D9.1 – CSC transnational access success story

Deliverable No.:	D9.1
Deliverable Name:	CSC transnational access success story
Contractual Submission Date:	30/04/2021
Actual Submission Date:	31/10/2018
Version:	v2.0

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 730897.

COVER AND CONTROL PAGE OF DOCUMENT	
Project Acronym:	**HPC-Europa3**
Project Full Name:	Transnational Access Programme for a Pan-European Network of HPC Research Infrastructures and Laboratories for scientific computing
Deliverable No.:	D9.1
Document name:	CSC transnational access success story
Nature (R, P, D, O):	R
Dissemination Level (PU, PP, RE, CO):	PU
Version:	V2.0
Actual Submission Date:	31/10/2018
Author, Institution: E-Mail:	Atte Sillanpää, CSC atte.sillanpaa@csc.fi
Other contributors	Miguel A Caro, HPC-Europa3 host Volker Deringer, HPC-Europa3 visitor Lykle Voort (SURFSara)

ABSTRACT:

This document describes the fruitful collaboration of Dr. Volker Deringer from the University of Cambridge, UK, and Dr. Miguel Caro, from the Aalto University, Finland, which started through a HPC-Europa3 transnational access visit. The collaboration has resulted directly in three high-impact journal publications and was motivated by the different skills which together enabled carrying out ground-breaking research on industrially and scientifically important amorphous carbon films. The work has also advanced the simulation methods in general. The visits have secured a strong international link and active collaboration between the two groups. The role of HPC-Europa3 in the collaboration is discussed.

KEYWORD LIST:
Materials Science, Machine Learning, DFT, international collaboration, transnational access

MODIFICATION CONTROL			
Version	**Date**	**Status**	**Author**
1.0	3.10.2018	Draft	Atte Sillanpää
1.1	8.10.2018	Draft	Atte Sillanpää, contribs from Caro, Deringer
1.2	9.10.2018	Draft	Atte Sillanpää, contribs from Caro, Deringer
1.3	29.10.2018	Draft	Atte Sillanpää, contribs from Lykle Voort
2.0	30.10.2018	Final	Final version ready

TABLE OF CONTENTS

Executive summary 4
1 First visit - join forces and solve a long standing dispute 5
2 Second visit - new atoms in the mix 7
3 Role of HPC-Europa3 9
4 Additional material 10

Executive summary

Dr. Volker Deringer from Cambridge, UK, has visited the group of Dr. Miguel Caro at Aalto University, Finland, several times during the last year. Two of the visits have been funded by HPC-Europa3. The collaboration has resulted directly in three high-impact journal publications. The collaboration has been motivated by the different skills which together enabled carrying out ground-breaking research on industrially and scientifically important amorphous carbon films. The work has also advanced the simulation methods in general. The visits have secured a strong international link and active collaboration between the two groups.

1 First visit - join forces and solve a long standing dispute

Volker Deringer from the University of Cambridge filed a transnational access application in summer 2017 in the very first call of HPC-Europa3 to visit Miguel Caro in Aalto University, Finland. Dr. Caro leads the computational simulation efforts in Prof. Tomi Laurila's group at Aalto. The group's work focuses on studying the properties of amorphous carbon, among other research topics. Together, the knowledge of the materials and access to new machine learning methods could be used to solve the long-lasting debate between competing growth mechanisms of amorphous carbon (a-C) films and to gain new understanding on the high-tech material that has enormous industrial and scientific importance.

To understand the properties of disordered materials, including the amorphous phases that are made in Laurila's lab, powerful computational tools are needed. They need to combine accuracy (to describe the diverse atomic structures) and speed (to achieve realistic simulation system sizes). The researchers use a novel simulation technique based on machine learning (ML) from quantum-mechanical data, called "Gaussian Approximation Potentials", or "GAP" in short. The GAP method uses Smooth Overlap Atomic Positions (SOAP) to represent the geometry. It is developed by Prof. Gábor Csányi and his group at Cambridge, with whom the team collaborates extensively. As a Leverhulme Early Career Fellow in the same department, Dr. Deringer aims to apply this methodology to study and understand a wide range of amorphous solids.

This mix of novel simulation tools and intriguing applications got Deringer and Caro talking.

During an initial visit in December 2017, they built the computational tools and designed experiments to simulate the amorphous carbon film growth process. There are two competing growth mechanisms for a-C. Due to the large fundamental and industrial importance of a-C films, there have been many attempts to solve the mechanism with classical (empirically fitted) interatomic potentials but they all have failed to quantitatively reproduce the experimental observables, such as the amount of sp^3 (fourfold bonded) carbon atoms. The present results, enabled by the accuracy and flexibility of the novel machine-learning based interatomic potential, have been a game-changer in this regard.

The mechanism was solved by molecular dynamics simulations consisting of slamming individual carbon atoms to an amorphous carbon film with different energies as shown in Fig 1. See also the additional materials link to videos on how the film grows. The resulting atomic trajectories enabled a detailed study of the surface reorganization dynamics.

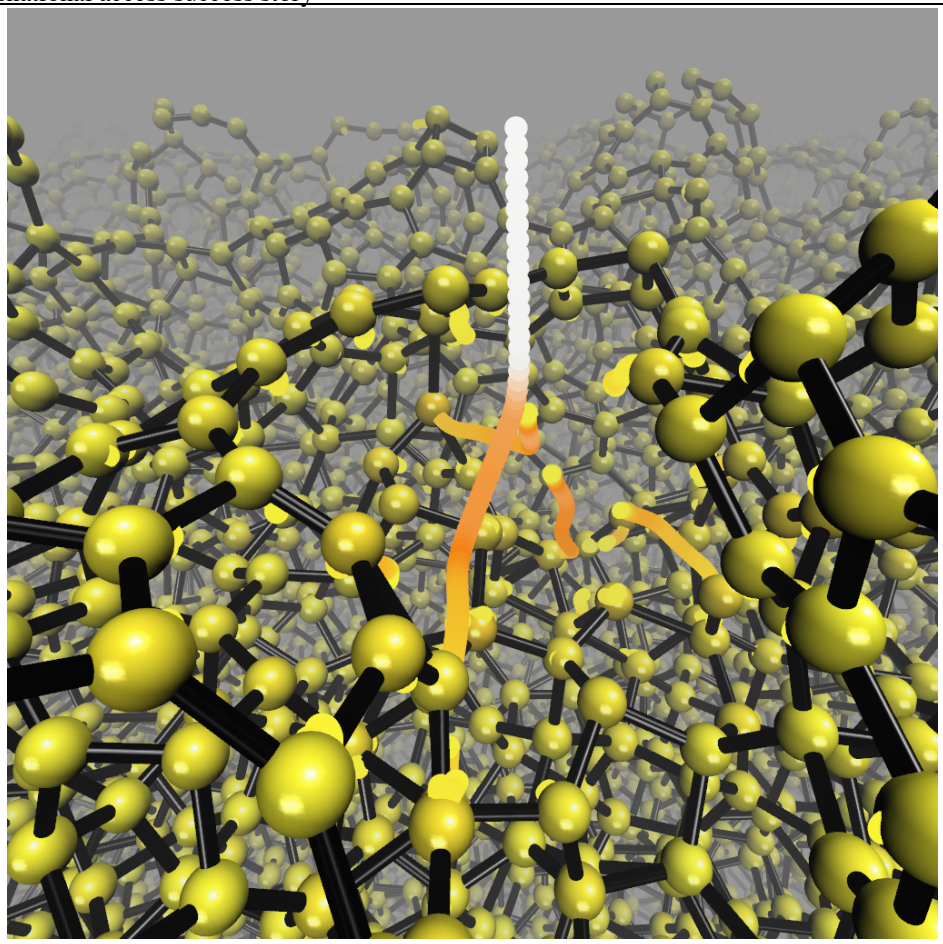

Figure 1. Amorphous carbon surface (yellow atoms) being hit by a fast carbon atom whose trajectory starts as white circles and gradually turns yellow with decreasing velocity. Collision to surface carbon atoms knocks few atoms out of their positions exerting local stress on the material.

The results unambiguously confirmed the less widely supported "peening" mechanism for the growth of high sp^3-hybridized carbon local structure, which is responsible for the mechanically superior properties of the carbon films. The article describing this work was published in Physical Review Letters already in April and was promptly selected by the American Physical Society as their feature article of the week. The results picked up further attention in science news outlets around the world, and the work achieved an Altmetric score (which measures the impact on news sites, Twitter, etc) of over 100, placing it in the top 5% of all research output scored by Altmetric[1].

It turned out that the collaboration and visit were so successful that Dr. Deringer applied for another HPC-Europa3 visit. The current version of the GAP at the time could be used to simulate materials composed only of carbon, but the team already had the next steps in mind to deepen the collaboration.

[1] https://aps.altmetric.com/details/38043907

2 Second visit - new atoms in the mix

There is only a finite amount of materials made of only carbon and a natural step forward was to include more atoms that the new model can describe. The next HPCE3 transnational access visit aimed to look at the surface reactivity of the amorphous carbon materials with oxygen and hydrogen. This was ground breaking science as the quantitatively accurate analysis of amorphous surfaces has not been possible before.

The authors created a large number of representative amorphous carbon (a-C) slabs with the GAP potential using a Monte Carlo algorithm. Armed with the very fast GAP potential, the authors determined the minimum system size which displays the converged a-C properties. These slabs were further functionalized with hydrogen and relaxed using Density Functional Tight Binding (DFTB) and oxygen using DFT-MD, respectively. The resulting large datasets of representative model systems are available for anyone to use in further analyses and parameterisations. The properties of the surfaces agree with available experimental data and earlier simulations.

The slabs were used to explore the local structure around the carbon atoms with a fingerprint method, namely defining the similarity to both sp^2 (graphite) and sp^3 (diamond) like structure. The different geometries were clustered and characterised to enable the analysis of representative motifs. The Figure 2 nicely highlights the characteristics of an amorphous material. It has been accepted on the Journal cover (November 2018 issue). The individual coloured points represent different *i.e.* unique local geometries or motifs. The large number of them is in contrast to crystalline systems with high symmetry where there are only a few discrete points.

The clustering was done with machine learning algorithms to build understanding from the raw data. The clusters were labelled with medioid geometries (shown with ball and stick representations). These help us humans to make sense of the chemical environment, while the individual geometries and their properties are needed for a quantitative description.

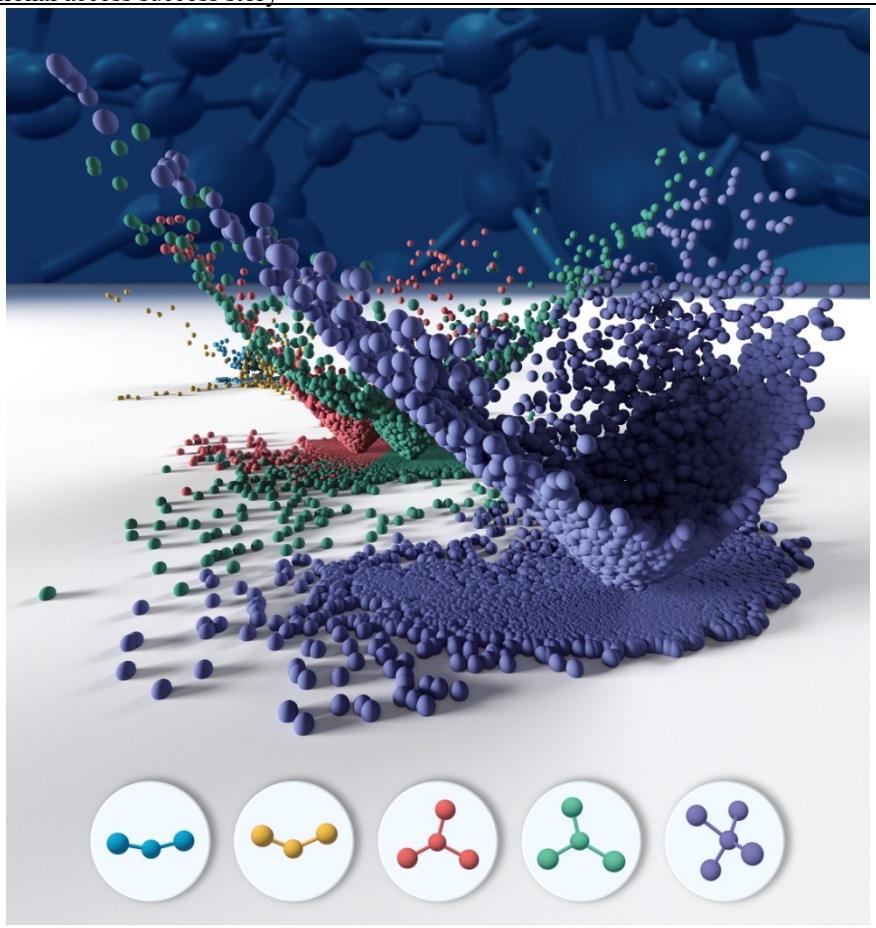

Figure 2. Categorization of the local amorphous carbon motifs (coloured dots) and a representative medioid for each of them at the bottom row. The grey plane containing the shadows is a two dimensional similarity representation of the motifs. The Z-axis (away from the surface) denotes the dissimilarity from the medioid. Some of the categories are seen to overlap. The illustration was created as collaboration with CSC's visualization expert Jyrki Hokkanen and the researchers.

The second aim of the paper was to design a representative set of geometries containing not only carbon but also oxygen and hydrogen, based on the pure amorphous carbon surfaces, which were then characterized with DFT. These structures will be used to parameterize the improved machine learned GAP interatomic potential in future studies. In the reported study the adsorption energy and thereby the reactivity of the different surface motifs was determined. Surface modifications included the relevant species containing hydrogen and oxygen.

Describing the surface chemistry (adsorption energies) could be described with the geometrical data (SOAP), but the adsorption energies were also found to correlate with the integrated Local Density of States (LDOS), which is suggested to imply propensity to form bonds. It was shown that additional parameterization, which was done also via machine learning using LDOS thereby resulting in a combined SOAP-LDOS method can be used to accurately predict adsorption energies with a fraction of the computational cost of the original DFT calculations.

The adsorption energies and reactivity correlate to the different motifs. Among other things, it turned out that while the conventional motifs overall map well on the results, more can be said about the relation of structure and energetics. The authors propose to split one traditional motif,

namely the sp^2 hybridised carbon local environment, into two subgroups with either short or long bonds.

Earlier the researchers had always used machine learning for making interatomic potentials only. In this paper this approach is made much broader, by introducing ML models for properties other than atomic energies and forces - for example the paper introduces an advanced ML scheme based on joint structural and electronic descriptors that can predict accurate hydrogenation energies (and thus chemical reactivity) of a-C films.

These two papers were accepted in Chemistry of Materials only some weeks after the second visit was over. As a further indication of the novelty and significance of the results the researchers estimate at least five additional publications in 2019.

3 Role of HPC-Europa3

Volker and Miguel briefly met by chance in a conference in Aalto University in 2017. The synergy of the two researchers and their groups was immediately obvious. The HPC-Europa3 programme likely speeded up getting into action and initiating the strong collaboration by offering funding for a research visit and computational resources. In addition to the two HPCE3-funded visits for Volker also other visits have taken place without HPCE3 support. While a reciprocal HPC-Europa3 visit by Miguel to Cambridge is being planned, Miguel or researchers from his team have already made three shorter visits to London and Cambridge to work on the project without HPC-Europa3 funding.

I think this is an excellent example of an EC funded programme successfully catalysing and promoting international collaboration where the particular strengths of the two groups complement each other. Overall, the HPC-Europe3 programme can be seen to enable these kinds of collaborations in Europe thereby improving the competitiveness by helping to bring together the leading experts to solve specific hard problems.

Parts of the research could have been done remotely but at times of analysing the results and making decisions on the next steps it has been crucially important to be at the same site and benefit of the maximal potential for communication. In practice Volker has flown in and out of Finland a few times to take care of his commitments at other locations, but luckily HPCE3 is flexible enough to allow this. The extra flights just have to be funded from other sources. On the other hand, another HPC-Europa3 visitor at Aalto (Laura, who is a PhD student) opted to visit for 3 months straight, allowing her to be fully integrated in the host research group and to perform a substantial piece of work on site; in contrast, more senior researchers (such as Volker and Miguel) can benefit strongly from shorter visits, allowing them to accommodate other commitments in between and regularly "catching up" on the progress of the overall project, which has a substantial cooperative effect (the benefit from the multiple visits is much more than the sum of individual durations.

"Nothing beats working with colleagues on site!", tweeted Dr. Deringer after the first visit confirming that in challenging projects with collaborators with complementing skills there really is added value to work together at the same location. Indeed, we think this idea is at the very core of the programme.

Visits have also enabled additional networking outside the host group *e.g.* via lectures and presentations at various events thus further spreading the latest knowledge of the state-of-the-art methods from another EU country.

The results published in high impact journals have been noted in CSC's and Aalto University's web pages and social media accounts, as well as, HCPE3 twitter account further promoting the programme but also giving additional publicity to the research itself, as could be verified from the Altmetric score above.

After a visit, the parties are asked for feedback. The following is one of the things Dr. Deringer wanted to say after the first visit: "This was a most successful visit - we have developed an initial collaboration, which led to a whole range of new ideas. We are planning and working on several publications, and hope to apply for HPC-Europa3 support again in one of the next rounds. "Dr. Caro highlights another benefit of the visit in his feedback: "The collaboration with Volker is invaluable to us. It expands our expertise in electronic structure methods by adding machine learning potentials. "

As described above, the new visits materialised and produced more high quality publications thus giving compelling evidence of new fruitful collaboration - supported by HPC-Europa3.

4 Additional material

- *Growth Mechanism and Origin of High Content in Tetrahedral Amorphous Carbon*, Miguel A Caro, Volker L Deringer, Jari Koskinen, Tomi Laurila, Gábor Csányi, (via APS)
- *Computational Surface Chemistry of Tetrahedral Amorphous Carbon by Combining Machine Learning and DFT*, Volker L Deringer, Miguel A Caro, Richard Jana, Anja Aarva, Stephen R Elliott, Tomi Laurila, Gábor Csányi, Lars Pastewka, Chemistry of Materials (via ResearchGate)
- *Reactivity of amorphous carbon surfaces: rationalizing the role of structural motifs in functionalization using machine learning*, Miguel A Caro, Anja Aarva, Volker L Deringer, Gábor Csányi, Tomi Laurila, Chemistry of Materials (via ACS)
- *CSC news item of the first article*: https://www.csc.fi/web/atcsc/-/kiistelty-hiilimateriaalin-kasvumekanismi-ratkesi-atomitason-simulaatioiden-avulla
- Videos showing the simulations of creating carbon films (via zenodo)