

EuroCC@Türkiye

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Designing Antireflective Glass Coatings using Quantum Chemistry Methods

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Company*	Türkiye Şişe ve Cam Fabrikaları A.Ş.
NCC Expert	
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Effort	
Subject Domain	Computational Materials Discovery
Infrastructure	TRUBA
Approved by	

*(Türkiye Şişe ve Cam Fabrikaları A.Ş.) accepts that the Use Case Report is shared with the EuroCC 2 Project and the community through the EuroCC@Türkiye awareness creation activities and platforms.

Date	Author	Comments	Version
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Instructions

- In case the form is being filled out for the initial time, solely the sections of “Abstract & Özet”, “Problem Identification”, “First Suggestions”, “HPC Usage Details”, and “HPC Usage Status” will be completed.
- During the process of PoC evaluation, the assessment will be based on three specific criteria. Therefore, it is advisable to create the content in alignment with these criteria. These are listed for reference:
 - The proposed PoC study is in line with the scope and objectives of EuroCC 2.

□ With this PoC study, the company can measurably increase its competencies in the fields of HPC/HPDA/AI. (HPC Ready --> HPC User, HPC User --> HPC Champ. The definitions are given in the Section-7.)

□ The HPC/HPDA/AI platform required for the operation can be provided by the TRUBA/UHeM infrastructure.

- This form must be updated every 3 months for monitoring the progress. During the updating process, the sections of “Solution Stage-1” and for the first results “Results and Achievements” will be completed.
- When the PoC project is completed, “Results and Achievements”, “Business Benefits” and “Challenges” parts will be completed.

1.

a-Abstract

This proof-of-concept study presented by Şişecam and ODTÜ focuses on implementing the use of *ab initio* calculations in glass production, particularly for large-area coatings, through high-performance computing. The project addresses two key areas: the design of functional layers in low-emissivity glass coatings and the optimization of precursors for chemical vapor deposition (CVD) processes. For low-emissivity coatings, computational modeling explores interfacial phenomena such as adhesion and lattice mismatch to improve silver crystallinity and thin-film performance. For CVD, quantum thermochemical calculations will aid in the selection of efficient, environmentally friendly tin precursors for fluorine-doped tin oxide coatings. By integrating quantum mechanical modeling into experimental insight, this study aims to establish a workflow for materials discovery, enhance energy-efficient coatings, and support Şişecam’s commitment to sustainable innovation in glass technologies.

b-Özet

Şişecam ve ODTÜ tarafından sunulan bu PoC çalışması, büyük alan kaplamaları için cam üretiminde *ab initio* metodlarının yüksek performanslı hesaplama yoluyla kullanılmasını amaçlamaktadır. Proje, düşük emisyonlu cam kaplamalarının fonksiyonel katmanlarının tasarımı ve kimyasal buhar biriktirme (CVD) süreçleri için öncül maddelerin optimizasyonu olmak üzere iki ana alana odaklanmaktadır. Düşük emisyonlu kaplamalar için, hesaplamalı modelleme, gümüş kristalliğini ve ince film performansını iyileştirmek amacıyla yapışma ve kafes uyumsuzluğu gibi arayüzey fenomenlerini incelemektedir. CVD için ise kuantum termokimyasal hesaplamalar, flor katkılı kalay oksit kaplamalar için verimli ve çevre dostu kalay öncüllerinin seçimine katkı sağlayacaktır. Kuantum mekaniksel modelleme ve deneysel öngörüü birleştiren bu çalışma, malzeme keşfi için bir iş akışı oluşturmayı, enerji verimli kaplamaları geliştirmeyi ve Şişecam’ın cam teknolojilerinde sürdürülebilir inovasyon taahhüdünü desteklemeyi hedeflemektedir.

2. Problem Identification

This proof-of-concept (PoC) study is aimed to elucidate molecular mechanics at the center of one of the most important technologies of modern industrial endeavors, namely the

production of glass. Centered around large area glass coating business, where high-performance computing is expected to make a notable impact by means of *ab initio* calculations, the goal of this PoC is to demonstrate that materials discovery for use in specific stages of production of glass coatings can be accelerated via *ab initio* calculations. We focus on two separate projects:

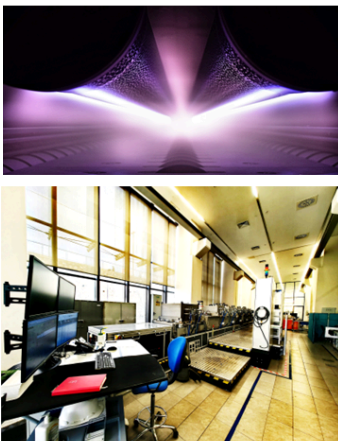
Project 1: Functional layers in low-emissivity (low-e) optical filters

The current state-of-the-art in large area glass coating applications is depositing multilayer stacks where one or more silver layers (typically 8-15 nm) are sandwiched between other materials (See Figure 1, left panel). The remaining layers in a low-e stack assume different functions, which can be summarized as follows:

- **Seed layer:** To minimize scattering losses, the seed layer is used so that silver can grow in the proper orientation.
- **Blocker layer:** this layer protects silver from the sputtering damage that can occur during the deposition of subsequent layers on top and mitigates corrosion by blocking oxidation.
- **Dielectric layer(s):** Dielectric layers (bottom and top) provide color and reflection control for the low-e layer stack
- **Top layer:** Top layer imparts extra mechanical resistance to the coating

Şişecam maintains a global scale large area coating operation based on magnetron sputtering technology and the annual production exceeds 20 million m² (soon to be upgraded to above 40 million m²). This project seeks to incorporate first-principles computational tools into material discovery in optical thin film applications in the flat glass business.

Deposition with Magnetron Sputtering



Resulting Soft Low-e Coatings

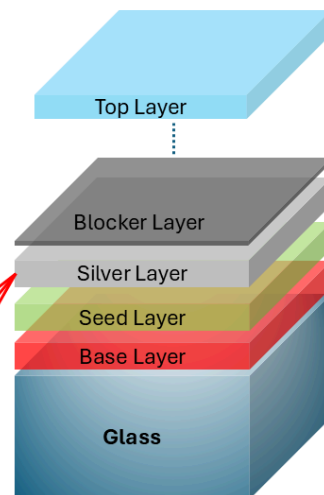
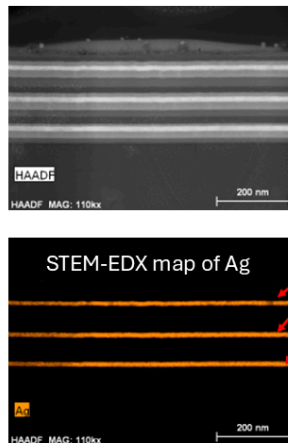


Figure 1: (Left) Large area deposition of low-e layerstacks with magnetron sputtering. (Right) Cross sectional image of a triple silver low-e layerstacks and a schematic representation of functional layers.

Understanding the interfacial phenomena and altering individual layers hold the key to performance improvements for coating systems with high and lasting commercial relevance. Despite the complexity of the deposited layer stacks, it is possible to define smaller but still relevant problems that are tractable with ab initio computational tools. Our primary motivation can be summarized as follows:

- Unlike the glass itself, the deposited thin film layers generally assume a crystalline state and the spatial periodicity assumption reduces the computational power needed.
- Understanding interfacial phenomena such as lattice mismatch and surface adhesion energies between dissimilar materials have been studied previously and linked with experiments. A more recent study based on DFT (Density Functional Theory) computations investigated the Ag adhesion on different oxides used as seed layers including the effect of defects/dopants [1-3]. We aim to extend these studies to other seed layer systems and the interfaces between other functional layers. In the current procedures, ZnAlO is used as the crystalline layer beneath Ag, which enhances the crystallinity of Ag and therefore all electrical and optical properties.
- It is a particular challenge to study the interfacial behavior of those materials with direct experimental means due to small system sizes which exceed the resolution of analytical instruments. Therefore, computational modeling can offer additional insights into such phenomena and complement the experimental studies.

Project 2: Chemical Vapor Deposition (CVD) and Precursor Selection

Fluorine-doped tin oxide, SnO₂:F, as a transparent conducting oxide (TCO), offers high optical transparency in the visible region ($\lambda=380-780$ nm) as well as the highest mechanical, thermal, chemical, and electrochemical stability properties among all known TCO materials [4-5]. Due to its unique advantages, it has a large commercial market spanning many sectors such as thin film photovoltaics and heat-reflecting glass applications for household appliances.

CVD-based high temperature techniques are generally used in the large-area coating of SnO₂:F thin films. CVD deposition of SnO₂:F is an online process (integrated on the glass production line) and occurs at temperatures typically exceeding 500°C on continuous glass ribbons as substrates. FTO coatings deposited by CVD exhibit high mechanical and chemical durability and can even be used as single glazing in architectural solar control applications.

CVD is a process based on introduction of a gas-phase stream of molecules flowing by a substrate. Through gas phase and surface reactions the desired material is obtained and deposited on the surface (See Figure 2). The idea of this part of the project is to expedite the selection of tin precursors for the CVD deposition of SnO₂:F thin films through computational quantum thermochemical calculations on small molecules.

We would like to identify precursors that can be used at lower deposition temperatures and assess their viability through the material properties of the resulting coating. In particular, the

team are interested in starting with exploring organometallic Sn compounds such as tetramethyl, dimethyl trichloride, Sn (IV) acetate, Sn acetoacetate, Sn (IV) tert-butoxide etc. The figures of merit to be calculated in this problem using a quantum chemistry code such as ORCA (<https://www.faccts.de/orca/>) are as follows:

- Chelation energy of the molecules
- Sn-ligand bond breaking energy and Sn-Ox bond formation energy
- Reaction free energies

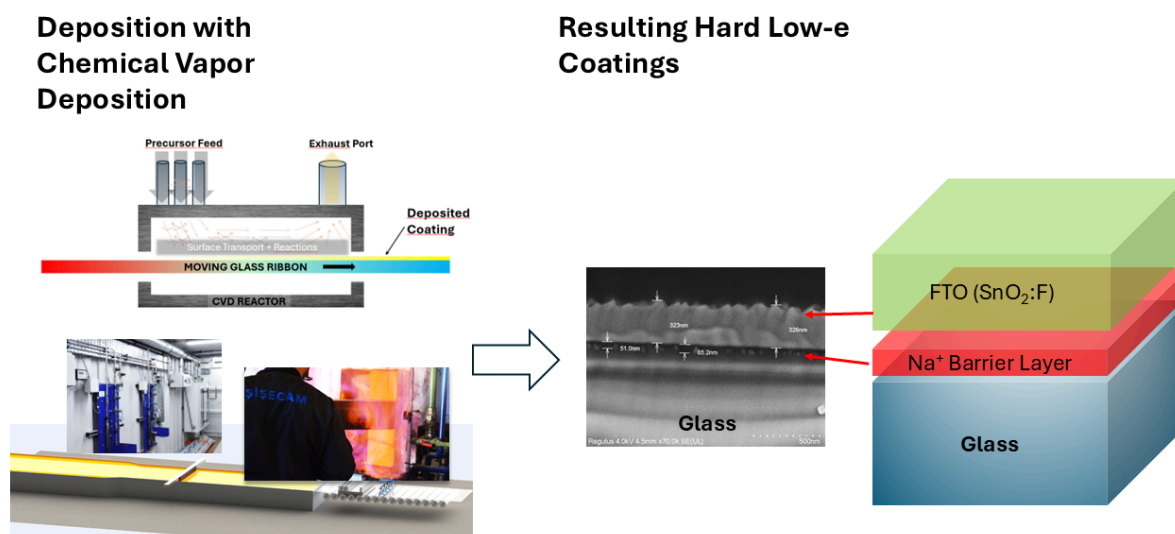


Figure 2: (Left) Large area deposition of low-e layerstacks with chemical vapor deposition. (Right) Cross sectional image of the resulting coatings and a schematic representation of functional layers.

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- Chelation energy of the molecules
- Sn-ligand bond breaking energy and Sn-Ox bond formation energy
- Reaction free energies

The objectives of this PoC study is to introduce the Şişecam team to HPC resources, and with the collaboration of the practitioners of academic (NCC) and practical materials science (Şişecam), develop a coherent work plan for scanning a very wide variety of materials for both of the projects suggested above. Much of today's materials discovery is built around ML algorithms, however, this does not replace a solid understanding of the quantum mechanical origins of the materials properties being explored. With this PoC, the academic expert will

help the team construct a computational workflow that they can then use as a part of their product development.

Stage 1: General training. This part of the PoC consists of training of the team in the usage of HPC resources, basics of DFT, and relevant software.

Stage 2: Preliminary calculations. In this stage of the project, the NCC expert will help the team pick a few materials for each of the two problems stated above and will guide them in completing an entire set of calculations. The expected results will include adhesion energy of oxides to Ag, structure of the interface, and any strain that may form in the interface.

Stage 3: The NCC expert and the Şişecam team will discuss possible ML-based materials discovery workflows, and finally they will work towards applying for an EU project to work on together.

Before closing this section, it must be mentioned that the R&D team are all composed of members with strong academic backgrounds, all with completed or ongoing postgraduate degrees in such areas as chemistry, materials science and various branches of engineering. All members are enthusiastic and dedicated to making computational work a part of their materials development toolkit.

3. First Suggestion

The tentative project plan is as follows:

Timeline	Tasks and Milestones
Weeks 1-2	Tasks: 1. Getting ready to hold a kickoff meeting 2. Kickoff meeting Milestone 1: A detailed plan of attack will be drafted.
Weeks 3-8	Tasks: 1. Training - Introduction to the basics of DFT 2. Training - Familiarizing the Şişecam team with the basics of running on HPC resources, Quantum Espresso, and CP2K 3. Finalizing the work plan Milestone 2: Training material will be written up, recorded and made available for the team to consult later as needed.
Weeks 9-15	Tasks : 1. In these weeks, we will attack Project 1. 2. As a preliminary study, team members and the consultant will Identify together a material to study the interface with Ag. 3. Adhesion energy and stress will be calculated using QE and CP2K. 4. A coherent work plan will be constructed for the team to work on by themselves to identify the best substrates for use in Ag growth. 5. ML applications will be discussed. Milestone 3: A summary of the results and a work plan for materials discovery will be written up.

Weeks 16-22	Tasks: 1. These weeks will be devoted to Project 2. 2. The team and the NCC expert are going to identify small molecules that will be relevant to the CVD process. 3. Using ORCA, bond-breaking and formation energies and chelation energies will be calculated. 4. A work plan will be developed to screen a large number of molecules that are relevant to CVD growth process. Milestone 4: A summary of the results and a work plan for materials discovery will be written up.
Weeks 22-24	Tasks: 1. These weeks will be devoted to planning for joint EU projects and finalizing our work. Milestone 5: At least one project will be identified.

4. Solution Stage – I

The plan of attack for the two abovementioned projects were formed within the first month. During our PoC work, the entire Şişecam team met with the expert a total of three times. In addition, we communicated often over email. Each meeting lasted at least an hour, with the following dates and agendas:

1. Kickoff meeting: 17.03.2025

During this meeting, we discussed the details of the two problems presented by Şişecam and decided which systems to include in our work. We specifically concentrated on the interface problem, for which we discussed what is already available in the literature and what needs to be done.

2. Interim Meeting: 22.04.2025

During this meeting, we concentrated on the CVD project and once again went over the available literature. The NCC expert demonstrated a preliminary calculation to the team to facilitate the decision process for how to proceed. The team composed a selection of molecules that they would like to concentrate on and shared it with the expert.

3. Training Meeting 1: 19.06.2025

This meeting lasted two hours with the entire focus being the demonstration of the usage of the software suites VASP and ORCA. In addition, the expert began to demonstrate how to log on to TRUBA and use the machines for calculations relevant to the two projects at hand. Due to the stimulating conversation that was catalyzed by the presentation, we decided to complete the training in a second meeting to be held in July.

In addition to the original code suites agreed upon, the Şişecam team expressed interest in learning the basics of Pymatgen (<https://pymatgen.org/>). The expert has prepared a Colab notebook with some relevant exercises and has shared it with the team. Although seemingly unrelated to using HPC resources, being fluent at Pymatgen reduces the job preparation time significantly, thereby rendering more efficient the use of said resources.

4. Closing Meeting: 02.09.2025

The PoC was concluded with a final meeting between the members of the team and the expert. During this meeting, the difficulties encountered as well as the benefits were discussed. The problem that was the most difficult to overcome was the inability of the R&D team members to long on to TRUBA systems. This is due to internal company policy that restricts internet usage from within the company network.

At the end of the meeting, the NCC expert and the team discussed opportunities for future collaborations. The expert presented a research plan that the company can use, which is detailed below.

5. HPC Usage Details

Please specify below HPC resource needs (CPUs and/or GPUs), partition names of relevant infrastructure, softwares which are utilized over the PoC study, size of the used storage

Requested Source	CPU	GPU	Other
	100000	0	0
Infrastructure	TRUBA	UHeM	Other
	100000		
Number of nodes	2		
Number of cores	110/node		
Software(s) used	Quantum Espresso, CP2K, LAMMPS, ORCA		
Storage (in TB)	1 TB		

Specify below special needs of the applications, libraries, compilers etc. that will be used, workflow tools, and other requirements. Please include the usage of multi nodes, multi-CPU, multi-GPUs etc.

Software: Quantum Espresso and CP2K will be used for the first problem of Ag growth. The systems involved in this problem are crystalline, therefore a periodic DFT code is more suitable. LAMMPS and ORCA on the other hand will be used for the second problem where molecules that are suitable for a particular CVD process will be identified.

Nodes/Cores and CPU Time: Two CPU cores on the ARF cluster with 110 nodes will be sufficient for running two jobs in parallel. Since this PoC is a mixture of training and preliminary work for future projects, we do not envision (at first) a large set of calculations. These resources should therefore be sufficient for the early stages.

6. Results and Achievements

The results and achievements can be summarized as follows:

1. The introduction of the Şişecam team to available computational software and resources: This part of the PoC was very successful, mostly due to the excellent scientific preparation of the team. The expert introduced the Physics behind the methods used and the main aspects of the software. The team responded very well and expertly designed the parameters to be used in the calculations. This facilitated our design of the workflow.
2. Preliminary calculations - Project 1: We have finished the preparation and the calculation of a sample job on the TRUBA resources. As mentioned in this report, the team is interested in understanding which metal oxide would adhere best to the Ag(111) surface. In order to improve adhesion, the oxide surface can also be doped. Our preliminary calculation is an interface between Al-doped ZrO₂ and Ag(111). In Figure 2, the supercell used for our calculations, prepared using Pymatgen can be seen. We have already completed the geometry optimization and are at the stage of calculating the adhesion energies. The R&D at Şişecam, at this point, have gained enough know-how to extend this example to any other system that they would be interested in. For all of the calculations in this project, we have used VASP.

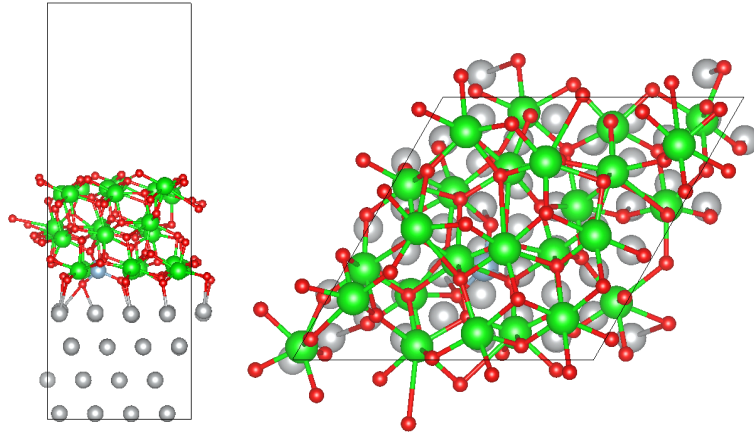
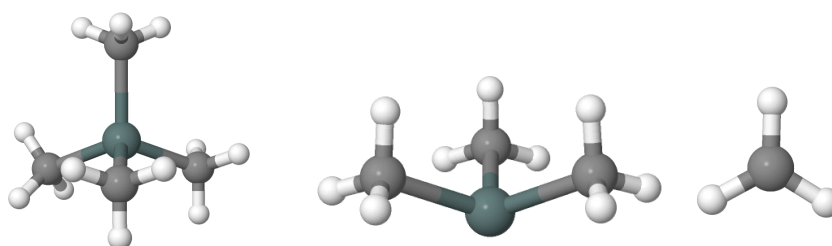


Figure 2: (Left) Side view and (right) top view of the the slab model used for the Al-ZrO₂/Ag interface. Blue, green, gray and red colors represent Al, Zr, Ag, and O atoms, respectively.

3. Preliminary Calculations - Project 2: The goal of the second project is to screen a really large number of molecules that can be used as a precursor in CVD experiments and find the ones that would yield the least amount of dissociation energy. As the prototypical example here, the NCC expert has used a common precursor, SnMe_4 , and using the software suite ORCA, has calculated the Sn-Me bond dissociation energy. (Figure 3) As requested by the Şişecam team, we have considered dissociation at finite temperature. Thus the calculations actually yield temperature-dependent free energies.



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INNER ENERGY
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The inner energy is: U= E(el) + E(ZPE) + E(vib) + E(rot) + E(trans)
E(el)  - is the total energy from the electronic structure calculation
        = E(kin-el) + E(nuc-el) + E(el-el) + E(nuc-nuc)
E(ZPE) - the the zero temperature vibrational energy from the frequency calculation
E(vib) - the the finite temperature correction to E(ZPE) due to population
        of excited vibrational states
E(rot)  - is the rotational thermal energy
E(trans)- is the translational thermal energy

Summary of contributions to the inner energy U:
Electronic energy      ... -373.70271985 Eh
Zero point energy      ...  0.14199969 Eh      89.11 kcal/mol
Thermal vibrational correction ... 0.00663379 Eh      4.16 kcal/mol
Thermal rotational correction ... 0.00141627 Eh      0.89 kcal/mol
Thermal translational correction ... 0.00141627 Eh      0.89 kcal/mol
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Total thermal energy    -373.55125383 Eh
  
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Figure 3: (Top panel) From left to right: Me_4Sn , Me_3Sn and Me , respectively. (Bottom panel) Excerpt from a typical ORCA output with the necessary thermodynamical data.

As of the writing of this report, our work plan has so far proceeded as planned. In the remaining two months, we will be completing our work as planned. The Şişecam team, once the PoC is completed will have access to all the input files, CoLab notebooks, meeting notes & recordings. This material will enable them to expand this work to any other material or molecule they would like to explore.

4. Workplan for the future: During the closing meeting of the PoC, the NCC expert presented the R&D team of the company with a work plan for materials discovery using the tools that the company has learned about. Since the company is interested, at the moment, to concentrate on the Ag/Metal oxide interface, the plan included

only this part of the PoC. To summarize, the NCC expert identified a diverse selection of metal oxides (TiO₂, Al₂O₃, SnO₂ etc) and dopants (Al, V etc.) that are all experimentally viable. The R&D team selected those materials that match their own goals and financial means. The expert then designed the calculations and shared with the team the necessary input files.

7. Business Benefits

Project 1: Thin film coatings are the primary way of adding value to glass, and computational modeling can provide insight into the suitability of new materials for use in functional coatings in glass. Low-e glass is the standard façade element in all new buildings and indispensable for energy economy. An Insulated Glass Unit bearing a solar low-e coating has similar heat insulation characteristics to that of a well-insulated wall. Better and more durable coatings will improve performance, reduce emissions and reduce material recycling needs owing to their longer lifespan.

Project 2: Greener chemicals that can be handled easily shall reduce the environmental footprint of the CVD technology while providing performance advantages.

8. Challenges

Project 1: Occasionally there are interfaces between amorphous (at least partially) and crystalline layers. Modeling of such interfaces might entail the use of larger unit cells

Project 2: High temperature deposition reactions for SnO₂:F are known to be very complex with a multitude of pathways and they are largely intractable by experimental or computational means [6]. Therefore, the experimental and theoretical literature on this chemistry still has many gaps. However, a careful study of bond enthalpies of candidate precursors can enable the identification of greener chemicals.

9. HPC Usage Status

Please discuss the HPC usage status before and after the PoC of the industry representative. The schema and the corresponding definitions are given below.

Şişecam is the largest glass producer in Türkiye. While HPC resources may be employed in different groups of the company, the R&D team who will work with our NCC has not used HPC resources before. From this point of view, the status can be considered digitalization-needed.

By the end of the project, the R&D team learned how to log on to the HPC system, the details of the software that are needed to perform the calculations, and to analyze the results. Unfortunately, due to the tight security regulations that the company has, the members of

the team are not freely able to log on to the system while they are on Şişecam premises. However, they are looking into resolving this problem.

10. References

1. W. Zhang, J.R. Smith, A.G. Evans, The connection between ab initio calculations and interface adhesion measurements on metal/oxide systems: Ni/Al₂O₃ and Cu/Al₂O₃, *Acta Materialia*, Volume 50, Issue 15, 2002, Pages 3803-3816, ISSN 1359-6454,
2. Cornil, D., Wiame, H., Lecomte, B., Cornil, J., & Beljonne, D. (2017). *Which Oxide for Low-Emissivity Glasses? First-Principles Modeling of Silver Adhesion*. *ACS Applied Materials & Interfaces*, 9(21), 18346–18354. doi:10.1021/acsami.7b03269
3. Cornil, D., Rivolta, N., Mercier, V., Wiame, H., Beljonne, D., & Cornil, J. (2020). *Enhanced Adhesion Energy at Oxide/Ag Interfaces for Low-Emissivity Glasses: Theoretical Insight into Doping and Vacancy Effects*. *ACS Applied Materials & Interfaces*. doi:10.1021/acsami.0c07579
4. Gordon, R.G. Criteria for Choosing Transparent Conductors. *MRS Bulletin* **25**, 52–57 (2000). <https://doi.org/10.1557/mrs2000.151>
5. Roy Gordon, Chemical vapor deposition of coatings on glass, *Journal of Non-Crystalline Solids*, Volume 218, 1997, Pages 81-91, ISSN 0022-3093.
6. Allendorf, M.D., van Mol, A.M.B. Gas-Phase Thermochemistry and Mechanism of Organometallic Tin Oxide CVD Precursors . In: Fischer, R.A. (eds) *Precursor Chemistry of Advanced Materials*. Topics in Organometallic Chemistry, vol 9. Springer, Berlin, Heidelberg. <https://doi.org/10.1007/b136141>