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Thesis Outline

The overall goal of this research work was to understand the physical properties of mineral surfaces at the nanoscale. Probing and characterizing physical properties and interactions (ligand-receptor binding) at the nanoscale using highly sensitive AFM was instrumental in accomplishing this goal.

Chapter 1 briefly discussed the existing knowledge that served as the background of this research work and presented the main goal and objectives. The role of minerals in prebiotic evolution and the importance of studying amino acid adsorption on mineral surfaces were discussed in the chapter.

Chapter 2, served as a general introduction to AFM, which was the main experimental approach used in this study. The working principles and the physical theories behind this instrument were presented at the beginning of the chapter. The discussion then moved to the introduction of the AFM force mode (dynamic mode). Through specific recognition processes, the force mode of AFM allows measurement of forces with piconewton sensitivity. Therefore, the basic and novel aspects of DFS were reviewed and the basic principles were described. The chapter finishes with the description of the immobilization strategies used for AFM tip in DFS experiments. The most common and efficient methods to immobilize amino acid molecules on AFM tips were reviewed with a particular emphasis on the use of molecular linkers to bind amino acids to the surfaces.

Chapter 3 was focused on surface force analyses of amino acids on pyrite. Glycine, lysine, and sulfur-containing amino acids were used to investigate the surface reactivity of this mineral. Sulfur vacancies on pyrite were characterized by Raman spectroscopy and found to augment amino acid adsorption by enabling the chemical bonding between Fe^{2+} of pyrite and the -COOH group of amino acids. The force measurements gave results in good agreement with the TDS results, in which glycine did not show chemical adsorption on the surface unlike the amino acids with side chains. These results suggest that the side chain of an amino acid may influence how it interacts with pyrite. However, more systematic experiments with various amino acids and under various environmental conditions are recommended for future studies to assess this mineral's ability to adsorb biomolecules.

Chapter 4 examined the adsorption of amino acids on the hematite surface. All amino acids used in this experimental section showed specific interactions with the hematite surface; however, the probability of specific events was found to be different among the amino acids. Specifically, glycine showed a much lower specific event probability compared to cysteine and lysine. In contrast, the adhesion forces between the three amino acids and the pyrite surface were similar. In our search for the reason underlying glycine's low probability of specific events in force measurements, we found that previous studies have also reported that glycine is less easily adsorbed than lysine. Moreover, it has been found that pH has a strong influence on amino acid adsorption on hematite.

Chapter 5 focused on titanium dioxide (TiO_2) as a representative clay mineral surface. Results from force analyses on surfaces with three different crystal orientations were summarized. Results suggested that the titanium dioxide (110) surface has unique characteristics for adsorbing glycine. Step structures in all crystals are believed to represent special bonding environments for adsorbing molecules. Hazen et al. reported such structures in TiO_2 (110). From our observations, it became clear that force curves on TiO_2 (110) showed different features compared to those on TiO_2 (001) and (100). To examine this difference, we investigated the interaction between the PEG cross-linker and the three TiO_2 surfaces. TiO_2 (110) interacted with PEG, while no specific interaction was observed on TiO_2 (001) and (100) surfaces. Although this will have to be verified by other techniques, our data suggest that the crystal shapes may have influenced the amino acid adsorption. Investigating the topography of this mineral's surfaces and identifying the sites where organic molecules attach should be pursued in future studies.