 10–13, 2018). A picture of the typical setup of the workshop can be seen in Figure 1. A subsequent RASPA workshop was organised in Wroclaw, Poland (July 1–4, 2019). During the covid pandemic, from 2021–2023 the RASPA workshops were organised as annual online 1-day events and in 2024 it was organised again as a physical event at Delft University of Technology, The Netherlands, from September 2–4, 2024. This was just before the Thermodynamics2024 conference in Delft, The Netherlands, from September 4–6, 2024. Typically, there are ca. 50 participants for physical RASPA workshops, and > 200 participants for the online workshops, clearly showing the demand of the community. The workshop is attended mainly by academics (PhD students and postdocs), and by a limited number of researchers from industry or research institutes. A part of the participants are experimentalists who would like to know the basics of RASPA



**Figure 1.** Second European RASPA school/workshop, Monday 25 June 2018 – Thursday 28 June 2018, Delft University of Technology, The Netherlands.

simulations to facilitate better collaborations with computer simulators. Randall Q. Snurr was one of the plenary speakers of Thermodynamics2024 and provided a large part of the lectures at the RASPA2024 workshop. Ca. 15 participants attended both Thermodynamics2024 and RASPA2024. Travel grants were provided to cover all costs (travel, food, registration) for two RASPA workshop participants from underrepresented countries (Brasil, India).

We found that online RASPA workshops have their advantages (e.g. no need to travel, less CO<sub>2</sub> footprint, cheaper to organise, lower barrier to participate) and disadvantages (e.g. more difficult to ask questions, to establish interactions between participants, and to create a community). For this reason we will plan to have both online and physical workshops in the future.

### 3. Learning objectives of the RASPA workshops

The learning objectives aim to guide students from basic knowledge to advanced simulation techniques. This is valuable for both experimentalists seeking to understand the fundamentals of simulation and researchers in modelling and simulation who wish to deepen their expertise in the latest advancements. The course begins with an introductory lesson on simulation, covering essential concepts necessary for performing Monte Carlo or Molecular Dynamics simulations. We emphasise a critical aspect of simulation: the understanding, development, and selection of functional forms, force fields, and models. Over the years, we have observed that many researchers treat molecular simulation as a black box, overlooking these fundamental considerations. This may lead to errors; for example, choosing inappropriate force fields in a simulation is analogous to improperly calibrating experimental equipment. In addition, careless selection or uninformed use of force fields, models, and simulation settings will significantly undermine the predictive accuracy of simulations.

The learning objectives of the workshop are as follows: (1) Providing the essential theoretical background. We feel strongly that for successful research on molecular simulation, it is essential to understand the basics of the simulation techniques, otherwise the software is used as a black box and one does not know whether or not the result of the simulation corresponds to the property that one would like to compute (for example, by incorrect setting of the simulation parameters one will obtain incorrect results). For this reason, we have devoted the morning parts of the RASPA workshop to teaching the fundamentals of the simulation techniques and algorithms, while in the afternoon the participants execute some pre-defined simulations on their own laptops. (2)

Understanding the simulation algorithms, their strengths and weaknesses, their efficiency, and how to select an algorithm for your simulation system. (3) Choosing force fields and framework models. Using a suitable force field is essential for conducting realistic simulations. We teach participants which force fields are available for describing guest-guest, intra-host, and guest-host interactions in nanoporous materials, and how to assess the quality of a force field. It is important to realise when framework flexibility of the host material is important and when not. (4) Understanding the connection between experiments and simulations. The chosen ensemble in the simulation should be convenient for the properties of interest. Periodic boundary conditions are important to be able to extrapolate the computed properties to bulk properties. The students will learn about finite size effects, their influence on computed properties, and how to minimise or correct for these. (5) Understanding simulation settings. Simulation settings like the equilibration period and the period for sampling statistics determine the error bars (and possible systematic drift) of properties. Settings in advanced Monte Carlo algorithms (especially for CBMC and CFCMC in open ensembles) have a major influence on the efficiency and required computational time to compute a certain property with a certain accuracy, so we feel that this understanding is important. (6) Learning which thermodynamic and transport properties can be computed from which simulation type (and which ones cannot). (7) Visualization of atomistic systems. We teach the students how to make graphs of the output data, and using iRASPA, how to make nice pictures of the system, and create movies. (8) Understanding the physics of your system. While making graphs and pictures, we focus on the mechanism how to elucidate the physics by scrutinising the graphs and molecular simulation snapshot pictures. In particular, we focus on understanding the effects of energetics and entropy. In adsorption, at low loading the adsorption is dominated by energy, while at high loading entropy starts to dominate (packing-effects become important) [18, 19]. Teaching this is important to understand mixture selectivity. (9) Learning how to run simulations from a computer terminal rather from a Python notebook. For large-scale simulation studies on supercomputers, one usually has to use a terminal. (10) Creating interactions and collaborations between participants and the developers of RASPA, thereby creating a community and a research network. An example of this is by providing lunch and dinner on all days to the participants. In Figure 2 a typical session at RASPA2024 is shown. After attending the RASPA workshop, the participants will be able to better understand the theory behind the simulations, understand the simulation settings, and have the ability to design, setup, execute, and



**Figure 2.** Participants hard at work at the RASPA2024 workshop/school in Delft, The Netherlands, September 2–4, 2024.

analyse simulations of nanoporous materials and fluids with RASPA.

#### 4. Content of the 2024 RASPA workshop in Delft

Typically, theory lectures are given in the morning, while participants practise with the RASPA software themselves in the afternoon. The main lecturers for the morning parts are Sofia Calero, Randall Q. Snurr, David Dubbeldam, and Thijs J.H. Vlugt. For RASPA2024, the content of these lectures was as follows (not in this order):

- Lectures Sofia Calero: General introduction on why molecular simulation of nanoporous systems are needed; key differences between the main simulation methods (Monte Carlo and Molecular Dynamics); functional forms and structures of force fields for guest molecules, guest-host interactions, and flexible host structures; electrostatics and the Ewald summation; how to select an appropriate force field; rare events and transition state theory (with dynamic corrections)
- Lectures Randall Q. Snurr: General introduction to porous materials and their classification and characterisation; BET analysis and surface area [20, 21],

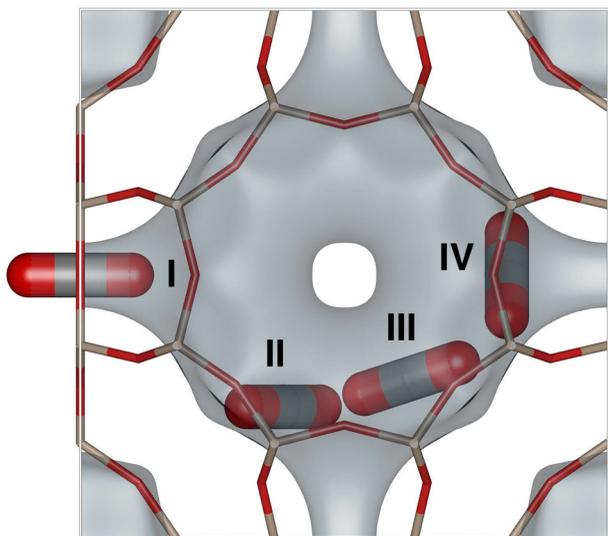
basics on Monte Carlo and Molecular Dynamics; detailed balance in Monte Carlo simulations; how to integrate the equations of motion in Molecular Dynamics simulations; thermostats and barostats in Molecular Dynamics; differences between static, thermodynamic properties and transport properties and how to compute these.

- Lectures David Dubbeldam: practical demonstration of RASPA2, RASPA3, RUPTURA, and iRASPA, and especially the integration between the software: Adsorption properties and adsorption isotherms are computed with RASPA. The structures and distribution of guest molecules inside the host material are visualised with iRASPA. RUPTURA can be used to simulate a breakthrough column experiments, so that one has insight in how adsorption and diffusion properties lead to a certain separation of components in a separation device.
- Lectures Thijs J.H. Vlugt: Monte Carlo simulations in various ensembles: NVT, NPT, Gibbs, grand-canonical ( $\mu VT$ ), and the reaction ensemble; Advanced simulation methods for molecule exchanges in open ensembles (such as the grand-canonical and Gibbs ensemble), CFCMC and CBMC; free energy calculations using Widom's test particle method, thermodynamic integration, and CFCMC.

In addition, there were three 1-hours guest lectures: Othonas Moulton (finite-size effects of computed transport properties [22]), Shrinjay Sharma (use of RUPTURA, isotherm models for mixture adsorption [23], and Ideal Adsorbed Solution Theory for estimating mixture adsorption from pure-component adsorption [24–26]), and Yongchul Chung (using the Core MOF database of MOF structures [27]). All lecture slides were provided to all participants. Due to privacy reasons, no video recordings were made of the lectures and exercises.

The topics of the exercises with RASPA3 of the afternoon sessions were as follows:

- Simulating a system of interacting methane molecules in the NVT ensemble; investigating the structure and structural properties of nanoporous frameworks; computing the adsorption properties of CO<sub>2</sub> in zeolite ITQ-29; energy and density distributions of CO<sub>2</sub> in MFI-type zeolite and ITQ-29.
- Simulating the radial distribution function of a fluid; self-diffusion of molecules in a fluid computed using the order-n algorithm [1, 28]; self- and collective diffusion of methane in MFI-type zeolite and ITQ-29
- Computation of the Vapor-Liquid Equilibrium of CO<sub>2</sub> in the Gibbs ensemble; computing the helium void



**Figure 3.** The four main minimum energy sites of CO<sub>2</sub> in ITQ-29 type zeolite: at low temperatures CO<sub>2</sub> is located predominantly in the windows (site I) connecting the cavities, but at higher temperatures the CO<sub>2</sub> molecules are driven out into the cavity. This CO<sub>2</sub> in ITQ-29 system is used to illustrate the effect of adsorption entropy.

fraction of a nanoporous structure; computing the adsorption isotherm of CO<sub>2</sub> in zeolite ITQ-29; computing isotherms using CFCMC and CBMC in the grand-canonical ensemble; computing the chemical potential and free energy of methane in MFI-type zeolite using various methods, e.g. thermodynamic integration, CFCMC, and Widom's test particle insertion method.

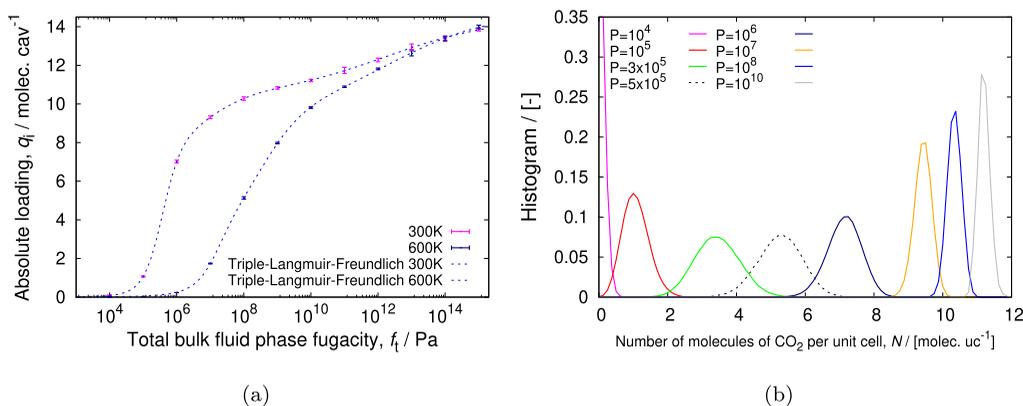
The exercises were setup in such a way that they could run sufficiently fast on a typical laptop. At the end of each day, detailed solutions were provided to all participants. A typical example of one of the exercises and the solutions is shown in Figures 3 and 4. The CO<sub>2</sub> in ITQ-29 system has a peculiar entropy effect where the CO<sub>2</sub>

molecule occupies the window-site at low temperatures but at higher temperatures is driven out into the cavities. This is used to illustrate the mechanism of adsorption as a function of temperature and loading. In the workshop, one of the main goals is to teach how the molecular mechanisms can be elucidated to explain the macroscopic, experimental results, and for example to connect the atomistic behaviour of CO<sub>2</sub> to the understanding of the isotherms.

The students compute the adsorption isotherm together in a group, where each student computes the loading at a separate fugacity using the grand-canonical ensemble. Afterwards, the students can combine their results to trace the full adsorption isotherm. This process illustrates to the students the concept of 'embarrassingly parallel' computations (a workflow that is very common in Monte Carlo simulations).

The students need to answer questions like: (1) What is the relation between chemical potential, fugacity, and pressure? (2) What are the advantages and disadvantages of using fugacity instead of pressure? (3) Why does the adsorption go down with temperature? (4) What are two ways of desorbing the structure? (5) Why does a triple Langmuir-Freundlich fit the data best? How does this relate to the adsorption sites that were found earlier by energy minimisation? How can the shape of the isotherm be explained? (6) Why does fixing the chemical potential imply that the amount of molecules fluctuates? (7) What is the relation between the steepness of the isotherm and the width of peaks of the number of molecules distribution? (8) Assume the computational force field is optimal, what are reasons why the computed loading could be higher than in experiments and what are reasons why the loading could be lower?

For RASPA2024, we used for the first time RASPA3 instead of RASPA2. The gain in speedup really helps in a school/workshop setting. For future workshops, also



**Figure 4.** Adsorption simulations of CO<sub>2</sub> in ITQ-29-type zeolite: (a) adsorption isotherms at 300 K and 600 K fitted with the triple Langmuir-Freundlich theoretical model, (b) the histogram of the number of CO<sub>2</sub> molecules at 300 K for various fugacities.

the installation process of the software will be dramatically simplified by using conda. Conda is available for macOS, Linux, and Windows, and allows you to create separate environments containing files, packages, and their dependencies that will not interact with other environments. RASPA3 is now available in conda for many supported architectures: linux-64, linux-aarch64, linux-ppc64le, win-64, osx-64, and osx-arm64.

## 5. Conclusions

We briefly described the content and history of the RASPA workshops. We plan to continue organising these workshops in the future. We feel that these workshops provide an important service to the simulation and porous materials communities. An online workshop is prepared and planned for the fall of 2025, while we aim at a physical workshop in 2026. These and other future RASPA workshops will be announced at <http://www.iraspa.org>. Online and physical workshops both have their advantages and disadvantages, so in the future we plan to organise both. We expect minor changes in the content of the workshop when migrating fully from RASPA2 to RASPA3. To date (March 2025), RASPA3 does not yet have the full functionality of RASPA2 and Brick-CFCMC, but this will change in the coming years. It will be a priority to (1) further integrate RASPA3, iRASPA and RUPTURA, (2) add support for machine learning potentials and polarisability, and (3) include advanced Monte Carlo algorithms for chain molecules [29]. Community-driven contributions are welcomed via ‘pull requests’ on the GitHub page.

## Acknowledgments

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## Disclosure statement

No potential conflict of interest was reported by the author(s).

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## References

- [1] D. Frenkel and B. Smit, *Understanding Molecular Simulation: from Algorithms to Applications*, 3rd ed. (Academic Press, Elsevier, UK, 2023).
- [2] D. Dubbeldam, S. Calero, D.E. Ellis and R.Q. Snurr, *Mol. Simul.* **42**, 81–101 (2016). doi:10.1080/08927022.2015.1010082
- [3] D. Dubbeldam, A. Torres-Knoop and K.S. Walton, *Mol. Simul.* **39**, 1253–1292 (2013). doi:10.1080/08927022.2013.819102
- [4] W.R. Smith and B. Triska, *J. Chem. Phys.* **100**, 3019–3027 (1994). doi:10.1063/1.466443
- [5] J.K. Johnson, A.Z. Panagiotopoulos and K.E. Gubbins, *Mol. Phys.* **81**, 717–733 (1994). doi:10.1080/00268979400100481
- [6] M.G. Martin and J.I. Siepmann, *J. Phys. Chem. B* **103**, 4508–4517 (1999). doi:10.1021/jp984742e
- [7] A. Rahbari, R. Hens, M. Ramdin, O.A. Moulto, D. Dubbeldam and T.J.H. Vlugt, *Mol. Simul.* **47**, 804–823 (2021). doi:10.1080/08927022.2020.1828585
- [8] W. Shi and E.J. Maginn, *J. Chem. Theory. Comput.* **3**, 1451–1463 (2007). doi:10.1021/ct7000039
- [9] A. Torres-Knoop, N.C. Burtch, A. Poursaeidesfahani, S.P. Balaji, R. Kools, F.X. Smit, K.S. Walton, T.J.H. Vlugt and D. Dubbeldam, *J. Phys. Chem. C* **120**, 9148–9159 (2016). doi:10.1021/acs.jpcc.5b11607
- [10] D. Dubbeldam, K.S. Walton, T.J.H. Vlugt and S. Calero, *Adv. Theory Simul.* **2**, 1900135 (2019). doi:10.1002/adts.v2.11
- [11] T.M. Becker, L.C. Lin, D. Dubbeldam and T.J.H. Vlugt, *J. Phys. Chem. C* **122**, 24488–24498 (2018). doi:10.1021/acs.jpcc.8b08639
- [12] D. Dubbeldam, S. Calero and T.J.H. Vlugt, *Mol. Simul.* **44**, 653–676 (2018). doi:10.1080/08927022.2018.1426855
- [13] S. Sharma, S.R.G. Balestra, R. Baur, U. Agarwal, E. Zuidema, M.S. Rigutto, S. Calero, T.J.H. Vlugt and D. Dubbeldam, *Mol. Simul.* **49**, 893–953 (2023). doi:10.1080/08927022.2023.2202757
- [14] Z. Li, K. Shi, D. Dubbeldam, M. Dewing, C. Knight, A. Vazquez-Mayagoitia and R.Q. Snurr, *J. Chem. Theory. Comput.* **20**, 10649–10666 (2024). doi:10.1021/acs.jctc.4c01058
- [15] Y.A. Ran, S. Sharma, S.R.G. Balestra, Z. Li, S. Calero, T.J.H. Vlugt, R.Q. Snurr and D. Dubbeldam, *J. Chem. Phys.* **161**, 114106 (2024). doi:10.1063/5.0226249
- [16] R. Hens, A. Rahbari, S. Caro-Ortiz, N. Dawass, M. Erdös, A. Poursaeidesfahani, H.S. Salehi, A.T. Celebi, M. Ramdin, O.A. Moulto, D. Dubbeldam and T.J.H. Vlugt, *J. Chem. Inf. Model.* **60**, 2678–2682 (2020). doi:10.1021/acs.jcim.0c00334
- [17] H.M. Polat, H.S. Salehi, R. Hens, D.O. Wasik, A. Rahbari, F. De Meyer, C. Houriez, C. Coquelet, S. Calero, D. Dubbeldam and T.J.H. Vlugt, *J. Chem. Inf. Model.* **61**, 3752–3757 (2021). doi:10.1021/acs.jcim.1c00652
- [18] B. Smit and T.L.M. Maesen, *Chem. Rev.* **108**, 4125–4184 (2008). doi:10.1021/cr8002642
- [19] R. Krishna, B. Smit and S. Calero, *Chem. Soc. Rev.* **31**, 185–194 (2002). doi:10.1039/b101267n
- [20] D.A. Gomez-Gualdrón, P.Z. Moghadam, J.T. Hupp, O.K. Farha and R.Q. Snurr, *J. Am. Chem. Soc.* **138**, 215–224 (2016). doi:10.1021/jacs.5b10266
- [21] J.W.M. Osterrieth, J. Rampersad, D. Madden, N. Rampal, L. Skoric, B. Connolly, M.D. Allendorf, V. Stavila, J.L. Snider, R. Ameloot, J. Marreiros, C. Ania, D. Azevedo, E. Vilarrasa-Garcia, B.F. Santos, X.-H. Bu, Z. Chang, H. Bunzen, N.R. Champness, S.L. Griffin, B. Chen, R.-B. Lin, B. Coasne, S. Cohen, J.C. Moreton, Y.J. Colón, L. Chen, R. Clowes, F.-X. Coudert, Y. Cui, B. Hou, D.M. D’Alessandro, P.W. Doheny, M. Dinca, C. Sun, C. Doonan, M.T. Huxley, J.D. Evans, P. Falcaro, R. Ricco, O. Farha, K.B. Idrees,

- T. Islamoglu, P. Feng, H. Yang, R.S. Forgan, D. Bara, S. Furukawa, E. Sanchez, J. Gascon, S. Telalovic, S.K. Ghosh, S. Mukherjee, M.R. Hill, M.M. Sadiq, P. Horcajada, P. Salcedo-Abraira, K. Kaneko, R. Kukobat, J. Kenvin, S. Keskin, S. Kitagawa, K.-I. Otake, R.P. Lively, S.J.A. DeWitt, P. Llewellyn, B.V. Lotsch, S.T. Emmerling, A.M. Pütz, C. Martí-Gastaldo, N.M. Padial, J. García-Martínez, N. Linares, D. Maspoch, J.A. Suárez del Pino, P. Moghadam, R. Oktavian, R.E. Morris, P.S. Wheatley, J. Navarro, C. Petit, D. Danaci, M.J. Rosseinsky, A.P. Katsoulidis, M. Schröder, X. Han, S. Yang, C. Serre, G. Mouchaham, D.S. Sholl, R. Thyagarajan, D. Siderius, R.Q. Snurr, R.B. Goncalves, S. Telfer, S.J. Lee, V.P. Ting, J.L. Rowlandson, T. Uemura, T. Iiyuka, M.A. van der Veen, D. Rega, V. van Speybroeck, S.M.J. Rogge, A. Lataire, K.S. Walton, L.W. Bingel, S. Wuttke, J. Andreo, O. Yaghi, B. Zhang, C.T. Yavuz, T.S. Nguyen, F. Zamora, C. Montoro, H. Zhou, A. Kirchon and D. Fairen-Jimenez, *Adv. Mater.* **34**, 2201502 (2022). doi:[10.1002/adma.v34.27](https://doi.org/10.1002/adma.v34.27)
- [22] A.T. Celebi, S.H. Jamali, A. Bardow, T.J.H. Vlught and O.A. Moulton, *Mol. Simul.* **47**, 831–845 (2021). doi:[10.1080/08927022.2020.1810685](https://doi.org/10.1080/08927022.2020.1810685)
- [23] S. Sharma, M.S. Rigutto, R. Baur, U. Agarwal, E. Zuidema, S.R.G. Balestra, S. Calero, D. Dubbeldam and T.J.H. Vlught, *Mol. Phys.* **121**, e2183721 (2023). doi:[10.1080/00268976.2023.2183721](https://doi.org/10.1080/00268976.2023.2183721)
- [24] A.L. Myers and J.M. Prausnitz, *AIChE J.* **11**, 121–127 (1965). doi:[10.1002/aic.v11:1](https://doi.org/10.1002/aic.v11:1)
- [25] C. Simon, B. Smit and M. Haranczyk, *Comput. Phys. Commun.* **200**, 364–380 (2016). doi:[10.1016/j.cpc.2015.11.016](https://doi.org/10.1016/j.cpc.2015.11.016)
- [26] K.S. Walton and D.S. Sholl, *AIChE J.* **61**, 2757–2762 (2015). doi:[10.1002/aic.v61.9](https://doi.org/10.1002/aic.v61.9)
- [27] Y.G. Chung, E. Haldoupis, B.J. Bucior, M. Haranczyk, S. Lee, H. Zhang, K.D. Vogiatzis, M. Milisavljevic, S. Ling, J.S. Camp, B. Slater, J.I. Siepmann, D.S. Sholl and R.Q. Snurr, *J. Chem. Eng. Data* **64**, 5985–5998 (2019). doi:[10.1021/acs.jced.9b00835](https://doi.org/10.1021/acs.jced.9b00835)
- [28] D. Dubbeldam, D.C. Ford, D.E. Ellis and R.Q. Snurr, *Mol. Simul.* **35**, 1084–1097 (2009). doi:[10.1080/08927020902818039](https://doi.org/10.1080/08927020902818039)
- [29] S. Consta, T.J.H. Vlught, J. Wichers Hoeth, B. Smit and D. Frenkel, *Mol. Phys.* **97**, 1243–1254 (1999). doi:[10.1080/00268979909482926](https://doi.org/10.1080/00268979909482926)