



Utilisation of Supercritical Critical Fluids in Adsorption Process: A Comprehensive Review

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Abstract

Conventional separation and purification processes in industries like pharmaceuticals and environmental remediation often face significant challenges, including high energy consumption, the use of large amounts of hazardous organic solvents, and limited efficiency in extracting or adsorbing target compounds. This review comprehensively examines the utilization of supercritical fluids SCFs in adsorption processes, highlighting their unique physicochemical properties such as tuneable density, low viscosity, and high diffusivity that bridge the gap between liquid and gas phases. The discussion covers fundamental adsorption mechanisms, thermodynamic principles, and the role of adsorption isotherms in supercritical conditions. Key applications in pharmaceuticals (e.g., drug encapsulation and controlled release), environmental remediation and separation processes are detailed. The review also addresses advanced adsorbent materials, including porous and nanostructured adsorbents, and their surface modification for enhanced performance. Furthermore, mass transfer mechanisms, kinetic modelling, and process integration such as supercritical fluid-assisted adsorption, extraction, and desorption are analysed. Despite significant advantages in efficiency, selectivity, and sustainability, challenges related to process optimization, material compatibility, and scalability remain. The review concludes with prospects, emphasizing the potential of SCFs in advancing green technologies and industrial applications through continued research and interdisciplinary innovation.

Keywords: Supercritical Fluid Adsorption (SFA); Supercritical Fluids (SCFs); Supercritical Carbon Dioxide (scCO₂); Supercritical Fluid Extraction (SFE), Adsorption Processes.

1.0 Introduction

SCFs represent a phase of matter for which specific substances or materials must be exposed to higher temperatures and pressures than those corresponding to their critical points. Its performance halfway between a liquid and gas depending upon its supercritical state makes this material useful in various applications. SCFs have distinct properties from those of normal liquids and gases. Their density is such that they can more easily dissolve materials, like liquids, while their viscosity is slightly higher than gasses, ensuring ready spreading and penetration into porous substrates. Furthermore, SCFs present higher diffusivity than liquids, making them excellent vehicles for fast mass transfer and efficient mixing in most applications. These properties are tunable; meaning by varying the temperature and pressure, you can specifically adjust a supercritical fluid's ability to dissolve different substances making them adaptable for various settings [1]. SFA is a novel technique combining adsorption with SCF technology, encompassing supercritical fluid chromatography (SFC) and supercritical fluid impregnation (SFI) [1]. SFA has applications in gas purification, fluid fractionation, and adsorbent regeneration [2]. Engineering models have been developed to describe physical adsorption from sub- to supercritical conditions, capable of quantitative fits over wide pressure and temperature ranges [3]. Lattice theory calculations have shown that SFA isotherms exhibit peculiar features, such as a maximum in adsorption with increasing pressure and two-layer adsorption over a wide range of densities [1]. These advancements in understanding and modelling SCF adsorption processes contribute to their growing industrial applications [4], [5], [6].

The adsorption behaviour of SCFs can be complex and is often characterized by non-ideal isotherms. For instance, the adsorption of volatile liquids in CO₂ has been shown to decrease with increasing temperature at constant concentration, contrary to typical exothermic adsorption behaviour. This phenomenon can be attributed to the interplay between fluid density and adsorption dynamics, indicating that the thermodynamic properties of SCFs significantly influence adsorption processes. Understanding these interactions is essential for optimizing adsorption systems, especially in applications involving mixed adsorbates or complex matrices. Adsorption isotherms play a pivotal role in characterizing the adsorption process. The Langmuir and Freundlich models are commonly employed to describe the relationship between the amount of adsorbate and its equilibrium concentration in the fluid phase. Recent studies have highlighted the importance of selecting appropriate isotherm models based on the specific characteristics of the adsorbent and the conditions of the adsorption process. In the context of drug delivery, the use of SCFs for the adsorption of pharmaceuticals onto carriers such as silica

aerogels has shown promise. This method allows for the encapsulation of poorly soluble drugs, enhancing their bioavailability while minimizing the risk of structural damage to the carrier material. The ability to control process parameters, such as pressure and temperature, further enables the tailoring of drug release profiles, making SCFs a valuable tool in pharmaceutical applications. In the context of supercritical fluid systems, this review sets itself apart by offering a thorough and holistic viewpoint that connects basic adsorption theory, thermodynamic modelling, and new material developments. This work critically synthesizes adsorption mechanisms, isotherm behaviour, and advanced adsorbent design under supercritical circumstances, in contrast to previous research that frequently address these topics separately. It also identifies important issues with scalability, material compatibility, and process optimization and emphasizes recent advancements in combining adsorption with SCF-assisted processes. The review seeks to improve knowledge of SFA and establish it as a feasible route for next-generation sustainable separation and processing technologies by using this multifaceted approach.

2.0 Fundamentals of in SCFs

2.1 Thermodynamic Properties of SCFs

SCFs possess unique thermodynamic properties arising from their position above the critical temperature (T_c) and pressure (P_c), blending characteristics of liquids and gases. These properties tunable via temperature and pressure adjustments underpin their utility in industrial processes like extraction, drying, and reactions. SCF heat capacity exceeds that of gases and approaches liquids, enabling efficient heat absorption or release with minimal temperature shifts. Near the critical point, fluctuates sharply due to elevated compressibility[7]. This trait aids thermal management in reactions, controlling exothermic or endothermic profiles. The thermal conductivity of SCF lies between gases (low) and liquids (high), promoting better heat transfer than gases but demanding precise gradient control to prevent hotspots. In reactor design, this influences uniform heating/cooling for extractions and syntheses. Enthalpy rises with temperature, yet shows pronounced variations near criticality from compressibility effects. Entropy undergoes abrupt shifts during gas-liquid-like transitions. These dynamics optimize energy efficiency in processes like supercritical drying and reactions by enhancing heat/work transfer[8].

2.2 Phase Behaviour and Solubility in SCFs

SCFs do not exhibit distinct phase boundaries between liquid and gas, which is a defining characteristic of the supercritical state. As a result, SCFs can transition smoothly between behaving more like a gas or more like a liquid based on temperature and pressure changes. This absence of a phase transition simplifies processes like extraction and separation, as there is no need to overcome surface tension between phases. Thus, the lack of phase separation allows solutes to dissolve and precipitate out more uniformly, making SCFs ideal for crystallization and material synthesis. Understanding the distinct properties of SCFs is a prerequisite to comprehending the solubility phenomenon in these fluids. Fig. 1 depicts the usual PT phase diagram of a pure material.

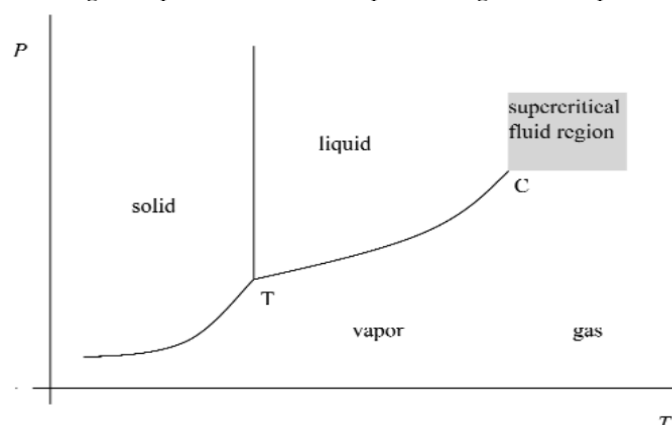


Figure 1: Pressure–temperature diagram for a typical pure substance[9].

The solubility of supercritical fluid is different from conventional organic compounds. SCFs usually demonstrate superior solubility for non-polar molecules as compared to standard organic solvents. Many factors, including pressure, temperature, solute type, and the nature of the supercritical fluid, can influence a solute's solubility[10].

At higher pressures, solubility often increases because the fluid has a higher density. Temperature also affects solubility, with higher temperatures typically being linked to higher solubility levels. One of the most often utilized SCFs is carbon dioxide (CO_2). Essential oils, flavors, and natural goods are examples of non-polar and lipophilic substances for which it has outstanding solvating potential[11]. Combining the two properties (density and diffusivity) appropriately makes SCFs much better solvents than expected. This is particularly true when the temperature and pressure are not too far above the critical temperature and pressure[9].

Table 1 lists the critical temperature, critical pressure, and acentric factors for several substances commonly used as supercritical solvents.

Table 1: Critical temperature and pressure and acentric factor for common SCF solvents [9]

Solvent	T_c/K	P_c/bar	ω
Carbon dioxide	304	73.7	0.225
Ethane	305	48.7	0.099
Ethene	282	50.4	0.087
Propane	370	42.5	0.152
Ammonia	405	114	0.257
Nitrous oxide	310	72.5	0.141
Trifluoro methane	299	48.4	0.267

The ability to tune solvent power is a major advantage of SCFs, enabling selective extraction, reaction, and purification of target compounds. Supercritical CO₂ is widely used as a solvent in extraction processes because of its mild critical temperature (31°C) and non-toxicity. By increasing pressure, its solvent power can be increased to dissolve a wide range of organic compounds, while lowering the pressure causes it to behave more like a gas, allowing for easy separation of solutes from the solvent.

2.3 Transport Properties and Mass Transfer in SCFs

SCFs have higher diffusivity than liquids but lower diffusivity than gases. Diffusion in SCFs occurs more readily than in liquids, allowing solutes to spread faster within the fluid. This property enhances the rate of mass transfer, making SCFs efficient solvents for extraction, reaction, and separation processes.

The higher diffusivity of SCFs, combined with their ability to dissolve solutes effectively, makes them particularly useful in processes where *rapid mass transfer* is critical, such as in catalysis or chemical reactions. When describing the mass transfer rate in supercritical fluid systems, the mass transfer coefficient (K) is frequently employed. Pressure, temperature, fluid velocity, and the composition of the solute and matrix are some of the variables that affect it.

3.0 Adsorption Mechanism in SCFs

Adsorption processes play a crucial role in various applications, such as catalysis [12], gas storage [13], separation and purification [14], sensors [15], chromatography [16], water/wastewater treatment [17], [18], environmental remediation [3], catalysis [19], [20], [21], [22], and drug delivery systems [23]. The process of adsorption involves the accumulation of molecules from a gas or liquid phase onto a solid surface (adsorbent), creating a concentration gradient that drives the process. The efficiency of adsorption is influenced by factors like the nature of the adsorbate and adsorbent, temperature, pressure, and the specific characteristics of the adsorption isotherm.

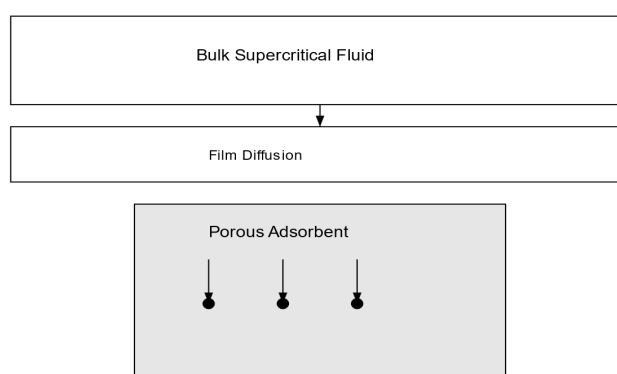


Figure 2. Schematic representation of adsorption in supercritical fluids, showing external film diffusion, intraparticle diffusion, and adsorption at active sites, enhanced by high diffusivity and tuneable solvent strength of SCFs.[24]

3.1 Mechanism and Factors Influencing Adsorption

Properties of Adsorbate and Adsorbent. The nature of both the adsorbate (the substance being adsorbed) and the adsorbent (the material onto which adsorption takes place) has a great influence in the process [4], [25], [26]. For example, the chemical composition, surface area, and porosity of the adsorbent can significantly affect its adsorption capacity and efficiency.

Temperature and Pressure: The effect of temperature in the adsorption process is generally manifested through its influence on the kinetic energy of molecules and the nature of adsorbate-adsorbent interactions. Influence of temperature on adsorption process may either be negative or positive, for example rise in temperature may either enhance or inhibit adsorption depending on the process type involve whether exothermic or endothermic. Pressure, particularly in the case of gases or SCFs, plays a critical role in determining the extent of adsorption.

3.2 Significance of SCFs in Adsorption

The utilization of SCFs in the adsorption process has gained attention due to their unique properties, which combine characteristics of both gases and liquids that cannot be found in any of either gas or liquid alone. These dual properties allow SCFs to effectively dissolve solutes and penetrate porous materials, enhancing efficiency of the adsorption processes. Enhancing the solubilization of active compounds is one of the fundamental benefits of using SCFs in adsorption processes. The mechanism of supercritical adsorption entails the solubilization of a solute within a supercritical medium, succeeded by its diffusion into a porous substrate, such as silica aerogel. Empirical evidence has demonstrated this technique to provide efficient absorption of drugs with poor water solubility, thereby fostering the development of novel drug delivery systems that overcome the constraints associated with conventional liquid solvent methods, which may jeopardize the structural integrity of the adsorbent ((Wang et al., 2023) Chai & Tassou, 2023). The addition of co-solvents, such as ethanol, significantly enhances the solubility of the active compound, thereby optimizing the adsorption process [27]. The behavior of adsorption under supercritical conditions is distinguished by mild operational parameters, which may facilitate more straightforward and efficient separation methodologies. Empirical studies suggest that the adsorption and desorption of organic compounds in supercritical carbon dioxide (scCO₂) can be conducted under conditions that reduce energy expenditure and enhance recovery efficiencies. Nevertheless, the equilibria and dynamics pertaining to co-adsorption phenomena involving scCO₂ and organic compounds are still inadequately investigated, indicating a pressing need for further research in this domain [28], [29].

The modelling of adsorption isotherms in supercritical environments presents additional challenges. Current theoretical frameworks are not fully developed, particularly concerning the mechanisms governing adsorption in porous materials under supercritical conditions. Understanding these mechanisms is crucial for accurately predicting adsorption behaviour and optimizing process parameter. The application of supercritical fluid technology has also been utilized in the field of material science to produce nanoparticles and other advanced materials. Techniques such as the Supercritical Antisolvent (SAS) method allow for the controlled precipitation and encapsulation of pharmaceutical compounds, enhancing their bioavailability and therapeutic efficacy [30]]. The ability to manipulate process parameters, such as scCO₂ density and solution concentration, provides researchers with the tools necessary to tailor the properties of the resulting materials [2]. SCFs offer unique properties that make them highly effective in adsorption processes, such as their ability to penetrate deep into porous materials and their tuneable solvating power. This makes them ideal for removing impurities, capturing desired compounds, and achieving high selectivity in separation processes. Additionally, the use of SCFs can lead to reduced energy consumption, shorter processing times, and the ability to recover and reuse solvents, further contributing to a more sustainable and efficient industrial practice. Overall, the significance of SCFs in adsorption lies in their potential to drive innovation, improve process efficiency, and promote environmental stewardship in various industries. One key advantage of using SCFs in adsorption is their unique properties that can be easily tailored by adjusting pressure and temperature, allowing for precise control over their adsorption capabilities. These fluids have a high diffusivity and low viscosity, which enables them to penetrate deep into porous materials and interact with a wide range of compounds with high efficiency. Their tuneable solvating power also makes them ideal for removing impurities, capturing desired compounds, and achieving high selectivity in separation processes. Additionally, the use of SCFs can lead to reduced energy consumption, shorter processing times, and the ability to recover and reuse solvents, further contributing to a more sustainable and efficient industrial practice. Overall, the significance of SCFs in adsorption lies in their potential to drive innovation, improve process efficiency, and promote environmental stewardship in various industries.

4.0 Thermodynamics and Adsorption Isotherms

Thermodynamics of SFA focuses on the study of the interactions between SCFs and solid surfaces to better understand the adsorption process at a molecular level. This review will explore the thermodynamic principles that govern the adsorption behavior of SCFs, including the factors that influence adsorption equilibrium, adsorption isotherms, and the determination of thermodynamic parameters such as enthalpy, entropy, and Gibbs free energy. By delving into the thermodynamics of SFA, researchers and industry professionals can gain valuable insights into optimizing process conditions, designing efficient adsorption systems, and ultimately achieving sustainable and environmentally friendly practices. This knowledge can also help in predicting the behavior of SFA systems under different operating conditions, allowing for more accurate modeling and simulation. Additionally, understanding the thermodynamic principles behind SFA can aid in the development of new

materials and technologies for various applications, such as separation processes, drug delivery systems, and environmental remediation. Overall, a deeper understanding of the thermodynamics of SFA can lead to significant advancements in the field and contribute to the development of more efficient and sustainable processes. SFA involves the interaction of SCFs with solid adsorbents, a process that is influenced by the unique thermodynamic properties of SCFs. Here's an overview of the thermodynamics involved in this process:

- i. **Adsorption Isotherms:** The relationship between the amount of adsorbate on the adsorbent and the pressure or concentration of the adsorbate at constant temperature is described by adsorption isotherms (e.g., Langmuir, Freundlich models).
- ii. **Gibbs Free Energy:** The spontaneity of adsorption can be evaluated using the Gibbs free energy change (ΔG). A negative ΔG indicates that the adsorption process is spontaneous.
- iii. **Enthalpy and Entropy Changes:** The changes in enthalpy (ΔH) and entropy (ΔS) during the adsorption process can provide insights into the nature of the interactions between the SCF and the adsorbent.
- iv. **Temperature and Pressure Effects:** The adsorption capacity and selectivity of SCFs can be significantly affected by temperature and pressure, due to changes in density and solvation power of the SCF.

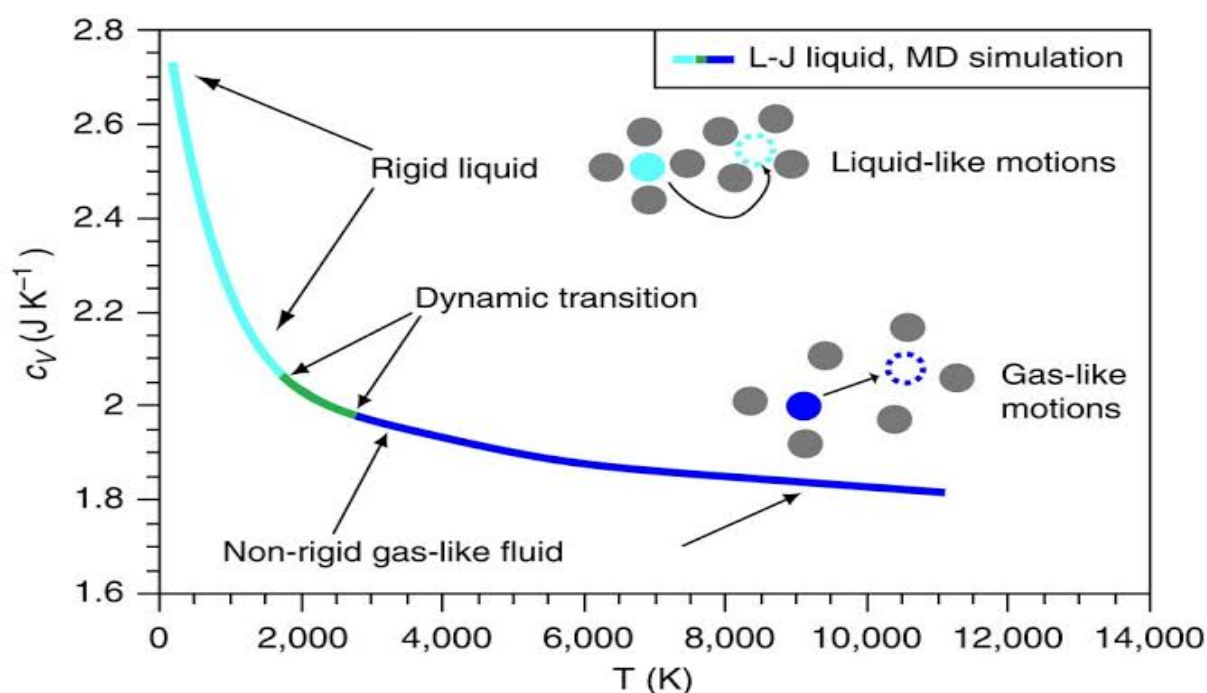


Figure 3: Thermodynamics of SCFs Adsorption

Adsorption equilibria in SCFs are influenced by a combination of factors, including temperature, pressure, and the nature of adsorbent material. By studying these equilibria, the adsorption efficiency can be enhanced through optimization of these conditions. This knowledge is crucial for the design and implementation of supercritical fluid-based processes in various industries, ultimately leading to more sustainable and cost-effective solutions. Additionally, new possibilities for innovation and the development of novel technologies in the future may open up by advancements in understanding adsorption equilibria in SCFs. For example, by fine-tuning the parameters of the system, such as temperature and pressure, researchers can increase the adsorption capacity of SCFs, making them more efficient for use in processes like extraction, separation, and purification. This can result in reduced energy consumption, waste generation, and overall costs for industries utilizing supercritical fluid technology. Overall, a deeper understanding of adsorption equilibria in SCFs holds great potential for revolutionizing traditional industrial processes and paving the way for more sustainable and environmentally friendly practices. By optimizing the conditions for adsorption in SCFs, the systems can be tailored to specific applications, thereby improving their effectiveness and efficiency. This enhances level of control that allows for more precise and selective extraction of desired compounds, minimizing the need for harsh solvents or extensive purification steps. Additionally, the ability to tune the adsorption capacity of SCFs opens up new possibilities for the development of novel separation techniques and the production of high-purity products. With further research and development in this field, supercritical fluid technology has the potential to transform various industries and contribute to a more sustainable future. However, it is important to note that SFE may not always be suitable for all compounds, as certain substances may not be easily soluble in SCFs. In such cases, traditional extraction methods may still be necessary to achieve the desired results. Additionally, the cost of implementing supercritical fluid technology can be a barrier for some industries, as the equipment and expertise required can be expensive.

Despite these challenges, the benefits of SFE, such as higher extraction yields and reduced solvent usage, make it a promising option for many applications. Research into optimizing the process parameters and exploring new applications for SCFs is ongoing, with the potential to revolutionize the way we extract and purify a wide range of compounds. As technology continues to advance, the possibilities for SFE are endless, offering a greener and more efficient alternative to traditional extraction methods.

4.1 Adsorption Isotherms and Models

Adsorption Equilibria in SCFs play a critical role in influencing the efficiency of the extraction process. Understanding the adsorption isotherms and models in SCFs provide researchers with an ability to predict and control the solutes behavior in the system, this will lead to improved extraction products. By studying the interactions between solutes and SCFs at different conditions, researchers can accurately develop models that can be utilized in the extraction process optimization for specific compounds. This knowledge can ultimately lead to more sustainable and cost-effective extraction methods that have the potential to revolutionize various industries. Some of the key factors that researchers consider when studying adsorption in SCFs include temperature, pressure, and the nature of the solute and solvent. Researchers can manipulate these variables and precisely determine the optimum conditions for extracting a specific compound in a more efficient way. Additionally, comprehending the adsorption behavior is also crucial for researchers to develop enhanced equipment and processes for extraction, ultimately enhancing the efficiency and sustainability of the extraction process.

4.1.1 Adsorption Isotherms

There are three basic models governing the behaviour of these fluid materials, which briefly described as follows.

- i. **Langmuir Isotherm:** This model is ideal for describing adsorption on surfaces with a finite number of identical sites, assuming no interactions between adsorbates. It's useful for scenarios where a monolayer of adsorbate forms on a surface [31].
- ii. **Freundlich Isotherm:** This model applies to heterogeneous surfaces and assumes multilayer adsorption. It's more flexible and can describe adsorption over a range of conditions but may be less precise for systems with well-defined surface sites [32].
- iii. **Non-Ideal Isotherms:** Adsorption in SCFs often shows non-ideal behaviour due to the interplay between fluid density and adsorption dynamics. For example, the adsorption of volatile liquids in scCO₂ decreases with increasing temperature, which contrasts with typical exothermic adsorption behaviour [33]

Understanding these models would enable the development of novel and energy-efficient extraction techniques with environmental sustainability and waste minimization [32]. The use of SCFs for extraction is growing in the food and beverage, cosmetics, and pharmaceutical industries due to the growing emphasis on sustainability and environmental impact. More innovation and efficiency in these sectors can be unlocked by researchers by carrying out research on and refining the adsorption process in SCFs. Researchers can also enhance the quality and purity of the extracted molecules, making them more appealing for application in a variety of goods, by comprehending the adsorption process in SCFs. This may result in finished goods of higher caliber that are both more effective and ecologically sustainable. their intended applications. In essence, there is immense potential for enhancing sustainability and fostering innovation across various industries through continuous research and optimization of SFE processes. For instance, in the pharmaceutical sector, scientists can precisely extract molecules from natural sources for medicine development using SFE, which lessens the need for synthetic alternatives and has a smaller environmental impact. Moreover, volatile flavors and fragrances from substances like herbs and spices can be extracted via SFE in the food and beverage industry, producing more concentrated and genuine goods without the need for chemical solvents [31].

4.2 Influence of Pressure, Temperature, and Density on SFE

The efficiency of extraction process using supercritical fluid is highly dependent on the pressure, temperature, and density of the fluid employed in the process. These parameters can be manipulated to enhance the solubility and selectivity of the extraction, thereby allowing for the precise isolation of desired compounds. Pressure plays a crucial role in increasing the density of the supercritical fluid, enhancing its solvent power and improving the extraction efficiency. Temperature, on the other hand, affects the fluid's viscosity and diffusivity, influencing the rate at which the extraction takes place. Additionally, the density of the supercritical fluid impacts its ability to penetrate the matrix of the material being extracted, ultimately determining the yield and quality of the final product. By carefully controlling these factors, researchers can tailor the SFE process to target specific compounds and achieve high purity levels [34] The versatility of SFE makes it a valuable technique in various industries, including pharmaceuticals, food and beverage, and environmental analysis. With its ability to extract compounds without the need for harsh solvents or high temperatures, SFE is a sustainable and efficient method for obtaining pure and high-quality extracts. scCO₂ is characterized by high diffusivity and low viscosity, which enable the

dissolution of solutes and their adsorption onto substrates. These properties enhance mass transfer, making scCO₂ effective for processes that require mild conditions. The unique physicochemical characteristics of SCFs significantly impact the adsorption equilibria of various molecules. SCFs possess adjustable density, viscosity, and diffusivity, leading to enhanced interaction with adsorbents and influencing adsorption behavior.

4.3 Influence of Temperature and Pressure

- The adsorption equilibria of molecules like propyl acetate on activated carbon are notably affected by temperature and pressure in supercritical CO₂ conditions, with optimal adsorption occurring at specific ranges (313 K–353 K and 10 MPa–20 MPa) [35]
- Higher pressures generally increase the solubility of adsorbates in SCFs, enhancing adsorption capacity[36]

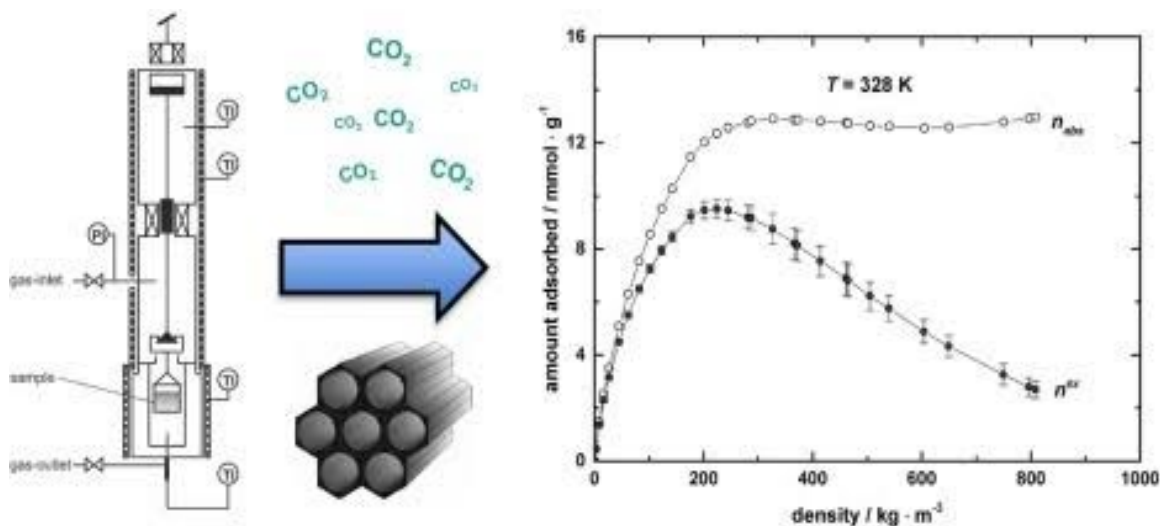


Figure 4: Effect of Density on SFA

Furthermore, the ability to adjust parameters such as pressure, temperature, and flow rate allows researchers to optimize the extraction process for different types of compounds. This flexibility is particularly advantageous in pharmaceutical applications, where the purity and potency of extracts are critical for ensuring the effectiveness and safety of medications. In the food and beverage industry, SFE is utilized to extract flavors, fragrances, and bioactive compounds from natural sources without altering their chemical composition. This gentle extraction method preserves the integrity of delicate compounds, resulting in higher quality products with enhanced sensory attributes. In environmental analysis, SFE is used to extract pollutants and contaminants from soil, water, and air samples, providing accurate and reliable data for monitoring and remediation efforts. Overall, the versatility and efficiency of SFE make it a valuable tool for researchers and industries seeking to obtain pure and high-quality extracts for a variety of applications.

4.4 Supercritical Fluid-Assisted Adsorption Processes

Supercritical Fluid-Assisted Adsorption Processes offer a promising avenue for enhancing the adsorption process. By utilizing SCFs as the solvent, researchers can overcome the limitations of traditional liquid-phase adsorption methods. The unique properties of SCFs, such as their high diffusivity and low surface tension, allow for faster and more efficient adsorption processes. This, in turn, can lead to higher adsorption capacities and shorter equilibrium times, ultimately improving the overall performance of the system. Additionally, supercritical fluid-assisted adsorption processes can be easily scaled up for industrial applications, making them a viable option for large-scale adsorption processes. Overall, the use of SCFs in adsorption processes holds great potential for advancing the field and addressing the challenges associated with traditional adsorption methods.

Key Aspects of Supercritical Fluid-Assisted Adsorption

- Technical Integration:** SFA combines supercritical fluid chromatography (SFC) and supercritical fluid impregnation (SFI), allowing for higher concentrations of target compounds and controlled release in applications like food packaging and drug delivery.
- Kinetics and Modeling:** Experimental studies on ibuprofen-silica aerogel systems reveal that pressure and temperature significantly influence the effective diffusion coefficient and adsorption capacity, which can be modeled using computational fluid dynamics (CFD).
- Competitive Adsorption:** Research indicates that sample solvents can affect the adsorption efficiency of solutes in SFC, necessitating careful consideration of solvent interactions during the process.

- iv. **Equipment Innovations:** New extraction equipment enhances the recovery of active substances from herbal medicines, improving extraction rates and reducing environmental impact.
- v. **Economic Viability:** Integrated processes, such as supercritical extraction and adsorption from passion fruit by-products, demonstrate promising economic outcomes, with cost analyses indicating competitive manufacturing costs.

While SFA shows significant potential, challenges remain in optimizing conditions and scaling processes for industrial applications. Further research is essential to address these issues and enhance the practicality of SFA in various sectors.

Effects of SCFs on Adsorption:

- a. **Non-Ideal Isotherms:** Adsorption in SCFs can deviate from ideal behaviour due to the interplay between fluid density, temperature, and adsorption dynamics. For example, the reduction in adsorption capacity with increasing temperature observed in scCO_2 can be attributed to changes in fluid density and interaction with the adsorbate.
- b. **Enhanced Mass Transfer:** The ability of SCFs to penetrate deeply into porous materials and dissolve a wide range of solutes enhances the overall efficiency of adsorption processes. This is especially beneficial for applications requiring fine control over process conditions.

One of the key advantages of using SCFs in adsorption processes is their ability to penetrate porous materials more effectively than conventional solvents. This enhanced penetration allows for a more thorough removal of contaminants from the surface of the adsorbent material, resulting in higher adsorption efficiencies. Furthermore, the unique properties of SCFs, such as their tunable density and solvating power, make them versatile and adaptable for a wide range of applications in adsorption processes. These properties allow for precise control over the adsorption process, leading to improved selectivity and efficiency in removing specific contaminants from a mixture. Additionally, the ability of SCFs to be easily recycled and reused makes them a more sustainable and environmentally friendly option compared to traditional solvents, making them a promising alternative for future advancements in adsorption technology.

4.5 SFE and Adsorption

One common application of SCFs in adsorption processes is in SFE (SFE) and adsorption. In SFE, SCFs are used to extract compounds from solid materials, such as plant materials or waste streams. The high solvating power of SCFs allows for efficient extraction of target compounds while leaving behind unwanted impurities. This process is often used in industries such as pharmaceuticals, food and beverage, and environmental remediation. Adsorption, on the other hand, involves the attachment of molecules onto a solid surface, often using SCFs as the medium. This method is highly effective in separating and purifying mixtures, as the high surface area and porosity of the adsorbent material allow for maximum contact with the supercritical fluid, leading to rapid and efficient adsorption of target compounds. SFE and adsorption are highly effective techniques for separation and purification, offering innovative solutions for industries such as pharmaceuticals, food and beverage, and environmental remediation. These methods selectively extract target compounds and ensure high levels of purity and efficiency. In the pharmaceutical industry, SFE is used to isolate active ingredients from natural sources, and the extracted compounds can be further purified through adsorption onto solid materials to ensure pharmaceutical product purity.

4.6 Supercritical Fluid Desorption and Regeneration

Desorption of the absorbed material and regeneration of the Supercritical Fluid desorption is critical step in the adsorption process. The step involves the removal of the absorbed materials and regeneration of the SCF adsorbent material for reuse in the subsequent application [37]. This step is essential in maintaining the efficiency and sustainability of the separation and purification process. By utilizing SCFs, desorption can be achieved at lower temperatures and pressures compared to traditional methods, reducing energy consumption and minimizing environmental impact. Additionally, the regenerated adsorbent can be used multiple times, making the overall process more cost-effective. Overall, desorption and regeneration using SCFs offer a more environmentally friendly and cost-effective solution for separation and purification processes. The ability to reuse the adsorbent multiple times not only saves on resources but also reduces waste. Furthermore, the lower temperatures and pressures required for desorption make the process more energy-efficient, further contributing to its sustainability. In conclusion, utilizing SCFs for desorption and regeneration is a win-win solution for both the environment and the economy [38]. By implementing this innovative technology, industries can significantly reduce their environmental impact while also saving on operational costs. The use of SCFs not only improves the efficiency of separation processes but also enhances the overall sustainability of industrial operations. This approach aligns with the growing trend towards more sustainable practices in the global marketplace. As more companies recognize the benefits of using SCFs for desorption and regeneration, we can expect to see a shift towards greener

and more cost-effective solutions in various industries. Ultimately, embracing this technology is a step towards a more sustainable future for our planet [39].

4.7 Supercritical Fluid Impregnation and Adsorption

Using SCFs for impregnating materials with desired compounds or adsorbing contaminants from surfaces is a promising application of this technology in industrial processes. Compared to traditional methods, companies can achieve more efficient and effective results by utilizing SCFs. This approach not only reduces the use of harmful chemicals and solvents but also enhances the overall quality and performance of the treated materials. The ability of SCFs to penetrate deep into porous structures allows for a more thorough and uniform distribution of additives or removal of impurities. Consequently, this results in products with improved properties and reduced environmental impact, making it a valuable tool for sustainable manufacturing practices. Furthermore, the use of SCFs in manufacturing processes has been shown to increase productivity and reduce production costs. By utilizing this advanced technology, companies can streamline their operations and achieve higher levels of precision and control over the final product. This level of efficiency is crucial in today's competitive market, where businesses are constantly seeking ways to stay ahead of the competition.

5.0 Mass Transfer in SFA

Efficiency of the extraction process in SFA is determined by mass transfer. The exploration and understanding the mechanisms of mass transfer in SFA is fundamental for the optimization of extraction conditions and yields maximization [40]. By studying the diffusion of solutes in SCFs, researchers can tailor extraction parameters to improve extraction efficiency and selectivity. Additionally, the interaction between the solute and the adsorbent surface plays a significant role in the adsorption process, influencing the rate and extent of extraction. By investigating the mass transfer phenomena in SFA, researchers can further enhance the capabilities of this powerful extraction technique for a wide range of applications. The mass transfer in SFA is governed by the unique properties of SCFs, including their high diffusivity, low viscosity, and tunable solubility. These characteristics enable efficient transport of solutes, making SCFs valuable in a range of applications from extraction to catalysis [41]. Understanding the interplay between thermodynamic conditions, adsorbent properties, and external influences is crucial for optimizing mass transfer processes in supercritical fluid systems.

5.1 Diffusion and Transport Mechanisms

Mass transfer in SFA is a critical area of study due to the unique properties of SCFs, which combine characteristics of both gases and liquids. This dual nature allows SCFs to exhibit high diffusivity and low viscosity, facilitating efficient mass transfer processes in various applications, including extraction, adsorption, and catalysis. SCFs, particularly scCO_2 , have been shown to possess excellent mass transfer capabilities due to their low surface tension and high diffusivity. This allows them to penetrate porous materials effectively, enhancing the extraction efficiency of solutes from solid matrices [42]. For instance, the ability of scCO_2 to diffuse into small pores is crucial for applications in the nutraceutical industry, where it can selectively extract bioactive compounds from plant materials [42]. The high diffusivity of SCFs also contributes to rapid mass transfer rates, which is essential for processes such as SFE (SFE) [43]. The mass transfer dynamics in SFA are influenced by several factors, including temperature, pressure, and the nature of the adsorbent material. For example, the diffusion coefficients of solutes in SCFs can vary significantly with changes in density and temperature, particularly near the critical point [44]. This sensitivity allows for tunable properties, making SCFs highly adaptable for specific applications. The solubility of SCFs is also affected by these parameters, which can be manipulated to enhance the extraction selectivity of various components [42]. Research has demonstrated that the mass transfer of solutes in SCFs can be significantly enhanced by external factors such as acoustic waves. These waves can promote the mixing of SCFs with the solute, thereby increasing the mass transfer rates during extraction processes [43]. Additionally, the structural characteristics of the adsorbent, such as pore size and surface interactions, play a crucial role in determining the efficiency of mass transfer. For instance, studies on mesoporous silica have shown that pore size can influence the sorption behavior of supercritical CO_2 , affecting the overall adsorption kinetics [45].

Moreover, the concept of the Widom line, which marks the transition between liquid-like and gas-like behavior in SCFs, is essential for understanding mass transfer phenomena. The changes in molecular interactions and diffusivity across this line can significantly impact the efficiency of adsorption processes [46]. As SCFs transition from a gas-like to a liquid-like state, their mass transfer properties also change, which can be exploited to optimize various industrial applications [46]. Understanding the complex diffusion and transport mechanisms in SFA is essential for optimizing the extraction process. By manipulating factors such as temperature, pressure, and solute and solvent properties, researchers can control the rate of solute movement and maximize extraction efficiency [47]. Additionally, exploring the use of co-solvents can enhance solubility and improve overall extraction efficiency, allowing for greater selectivity and expanded applications in industries such as pharmaceuticals and food

production. With further research and development, SFE combined with co-solvent utilization has the potential to revolutionize extraction processes across various sectors.

5.2 Intraparticle and Film Mass Transfer

Intraparticle and film mass transfer play a crucial role in the efficiency and effectiveness of SFE. Understanding the mechanisms involved in mass transfer within the solid matrix and at the interface between the solid and supercritical fluid is essential for optimizing extraction processes. By studying these processes, researchers can identify ways to enhance the extraction efficiency and increase the yield of desired compounds [48]. Additionally, improving our understanding of intraparticle and film mass transfer can lead to the development of new extraction techniques and technologies that further enhance the capabilities of SFE.

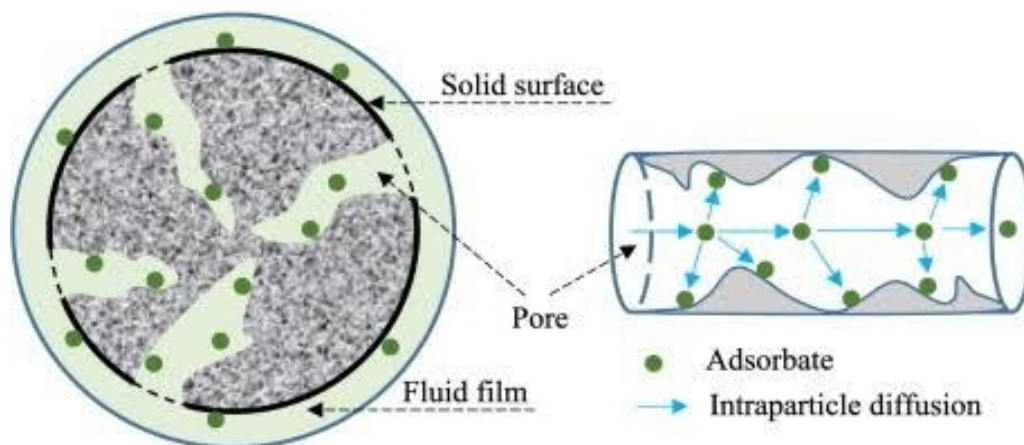


Figure 5: Intraparticle Mass Transfer

For instance, to boost extraction yields and improve mass transfer rates [49], researchers have been investigating the application of innovative materials with improved surface qualities. By offering a greater surface area for interaction with the supercritical fluid, these materials can facilitate the extraction of target molecules more effectively. In addition, developments in modeling and simulation approaches have enabled researchers to better forecast and optimize extraction conditions, resulting in more sustainable and cost-effective extraction procedures. SFE has a bright future if mass transfer mechanisms and extraction technologies are researched further. For instance, to enhance the surface area that may be extracted, scientists have created nanoparticles coated with a substance that is compatible with supercritical fluid. Higher extraction yields can be achieved by combining these nanoparticles with SFE. of bioactive compounds from plant materials have been achieved. This innovative approach not only improves efficiency but also reduces the amount of solvent and energy required for the extraction process, making it more environmentally friendly. In general, the rate of adsorption involves both rates of liquid film mass transfer and intraparticle diffusion. Researchers have proposed techniques to determine both intraparticle diffusivity ($D(S)$) and liquid mass transfer coefficient ($k(F)$) simultaneously from a single-component adsorption uptake curve. The Biot number (Bi) is used to represent the ratio of transport rates across liquid film and intraparticle mass transfer. This method can be applied in systems where both liquid film resistance and intraparticle diffusion are significant [50]. One key aspect of optimizing adsorption systems is understanding the interactions between the adsorbate and adsorbent materials. This involves studying factors such as surface area, pore size distribution, and chemical composition of the adsorbent. By fine-tuning these parameters, researchers can enhance the adsorption capacity and efficiency of the system. Additionally, the kinetics of adsorption play a crucial role in determining the overall performance of the system. By studying the rate at which adsorption occurs and the mechanisms involved, scientists can identify potential bottlenecks and improve the overall efficiency of the process.

5.3 Kinetic Modelling and Rate Limiting Steps

Mathematical models are used in kinetic modeling to describe the process of adsorption [51] and identification of the rate-limiting step [52]. Study of the adsorption kinetics provides a better understanding of the mechanisms involved and ability for design adsorption-systems with optimize efficiency [53]. Different models, such as pseudo-first order, pseudo-second order, and intraparticle diffusion models, can be used to analyze the experimental data and determine the rate constants and mechanisms of adsorption [54]. These models can provide valuable insights into the factors that influence the adsorption process and help in the development of more efficient adsorption systems. Overall, the study of adsorption kinetics plays a crucial role in improving the efficiency and effectiveness of adsorption systems. By utilizing different models and analyzing experimental data, scientists can pinpoint the

rate-limiting steps and optimize the design of these systems. This knowledge can lead to the development of more sustainable and cost-effective adsorption processes that can be utilized in various industries, such as wastewater treatment, air purification, and gas separation. Ultimately, a better understanding of adsorption kinetics can help address environmental challenges and contribute to the advancement of green technologies .

5.4 Mass Transfer Model in Supercritical Adsorption

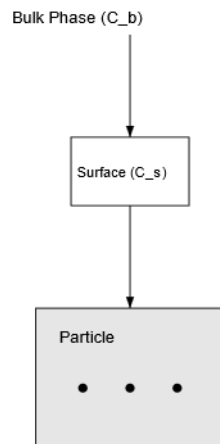


Figure 6. Conceptual mass transfer model for supercritical adsorption showing external film resistance and intraparticle diffusion coupled with adsorption equilibrium [24]

5.4.1 Governing Model

The adsorption process in SCFs is typically described using a **film–pore diffusion model** coupled with adsorption equilibrium.

(a) External Mass Transfer

$$N = k_f (C_b - C_s) \tag{1}$$

Where:

- N: mass flux
- k_f : film mass transfer coefficient
- C_b : bulk concentration
- C_s : surface concentration

(b) Interparticle Diffusion

$$\frac{\partial q}{\partial t} = D_e \nabla^2 q \tag{2}$$

Where:

- q: adsorbed phase concentration
- D_e : effective diffusivity

(c) Adsorption Equilibrium (Langmuir-type)

$$q = \frac{q_{max} bC}{1 + bC} \tag{3}$$

6.0 Adsorbent Materials

Adsorbent Materials for SFA are being developed to further improve the efficiency and effectiveness of this process. These materials have shown promising results in initial testing. Thus, they have the potential to revolutionize the way SCFs are utilized in industrial applications. The potential benefits include increased selectivity, faster reaction rates, and improved product purity. As research continues and more advancements are made, it is likely that Adsorbent Materials for SFA will become even more refined and effective. This could lead to even greater benefits for manufacturers, such as reduced production costs and increased overall productivity. Ultimately, the widespread adoption of these materials could have a significant impact on various industries, leading to more sustainable and efficient manufacturing processes.

6.1 Porous Adsorbents

Porosity is one of the key requirements for an adsorbent material, the use of porous adsorbent materials for SFA such as metal-organic framework (MOFs), activated carbon and zeolites [55]. These materials possessed high surface areas and specific pore structures that made them perform excellently for application in capturing and separating target molecules in supercritical fluid environments [56]. Activated carbon, for example, is known for its high adsorption capacity and ability to remove impurities from gases and liquids. Zeolites, on the other hand, have well-defined pore structures that can selectively adsorb certain molecules based on their size and shape. MOFs are a newer class of porous materials that offer tunable pore sizes and functionalities, making them highly versatile for a wide range of adsorption applications. These materials are extensively utilized in industries such as gas separation, water purification, and drug delivery. Their ability to tailor their properties for specific purposes has sparked growing interest in their application across various fields. Researchers are continuously exploring new methods to enhance the adsorption capabilities of these porous materials, including modifying their surfaces with diverse chemical groups or integrating metal ions into their structures. Overall, the unique properties of these materials make them valuable tools for capturing and isolating target molecules in diverse settings. These advancements in porous materials have opened up new opportunities for improving efficiency and sustainability in industries reliant on separation and purification processes.

By precisely adjusting the properties of these materials, researchers can achieve higher levels of selectivity and efficiency in capturing specific molecules. As the demand for cleaner and more effective separation processes continues to grow, the development of advanced porous materials will play a crucial role in shaping the future of various industries. With the ability to customize pore size, surface chemistry, and other characteristics of porous materials, scientists can create materials that specifically target pollutants in water treatment plants, separate various gases in industrial processes, and purify pharmaceuticals in the healthcare sector [57]. These advancements not only enhance the overall effectiveness of separation processes, but also reduce energy consumption and waste production, leading to more sustainable practices across industries. As research in this field continues to advance, we can expect the emergence of even more innovative solutions, revolutionizing our approach to separation and purification challenges in the future.

6.2 Nanostructured Adsorbents

Nanostructured adsorbents, including nanoparticles and nanocomposites, have emerged as promising materials for environmental remediation [58], [59], [60], [61], particularly in wastewater treatment and pollutant removal [62], [63]. These materials offer enhanced adsorption capacity and efficiency compared to traditional adsorbents like activated carbon [64]. Surface modification of nanostructured adsorbents can improve their affinity towards specific pollutants, addressing key challenges in nanomaterial utilization for water treatment [65]. Chitosan-based nanocomposite membranes have shown effectiveness in removing dyes and heavy metals from aqueous environments, with various nanoparticles used to enhance their properties [66]. Nanoparticle adsorbents, such as nano titanium oxide, have demonstrated potential for arsenic removal from drinking water [67]. Despite their advantages, challenges remain, including high costs, stability issues, and potential nanotoxicity [64]. Addressing these limitations is crucial for the commercial viability and large-scale application of nanostructured adsorbents in environmental remediation. Nanostructured adsorbents, including nanoparticles and nanocomposites, have emerged as effective solutions for wastewater treatment, particularly in removing heavy metals and other pollutants. Their unique properties, such as high surface area and tunable functionalities, enhance adsorption efficiency, making them superior to traditional materials.

i. Efficiency in Pollutant Removal

- Nanostructured adsorbents exhibit significantly higher adsorption rates due to their large surface areas, which facilitate interactions with contaminants.
- Surface engineering of these materials can enhance their affinity for specific pollutants, addressing challenges in wastewater treatment.

ii. Types of Nanocomposites

- Various nanocomposites, such as those incorporating manganese dioxide, have shown excellent performance in removing specific ions like strontium.
- Magnetic nanocomposites are particularly promising for environmental applications, allowing for easy recovery and reuse after adsorption processes.

These materials offer a high surface area-to-volume ratio, allowing for increased adsorption capacity and efficiency in removing contaminants from various fluids and gases [68]. Nanostructured adsorbents have shown great potential in applications ranging from water treatment to gas separation, offering a more sustainable and cost-effective alternative to traditional separation methods [69]. Researchers are actively exploring the use of these materials in different industries, seeking to harness their unique properties for improved separation and purification processes. While nanostructured adsorbents present significant advantages in pollutant removal, their

commercial viability is hindered by economic and environmental concerns that require further investigation and innovation.

6.3 Surface Modification and Functionalization

Nanostructured adsorbent materials can be flexibly modified and functionalized to enhance their efficiency in various applications [70]. The modification and functionalization increase the selectivity and adsorption capacities efficiencies of the materials in the employed application such wastewater treatment. This process involves tailoring the surface properties of nanomaterials to improve their interaction with specific pollutants, thereby increasing their adsorption capacity and selectivity [71]. Surface modification and functionalization of nanostructured adsorbents have gained significant attention for enhancing their performance in wastewater treatment and pollutant removal. Modified nanoparticles, including silica, iron, and alumina-based materials, demonstrate superior adsorption capacities compared to their unmodified counterparts, particularly for heavy metal ions [72]. Surface engineering techniques improve the affinity of nanostructured adsorbents towards targeted pollutants, addressing a key challenge in nanomaterial utilization [65]. Various modification methods, such as hydrothermal synthesis, have shown a close relationship with adsorption capacities [72]. While surface modification may decrease specific surface area, it generally increases adsorption capacity with the degree of functionalization [73]. However, challenges like high cost, low stability, and potential nanotoxicity need to be addressed for large-scale applications. Proper surface functionalization is crucial for determining nanoparticle interactions with the environment and their colloidal stability [64].

iii. Surface Modification Techniques

- **Chemical Functionalization:** Techniques such as grafting glycidyl methacrylate (GMA) onto polypropylene surfaces have been shown to enhance the affinity for sticky particles in papermaking whitewater, achieving up to 58.3% removal efficiency.
- **Hybrid Nanostructures:** The combination of graphene with inorganic nanoparticles like Fe_3O_4 and TiO_2 has been effective in removing pharmaceutical pollutants, demonstrating high adsorption capacity and photocatalytic properties.

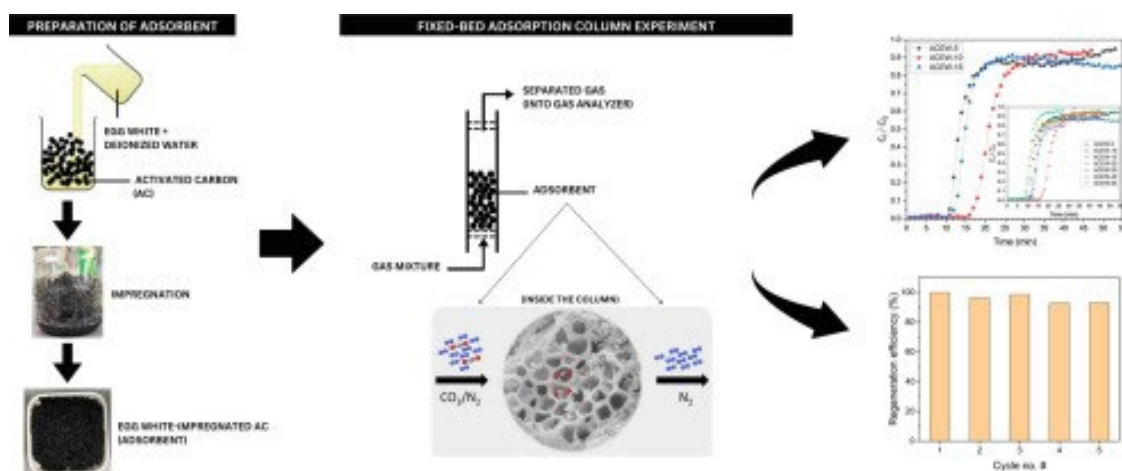


Figure 7: Surface Modification and Functionalization.

While surface modification significantly enhances the performance of nanostructured adsorbents, challenges such as cost, stability, and potential toxicity remain critical considerations for their practical application. Moreover, integrating SCFs into developing nanostructured adsorbents offers a promising avenue for enhancing pollutant removal efficiency. Utilizing SCFs during the synthesis process can lead to the formation of highly porous materials with tailored surface characteristics that improve adsorption capacity and kinetics significantly. For instance, recent studies have demonstrated that employing carbon dioxide as a supercritical solvent in preparing activated carbon composites not only optimizes pore structure but also enhances the interaction between the adsorbent and contaminants through increased local density effects. Additionally, this method has been shown to facilitate the uniform distribution of functional groups on the adsorbent's surface, further improving its selectivity towards specific pollutants such as heavy metals or pharmaceuticals. As research progresses, these innovative approaches may bridge the gap between theoretical advancements in thermodynamics and practical applications in environmental remediation, ultimately leading to more sustainable solutions for wastewater treatment challenges. Table 2 provide Summary of adsorbents used in supercritical fluid adsorption systems, highlighting material properties and performance characteristics.

Table 2: Adsorbents Used in SCF Systems [24]

Adsorbent	Type	Key Properties	SCF Applications	Advantages	Limitations
Activated Carbon	Carbonaceous	High surface area, microporous	CO ₂ adsorption, organic removal	Low cost, high capacity	Low selectivity
Silica Gel	Inorganic	Polar surface, mesoporous	Polar solute adsorption	affinity for polar compounds	Moisture sensitivity
Zeolites	Aluminosilicates	Uniform pores, high selectivity	Gas separation	selectivity	Diffusion constraints
MOFs	Hybrid porous	Ultra-surface area, tuneable	Advanced CO ₂ capture	Exceptional capacity	Stability issues
Alumina	Metal oxide	Thermally stable	Catalyst support	Robust	Moderate adsorption
Polymer Resins	Organic	Functionalizable	Pharmaceutical SCF extraction	Selective	Swelling
CNTs	Nanomaterial	High interaction energy	Advanced adsorption	High efficiency	Cost

7.0 Industrial Applications

SFA is a versatile technique with numerous applications across various fields, particularly in natural extracts, environmental remediation, and pharmaceuticals. This method leverages the unique properties of SCFs to enhance adsorption processes, leading to improved efficiency and effectiveness. SFA is an emerging technique combining adsorption with supercritical fluid technology, with applications in chromatography (SFC) and impregnation (SFI) [74]. SFA has been applied to various fields, including natural product extraction, pharmaceuticals, and polymer processing [1]. The technology has evolved from primarily extraction and fractionation processes to diverse industrial applications [75]. These include mass-transfer processes, phase transitions, reactive systems, and materials processing [76]. Some applications, such as extraction, have reached industrial scale, while others are still under development. SFA's versatility extends to impregnation, cleaning, particle formation, and coating [76]. The integration of SFA with other high-pressure processes shows promise for biorefinery applications [2]. As research continues, a deeper understanding of supercritical fluid properties and their interactions with processed materials is crucial for further industrial exploitation [77]. Moreover, the integration of supercritical fluid technology with advanced computational modeling can significantly enhance our understanding of adsorption mechanisms and optimize process conditions. By employing multicomponent potential theory (MPTA) alongside molecular dynamics simulations, researchers can predict complex interactions between SCFs and various adsorbents under high-pressure scenarios. This approach not only aids in elucidating the thermodynamic parameters governing adsorption equilibria but also allows for the customization of adsorbent materials tailored to specific applications, such as targeted pollutant removal or selective extraction of bioactive compounds. As a result, this synergy between theoretical frameworks and experimental techniques may pave the way for innovative methodologies that maximize efficiency while minimizing environmental impact, ultimately leading to more sustainable industrial practices.

7.1 Applications in Drug Delivery

7.1.1 Encapsulation of Pharmaceuticals:

Silica Aerogels: scCO₂ is used to adsorb pharmaceuticals onto carriers such as silica aerogels, improving the solubility and stability of poorly soluble drugs. This method minimizes damage to carrier materials and allows for controlled drug release.

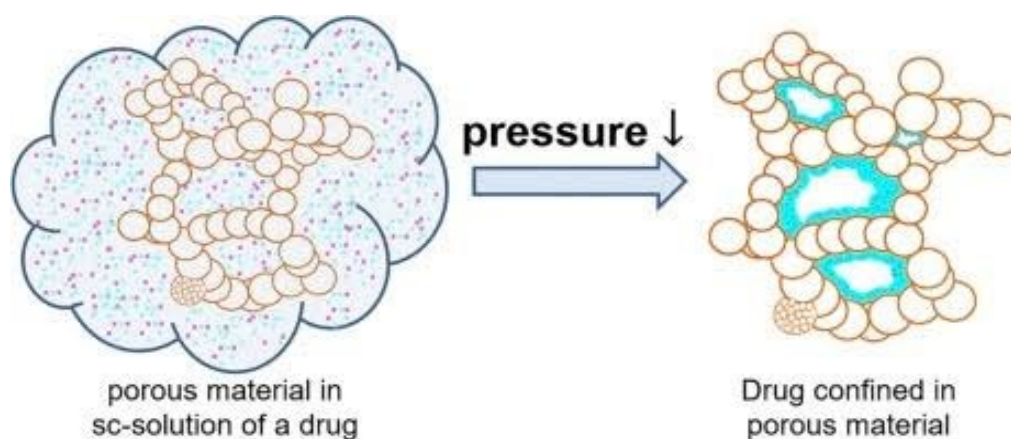


Figure 8: Encapsulation of Pharmaceuticals

7.1.2 Control over Drug Release

The ability to adjust pressure and temperature during scCO₂ processing enables precise control over drug release profiles, enhancing the efficacy of drug delivery systems.

7.1.3 Advantages of SCF-Based Methods over conventional methods

SCF processes offer several advantages over traditional methods, including reduced processing temperatures, minimal use of organic solvents, and the ability to achieve high precision in drug encapsulation and release.

7.2 Separation and Purification

The unique properties of SCFs has made them indispensable in separation and purification processes, combining liquid-like density with gas-like viscosity and diffusivity [9]. SCFs, particularly CO₂, are widely used in extraction, fractionation, and chromatography for various industries, including pharmaceutical, food, and chemical sectors [76]. These applications leverage SCFs' ability to selectively extract and separate compounds by adjusting pressure and temperature [77]. In the pharmaceutical industry, SCFs are employed for extraction from natural materials, purification of active substances, and innovative drug formulations to enhance bioavailability and controlled release [77]. Adsorptive separations using SCFs show promise for multicomponent systems, offering potential advantages in processing pharmaceuticals and food products [78]. Despite the numerous laboratory-scale studies, further research is needed to address scale-up challenges and improve commercial viability of SCF technologies [75], [76]. SFA has emerged as a versatile technique for separation and purification across various industries, particularly in pharmaceuticals and environmental applications. The unique properties of SCFs, such as tuneable density and low viscosity, facilitate efficient sorption processes, making them ideal for diverse applications.

7.3 Environmental Remediation

The application of SFA in environmental remediation has presented new innovative method in environmental sustainability, particularly in toxic contaminants removal and resource recovery [3]. This technique leverages the unique properties of SCFs, such as supercritical carbon dioxide (scCO₂), to enhance adsorption processes while minimizing environmental impact.

i. Environmental Remediation

- **Contaminant Removal:** SCFs are effective in separating and removing pollutants from wastewater, including heavy metals, due to their adjustable physicochemical properties.
- **Biosorbents:** The integration of biosorbents with SFA enhances the efficiency of contaminant capture, promoting sustainable practices in waste management.

ii. Resource Recovery

- **Natural Extracts:** SFA is utilized to extract valuable compounds from natural sources, facilitating biorefinery processes that convert biomass into useful products.
- **Energy Efficiency:** The use of SCFs reduces the need for harsh chemicals and high temperatures, leading to lower energy consumption in industrial applications.

While SFA shows promise, challenges remain in optimizing these processes for broader industrial adoption, particularly in scaling up and ensuring economic viability. The integration of supercritical fluid (SCF) technology with advanced nanomaterials not only enhances pollutant removal efficiency but also opens avenues for the development of multifunctional adsorbents. For instance, recent studies have explored the use of hybrid nanostructures that combine SCFs with materials like metal-organic frameworks (MOFs), which exhibit exceptional surface areas and tunable pore sizes, significantly improving adsorption capabilities for a variety of

contaminants. Moreover, the application of multicomponent potential theory in these systems allows for better predictions of adsorption behavior under varying conditions, thereby optimizing design parameters for specific environmental challenges. As researchers continue to innovate in this space, the potential for creating tailored solutions that address both efficacy and sustainability in wastewater treatment becomes increasingly viable, paving the way for more effective remediation strategies in industrial applications

8.0 Challenges and Future Prospects

The use of SCFs in adsorption processes presents both significant challenges and promising future prospects. SCFs, with their unique properties, offer innovative solutions for various applications, yet they also face technical and practical hurdles that need addressing.

- ✓ **Process Optimization:** Achieving optimal conditions for SFA requires precise control over parameters like pressure and temperature, which can complicate industrial applications.
- ✓ **Material Compatibility:** The interaction between SCFs and adsorbent materials can lead to degradation or ineffective adsorption, necessitating the development of more resilient materials.
- ✓ **Integration with Other Technologies:** Combining SFA with techniques like supercritical fluid chromatography (SFC) and supercritical fluid impregnation (SFI) can enhance the efficiency of extracting and utilizing bioactive compounds.
- ✓ **Sustainability:** SCFs are considered green solvents, making them attractive for environmentally friendly extraction processes, particularly in the recovery of valuable compounds from algae.

The process design and optimization entail identifying the best solution from numerous possibilities, taking into account objective functions, constraints, and design variables. Constraints serve as boundaries that solutions must adhere to, while design variables encompass the available approaches for developing solutions. Given the complexity of real-world engineering tasks, multidisciplinary design optimization (MDO) and robust computational tools are often necessary. Process design plays an increasingly crucial role in optimizing processes amid economic constraints, although it encounters unresolved challenges. Recent progress includes feasibility/flexibility-based optimization, which effectively integrates multiple constraints and avoids unnecessary exploration of infeasible spaces in black-box optimization. This approach finds applications in pharmaceutical design and process scheduling. The link between flexibility analysis and robust optimization offers promising avenues for further exploration [78] Overall, process design and optimization are continuously evolving, addressing limitations and constraints through diverse methodologies and computational approaches. Process design and optimization face several limitations and constraints that can hinder efficiency and effectiveness. Key challenges include the integration of sustainability, complexity in mathematical modeling, and operational feasibility.

Thus, future studies should concentrate on creating robust, application-specific adsorbents as well as predictive, multi-scale models that combine thermodynamics and transport phenomena. To move SCF-based adsorption from laboratory-scale innovation to industrial reality, further focus on process integration, techno-economic analysis, and lifetime sustainability will be necessary. SCFs can significantly advance effective, selective, and environmentally friendly separation technologies by tackling these issues through interdisciplinary cooperation.

9.0 Conclusion

SFA has gained attention for its applications in clean energy technology, gas purification processes, and separation science. This offered a compelling alternative to conventional techniques constrained by high energy demands, solvent toxicity, and limited selectivity. The unique and tuneable physicochemical properties of SCFs which bridge the liquid-like solvation with gas-like transport behaviour enable enhanced control over adsorption equilibria, mass transfer, and process efficiency. Research has focused on understanding the adsorption mechanism, which involves monomolecular coverage and two-layer adsorption over a wide range of densities. This understanding has led to the development of improved materials for gaseous fuel storage and separation technologies. SCFs can alter the gas-solid interface, facilitating the removal and migration of adsorbates, which is useful in adsorbent regeneration and chromatographic separation. Engineering models have been developed to describe physical adsorption from sub- to supercritical conditions on flat surfaces and clustering in SCFs. These models can provide quantitative fits over wide pressure and temperature ranges, making them valuable tools for predicting adsorption behaviour in various applications, including gas purification, supercritical extractions, and critical point drying.

References

- [1] E. Sadatshojaei and D. A. Wood, "Applications of supercritical fluids in environmental remediation," in *Green Sustainable Process for Chemical and Environmental Engineering and Science*, Elsevier, 2021, pp. 257–265. doi: 10.1016/B978-0-12-821884-6.00003-6.

- [2] R. Camarillo, S. Tostón, F. Martínez, C. Jiménez, and J. Rincón, "Preparation of TiO_2 -based catalysts with supercritical fluid technology: characterization and photocatalytic activity in CO_2 reduction," *Journal of Chemical Technology & Biotechnology*, vol. 92, no. 7, pp. 1710–1720, Jul. 2017, doi: 10.1002/jctb.5169.
- [3] M. Namakka, Md. R. Rahman, N. bt M. Sa'don, M. K. Bin Bakri, and A. Z. Sueraya, "Chemisorption and physisorption of polymer nanocomposite membranes," *Polymer Nanocomposite Membranes in Water Treatment and Desalination*, pp. 423–446, Jan. 2025, doi: 10.1016/B978-0-443-23879-6.00011-7.
- [4] M. Namakka, Md. R. Rahman, K. A. M. Bin Said, M. Abdul Mannan, and A. M. Patwary, "A review of nanoparticle synthesis methods, classifications, applications, and characterization," *Environ. Nanotechnol. Monit. Manag.*, vol. 20, no. November, p. 100900, 2023, doi: 10.1016/j.enmm.2023.100900.
- [5] A. Z. Sueraya, M. R. Rahman, A. A. James, M. Namakka, and M. K. Bin Bakri, "Biocomposites utilization in drug delivery applications," *Smart Biocomposite Materials: Fabrication, Applications and Sustainability*, pp. 199–226, Jan. 2026, doi: 10.1016/B978-0-443-33643-0.00014-4.
- [6] L. Chai and S. A. Tassou, "Recent Progress on High Temperature and High Pressure Heat Exchangers for Supercritical CO_2 Power Generation and Conversion Systems," *Heat Transfer Engineering*, vol. 44, no. 21–22, pp. 1950–1968, 2023, doi: 10.1080/01457632.2022.2164683.
- [7] Q. Wang, J. Xu, C. Zhang, B. Hao, and L. Cheng, "A Critical Review on Heat Transfer of Supercritical Fluids," *Heat Transfer Engineering*, vol. 44, no. 21–22, pp. 1969–1994, Dec. 2023, doi: 10.1080/01457632.2022.2164684.
- [8] D. E. Knox, "Solubilities in supercritical fluids," *Pure and Applied Chemistry*, vol. 77, no. 3, pp. 513–530, Jan. 2005, doi: 10.1351/pac200577030513.
- [9] J. I. Cacho, N. Campillo, P. Viñas, and M. Hernández-Córdoba, "In situ ionic liquid dispersive liquid-liquid microextraction coupled to gas chromatography-mass spectrometry for the determination of organophosphorus pesticides," *J. Chromatogr. A*, vol. 1559, pp. 95–101, Jul. 2018, doi: 10.1016/j.chroma.2017.12.059.
- [10] H. S. Samuel, U. Nweke-Maraizu, and E. E. Etim, "Supercritical Fluids: Properties, Formation and Applications," *Journal of Engineering in Industrial Research*, vol. 4, no. 3, pp. 176–188, Sep. 2023, doi: 10.48309/JEIRE.2023.3.5.
- [11] W. Xie, S. Chen, M. Wang, Z. Yu, and H. Wang, "Progress and Prospects of Supercritical CO_2 Application in the Exploitation of Shale Gas Reservoirs," *Energy & Fuels*, vol. 35, no. 22, pp. 18370–18384, Nov. 2021, doi: 10.1021/acs.energyfuels.1c02944.
- [12] L. Santos-Zea, J. A. Gutiérrez-Urbe, and J. Benedito, "Effect of ultrasound intensification on the supercritical fluid extraction of phytochemicals from Agave salmiana bagasse," *J. Supercrit. Fluids*, vol. 144, pp. 98–107, Feb. 2019, doi: 10.1016/j.supflu.2018.10.013.
- [13] C. Steffens, E. Franceschi, F. C. Corazza, P. S. P. Herrmann, and J. V. Oliveira, "Gas sensors development using supercritical fluid technology to detect the ripeness of bananas," *J. Food Eng.*, vol. 101, no. 4, pp. 365–369, Dec. 2010, doi: 10.1016/j.jfoodeng.2010.07.021.
- [14] C. West, "Current trends in supercritical fluid chromatography," *Anal. Bioanal. Chem.*, vol. 410, no. 25, pp. 6441–6457, Oct. 2018, doi: 10.1007/s00216-018-1267-4.
- [15] R. K. Kankala, Y. S. Zhang, S. Wang, C. Lee, and A. Chen, "Supercritical Fluid Technology: An Emphasis on Drug Delivery and Related Biomedical Applications," *Adv. Healthc. Mater.*, vol. 6, no. 16, Aug. 2017, doi: 10.1002/adhm.201700433.
- [16] M. Namakka, M. R. Rahman, M. K. B. Bakri, et al., "Advancements in acoustic properties of natural waste biocomposites: current trends, applications, and future perspectives," *Adv. Compos. Hybrid Mater.*, vol. 8, art. no. 386, 2025, doi: 10.1007/s42114-025-01427-6.
- [17] M. Namakka, M. R. Rahman, K. A. Bin Mohamad Said, and A. Muhammad, "Insights into micro- and nano-valent iron materials: synthesis methods and multifaceted applications," *RSC Adv.*, vol. 14, no. 41, pp. 30411–30439, 2024, doi: 10.1039/d4ra03507k.
- [18] K. Fatema, A. A. Sharmin, J. F. Sharna, M. A. Haque, M. Rahman, S. Sarker, M. Kazi, M. R. Rahman, M. Namakka, M. Uzzaman, and M. A. M. Patwary, "Antioxidant and antidiabetic effects of *Flemingia macrophylla* leaf extract and fractions: In vitro molecular docking, dynamic simulation, pharmacokinetics, and biological activity studies," *BioResources*, vol. 19, no. 3, pp. 4960–4983, 2024, doi: 10.15376/biores.19.3.4960-4983.
- [19] M. R. Rahman, A. James, K. A. Mohamed Said, M. Namakka, M. U. Khandaker, W. H. Jiunn, J. Y. Al-Humaidi, R. H. Althomali, and M. M. Rahman, "A TiO_2 grafted bamboo derivative nanocellulose polyvinylidene fluoride (PVDF) nanocomposite membrane for wastewater treatment by a photocatalytic process," *Mater. Adv.*, vol. 5, no. 19, pp. 7617–7636, 2024, doi: 10.1039/D4MA00716F.
- [20] A. James, M. Rezaur Rahman, K. Anwar Mohamed Said, M. Namakka, K. Kuok Kuok, M. Uddin Khandaker, J. Y. Al-Humaidi, R. H. Althomali, and M. M. Rahman, "Lithium chloride-mediated

- enhancement of dye removal capacity in Borneo bamboo derived nanocellulose-based nanocomposite membranes (NCMs)," *J. Mol. Liq.*, vol. 413, art. no. 125973, Apr. 2024, doi: 10.1016/j.molliq.2024.125973..
- [21] M. Namakka, M. R. Rahman, K. A. Mohamad Bin Said, K. K. Kuok, F. A. Md Yusof, M. S. M. Al-Saleem, J. Y. Al-Humaidi, and M. M. Rahman, "Unveiling the synergistic effect of an nZVI-SiO₂-TiO₂ nanocomposite for the remediation of dye contaminated wastewater," *Mater. Adv.*, vol. 5, pp. 9292–9313, Oct. 2024, doi: 10.1039/D4MA00853G.
- [22] A. Lebedev, D. Lovskaya, and N. Menshutina, "Experimental Investigation and CFD Modeling of Supercritical Adsorption Process," *Polymers (Basel)*, vol. 12, no. 9, p. 1957, Aug. 2020, doi: 10.3390/polym12091957.
- [23] S. Lucas, M. P. Calvo, C. Palencia, and M. J. Cocero, "Mathematical model of supercritical CO₂ adsorption on activated carbon: Effect of operating conditions and adsorption scale-up," *J. Supercrit. Fluids*, vol. 32, no. 1–3, pp. 193–201, Dec. 2004, doi: 10.1016/J.SUPFLU.2004.02.008.
- [24] M. R. Rahman, A. Z. Sueraya, K. A. B. M. Said, M. Namakka, A. James, I. M. M. Rahman, M. S. M. Al-Saleem, J. Y. Al-Humaidi, and M. M. Rahman, "Impact of graphene/nanocellulose on nanocomposite membrane for methylene blue dye removal and antifouling performance," *J. Appl. Polym. Sci.*, pp. 1–21, 2025, doi: 10.1002/app.57648.
- [25] A. Z. Sueraya, M. R. Rahman, K. A. B. M. Said, M. Namakka, D. Kanakaraju, J. Y. Al-Humaidi, S. M. Al-Baqami, M. M. Rahman, and M. U. Khandaker, "Impact of titanium dioxide/graphene in polyvinylidene fluoride nanocomposite membrane to intensify methylene blue dye removal, antifouling performance, and reusability," *J. Appl. Polym. Sci.*, pp. 1–14, Apr. 2024, doi: 10.1002/app.56257
- [26] G. Caputo, "Fixed Bed Adsorption of Drugs on Silica Aerogel from Supercritical Carbon Dioxide Solutions," *International Journal of Chemical Engineering*, vol. 2013, pp. 1–7, 2013, doi: 10.1155/2013/752719.
- [27] K. Chihara, S. Ito, H. Nagashima, M. Hikita, and R. Suzuki, "Adsorption of organics on MSC5A in supercritical CO₂, chromatographic measurements & stop & go simulation," *Adsorption*, vol. 18, no. 5–6, pp. 417–422, Dec. 2012, doi: 10.1007/s10450-012-9425-z.
- [28] D. J. Laky, D. Casas-Orozco, C. D. Laird, G. V. Reklaitis, and Z. K. Nagy, "Simulation–Optimization Framework for the Digital Design of Pharmaceutical Processes Using Pyomo and PharmaPy," *Ind. Eng. Chem. Res.*, Oct. 2022, doi: 10.1021/acs.iecr.2c01636.
- [29] M. Al-Jabari, "Kinetic models for adsorption on mineral particles comparison between Langmuir kinetics and mass transfer," *Environ. Technol. Innov.*, vol. 6, pp. 27–37, Nov. 2016, doi: 10.1016/j.eti.2016.04.005.
- [30] E. L. G. Oliveira, A. J. D. Silvestre, and C. M. Silva, "Review of kinetic models for supercritical fluid extraction," *Chemical Engineering Research and Design*, vol. 89, no. 7, pp. 1104–1117, Jul. 2011, doi: 10.1016/j.cherd.2010.10.025.
- [31] U. Guven and G. Ribert, "Impact of non-ideal transport modeling on supercritical flow simulation," *Proceedings of the Combustion Institute*, vol. 37, no. 3, pp. 3255–3262, 2019, doi: 10.1016/j.proci.2018.05.013.
- [32] S. S. Hanim, R. Norsyabilah, M. H. N. Suhaila, A. Noraishah, and A. K. S. Kartina, "Effects of Temperature, Time and Pressure on the Hemicelluloses Yield Extracted Using Subcritical Water Extraction," *Procedia Eng.*, vol. 42, pp. 562–565, 2012, doi: 10.1016/j.proeng.2012.07.448.
- [33] M. de Andrade Lima, D. Charalampopoulos, and A. Chatzifragkou, "Optimisation and modelling of supercritical CO₂ extraction process of carotenoids from carrot peels," *J. Supercrit. Fluids*, vol. 133, pp. 94–102, Mar. 2018, doi: 10.1016/j.supflu.2017.09.028.
- [34] C. P. Passos, R. M. Silva, F. A. Da Silva, M. A. Coimbra, and C. M. Silva, "Supercritical fluid extraction of grape seed (*Vitis vinifera* L.) oil. Effect of the operating conditions upon oil composition and antioxidant capacity," *Chemical Engineering Journal*, vol. 160, no. 2, pp. 634–640, Jun. 2010, doi: 10.1016/j.cej.2010.03.087.
- [35] S. Mohamed Nasser, M. Abbas, and M. Trari, "Understanding the rate-limiting step adsorption kinetics onto biomaterials for mechanism adsorption control," *Progress in Reaction Kinetics and Mechanism*, vol. 49, Jan. 2024, doi: 10.1177/14686783241226858.
- [36] M. F. de Aguiar and G. L. V. Coelho, "Adsorption of sulfur compounds from natural gas by different adsorbents and desorption using supercritical CO₂," *J. Environ. Chem. Eng.*, vol. 5, no. 5, pp. 4353–4364, Oct. 2017, doi: 10.1016/j.jece.2017.07.079.
- [37] B. Rezai and E. Allahkarami, "Desorption/regeneration of adsorbents and their performance," in *Sustainable Technologies for Remediation of Emerging Pollutants from Aqueous Environment*, Elsevier, 2024, pp. 263–283. doi: 10.1016/B978-0-443-18618-9.00021-8.
- [38] Z. Huang, "Mass Transfer Models for Supercritical Fluid Extraction," 2015, pp. 77–115. doi: 10.1007/978-3-319-10611-3_3.
- [39] L. Li, M. Chen, Y. Zeng, and G. Liu, "Application and Perspectives of Supercritical Fluid Technology in the Nutraceutical Industry," *Adv. Sustain. Syst.*, vol. 6, no. 7, Jul. 2022, doi: 10.1002/adsu.202200055.
- [40] N. Hasan and B. Farouk, "Mass transfer enhancement in supercritical fluid extraction by acoustic waves," *J. Supercrit. Fluids*, vol. 80, pp. 60–70, Aug. 2013, doi: 10.1016/j.supflu.2013.04.002.

- [41] M. Musah, Y. Azeh, J. Mathew, M. Umar, Z. Abdulhamid, and A. Muhammad, "Adsorption Kinetics and Isotherm Models: A Review," *Caliphate Journal of Science and Technology*, vol. 4, no. 1, pp. 20–26, Feb. 2022, doi: 10.4314/cajost.v4i1.3.
- [42] M. S. Gruszkiewicz, G. Rother, D. J. Wesolowski, D. R. Cole, and D. Wallacher, "Direct Measurements of Pore Fluid Density by Vibrating Tube Densimetry," *Langmuir*, vol. 28, no. 11, pp. 5070–5078, Mar. 2012, doi: 10.1021/la204517v.
- [43] U. Ranieri *et al.*, "Crossover from gas-like to liquid-like molecular diffusion in a simple supercritical fluid," *Nat. Commun.*, vol. 15, no. 1, p. 4142, May 2024, doi: 10.1038/s41467-024-47961-7.
- [44] M. R. Rahman, M. K. Bin Bakri, and M. Namakka, "Introduction to nanocarbon biocomposites," *Woodhead Publishing in Materials*, pp. 1–14, 2024, doi: <https://doi.org/10.1016/B978-0-443-13981-9.00001-6>.
- [45] K. K. Kuok, M. K. B. Bakri, P. Chan, M. Namakka, A. Mohamad, M. Y. Chin, and M. R. Rahman, "Merits of bamboo utilization in earth preservation, water, and wastewater treatment: A mini review," *BioResources*, vol. 19, no. 2, pp. 3921–3944, 2024, doi: 10.15376/biores.19.2.Kuok.
- [46] M. Namakka, M. R. Rahman, and M. K. Bin Bakri, "Waste biomass pellets for green energy production - A sustainable alternative for energy security," *Renewable and Sustainable Energy Reviews*, vol. 226, no. September 2025, 2026, doi: 10.1016/j.rser.2025.116315.
- [47] E. Matsumoto, M. M. Azim, S. Takishima, and I. Ushiki, "Adsorption Equilibria of Propyl Acetate on Activated Carbon Under Supercritical CO₂ Conditions," *Journal of Chemical Engineering of Japan*, vol. 56, no. 1, Dec. 2023, doi: 10.1080/00219592.2023.2256366.
- [48] P. L. N. Khui, M. R. Rahman, K. A. B. M. Said, M. Namakka, M. Shahabuddin, M. S. M. Al-Saleem, J. Y. Al-Humaidi, and M. M. Rahman, "Enhanced flux and surface modification of PVDF hollow fiber membranes using bamboo cellulose/PVA composite coatings," *Polym. Bull.*, vol. 82, no. 18, pp. 12909–12927, 2025, doi: 10.1007/s00289-025-06023-8.
- [49] M. Askarizadeh, N. Esfandiari, B. Honarvar, S. A. Sajadian, and A. Azdarpour, "Kinetic Modeling to Explain the Release of Medicine from Drug Delivery Systems," *ChemBioEng Reviews*, vol. 10, no. 6, pp. 1006–1049, Dec. 2023, doi: 10.1002/cben.202300027.
- [50] M. Askarizadeh, N. Esfandiari, B. Honarvar, S. A. Sajadian, and A. Azdarpour, "Kinetic Modeling to Explain the Release of Medicine from Drug Delivery Systems," *ChemBioEng Reviews*, vol. 10, no. 6, pp. 1006–1049, Dec. 2023, doi: 10.1002/cben.202300027.
- [51] A. James, M. R. Rahman, K. A. B. M. Said, M. Namakka, M. Shahabuddin, M. S. M. Al-Saleem, J. Y. Al-Humaidi, M. M. Rahman, and M. A. Salam, "Multifunctional PVDF membranes incorporating graphene, TiO₂, and nanocellulose: Synergistic effects on filtration and antifouling performance," *RSC Adv.*, vol. 15, no. 38, pp. 31471–31497, 2025, doi: 10.1039/d5ra04672f.
- [52] J. E. Mosquera, M. L. Goñi, R. E. Martini, and N. A. Gañán, "Mass transfer kinetics of CO₂ and eugenol in the supercritical impregnation of polyamide fibers: Experimental data and modeling," *J. Supercrit. Fluids*, vol. 166, p. 105030, Dec. 2020, doi: 10.1016/j.supflu.2020.105030.
- [53] A. E. Costa, A. Santana, M. B. Quadri, R. A. F. Machado, F. Recasens, and M. A. Larrayoz, "Glycerol desorption from ion exchange and adsorbent resin using supercritical fluid technology: An optimization study," *J. Supercrit. Fluids*, vol. 58, no. 2, pp. 226–232, Sep. 2011, doi: 10.1016/j.supflu.2011.06.012.
- [54] M. Namakka, R. Rahman, K. Anwar, B. Mohamed, S. Bavya, and D. Karuppasamy, "PVDF-TiO₂-nZVI Nanocomposite Membranes for the Remediation of Heavy Metal Contaminated Wastewater," 2026.
- [55] M. Namakka, M. R. Rahman, and M. K. Bin Bakri, "Investigation into self-healing in biocomposites," *Smart Biocomposite Materials: Fabrication, Applications and Sustainability*, pp. 227–255, Jan. 2026, doi: 10.1016/B978-0-443-33643-0.00006-5.
- [56] M. Namakka, M. R. Rahman, and M. K. Bin Bakri, "Natural fiber-reinforced materials: towards biosustainability," *Smart Biocomposite Materials: Fabrication, Applications and Sustainability*, pp. 23–57, Jan. 2026, doi: 10.1016/B978-0-443-33643-0.00009-0.
- [57] M. Namakka, M. R. Rahman, K. A. Bin Mohamed Said, and B. D. Karuppasamy, "Enhanced hydrophilic properties and performance evaluation of PVDF-TiO₂-nZVI-SiO₂nanocomposite membranes for the remediation of heavy metal contaminated wastewater," *RSC Adv.*, vol. 16, no. 5, pp. 4392–4419, 2026, doi: 10.1039/d5ra08723f.
- [58] A. Z. Sueraya, Md. R. Rahman, N. bt M. Sa'don, A. Anak James, M. K. Bin Bakri, and M. Namakka, "Polymer nanocomposite membranes for dye removal," *Polymer Nanocomposite Membranes in Water Treatment and Desalination*, pp. 187–207, Jan. 2025, doi: 10.1016/B978-0-443-23879-6.00006-3.
- [59] M. R. Rahman, A. Z. Sueraya, K. A. B. M. Said, M. Namakka, A. James, I. M. M. Rahman, M. S. M. Al-Saleem, J. Y. Al-Humaidi, and M. M. Rahman, "Impact of graphene/nanocellulose on nanocomposite membrane for methylene blue dye removal and antifouling performance," *J. Appl. Polym. Sci.*, pp. 1–21, 2025, doi: 10.1002/app.57648.

- [60] L. Sun, M. Gao, and S. Tang, "Porous amino acid-functionalized poly(ionic liquid) foamed with supercritical CO₂ and its application in CO₂ adsorption," *Chemical Engineering Journal*, vol. 412, p. 128764, May 2021, doi: 10.1016/j.cej.2021.128764.
- [61] R. Chokkareddy, J. C. Kilele, S. Kanchi, G. G. Redhi, B. Kabane, and N. K. Katari, "Novel magnetic nanocomposites and their environmental applications," in *Magnetic Nanoparticles and Polymer Nanocomposites*, Elsevier, 2024, pp. 403–414. doi: 10.1016/B978-0-323-85748-2.00015-3.
- [62] J. K. Sahoo, J. Das, H. S. Mohanty, D. Panda, J. Rath, and H. Sahoo, "Surface functionalization and modification strategies of magnetic nanosystems," in *Functionalized Magnetic Nanosystems for Diagnostic Tools and Devices*, Elsevier, 2024, pp. 79–113. doi: 10.1016/B978-0-443-19012-4.00005-9.
- [63] S. Katiyar and R. Katiyar, "A comprehensive review on synthesis and application of nanocomposites for adsorption of chromium: status and future prospective," *Appl. Water Sci.*, vol. 14, no. 1, p. 11, Jan. 2024, doi: 10.1007/s13201-023-02062-6.
- [64] M. K. Bin Bakri, M. R. Rahman, and M. Namakka, "Introduction to intelligent biocomposite materials," *Smart Biocomposite Materials: Fabrication, Applications and Sustainability*, pp. 1–22, Jan. 2026, doi: 10.1016/B978-0-443-33643-0.00008-9.
- [65] G. Yunusa, B. Dalhatu, A. A. Shehu, and K. Suleiman, "2 3 4 5," vol. 9, no. 9, pp. 243–249, 2025.
- [66] X. Lu, Z. Liu, W. Wang, X. Wang, H. Ma, and M. Cao, "Synthesis and Evaluation of Peptide–Manganese Dioxide Nanocomposites as Adsorbents for the Removal of Strontium Ions," *Nanomaterials*, vol. 14, no. 1, p. 52, Dec. 2023, doi: 10.3390/nano14010052.
- [67] K. Ikeda, Y. Shimoyama, and Y. Orita, "Efficient purification of surface modified nanoparticles from its nanosuspension by using supercritical CO₂ technology," *J. Supercrit. Fluids*, vol. 199, p. 105966, Aug. 2023, doi: 10.1016/j.supflu.2023.105966.
- [68] T. Wijakmatee, Y. Shimoyama, and Y. Orita, "Enhanced extraction of surface-modified iron oxide nanoparticles using supercritical carbon dioxide," *Sep. Purif. Technol.*, vol. 354, p. 129067, Feb. 2025, doi: 10.1016/j.seppur.2024.129067.
- [69] T. Wijakmatee, Y. Shimoyama, and Y. Orita, "Enhanced extraction of surface-modified iron oxide nanoparticles using supercritical carbon dioxide," *Sep. Purif. Technol.*, vol. 354, p. 129067, Feb. 2025, doi: 10.1016/j.seppur.2024.129067.
- [70] K. Xu, W. Mo, F. Zhang, C. J. Houtman, J. Wan, and B. Li, "Surface Modification of Adsorbents for Removing Microstickies from Papermaking Whitewater," *Ind. Eng. Chem. Res.*, vol. 63, no. 17, pp. 7950–7957, May 2024, doi: 10.1021/acs.iecr.4c00483.
- [71] J. Oliveras, L. Marcon, N. G. Bastús, and V. Puentes, "Functionalization of graphene nanostructures with inorganic nanoparticles and their use for the removal of pharmaceutical pollutants in water," *Frontiers in Chemical Engineering*, vol. 4, Dec. 2022, doi: 10.3389/fceng.2022.1084035.
- [72] M. G. Montalbán and G. Vllora, "Supercritical Fluids: Properties and Applications," in *Phase Equilibria With Supercritical Carbon Dioxide - Application to the Components of a Biocatalytic Process*, IntechOpen, 2022. doi: 10.5772/intechopen.105485.
- [73] K. A. Kravanja, M. Finšgar, Ž. Knez, and M. Knez Marevci, "Supercritical Fluid Technologies for the Incorporation of Synthetic and Natural Active Compounds into Materials for Drug Formulation and Delivery," *Pharmaceutics*, vol. 14, no. 8, p. 1670, Aug. 2022, doi: 10.3390/pharmaceutics14081670.
- [74] G. Brunner, "Applications of Supercritical Fluids," *Annu. Rev. Chem. Biomol. Eng.*, vol. 1, no. 1, pp. 321–342, Jun. 2010, doi: 10.1146/annurev-chembioeng-073009-101311.
- [75] H. Liu, X. Liang, Y. Peng, G. Liu, and H. Cheng, "Supercritical Fluids: An Innovative Strategy for Drug Development," *Bioengineering*, vol. 11, no. 8, p. 788, Aug. 2024, doi: 10.3390/bioengineering11080788.
- [76] A. Jamshed, A. Iqbal, S. Ali, S. Ali, and M., "A quick review on the applications of nanomaterials as adsorbents," *MOJ Ecology & Environmental Sciences*, vol. 8, no. 3, pp. 86–89, Jun. 2023, doi: 10.15406/mojes.2023.08.00278.
- [77] V. R. Kandula, L. Shaikh, and G. Samanta, "Utilization of Quality by Design, Kinetic Modeling, and Computational Fluid Dynamics for Process Optimization and Scale-Up," *Org. Process Res. Dev.*, vol. 27, no. 5, pp. 875–889, May 2023, doi: 10.1021/acs.oprd.3c00015.
- [78] H. Tian, J. S. Jagana, Q. Zhang, and M. Ierapetritou, "Feasibility/Flexibility-based optimization for process design and operations," *Comput. Chem. Eng.*, vol. 180, p. 108461, Jan. 2024, doi: 10.1016/j.compchemeng.2023.108461.