

D3.1 – Surface treatment model calibrated

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List of abbreviations

HTS	High-Throughput Screening
MD	Molecular Dynamics
CVs	Collective Variables
CSH	Calcium Silicate Hydrate
MBL	Methylene butyrolactone
LAMMPS	Large-scale Atomic/Molecular Massively Parallel Simulator
PBC	Periodic Boundary Conditions
PPPM	Particle-Particle Particle-Mesh
CG	Conjugate Gradient
DIPC	Donostia International Physics Center





1 Executive Summary

This report details the development and calibration of a surface treatment modeling tool designed specifically to identify optimal bio-based monomers for coatings with antifouling and anticorrosive properties. This tool employs a metadynamics approach to evaluate the affinity of diverse monomers to concrete surfaces. Through this methodology, a comprehensive high-throughput screening (HTS) of large set of bio-based monomers can be performed to identify those with the highest binding affinity for the substrate.

2 Introduction

Effective protection of concrete structures in marine environments is a central concern in civil engineering. Corrosion and biofouling can compromise the integrity and durability of these structures, posing a significant challenge to the sustainability and safety of coastal and offshore infrastructures. Therefore, the development of advanced coatings with antifouling and anticorrosive properties is a priority in material science research, as they not only protect concrete structures against corrosion and biofouling but also contribute to extending their service life and reducing maintenance and repair costs.

A key aspect in this field is the use of bio-based materials in coating formulations, aiming to reduce environmental impact and enhance the overall sustainability. However, achieving a bio-content percentage of over 80% in these coatings, as committed in this project, is a considerable challenge. It requires the careful identification and selection of bio-based monomers with specific antifouling and anticorrosive properties, which also exhibit adequate adhesion to the substrate to prevent delamination and ensure long-lasting protection of structures in demanding marine environments.

In this context, Task 3.2 (Modelling of surface treatments) involves the development and calibration of a surface treatment modelling tool designed to assist in the process of identifying optimal biobased monomers for coating formulations. This tool, based on a metadynamics approach, allows for the evaluation of the affinity of a large database of monomers to concrete surfaces at the atomic level, thus facilitating the selection of the most suitable materials for coating. This tool provides valuable guidance for monomer selection and coating formulation with high bio content.

Finally, it is noteworthy that this surface treatment modelling tool is not only applicable to the evaluation of monomers for bio-based coatings but also sufficiently flexible to be used in assessing other materials and coating systems. For example, it can be adapted to study the interaction of petroleum-based monomers, small polymers, or organic molecules with a variety of substrates, such as metals, zeolites, or clays. This versatility further expands its utility in materials research and the design of coatings for various industrial and environmental applications.





3 Methodology

In the present project, the evaluation of the affinity of various monomers to concrete surfaces has been proposed. A metadynamics approach has been considered to do so since this technique is a powerful tool for exploring the chemical free energy landscape of the system, enabling the evaluation of the affinity between the monomer and the surface at atomic-level to identify optimal monomers for surface coating applications. Nevertheless, to develop a robust tool capable of analyzing large sets of monomers, it is essential not only the implementation and calibration of the metadynamics simulation for accurate evaluation of monomer-surface affinity, but also the design of the scripts that facilitate the creation and equilibration of the monomer-surface to be analyzed. Moreover, it must be flexible enough to allow the exploration and study of different monomers by only changing the specification for these molecules, thus enabling comprehensive analysis and identification of optimal candidates for surface coating applications.

3.1 Metadynamics

Metadynamics is a sampling technique used in computational chemistry to explore the free energy landscape of a molecular system. It operates by adding an external bias potential to the potential energy of the system at regular intervals during the simulation to effectively accelerate the sampling of energetically unfavorable states ¹. The aim of applying the bias potential is to encourage the system to visit unexplored regions of the configuration space, thereby driving the exploration towards new areas and effectively accelerating the sampling of energetically unfavorable states. This allows the characterization of complex molecular processes that occur over long timescales in traditional molecular dynamics (MD) simulations.

The applied bias potential allows that the energy barriers hindering the transitions between different states to be lowered, allowing the system to explore a broader range of conformations and pathways. To effectively do it, over the course of the simulation, the bias potential is incrementally built up, creating a history-dependent bias that evolves with time ¹. This ensures that the system continuously explores new regions of the free energy landscape. The bias potential is constructed based on the collective variables (CVs) that describe the relevant degrees of freedom of the system. These CVs can represent structural or dynamical properties of the system, such as distances between atoms, angles, torsions, or more complex order parameters.

Focusing on the adhesion of monomers and the concrete surface, metadynamics allows for investigating how monomers interact with the surface and what are the most stable and energetically favorable binding modes and the calculation of their associated free energies. Metadynamics achieves this by introducing an additional "bias potential" into the equation of motion of the particles of the monomers. This potential is gradually built up during the simulation, adding small contributions as the simulation progresses to push the monomers towards regions of the configuration space that have not yet been explored, facilitating the search for energetically favorable configurations. By periodically adding Gaussian-shaped hills to the bias potential centered around the observed states, metadynamics effectively fills in the free energy minima and calculates the free energy profile along the chosen collective variables.

Once the metadynamics simulation is complete, the data is analyzed to reconstruct the free energy profile of the system. The resulting free energy landscape provides valuable insights into the





thermodynamics and kinetics of the molecular process under investigation. It identifies the most stable states of the system (energy minima), as well as the transition pathways and energy barriers to be overcome for certain interactions between the monomers and the surface to occur.

The considered metadynamics not only offers an efficient exploration of the free energy landscape of chemical systems, but also exhibits allowing for thorough sampling of various conformations and energy states. Its flexibility extends to handling complex chemical systems, including those with large surfaces and diverse molecular compositions ². Consequently, it serves as a suitable tool for investigating the interaction between monomers and concrete surfaces, which present intricate molecular environments.

3.2 System

To analyse the interaction between the monomers and the surface, first it is necessary to define and model the system under study. This system comprises two blocks of calcium silicate hydrate (CSH), the main hydration product of cement, that sandwich the monomer under study. To construct the atomic structure of the CSH with a Ca/Si ratio of 1.67, which is representative of typical proportions found in CSH formed during the hydration process of Portland cement ³, the pyCSH code ⁴ was used. Each CSH block measures 5.4 nm, 5.2 nm, and 2.5 nm in the x, y, and z directions, respectively.

Between these two blocks, a pore filled with water of approximately 6 nm wide in the z-direction is present. To allow the study the interaction between the CSH surface and the monomer, this pore is opened, leaving a central cavity with dimensions of 3 nm in the z direction (see Figure 1a). This cavity is large enough to accommodate any of the bio-based monomers listed in





Table 1, initially proposed as potential candidates for the experimental development of the coating in WP2. After the insertion of the monomer, the system is equilibrated by performing molecular dynamics simulations to close this pore and relax all the atomic positions, as shown in Figure 1b. The methylene butyrolactone (MBL) monomer has been employed in the calibration of the tool proposed in this project. Nevertheless, the final width of the pore and the dimensions of the whole system will be influenced by the specific size monomer introduced into the system, although, due to the small size of the monomer in comparison with pore size, slight differences in the final equilibrated dimensions of the pore are expected.



Figure 1 Snapshot of the MBL-CSH system (a) before and (b) after MD equilibration. The yellow tetrahedra correspond to silicates, while the light blue balls illustrate the Ca atoms. Water molecules are represented by red and white sticks, while in the MBL molecule, the C, O, and H atoms are shown in black, red, and white respectively.





Table 1 Initial proposed bio-based monomers for the coating development in WP2.

Name	Structure	
2-Octyl acrylate (2-OA)		
2-Octyl methacrylate (2-OMA)		
Tetradecyl methacrylate (TdMe)	$\int_{12}^{0} (\cdot)_{12}$	
Tetrahydrogeraniol acrylate (ThgA)		
Tetrahydrogeraniol methacrylate (ThgMe)		
Isobornyl acrylate (IBOA)		
Isobornyl methacrylate (IBOMA)		
Tetradecyl acrylate (TdA)		
Methylene butyrolactone (MBL)		
Piperonyl methacrylate (PIPEMA)		
Dimethyl ithaconate (DMI)		
Isosorbide methacrylate (ISOMA)		
Isosorbide dimethacrylate (ISODMA)		
Ecomer		





3.3 Scripts

To conduct both molecular dynamics and metadynamics simulations, the LAMMPS software package ^{5,6} is employed. The structural information of the CSH surface and the monomer is described in separate data files, specifying the molecular systems to be simulated and the interaction potential between particles. Additionally, two primary scripts have been developed for performing the molecular dynamics equilibration and the metadynamics simulation. These scripts have all the parameters for the simulation run, encompassing the initial equilibration of the system and the exploration of monomer-surface interactions to assess the binding affinity of the monomers to the concrete surfaces.

Data files

The data files provide essential structural information to define the simulated system and include parameters describing particle interactions. Specifically, they contain details such as the type and number of atoms, bonds, angles, dihedrals, and impropers, as well as atomic masses, charges, pair coefficients, bond coefficients, angle coefficients, dihedral coefficients, and improper coefficients. These latter parameters are derived from the LigParGen force field ^{7–9} for the monomers, while the CSHFF ¹⁰ has been utilized for the CSH surfaces.

The LigParGen force field ^{7–9} is a versatile tool for modeling the interactions of organic molecules in various conditions and environments. It is widely used due to its accuracy and flexibility in representing a wide range of molecular structures and chemical environments. By utilizing LigParGen, researchers can accurately describe the behavior of monomers in different solvents, temperatures, and pressure conditions, making it suitable for simulating monomer-surface interactions in diverse settings.

On the other hand, the CSHFF ¹⁰ is a specialized force field specifically developed to model the complex interactions within CSH surfaces. The CSHFF accurately captures the structural and energetic properties of CSH surfaces, allowing for precise simulations of interactions between CSH surfaces and other molecules or materials.

By employing two separate data files for monomers and CSH surfaces, a broad spectrum of monomers can be analyzed simply by swapping out the data file corresponding to the monomers, while maintaining the data file for CSH. This modular approach allows a more efficient exploration of the monomer candidates without the need to modify the parameters for the whole system for each simulation. Additionally, it also ensures consistency and accuracy in the simulation results by maintaining the integrity of the CSH surface representation throughout the analysis of different monomer candidates.

Input files

The input files provide the configuration for the simulations to be performed, covering various aspects including the loading of data files, specifications for the force-field, parameters for energy minimization and molecular dynamics simulation, and settings for metadynamics simulation. In particular, the input file indicates that periodic boundary conditions (PBC) were applied in all three directions, allowing for the simulation of an infinite system by replicating a unit cell. The interaction between the monomers and CSH surfaces is modeled using electrostatic and dispersive forces. The long-range Coulombic interactions are treated using the Particle-Particle Particle-Mesh (PPPM)





method, while the dispersive forces are computed by a geometric combination of the Lennard-Jones parameters of each atomic pair.

The energy of the system is initially minimized using the Conjugate Gradient (CG) method to relax both the simulation box and the atomic positions. Subsequently, molecular dynamics in the isobaric–isothermal ensemble (NPT) is performed for 5 ns to equilibrate the atomic positions and volume under room temperature and pressure conditions (298K and 1 atm) using a thermostat and barostat coupling constants of 0.1 ps and 1 ps respectively. The equations of motion are integrated using a Verlet algorithm with a time step of 1 fs.

Following the equilibration phase, metadynamics simulation is initiated to explore the free energy landscape of the system for 50 ns under NVT ensemble at 298K, with a timestep of 1 fs. The metadynamics simulation utilizes collective variables defined to monitor the distance between two key components in our system: the monomer and the surface of CSH along the z-axis, perpendicular to the surface. Tracking changes in this distance provides insight into how the monomer interacts with the cement surface throughout our simulation, focusing on the monomer's binding affinity to the surface. It is important to note that while the focus is on the collective coordinate along the z-axis, relevant to the monomer-CSH surface interaction, exploration also occurs in the x and y directions, parallel to the CSH surface. This exploration in the x and y axes progresses as molecules move and explore different configurations during the metadynamics simulation. The collective variables also define the bias potential applied to drive the evolution of the system towards various configurations of the monomer-surface distance, aiming to sample a broad range of distances and understand how they evolve over time under different conditions and external forces.

The calibration of the parameters to perform the metadynamics simulations is crucial to ensure the effectiveness and accuracy of the sampling process. One key aspect is the correct specification of the boundaries within the system which can evolve along the defined collective coordinates. These boundaries restrict the system's movement, ensuring the exploration of relevant regions of the collective coordinate space while avoiding irrelevant or unphysical states that could lead to inefficient exploration of the conformations of the system or compromise the quality of the obtained data. For the MBL-CSH system employed in the calibration, the lower and upper boundary limits have been set between 0 and 36. This corresponds to the distance spanning from the center of the pore (z = 0 Å), where the monomer is initially placed, to the CSH, whose surface is located at an approximate distance of z = 36 Å after equilibration with MD, during which the pore closure and its stabilization occur. While the width of the pore may vary slightly depending on the size of the monomer employed, the variation is not significant, as size of the monomers is relatively small and the pore initially contains enough water to stabilize at around 7 nm.

The definition of Gaussian hills added to the bias potential during metadynamics simulations is also critical. These hills act as energy barriers that modify the potential energy landscape, encouraging the system to explore new configurations in the collective coordinate space. The height, width, and frequency of these hills must be carefully adjusted to balance the exploration and maintain an accurate representation of the system's energy landscape, preventing them from prematurely dominating the energy landscape and distorting the simulation results.

The height of the Gaussian hills determines the strength of the biasing effect, influencing the rate at which the system explores new configurations. A higher hill height accelerates the sampling





process but may lead to premature convergence or biasing toward specific states. Conversely, a lower hill height slows down exploration but ensures a more thorough sampling of the energy landscape. Similarly, the width of the Gaussian hills defines the extent of the perturbation introduced to the system. Narrower hills provide localized perturbations, facilitating exploration of specific regions, while broader hills offer more diffuse influences, allowing for broader exploration of the collective coordinate space. Finally, the frequency at which Gaussian hills are added to the bias potential also influences the sampling efficiency. Higher frequencies result in more frequent updates to the bias potential, promoting rapid exploration, but it may lead to oversampling of certain regions, while lower frequencies ensure more thorough sampling and prevent premature convergence, but may slow down the exploration excessively.

Proper calibration of these parameters is essential to achieve simulation convergence, ensure thorough exploration of conformational space, and optimize the computational efficiency. To this end, we have conducted a comprehensive analysis of various parameter values, including height, width, and frequency of the Gaussian hills, as well as the number of processors utilized to run the simulations. Through meticulous testing and iteration, we evaluated the impact of varying parameter configurations on the accuracy of the results. Specifically, we incrementally adjusted the height, width, and frequency of the Gaussian hills, monitoring changes in the free energy landscape in order to select optimal parameter values that ensure that free energy landscape does not significantly change when increasing them to minimize computational cost. Table 2 below summarizes the values of parameter tested and the resulting free energy after convergence.

Height	Width	Frequency	Free Energy (kcal/mol)
0.1	1.0	100	-18.2
0.5	1.0	100	-18.4
1.0	1.0	100	-18.3
1.5	1.0	100	-18.1
2.0	1.0	100	-18.9
2.5	1.0	100	-19.5
3.0	1.0	100	-20.3
1.5	0.1	100	-18.2
1.5	0.5	100	-18.0
1.5	1.0	100	-18.1
1.5	1.5	100	-18.4
1.5	2.0	100	-18.3
1.5	2.5	100	-18.8
1.5	3.0	100	-19.2
1.5	2.0	50	-18.3
1.5	2.0	100	-18.3
1.5	2.0	150	-18.2
1.5	2.0	200	-18.5
1.5	2.0	250	-18.5

Table 2 Summary of Parameter Calibration Analysis.





Based on the analysis of the results shown in Table 2, we have concluded that the parameters: height 1.5, width 2.0, and frequency 150 offer an optimal balance between result accuracy and computational efficiency. This determination is based on the observation that while lower values of height, width, and frequency typically yield more accurate results, the outcomes obtained with these lower values closely resemble those obtained with the selected parameters, yet they demand fewer computational resources. For the simulations presented in Table 2, we utilized 8 processors available on the Atlas supercomputer at DIPC (Donostia International Physics Center) facilities. To further optimize simulation time, we conducted an analysis spanning from 4 to 32 processors, as depicted in Figure 2. Our findings reveal that while the simulation speed increases with the use of additional processors, the rate of improvement diminishes notably beyond 8 processors. As such, we have chosen to employ 8 processors to avoid excessive resource consumption and cluster saturation.



Figure 2 Relationship between simulation speed and the number of processors utilized in the computational simulations. The simulation speed is measured in simulation time (in nanoseconds) per real time (in hours).

Output files

During the simulation, several output files are generated and they provide comprehensive insights into the temporal evolution of the system, encompassing atomic coordinates, potential and kinetic energies, temperature, pressure, and other relevant thermodynamic quantities. In addition, during the metadynamics simulation, two key output files are generated, providing valuable insights to assess the binding affinity of the monomers to the concrete surfaces, and to identify preferred binding configurations.

One of the output files records the trajectory of Gaussian hills added to the bias potential. This file tracks the variations in the position of each Gaussian hill along the z-axis throughout the simulation. The hills trajectory file provides valuable insights into the evolution of the bias potential landscape over time since it illustrates how the positions of Gaussian hills change as the simulation progresses. Thus, when multiple Gaussian hills accumulate at a certain position along the z-axis, it indicates a minimum in the energy landscape since in those regions, more Gaussian hills are needed to fill the energy well.

Figure 3 depicts the Hills Trajectory for the MBL-CSH system used in tool calibration using the selected parameters for the Gaussian hills. This graph illustrates the distribution of Gaussian hills along the z-axis, providing a visual representation of the energy landscape explored during the





metadynamics simulation. The Gaussian hills accumulate across all regions, facilitated by the additional energy that enables transitions to alternative configurations throughout the entire simulation period. Consequently, it ensures comprehensive exploration of the interlayer space, encompassing configurations with varying degrees of stability.



Figure 3 Evolution of the position of the Gaussian hills along the z-axis over time during the metadynamics simulation.

During metadynamics simulation, another output file is generated periodically containing information about the free energy landscape of the system. This landscape is given as a function of the defined collective variable, the distance between the monomer and the surface of CSH along the z-axis. In this way, this output offers a detailed profile of the free energy of the system in relation to this distance. Consequently, it facilitates the identification of stable binding configurations between the monomer and the CSH surface, corresponding to local minima in the free energy profile, along with energy barriers or less favorable states, associated with local energy maxima and indicating the energy required to overcome specific interactions. In this output, the free energy values are normalized for direct comparison between systems with different monomers, facilitating the assessment of relative stability and adhesion strength across various the set of monomers to be analyzed, allowing to discern the most favorable monomers and configurations for binding to the CSH surface.

As a periodically generated output file, the free energy in the output file represents an accumulated average of values obtained during the simulation time up to the file generation moment. Therefore, each point of free energy along the z-coordinate does not reflect the free energy at a specific instant but rather a cumulative estimate over the simulation time.

Figure 4 illustrates the evolution of free energy along the z-axis as the MBL monomer approaches the surface of CSH once the simulation has converged. The y-axis represents the normalized free energy associated with each position along the perpendicular direction of the CSH surfaces (z-axis), while the x-axis represents the distance between the monomer and the CSH along the z-axis. The monomer was initially located at the center of the pore, with z = 0Å. Therefore, as the distance along the z-axis increases, the monomer approaches the CSH surface, which is located approximately 18 Å from the center of the pore after equilibration.





The graph depicts fluctuations in free energy, indicating different levels of stability in the interaction between the monomer and the CSH surface. Peaks in the free energy profile indicate less stable configurations or energy barriers that the system must overcome to reach more stable states. Conversely, local minima in the free energy profile correspond to relatively stable binding configurations between the monomer and the surface, suggesting energetically favorable states. The depth of these minima reflects the strength of interactions between the monomer and the surface. Thus, the absolute minimum in the vicinity of the CSH surface indicates that the most favorable position or configuration exists at that specific short distance from the surface. This absolute minimum represents the most stable binding configuration between the monomer and the CSH surface, suggesting a highly favorable interaction at this particular distance.



Figure 4 Evolution of the Free Energy along the z-axis over time during the metadynamics simulation.

Expanding this analysis to other monomers, their respective affinities for CSH can be determined by assessing the depth of the potential well in the vicinity of the surface and inferring the strength of interaction between each monomer and the CSH. Monomers exhibiting deeper potential wells near the surface are indicative of stronger affinities for CSH, suggesting enhanced binding capabilities and potentially superior performance as a coating. On the contrary, those monomers showing less deep potential wells near the surface may indicate a weaker affinity for CSH, leading to a reduced binding capacity and potentially lower performance as a coating.





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