Fluidized Bed Scale-Up for Sustainability Challenges. 1. Tomorrow's Tools

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Review

Fluidized Bed Scale-Up for Sustainability Challenges. 1. Tomorrow's Tools

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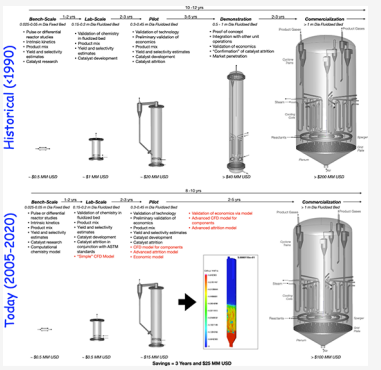
ABSTRACT: The scaling up of fluidized beds has been purposefully pursued for more than 100 years. Yet, over that time, scale-up tools have not significantly changed. Data analysis is typically a standard analysis of variances statistical exercise, perhaps reinforced with a design of experimental procedure. Flowsheeting and equipment design are based on institutional knowledge, albeit graphical user interface-based process flow models make that job more manageable. Advanced models such as computational fluid dynamics are used but often as a supplement and not a primary driver. As a result, the scale-up process for a fluidized bed can take more than 10 years. Fluidized beds remain at the forefront of the present time-critical sustainability challenges, e.g., carbon capture by particulate sorbents, methane-to-hydrogen, plastic-to-chemicals, etc. In view of the exigency toward net zero, today's scale-up efforts need to be accelerated, leveraging the advanced new tools that have become readily available. The problem is that such tools are often neglected, inadequately implemented, ineffectively resourced, and/or poorly understood. This motivated the current effort, which is targeted at reviewing how scale-up tools have evolved over the years and the promising new tools, addressing some of the barriers of these tools in the design and scale-up of fluidized beds, as well as contemplating what can be done to circumvent these barriers. As a follow up, a companion part 2 (Cocco, R. A.; Chew, J. W. *Ind. Eng. Chem. Res.*, submitted for publication) proposes a new scale-up path leveraging the advanced tools to achieve timely implementation of the new green fluidized bed processes.



INTRODUCTION

The year 2022 represents a significant milestone in fluidization, namely, the 100th anniversary of the commercial fluidized bed reactor.[1] The persistent interest today in using the fluidized bed to tackle time-critical sustainability challenges (e.g., carbon capture by particulate sorbents, methane-to-hydrogen, plastic-to-chemicals, etc.) is not surprising, considering the proven efficacy of such unit operations over 100 years in wide-ranging applications (Figure 1).

With today's urgent climate-change-related processes and the need for sustainability and circularity, expediting the commercialization of these new concepts is all the more critical. Carbon-zero goals have been set for as soon as 2025,[2] so the timeline is akin to the same urgency during World War II that drove the operation of 35 commercial fluidized catalyst cracking units (FCCUs) within five years from conception.[1] We need to move faster without compromising today's and tomorrow's energy conservation, safety, waste reduction, and emissions reduction objectives.

Fluidized bed technology and scale up play a pivoting role in this need. As shown in Figure 2, new green-transition technologies with biomass gasification and pyrolysis, plastic pyrolysis, photovoltaics, batteries, and methane pyrolysis are based on fluidization. Fortunately, current capabilities now allow scale-up efforts to move beyond the cost- and time-consuming Edisonian approaches of the past. Concept tools such as Design for Six Sigma,[3] TRIZ (Theory of Inventive Problem-Solving),[4] NICE (Nature-Inspired Chemical Engineering),[5] SCAMPER (Substitute, Combine, Adapt, Modify/Magnify, Purpose, Eliminate/Minimize, and Rearrange/Reverse),[6] CPS (Creative Problem Solving),[7] SWOT (Strength, Weaknesses, Opportunities, and Threats),[8] RAP (Risk Assessment Process),[9] and Flowsheet Synthesis[10] have become available to get the design team on the optimized track in a much shorter time. Modeling tools such as computational fluid dynamics (CFD), computational fluid dynamics−discrete

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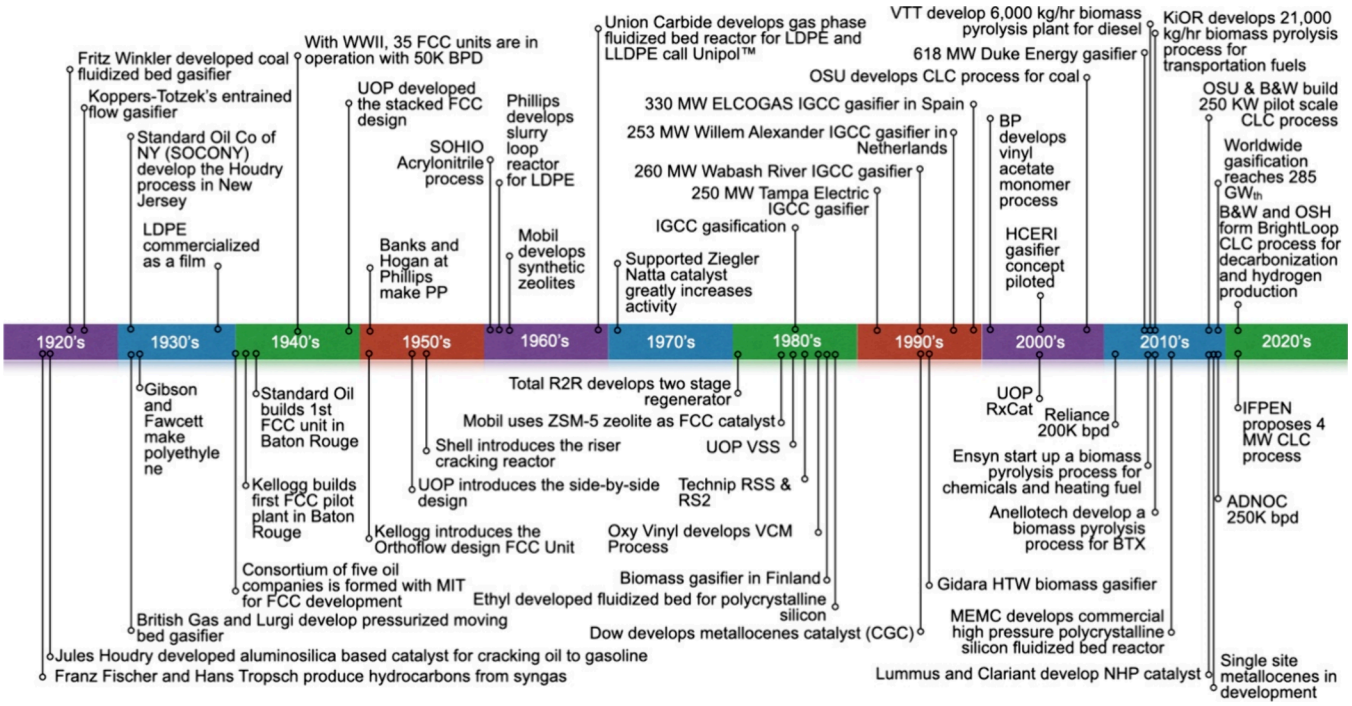

Figure 1. Key milestones of fluidization. Details taken from ref 1. Copyright 2022 Elsevier B.V.

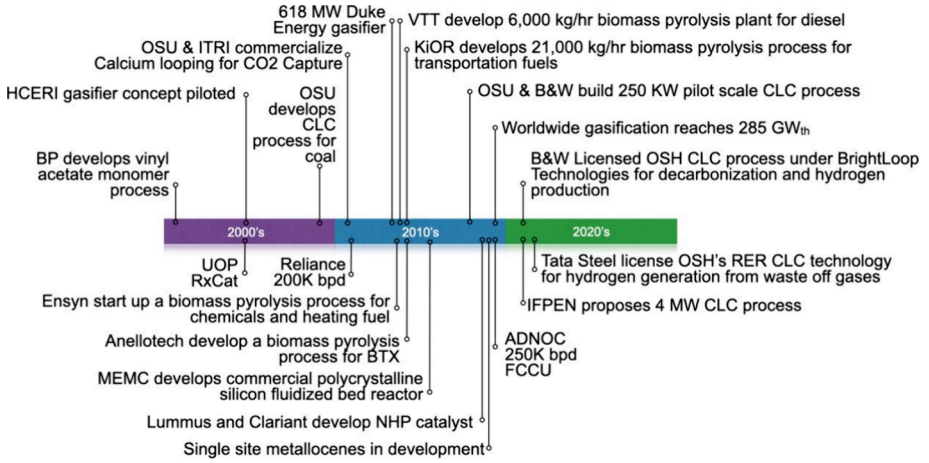

Figure 2. Time-critical sustainability challenges.

element method (CFD-DEM), and hybrid multiphase particle-in-cell (MP-PIC) have been refined over the past 30 years for fluidized bed applications.[11−14]

Today's codes can model commercial-scale units (including chemical reactions and heat transfer) in weeks, often much faster than setting up and collecting data from a lab-scale unit, and certainly faster than pilot units or process development units. Advanced statistics, along with artificial intelligence and machine learning, all have a role to play here as well. However, it is more than just having access to these tools; how, when, why, and how often need to be defined at the very start of the scale-up project. Here, the tools and the scale-up path for using those tools are defined in two parts, with this first part giving a perspective on tools and the subsequent part[15] focusing on the path.

HISTORICAL TOOLS (<1990)

The success of the FCCU process during World War II garnered keen interest in the new gas−solid contacting method. As a damper on the success, shortly after in 1950, two 5 m diameter Fischer−Tropsch reactors (i.e., bubbling fluidized beds) in Texas were found to have much lower yields than that in the pilot 0.3 m diameter pilot reactors, which has been tied to slugging at the pilot reactor that led to much shorter gas residence time in the nonslugging commercial reactor.[16] This failure underscores the need for judicious scale up to leverage the benefits of fluidized bed reactors, leading to extensive studies to bridge the gaps. The first phenomenon earnestly studied was the gas−solid heat transfer rate, followed by drawing analogies to plug versus mixed flow models, resulting in at least 34 models by 1970 that contradict one another.[17] The scale up of this era was largely Edisonian, as extensive efforts in the background proceeded in parallel to contribute toward more mechanistic understanding.

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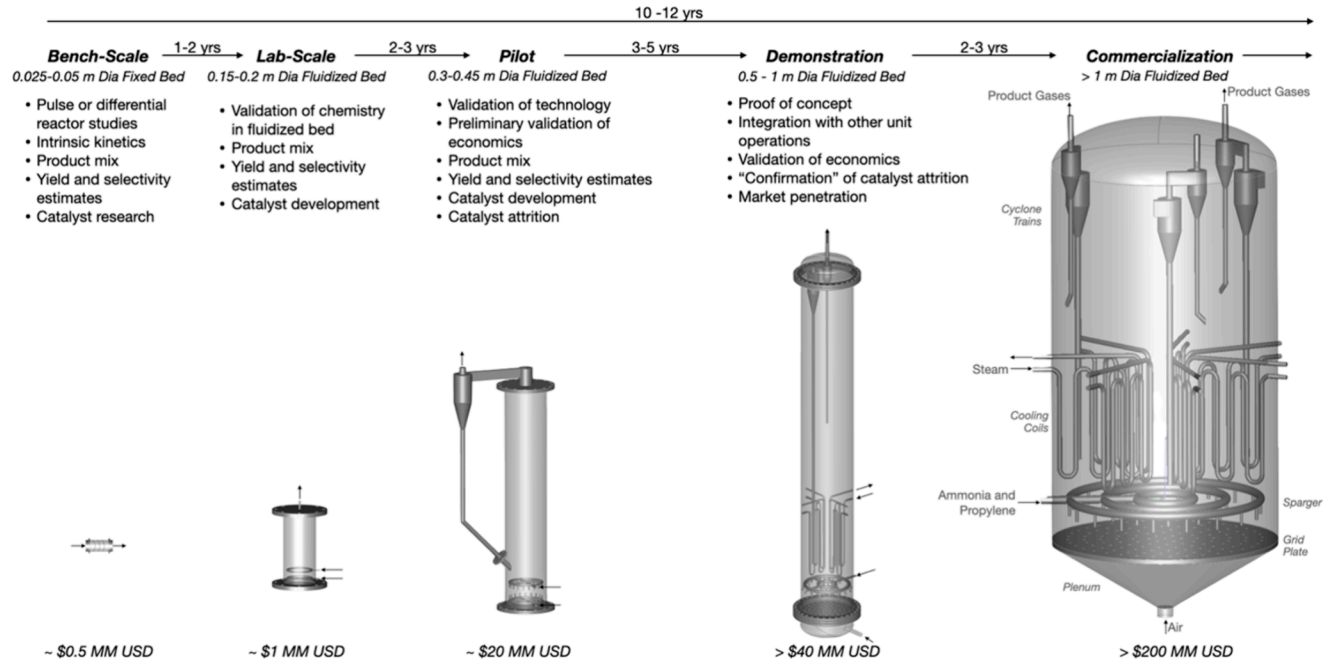

Figure 3. Historical tools (<1990): Stochastic (Edisonian) scale up.

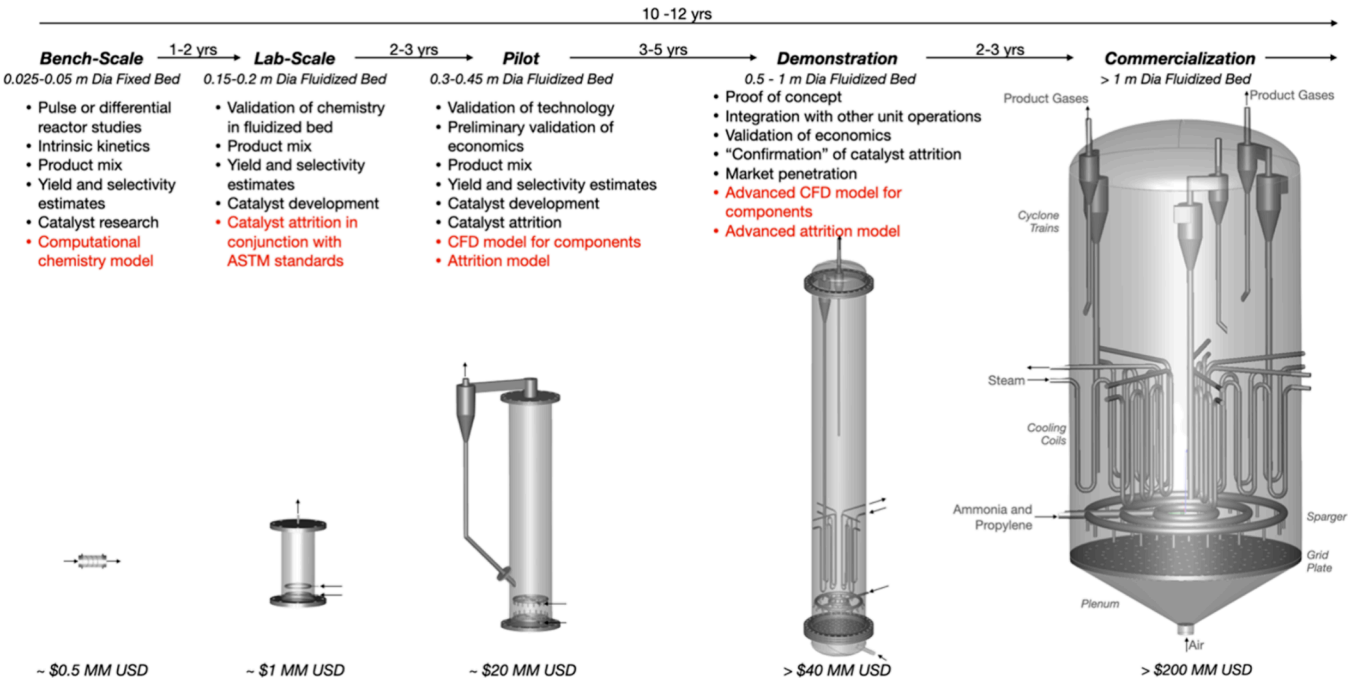

Figure 4. Yesterday's tools (1990−2005): Semi-deterministic scale up.

Figure 3 illustrates a typical traditional scale-up procedure. The first step is the bench-scale unit, typically a fixed bed with a diameter of 0.025−0.05 m. Conventionally, this has been a trial-and-error process to establish the efficacy of the desired reaction at this scale, including preliminary data on the reaction kinetics, product mixture, yield, selectivity, and catalyst type. The cost incurred at this stage is approximately $0.5 MM. In about 1−2 years, a lab-scale unit is tested using a more practical fluidized bed with a diameter of about 0.15−0.2 m. Validation of the chemical reaction is obtained at this point, and the behavior and properties of the catalyst are critically assessed and improved upon. As with the bench-scale test, this step costs similarly $1 MM. In another 2−3 years, a pilot-scale fluidized bed with a diameter of about 0.3−0.45 m is set up and used to validate the technology and economics, as well as develop models for catalyst attrition and scaling up. The price tag of this stage is much steeper at $20 MM due to the larger scale. Then, 3−5 years later, the demonstration unit, which is a 0.5−1 m diameter fluidized bed, is used to provide proof-of-concept at the targeted operating conditions and integrated with associated unit operations. Catalyst attrition and scaling models are further validated. The economics of the process and predicted market penetration are analyzed at this stage. The demonstration step is estimated at upward of $40 MM but

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is a cost considered necessary as a final check before commercialization. Finally, in about 2−3 years, the commercial fluidized bed with a diameter of more than 1 m is built at a cost of more than $200 MM. Beyond this, further refinements and design iterations continue.

The risk involved is different at each step, with the scaling from small to large reported to be relatively more risky than that from large to larger.[18] From bench scale to commercial scale, the total duration is approximately 10−12 years, and the total cost is upward of $500 MM. The time-critical climate-change challenges cannot wait 10 years.

Clearly, the scale-up of this era is driven by expensive (time-wise and cost-wise) experiments. It was highlighted in 1985 that, although the oil and chemical industries have successfully transformed chemistry concepts into commercial technology and products, chemical plants processing solids have been underperforming with respect to that designed for the past two decades.[19] This deficiency was attributed to the focus of industrial research and development (R&D) on chemistry rather than physics. In particular, the lack of focus on fluidization hydrodynamics significantly diminished the scale-up ratio at each step.[1] Specifically, when plug-flow reactors (PFRs) and continuous stirred tank reactors (CSTRs) scaled at 1000−10000 times, fluid catalytic cracking (FCC) reactors and fluid coking scaled at roughly 20−150 and 40 times, respectively, from pilot to commercial scales in the 1940−1950s. Correspondingly, the demand for faster and cheaper scale up catalyzed extensive academic research on more fundamental mechanisms, representative efforts of which are bubble hydrodynamics models in the 1960s,[20−23] entrainment and transport disengaging heights in the 1970s and 1980s,[24−27] and scaling laws[28−31] and compartment models[32−34] in the 1990s.

As Gidaspow's 1986 review[35] indicated, hydrodynamic models programmed on supercomputers of that time have become available to predict bubbles, heat transfer coefficients, and product distributions in various fluidization regimes. However, simulation tools are not involved in scale up yet due to the lack of maturity of the models, lack of computational capacity to obtain results expeditiously, and lack of confidence in any of the results generated.[36] At that point, no multiphase reactor model is adequate to either enhance existing units or scale up new ones, which is tied to the poor understanding of the macroscale structures that impact interphase contact, and thus unsurety of heat and mass transfer, and pressure and temperature effects.[37]

YESTERDAY'S TOOLS (1990−2005)

Figure 4 represents an update of Figure 3 based on the advances made. In particular, although the number of units and durations remained the same, the red fonts in Figure 4 highlight the advent of better models (namely, computational chemistry, CFD, attrition) and better testing methods (e.g., attrition[38]) to enhance the information obtained at each step. Nonetheless, seven decades since the first commercial fluidized bed reactor back in the 1920s, scale up remained "that mix of mathematics, witchcraft, history and common sense which we call engineering" such that managing the uncertainties was deemed more crucial than improving calculation accuracy.[39] The challenges related to scaling up the fluidization behavior have been highlighted as different phenomena between the scales, including particle sintering, erosion, solids flow, particle size distribution (and evolving changes due to growth, attrition, etc.), and entrainment (and thus particle losses).

CFD modeling remained limited.[40] Stemming from the U. S. Department of Energy's (DOE) Technology Vision 2020 in the mid-1990s,[41] the Chemical Industry laid out the goals for reliable simulation tools with detailed chemistry and transport models, including that for granular-fluid unit operations. Part of that effort was the Multiphase Fluid Dynamics Research Consortium (MFDRC), which consisted of universities (namely, University of Colorado at Boulder, Iowa State, Illinois Institute of Technology, University of Michigan, Princeton, Purdue, and Washington University at St. Louis), industries (namely, Dow, Dow Corning, DuPont, Millennium, Fluent, Chevron, AEA, ExxonMobil, Siemens, and PSRI), national laboratories (namely, Ames, National Energy Technology Lab, Los Alamos, Oak Ridge, Pacific Northwest, and Sandia), and government agencies targeted at fundamental multiphase fluid dynamics research to answer the call for experimentally validated computational tools. MFDRC set out a three-pronged research approach: (i) numerical methods, (ii) phenomenology and constitutive relations, and (iii) experimental validation. The specific goals were that CFD tools need to be versatile, fundamentally based, experimentally verified, computationally efficient, and user-friendly. Its five year charter in 1999 focused on developing CFD models that could address deficiencies in fluidized beds and circulating bed risers. This resulted in full algebraic and differential granular temperature equations in the Fluent and AEA CFX commercial CFD codes, as well as advanced experimental techniques, including γ-ray tomography, capacitance tomography, computer-aided radioactive particle tracking, and millimeter wave velocimeter for riser hydrodynamics.[42]

Also based on the U. S. DOE Technology Vision 2020 platform that calls for reliable simulation tools,[41] 1999 brought a dedicated initiative named "Technology Roadmap for Computational Fluid Dynamics" targeted at chemical industries.[43] MFDRC identified the path forward for designing fluidized beds. At that time, CFD was a relatively straightforward modeling approach based on the two-fluid model, whereby each phase is treated as a continuum and continuity and momentum equations are solved using closure equations. To enhance the resolution of the particle phase at a reasonable computational cost, a multiphase particle-in-cell (MP-PIC) technique was developed for fluidized bed applications by ArenaFlow (now CPFD-Software). The fluid phase is handled similarly to the two-fluid model, but the particle phase is mapped on a Lagrangian framework.[44] Particles or parcels of particles could be tracked, but collisions are handled as in the two-fluid model whereby the solids stresses are numerically determined from a packing fraction model or the kinetic theory of granular flow.[45]

Toward improving predictive capability, this era brought about efforts directed at first-principles understanding underpinning the governing equations that describe the various fluidization phenomena,[46] particularly with respect to bridging the microscale phenomena to the macroscale challenges in practical applications.[47] The overarching goal has been to achieve a fully specified model that runs efficiently. As evident in the discrepancies among empirical correlations for various phenomena (e.g., entrainment rate,[48] transport disengaging height,[49] and minimum fluidization velocity[50]) of multiple orders of magnitude, many deficiencies still exist that compromise confidence in the predictions and thereby the

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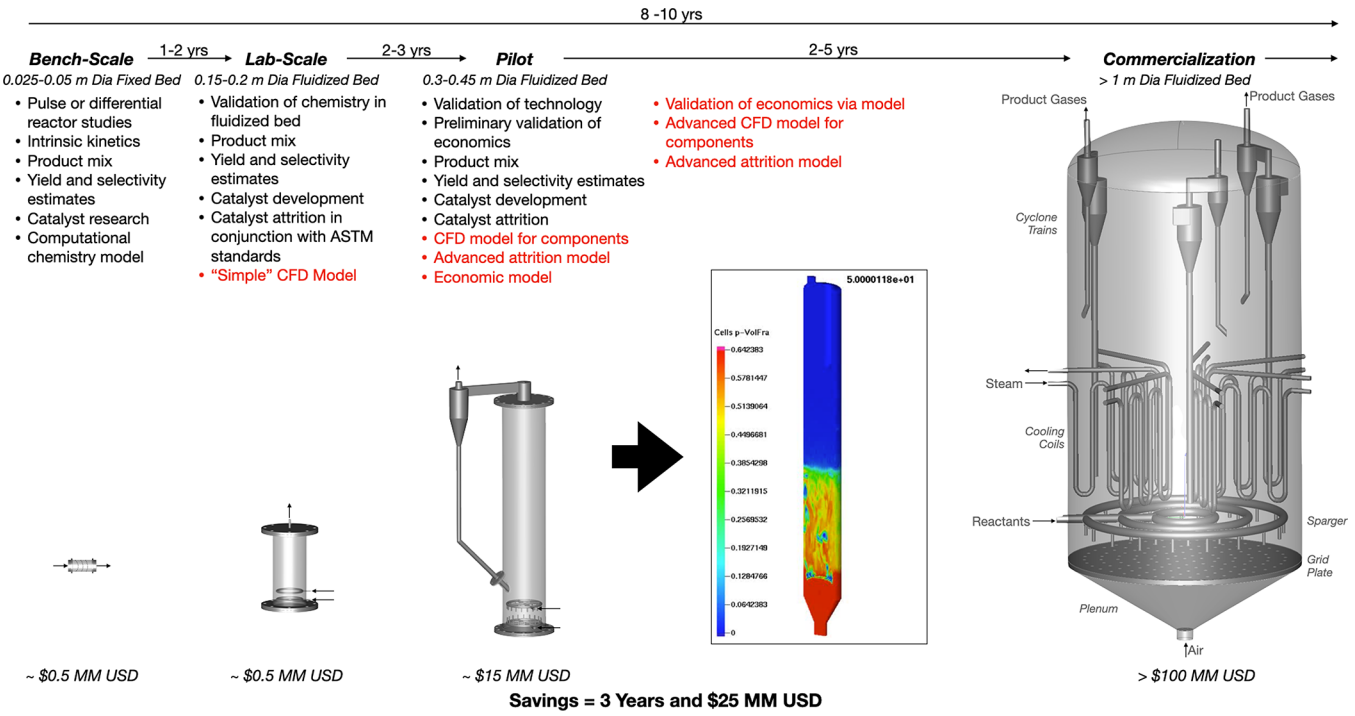

Figure 5. Today's tools (2005−2020): Semi-deterministic scale up.

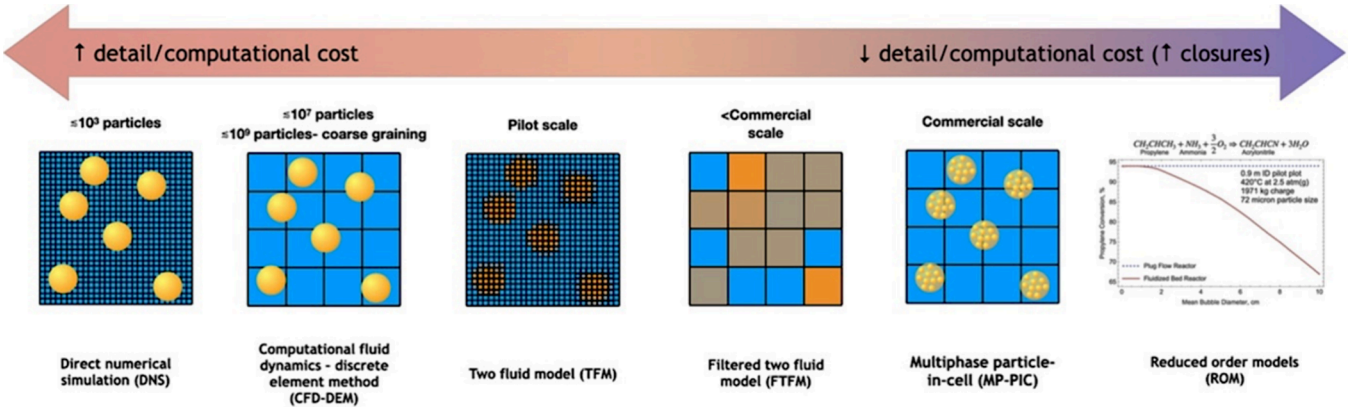

Figure 6. Summary of hydrodynamic models. The number of particles specified is based on current capabilities. Reproduced with permission from ref 1. Copyright 2022 Elsevier.

models. For instance, although the particles are inevitably nonmonodisperse, the understanding of the effect of polydispersity was and remains somewhat imperfect. Extensive efforts were directed to the corresponding incorporation of polydispersity into the models.[51,52] As another example, the convention was homogeneous grids, which may not be as accurate. Specifically, each set of equations describes a grid whose size is chosen based on the resolution targeted. It is now well-known that subgrids can be used to resolve small mesoscale structures (i.e., bubbles and clusters), which are constantly evolving. However, subgrid models were not available yet in 2000,[37] and thus, smaller grids have to be used throughout the system to factor in the smaller structures. Coupled with energy and species balances, the magnitude of the task is not trivial and thus not commonly carried out back then.

TODAY'S TOOLS (2005−2020)

As models improved and confidence in model predictions grew, the possibility of eliminating a step in the scale up of fluidized bed processes became a reality. Figure 5 shows that the maturation of multifaceted models (i.e., economics, computational fluid dynamics, and attrition) has allowed for reduced tests needed at the lab and pilot cales as well as the bypassing of a step (e.g., the pilot plant or the market development unit (MDU)), resulting in savings of approximately three years and $25−50 MM.

Leveraging more stringent scaling laws and similitude of incoherent output power spectral density (IOP) of pressure fluctuations, successful scale up from lab scale to PDU scale has been proven.[53] Advanced analysis methods made possible the extraction of more valuable information from pressure data. For example, while pressure data are typically only used for extracting information on bed density or for calculating standard deviation as an imprecise indicator of fluidization

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state, more sophisticated tools like wavelet decomposition of pressure signals made possible a direct indicator of fluidization quality in a commercial fluidized bed application.[54] A sectoral scaling method has also been shown to work well to scale up bubbling fluidized beds from the lab scale to market development unit (MDU) scale.[53]

In a 2017 article, a poll of CFD and CFD-DEM champions from various industries showed that 39% of respondents consider CFD-DEM to be a valuable industrial tool, and that percentage ballooned to 83% by 2022.[55] Regarding the simulation of hydrodynamics, Figure 6 presents the trade-off between resolution and computational cost;[1] readers are referred to a recent review for more details on the various methods.[56] Direct numerical simulation (DNS), which resolves the gas phase at the required spatiotemporal scale and tracks the particle motion individually, gives high-resolution information but can only simulate 10^3 particles at present. While the number of particles is inadequate to match even that of bench-scale tests, it is an important technique for enhancing correlations (e.g., drag, heat, and mass transfer) and providing mechanistic understanding. Compared to DNS, the computational fluid dynamics−discrete element method (CFD-DEM) accounts for at least 4 orders of magnitude more particles, i.e., 10^7 particles, through using averaged equations for the gas phase. Various coarse-graining methods allow for 10^9 particles to be simulated by using parcels to represent numerous similar particles, thus reducing the computation load. This is still insufficient for the pilot scale, much less the commercial scale. The two-fluid model (TFM), which presumes the gas and particle phases to be interpenetrating continua, compromises particle-phase resolution to enable pilot-scale fluidized beds to be simulated. The earlier embodiments of TFM assume homogeneity within each computation grid, which thus requires smaller grids to resolve gradients of instabilities, which in turn increases the computational load. To address this, subgrid models were devised to account for local variations in particle concentration and velocity within each grid, thereby facilitating the use of larger grids, and hence lower computation costs. Well-established embodiments are the filtered-TFM and energy minimization multiscale system (EMMS), almost permitting commercial-scale fluidized bed behaviors to be predicted.

All of these developments directly or indirectly led to the general embracing of commercial software by around 2005 due to significant improvements to hardware that made possible simulations of commercial units for the simulations of commercial fluidized bed reactors. Figure 7 shows the steep increase in publications at this transition. The two most popular ones are arguably Fluent and Barracuda, both of which are based on similar coarse-graining CFD-DEM principles to enable the simulation of fewer particles, use of larger time steps, and using force terms to represent parcel−parcel interactions. Fluent's method is termed dense discrete particle model (DDPM), while Barracuda's is the multiphase particle-in-cell (MP-PIC), whose theoretical foundation is the kinetic equation for the particle phase. Due to the retention of the discrete nature of the particle phase, the actual particle size distribution can be used to enhance the accuracy of the predictions.

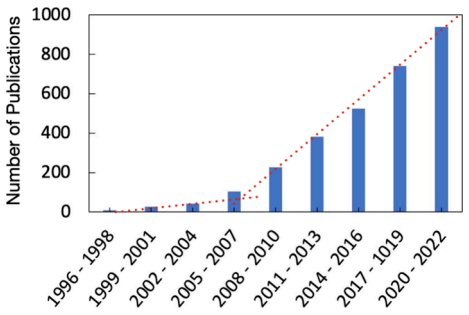

Figure 7. Number of CFD publications on fluidization through the years. Search term = "CFD fluidiz*" in "All fields". Source = Web of Science. Date retrieved = October 21, 2023.

Since ca. 2010, CFD has been undergoing rapid advances in three key areas,[57] namely, (i) algorithm with more accurate description of the physical phenomena that leverages the state-of-the-art processor speed and capacity; (ii) computer architecture evolving from single CPU (central processing unit) to 100s of CPUs to 1000s of graphical processing units (GPUs) to grid or cloud computing and massive parallel systems with multicore processors; and (iii) CFD as an embedded application to focus on specific design functions rather than on CFD per se. However, the limitations plaguing CFD highlighted in 2012[57] were acknowledged to have persisted:[58] (1) grid selection, (2) ease of use, (3) computation speed, (4) robustness, (5) incomplete physics, (6) accounting for uncertainty, and (7) confidence in predictions.

Typical tools used for fluidized beds are two-fluid CFD models (TFM), CFD-DEM models, or hybrid MP-PIC models.[56,59] Today's CFD models are sophisticated and robust and able to provide simulation results of a commercial process in a few weeks or less.[60] Most commercial CFD models directed toward granular-fluid flow can capture the particle size distribution instead of using one "representative" particle size,[61] which is inherent in the Lagrangian framework of CFD-DEM and MP-PIC codes. For the two-fluid model, moments can be used to represent that particle size distribution.[62,63]

Commercial codes have been shown to be accurate within the confines of the code's capabilities.[64] Indeed, both well-established engineering companies[65,66] and start ups[67] have embraced such commercial tools. It should be noted that these codes are limited when other factors such as shape, interparticle forces, agglomeration, clustering, attrition, and particle growth or shrinkage come into play. Capturing these effects requires adaptations to the constitutive equations involving the drag and the collisional stresses, along with numerical operations with spatial and temporal resolution.

Drag. There are several issues with drag. First, it assumes all particles are independent and not interacting with each other. For Geldart Group A and C particles, this is often far from true. Second, many drag correlations were developed from a single-particle terminal velocity or packed-bed pressure drops where solids concentration in fluidized beds is between these two limits. Efforts have been dedicated to using direct numerical simulation (DNS) to determine the corresponding drag relationship.[68−72] While much of this work is promising, many of the DNS models have their own limitations such as stationary particles, nonrotating particles, dynamic clustering, frictionless surfaces, etc. More recent studies have addressed some of these issues.[70,72] Nevertheless, this means that grid resolution can affect the calculated drag if grid resolution is not resolved enough.[73] Details of these drag models and how

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clustering and grid resolutions matter are addressed in a recent review.[74]

Still, many of today's commercial codes do not capture interparticle forces. These forces are presumed to play roles in the particle clustering in fluidized beds. Whether it is van der Waals forces, Coulombic forces, or liquid bridging is still up for debate.[75−77] This omission can be significant with Geldart Groups A and C particles,[78−80] especially with respect to predicting the entrainment rates. Indeed, even the wall material can have a pronounced effect on the hydrodynamics in a fluidized bed.[81] Addressing interparticle forces is complex and dependent on the environment to which van der Waals, Coulombic, and/or adsorbate bridging forces are sensitive too. It is known that Coulombic forces tend to decrease with increasing temperature due to a lower conductivity (i.e., increased electron scattering).[82−85] LaMarche et al.[86] showed experimentally that the electric field created by a tribocharged Teflon sheet was enough to cause significant dielectrophoretic interactions and suggested that triboelectrification present in granular flows would be enough to cause similar effects. In contrast, van der Waals forces increase with increasing bed temperatures.[87] Adsorbates (i.e., hydroxyl groups) on the particle's surface may change in concentration or functionality, which affects a particle's behavior. All of this can lead to increased or decreased particle clustering or microclustering (i.e., on the order of 10s of particles loosely held together[88]), a known factor affecting scale-up behavior.[89]

Waitukaitis et al.[90] quantified such particle interactions on a microscopic scale. Their research showed that surface roughness significantly reduced particle clustering, as determined by adding fumed silica nanoparticles to 300 μm glass spheres. Cocco et al.[91] hypothesized that changes in surface roughness lead to different particle rotational and collision dynamics. A later study[92] showed that the degree of particle clustering correlated to the consolidation index and the basic flowability energy was determined from powder rheometer measurements (i.e., FT4 powder rheometer). Similarly, Mishra et al.[93] found that the characteristic velocities obtained from powder rheometry provided a relative gauge to particle-level cohesion. Affleck et al.[94] measured the over- and undershoot with the pressure drop and bed voidage curves obtained from fluidization and defluidization experiments to determine the granular Bond number. Soleimani et al.[95] were able to quantify the degree of interparticle forces with a dynamic Hausner ratio (i.e., tapped bulk density to freely settled bulk density), which is an extension of the work of Manuel Valverde et al.[96]

Tenneti and Subramaniam[97] used DNS of granular fluid flow to develop a fluid−particle and particle−particle drag force, which was then applied to CFD models. Sundaresan[98] proposed that DNS can be used to formulate a particle-phase stress model that captures the PSD and interparticle forces via the Bond number. Even pure CFD-DEM models are limited. Drag is still approximated with the same single-particle or packed-bed models as that used with CFD. Collisional stresses are captured on a singular particle-to-particle or particle-to-wall event, but rigorously capturing that collision is CPU-prohibitive for large commercial applications. For large DEM computation domains, collisions are soft and captured with a collision stress model of some combination of elasticity and viscous dissipation. Still, it does allow for cohesion directly using a Johnson−Kendeal−Roberts (JKR)-type model. Yet, the JKR needs a value for the Hamaker constant, which is difficult to measure in the presence of particle rotation.[99,100]

Collisional Stresses. Commercial CFD codes capture collisional stresses using a packing fraction relationship[101] or the kinetic theory of granular fluids (KTGF).[102−104] For lower-velocity fluidized beds, the hydrodynamics is drag-dominated and collisional stress models have less impact on the modeling results. For circulating fluidized bed risers, the duality of particle concentrations between the wall and core[105] requires a sufficient collisional stress model, typically using the differential form of the granular temperature equation.[106−109]

The KTGF formulation requires the coefficient of restitution and the specularity coefficient as inputs. The coefficient of restitution captures the elasticity of normal particle impact (particles or walls) while the specularity coefficient captures the shear impact of a particle with the wall. Both are complicated to measure. Marinack et al.[110] determined the coefficient of restitution using a particle drop test with high-speed video images. A similar method was used by Tang et al.[111] Jiang et al.[112] extended this technique to capture the coefficient of restitution of irregular particles (i.e., maltodextrin). Oesau et al.[113] used magnetic particle tracking for paramagnetic particles with a similar particle-drop procedure. Measuring the specularity coefficient is not much different than that used for coefficient of restitution. Instead of a particle drop, particles are ejected at an angle to the surface, and the resulting speed and angle of impact were captured with high-speed videoing.[114] Given the complexity of these experiments, numerical solutions have been tested. Gu et al.[115] used CFD-DEM OpenFOAM solver with LIGGGHTS integration (i.e., two-way coupling) to obtain the required inputs for the particle phase stress model. However, the model is limited to smooth, spherical particles.

Thus, the collisional stress input parameters are often relaxed to technical experience or used as a tuning parameter at the model validation step. For lower-velocity fluidized beds, this may be effective; but, for riser simulations, such efforts should be done with caution. For most CFD simulation efforts, this remains a gap.

Grid Resolution. Grid resolution is an important parameter in CFD simulations of fluidized beds. First, grids should be configured to a 3D computational domain. The asymmetric characteristics of fluidized bed hydrodynamics can only realistically be obtained in a 3D framework. Notably, 3D simulations have been shown to obtain solution convergency faster (normalized) than that of 2D simulations.[116] To sufficiently capture particle drag, grid size should be on the order of 10 times the particle size.[73,117] For Geldart Group A particles are on the order of 1 mm, which is unrealistic for commercial-scale simulations. Thus, numerical assumptions also need to be considered with large-scale simulations, mainly by using subgrid models to accelerate solution times. As a result, a filtered drag expression has been used where drag is first evaluated in a much smaller, more resolved computational domain gridded in accordance with this filtered size. In the filtered model, the drag coefficient is obtained by filtering fine-grid simulations. Presumably, the filtered drag accounts for the less resolved grids in the larger computational domain.[118,119] A dynamic grid adjustment approach, whereby the impact of unresolved scales is predicted from the large resolved scales, has also been demonstrated;[120,121] this still remains within academic realms.

Coarse Graining. For Lagrangian-based models, the number of particles in a commercial-scale simulation is prohibitive. Today, CFD-DEM or MP-PIC models are capable

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of modeling $O(10^7)$ particles whereas commercial fluidized bed systems have $O(10^{14})$ particles.[55] Thus, numerical coarse-graining assumptions are typically used for CFD-DEM and MP-PIC simulations of commercial-scale computational domains. The number of particles and the equations tied to them can be reduced by grouping similar particles in a cell into one larger or more dense particle or both. As long as the particle properties are similar, they can be grouped within a computational cell. The level of this grouping, called a cloud or parcel, needs to be resolved much like the grid resolution. Interestingly enough, the effects of this type of coarse-graining could be minimized by using a cluster-based drag model.[122]

TOMORROW'S TOOLS (>2020)

Artificial Intelligence (AI) has become the tool for everything. Currently, that is a stretch, but its usefulness in the concept and development stages can also be valuable. A perspective article in 2019 pointed out that AI in chemical engineering is about 40 years old.[123] Notably, the first AIChE (American Institute of Chemical Engineers) session on AI was in 1985, and the first articles on AI in process engineering appeared in 1986. More importantly, AI has already been implemented in industry with respect to process operations and diagnosis, and thus is a proven tool. For example, British Petroleum (BP) and General Electric monitor oil wells using ML to augment performance.[124]

With enhanced computational power and advanced algorithms, AI, particularly machine learning (ML), has gained significant traction in recent years. Tomorrow's tools for scale up will rely heavily on AI. As AI (especially ML and deep learning) proliferates in nearly all disciplines of science and technology,[125−129] including chemical engineering,[123,130] fluid mechanics[131,132] and multiphase flow,[133−136] AI is expected to lead significant changes in the research and development of gas−solid fluidization, specifically in data analysis, generative equipment design, flow sheet synthesis, modeling, and even risk analysis.

ML, a subfield of AI, focuses on developing algorithms and statistical models that enable computers to learn and make predictions or decisions without being explicitly programmed to perform a specific task. In other words, it is a computational approach to teach machines how to learn and improve from data, allowing them to recognize patterns, make decisions, and adapt to new information. No physical governing equation is needed, which cautions against use without sufficient domain knowledge.

The fundamental idea behind ML is to use available data sets to train a ML model, which learns to recognize patterns and relationships in the data and can then generalize from that data to make predictions or decisions about new, unseen data. The data sets typically include input features and corresponding target labels or outcomes. During training, the model adjusts its internal parameters to minimize the difference between its predictions and the target values. A separate data set is then used to validate the predictions of the first training set and the ML model. Common evaluation metrics include accuracy, precision, recall, and F1-score (i.e., model accuracy), depending on the specific problem. ML encompasses a wide range of techniques and algorithms, including supervised learning (whereby models are trained on labeled data) and unsupervised learning (whereby models discover patterns and structures in unlabeled data). Principle component analysis is a good simple example of unsupervised learning where data clusters are the product.

AI encompasses ML but has additional components such as reinforcement learning, deep learning, natural language processing, image recognition, expert systems, etc. Reinforcement learning uses agents to make decisions by taking actions in an environment and receiving rewards or penalties based on those actions. It is commonly used in applications like game-playing and robotics. Data are classified, the classification is tested, and the cycle repeats if it is incorrect. Deep learning is a specialized form of ML that uses artificial neural networks (ANNs) inspired by the structure of the human brain. Deep learning has been particularly successful in tasks like pattern recognition, data mining, image recognition, and natural language processing. Accurate physical models, together with supercomputing and the capability of handling big data, will enable the virtual reality of gas−solid fluidization technology.[137]

Data Analysis. Data analysis can now readily reveal multidimensional relationships and trends using ML tools such as self-organizing map (SOM), neural nets, and Gaussian process reduction.[138] For example, Patel et al.[139] used the ML tools of SOM and random forest (RF) to discern key parameters tied to macroclusters and streamers in a CFB riser with 1188 data sets. The data's multidimensional aspect precluded traditional statistical models from being useful. Notably, SOM revealed that broad particle size distributions or bimodal distributions may hinder the growth of macroclusters and streams. A similar study was done with bubble hydrodynamics in a fluidized bed of Geldart Group B powders, further revealing the importance of polydispersity.[140] Fu et al.[141] used an ANN model to predict the best conditions (i.e., pressure drop and expansion ratio) for fluidized bed reactors. Kim et al.[142] used RF and ANN to optimize syngas production from a fluidized bed biomass gasifier. A similar approach was employed by Lian et al.[143] for the production of hydrogen from a fluidized bed reactor. Indeed, ML methods for process monitoring, fault detection, and soft sensing are already commercially implemented.[144] In short, ML tools could very well highlight key scale-up relationships missed by a more traditional data analysis process.

Design of Experiments. The first step of this stage is to get reactor data such as productivity, extrinsic kinetics, bed density profile, entrainment rates, and attrition rates. It would also be advantageous to characterize the unit with respect to the bubble hydrodynamics and the gas residence time distribution. Although the sizes of fluidized bed lab-scale units are typically too small to contribute to realistic scale-up parameters, it is advisible to have such characteristic properties to relate the finding of the lab-scale unit with future larger test units. It also provides a higher level of validation data for any mathematical models. Test factors include varying temperature, pressure, feed concentration, and superficial gas velocity, and could involve 100s of experiments in a full factorial configuration. Design of experiments (DOEs) can reduce the number of experiments needed while still maintaining statistical significance with a predetermined confidence level (e.g., 95%). However, new DOE tools can make that easier and faster.

AI-assisted DOE is available today. A Latvian start up has developed an artificial intelligence-driven Design of Experiments (DOE) software called xT SAAM.[145,146] This adaptive DOE with deep learning assesses the data as it is being

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collected, instead of having all the experiments completed before such an assessment is performed. The result is a faster turnaround without compromising the data integrity. In short, the process development stage uses AI to augment solutions around previous obstacles. Leveraging AI-directed DOEs, experiments can now be more focused and require fewer resources.[147,148]

Generative Equipment Design. Using the optimization algorithms with AI, components can be designed and then tested with a model. AI is even being used to "simulate" kinetic and equilibrium patterns even if enough data is not available to develop a kinetic model.[149] A design engine starts with a CAD which is then submitted to a CFD or CFD-DEM model to calculate the grade efficiency curve and corresponding solids loss rates and the cyclone pressure drop. If those results meet the predefined objective function, a new design is generated by the AI engine. One such engine is Siemens' Simcenter HEEDS, which can interface with just about any software, thereby allowing for automation. In addition, HEEDs uses a hybrid-adaptive search framework (SHERPA) that can distinguish local versus global maxima or minima, something gradient-based methods are less effective at. Dow and Siemens used such a methodology for the design and operation of a distillation column.[150] Similarly, Roach and Eldridge from University of Texas with Barsotti of Siemens PLM used HEEDS for the optimization of a dividing wall distillation column.[151] Mihailova et al.[152] used Star CCM+ with HEEDS for optimizing the design of a helical ribbon mixer, results of which were validated with electrical resistance tomography.

For lower-velocity fluidized bed design, CFD integrated with a HEEDS or similar optimizer could address the design of the gas distributor, liquid injection points, bed internals, and cyclone. For circulating fluidized beds, such efforts would be beneficial with aeration strategies, especially with standpipe design and operations.[153]

Flowsheet Synthesis. Flowsheet synthesis accelerates the development of a process flowsheet using AI.[123] What traditionally takes weeks can be done in hours, thereby allowing a flowsheet to be developed early in the scale-up process using a minimal amount of data. Using a generative flowsheet transformer, a simple but incomplete flowsheet can be readily optimized into a completed process flow diagram, as illustrated in Figure 8.[154] The Aspen Hybrid Modeler is a good example of this capability. In 2021, Dow and Siemens PLM coupled ASPEN Plus with HEEDs to optimize condensation

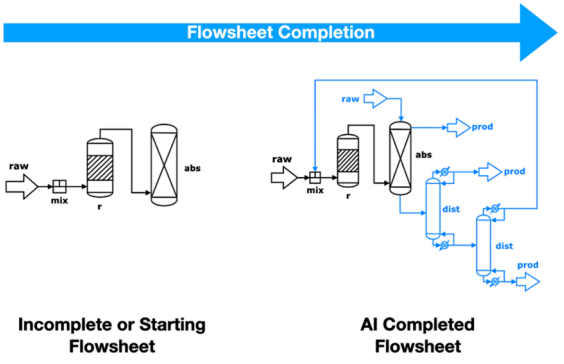

Figure 8. Flowsheet completion with the generative flowsheet transformer. Adapted with permission from ref 10. Copyright 2021 Wiley.

polymerization reactors that produce polydimethylsiloxanes (PDMSs for sealants, adhesives, coatings, and emulsions.[150] Such an exercise is even more critical for fluidized bed processes, which can have less flexibility than the more traditional reactors (e.g., superficial gas velocities, turn-downs, and particle emissions).

Thus, even in the conceptional stage, a detailed flowsheet can be obtained for mass and thermal flows. This useful tool is available to explore several strategies that could affect the fluidized bed reactor design, performance, turn-down, emissions, etc. at this early stage and illuminate gaps, limits with systems integrations, and limits with reliability. Such information is valuable for the first-stage estimation of economics along with subsequent economic evaluations.

Risk Assessment Process. The Risk Assessment Process (RAP) stems from Quantitative Risk Assessments (QRA) with the quantifying environmental and safety risks. However, QRA has been leveraged toward process risks as related to reliability and asset management.[155] The failure mode probability for each component is documented within the team and perhaps with external experts. The likelihood of failure and the consequence of that failure is quantified within the team. It is important that this process has a systems integration approach whereby how the failure affects up- and downstream performance is considered. For each operation, a matrix of probabilities can be used in an AI Bayesian network model to quantify the process reliability.[156] AI can add anomalies to the model, further test the perceived risks, as well provide advanced analysis to understand the underlying causes of any anomaly.[157,158] This exercise will define weak links in the process flow and equipment design. With RAP being done early and frequently, any weakness can be addressed much earlier in the scale-up path.

Assisted CFD. In a book chapter divining CFD for 2025 and beyond,[58] it is stated that deep machine learning will be used to improve the speed, accuracy, and user-friendliness of CFD software and be routinely used to generate digital twins/reduced order models that will profoundly impact the utilization of CFD.

Constitutive equations needed to capture the microscale physics for CFD can be refined with AI-directed experiments and experimental fitting.[159] With the state of technology today, developing the constitutive equations and the parameters for those constitutive equations will likely result from well-tailored experiments coupled with AI or, at the least, the ML part of AI. Sundaresan et al.[133] suggested that the scale dependency of multiphase flows can be quantified through deep learning methods to improve constitutive models for momentum, species, and energy transfer. Furthermore, Sundaresan[98] noted that modeling efforts would start with the experimental measurements of the PSD along with proxy experiments for the calibration of constitutive equations. Direct measurements of the actual particulate system may not be resource-effective. For instance, fluidization and defluidization tests have been used to calibrate drag models.[160] Rheological and flow testing can be used to validate the collisional stress model.[93,161]

Jiang et al.[136] gave an example using a neural network ML tool for optimizing a filter drag constitutive model for fTFM (filtered two-fluid model), which for gas-particle flows require closures for the subfilter scale corrections to interphase drag force and stresses, the former being more significant. It allowed them to verify that an algebraic drift flux model can be used to capture the micromixing observed in the highly resolved, fine-

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grid simulations. Yang et al.[162] coupled neural network with the EMMS drag model for predetermining the heterogeneity index. Similarly, Lu et al.[163] used ANN in PyTorch coupled with MFiX to develop a filtered drag expression.

The time step and grid resolution can be made more adaptive with AI, as proposed by Lorsung and Fairimani.[164] Existing methods for adaptive meshing need additional functionality out of solvers and/or many training simulations. Their Mesh Deep Q Network (MeshDQN) is designed as a general-purpose deep reinforcement learning framework to coarsen meshes while preserving target property calculation iteratively. A neural network-based deep Q network (deep learning neural network) is used to selectively remove mesh vertices, while solution interpolation bypasses costly simulations at each step during the improvement process. MeshDQN needs one simulation before mesh-coarsening without any assumption about flow regime, mesh type, or solver, and only requiring the ability to modify meshes directly in a CFD pipeline.

AI Reactor Models. Mutlu and Yucel[165] employed AI methods to predict syngas composition from downdraft biomass gasification. The conclusion that the temperature distribution is more influential than fuel parameters informs that optimizing syngas composition relies on controlling the temperature well. Krzywanski et al.[166] developed AI-based models for hydrogen production through calcium oxide (CaO) sorption in two types of gasifiers, namely, bubbling fluidized bed (BFB) and circulating fluidized bed (CFB). The results indicate that CFBs are more effective than BFBs, and the developed model can be further used for optimization. In both cases, the use of AI significantly reduced the development and possible optimization of a preliminary process model.

The question here is, how can AI play a role in speeding up the scale up of fluidized beds? Purely data-driven models may fall short because chemical engineering systems are governed by fundamental laws and principles, which are valuable in making sense and imposing rigor on data-driven models.[30] Also, the large amount of high-quality fluidization data needed for these black-box models may not be practical. To address this, gray-box physics-informed ML and AI have been quickly gaining traction.

PINN (Physics-Informed Neural Network) Reactor Models. AI models are black boxes that work according to a regression of the input factors to an unspecified, hidden layer or layers of weights. The model results are only as good as the quantity and quality of the data used in the training and testing sets. In a way, ANN may be too good at approximating functions that they can overfit the training sets. As a result, for them to be effective at generalizing and not just learning the training set, one needs lots and lots of data as well as some clever tricks such as batching.[167] To address this shortcoming, PINN regularizes a neural network to conform to physics by having both parts contribute to a loss function, as depicted in Figure 9. An additional layer is added that relates to the expression for the physics.[168] Essentially, the physics layer helps the neural network function have the right physics-governed trend. To do this, the ANN is embedded with information in the form of a differential equation. For example, if a model was being developed for a convection−diffusion problem using the Burgers' equation, that extra layer would consist of nodes for the time gradient ($\frac{\partial}{\partial t}$), the axial gradient ($\frac{\partial}{\partial x}$), and the gradient of the axial gradient ($\frac{\partial^2}{\partial x^2}$).[169]

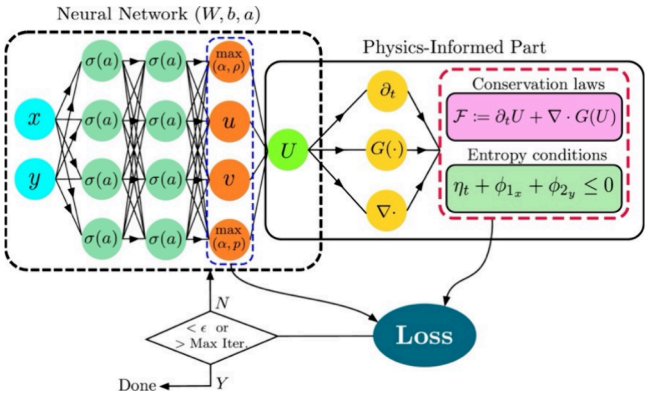

Figure 9. PINN construct for the Euler equations. The neural network is hybridized to satisfy laws of conservation and entropy conditions. Reproduced with permission from ref 170. Copyright 2022 Elsevier.

A similar approach can be taken with the Navier−Stokes equations. Sun et al.[171] used PINN to model and optimize the flow around an airfoil to maximize the lift to drag ratio. The parameters related to the airfoil shape were input to the PINN, and the multidimensional search space of shape parameters was populated with collocation points to satisfy the Navier−Stokes equations as much as possible throughout. Amalinadhi et al.[172] used PINN for solving the velocity, pressure and stress tensor for Poiseuille, Couette, and lid-driven cavity flows. Eivazi et al.[173] used PINNs to model incompressible turbulent flow without a specific turbulence model. They only used the data from the boundary domain and reported good accuracy even for the Reynolds stress component.

Qiu et al.[174] applied PINNs to two-phase flows. The Cahn−Hillard equation and Navier−Stokes equations were encoded directly into the residuals of a fully connected neural network. Lu and Christov[175] used PINNs for modeling particle migration in a non-Brownian suspension from Couette flow, with results revealing that the inferred values of the empirical model's parameters vary with the shear Peclet Number as well as particle bulk volume fraction of the suspension.

All these applications of PINNs suggest that such a model may soon be possible for the hydrodynamics and reactions in fluidized beds. At the very least, PINN-based models are possible for the constitutive equations used with granular-fluid CFD or CFD-DEM models. The benefit would be a more robust and accurate drag and collisional stress model while relaxing the grid and time step resolution needed for the more traditional models. Also, before the possibility for accurate quantitative descriptions of the various fluidization phenomena, PINN promises to supersede the current empirical correlations that have been known to make predictions that are different orders of magnitude (e.g., entrainment[48]). In particular, symbolic regression hybridized with genetic programming has been shown to provide analytical equations from data.[176]

CONCLUDING REMARKS

The vast resources and time needed to scale up fluidized beds represent a key obstacle in tackling time-critical sustainability challenges. AI with other modeling efforts (ROM, PBM, CFD, CFD-DEM, generative design, flowsheet synthesis, risk analysis process) offer invaluable tools to circumvent this limitation. While many of these tools may already be in place for some

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recent scale-up projects, such tools are not used typically, used too infrequently, or only used in a reactive setting. This restricts the effectiveness of the tools, and scale-up decisions remain primarily based on traditional methods merely because this has been the way for decades. For instance, many scale-up programs for lower-velocity fluidized beds or circulating fluidized beds rely on large-scale cold-flow experiments instead of more well-tailored, lab-scale experiments coupled with effective modeling efforts. Often, these cold-flow models are only implemented after an unexplained result in the mini plant or pilot plant, if not even later. This can significantly compromise the economics of the project. Large cold-flow experiments take months (6−12 months) and are expensive (> $500 MM), plus a wide range of physics can be convoluted in the data collected. Specific physics, such as wall stresses, shear stresses, interparticle forces, wetting, attrition, etc., are difficult to capture in such units, limiting the information obtainable with such a big time and financial investment.

A more systematic approach whereby experiments are integrated with models to capture specific physics should be used, with the results subjective to additional flowsheeting, risk analysis, and financial analysis. This will provide a higher confidence level with respect to the unit and process model, provide better understanding of the dominant physics involved, and lay the foundation for process control (i.e., feedforward, real-time optimization, AI) and troubleshooting for the commercial plant. This is not to say that large cold flow experiments are not valuable; it is just that the value-add may not be as significant as well-tailored, lab-scale experiments coupled with AI (including ML), ROM, PBM, CFD, DEM, generative design, flowsheet synthesis, risk analysis process tools. Much of this is already part of many scale-up programs, but these modeling efforts need to be formalized early in the project and managed proactively, not reactively. These advanced tools were not readily available before, but they are now well within reach to significantly expedite the scale up of fluidized beds for today's urgent sustainability goals.

Leveraging the advanced tools that have become readily available, a companion part 2[15] aims to put forth a new pathway for scale-up to enable confident exploitation of the benefits of fluidized beds more swiftly and economically.

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Notes

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