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Analytical Multi-Scale Methodology for Fluidization Systems - Retrospect and Prospect

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ANALYTICAL MULTI-SCALE METHODOLOGY FOR FLUIDIZATION SYSTEMS — RETROSPECT AND PROSPECT

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ABSTRACT

Understanding the spatio-temporal multi-scale structure of fluidization is a challenging problem. This presentation reviews our 20-year efforts on this subject, showing the roadmap that has gradually evolved from a simple idea to a systematic methodology inclusive of subsidiary, related systems and industrial applications. The strategy of establishing stability conditions through analyses of the compromise between dominant mechanisms is emphasized. The presentation concludes with prospects for further theoretical explorations and industrial applications.

1. **Understanding spatio-temporal multi-scale structures is not only a challenge for fluidization engineering, but also an opportunity to contribute to complexity science:**

Gas-solid fluidization systems are non-linear and non-equilibrium in nature and feature spatio-temporal multi-scale structures, showing alternately a solid-rich dense phase and a gas-rich dilute phase at any local point and the co-existence of these two phases in space at any instance. Globally, however, a dilute region and a dense region co-exist in both the radial and the axial directions, leading to the overall heterogeneity in these systems. In addition, state bifurcation in fluidization systems can occur under certain critical conditions, showing a sudden change in structure with a slight change in operating conditions. This is commonly known as “choking” in engineering.

Due to the crucial influence of structural changes on transport properties and reaction processes, quantification of these structural changes has become a core task for our fluidization community. However, theoretical prediction of these changes remains undeveloped, and contradictions continue to exist between different attempts due to insufficient understanding. Particularly, the stability condition for the heterogeneous structures and the critical condition for choking have long been perplexing, since a general and single variational criterion is not available for nonlinear and non-equilibrium systems. Therefore, new concepts need to be introduced. This represents an opportunity for our fluidization community. Our investigations of this problem have shown that any progress on this *particular* subject of fluidization has provided, at the same time, a paradigm and a driving force for research on complex systems in *general*.

2. **Stability and critical conditions are the key to under- standing spatio-temporal multi-scale structures, which are both subject to the compromise between dominant mechanisms and the correlation between different scales. These are also important topics in complexity science:**

Our study on spatio-temporal multi-scale structures using the multi-scale strategy started in 1984 when the first author came to Prof. Kwauk for Ph. D program (1). First, we believed that traditional average approaches were not sufficient for understanding structures, and micro-scale analysis for a whole system was not realistic at that time. Therefore, we selected the multi-scale strategy between the average and the micro-scale analyses, aiming at capturing the dominant features of fluidization systems — co-existence of a gas-rich dilute phase and a solid-rich dense phase. That is, the effect of particle clusters on particle-fluid interaction was taken into account as a new entity of significance in the study of structure, as shown in Figure 1.

To describe the two-phase structure in fluidization systems, 8 parameters were required, from which only 6 equations could be formulated, which would result in multiplicity apparently. Therefore, we had to look for some stability condition or something equivalent to provide a closure and define the steady state of the system. This was a real challenge for us primarily because fluidization systems with multi-scale structures are featured by a non-linear and non-equilibrium behavior, which could not be described by theories available at that time.

In looking for solutions, we recognized that both particles and fluid had to compromise with each other in realizing their respective extremum tendencies. This situation is totally different from linear non-equilibrium systems dominated by only a single mechanism, thus making the stability of non-linear and non-equilibrium systems more complicated.

In order to provide closure, we postulated two new relationships. The extremum tendency of the fluid was represented by the minimization of volume-specific energy consumption for suspending and transporting particles, W_{st}=min, while that of the particles was represented by the minimization potential energy, ε=min. Stability of the two-phase system calls for mutual compromise, as much as possible, between the fluid and the particles in following their respective tendencies, that is, $W_{st}=\min|_{\varepsilon=\min}$, leading to the establishment of the stability condition, N_{st}=min, where N_{st} is the mass-specific energy consumption for suspending and transporting particles, that is, $N_{st}= W_{st}/(1-\varepsilon)\rho_p$. In fact, this stability condition also defined the correlation between the micro-scale interaction and the meso-scale interaction, leading to a closure of the hydrodynamic description consisting of 6 equations, and providing a critical element of the local energy-minimization multi-scale (EMMS) model, as shown in Figure 1. Numerical solution of the EMMS model can yield the 8 parameters under specified operating conditions (2).

Although this particular compromise between the fluid and the particles was not fully understood and the physical essence of N_{st}=min was not revealed to a convincing extent in the early stage of our work, a mathematical analysis of the EMMS model at

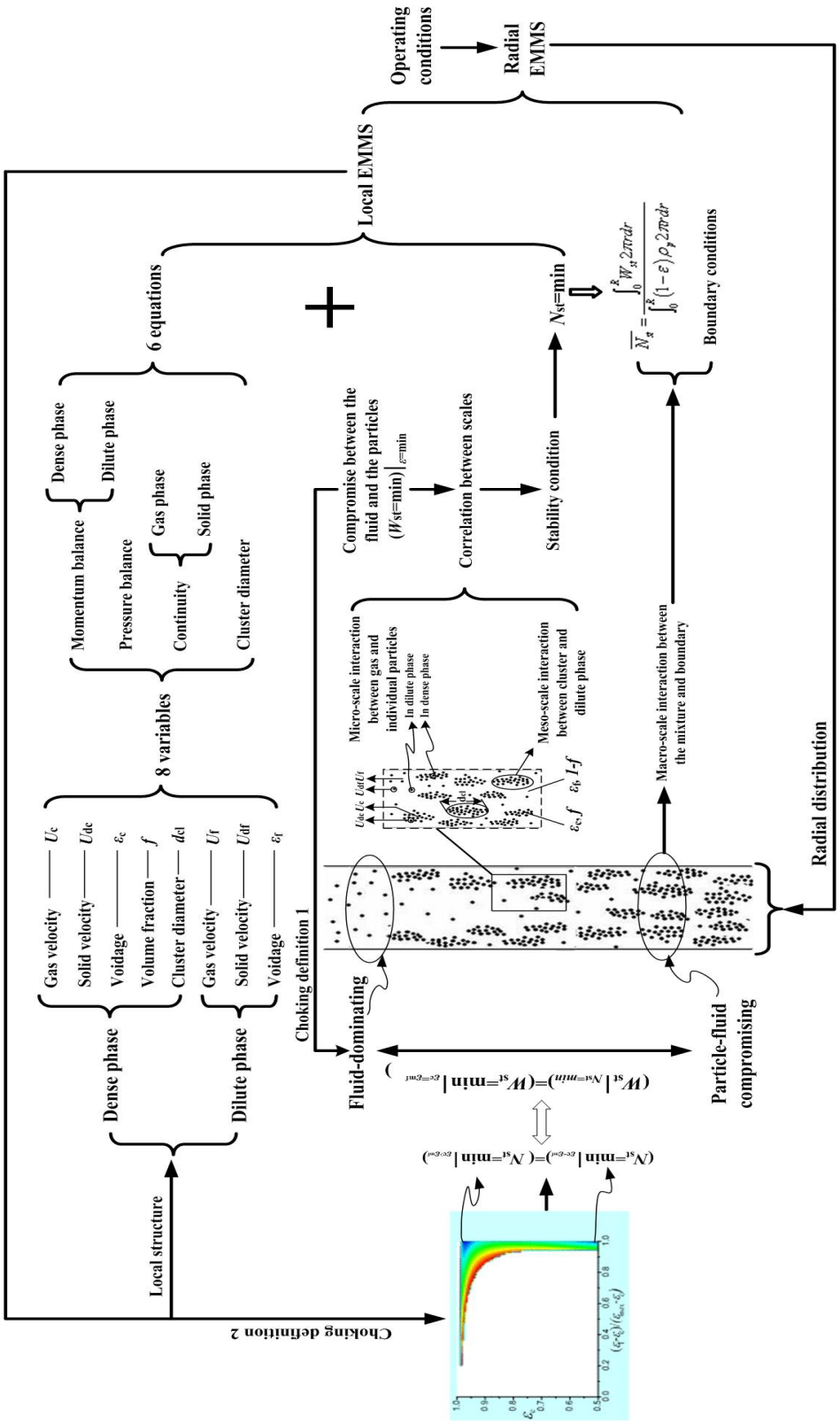


Fig. 1 Diagram illustrating the framework of the EMMS model

that time indicated that N_{st} =min corresponded to a heterogeneous two-phase flow structure consisting of a particle–rich dense phase and a gas-rich dilute phase, well known from fluidization experiments, thus giving us confidence in the potential validity of N_{st}=min. Following the preliminary exploration of the EMMS model, the first author studied gas backmixing in different flow regimes of gas-solid fluidization at Prof. H. Weinstein's Lab of CCNY, New York, and measured the dynamic changes of heterogeneous structures in fluidized beds using optical probes developed at Prof. L. Reh's Lab of ETH, Zurich. The alternating changes in voidage between two approximately constant values verified by these experimental studies (3), furthered our belief in the rationality of N_{st} =min. The down-flow of the dense phase near the wall and up-flow of the dilute phase at the center of a fluidized bed (4) sparked the extension of N_{st}=min to $\overline{N_{st}} = \int_0^R W_{st} 2\pi r dr / \int_0^R (1-\varepsilon)\rho_p 2\pi r dr$ as the stability condition for radial distribution, leading to the formulation of the radial EMMS model based on a two-fold optimization for calculating the radial distribution (5), as shown in Figure 1. The solution of the radial EMMS model showed a maximum gas-solid slip velocity near the wall. Later on, Tanner, Li & Reh (6) detailed this research by carrying out many more case studies.

Success in applying N_{st}=min for local structures and $\overline{N_{st}} = \min$ for radial heterogeneity encouraged us to tackle the choking phenomenon where the concept of *particle-fluid compromise* is extended to two extremum cases: *particle-dominating* and *fluid-dominating.* That is,

- ***particle-dominating (PD)*** when the fluid cannot induce the movement of particles, as in fixed beds, or inside a dense-phase cluster;
- ***fluid-dominating (FD)*** when the fluid flow acquires sufficient force to move the particles to follow its movement tendency, as in dilute transport, or in the dilute-phase broth;
- ***particle-fluid compromising (PFC)*** when neither the fluid nor the particles can dominate the other in displaying their tendencies, as in fluidization.

The analysis of these three different mechanisms of particle-fluid interaction led to the establishment of the critical condition for choking which occur when the fluid gains the capability of destroying the two-phase structure and reforming it to the fluid-dominating state with a more dilute and uniform structure (2, 7). At this condition, with increasing fluid velocity, the W_{st} (whose minimum features the fluid movement tendency), corresponding to the fluid-dominating interaction, just decreases to the value corresponding to the particle-fluid- compromising state (N_{st}=min), that is

$$(W_{st} = \min)_{N_{st}=\min} = (W_{st} = \min)_{\varepsilon_c=\varepsilon_{mf}}, \qquad (1)$$

as indicated in Figure 1 by "choking definition 1".

At this critical condition, two different states, fluid-dominating and particle-fluid compromising, can co-exist in a system if operating conditions allow. Beyond this condition, the system shows only the fluid-dominating state, as shown in Figure 1. However, this definition is difficult to understand for those not deeply engaged in such studies. Later, we realized (8), not without surprise, that this critical condition was, in fact, already implicit in the original EMMS model. That is, if we solve the EMMS model numerically, we obtained at the critical condition two solutions with the same value of

N_{st}, shown in Fig. 1 as "choking definition 2", and giving the same choking point as Equation 1. We had not recognized this important feature of the EMMS model, due to the limitation of numerical tools used in identifying multiple minima of N_{st} in the early stages of our work. That was a pity, but also fortunately, a convincing reflection of the physical consistency of this model, because we had now arrived at the same result from two independent directions.

3. **N_{st}=min represents something different from stability conditions for equilibrium and linear non-equilibrium systems. Theoretical support or proof is not available, making N_{st}=min an interesting puzzle. We have spent great efforts to solve this puzzle in the last two decades:**

Between 1992 and 1995, the EMMS research was slowed down — On the one hand, we had no clear idea as to how to continue the work, and on the other, we could not theoretically and satisfactorily answer the question as to why N_{st}=min should be valid, primarily because of the lack of available theories. In fact, our understanding of N_{st}=min was limited to the consideration of both the particle movement tendency and the fluid movement tendency. There was no clarification of the spatial and temporal scale on which the stability condition is applicable. Moreover, the inclusion of "energy-minimization" in its name (EMMS) also led to unfortunate confusion regarding this concept since N_{st} represents a kind of energy consumption rather than energy. To clarify this confusion, we made the following efforts:

- We wrote a monograph to summarize the EMMS concept (2), in which we clarified the importance of the compromise between dominant mechanisms, leading to the exploitation of the same strategy in analyzing other systems, such as single phase turbulent flow. Up to now, we have successfully studied 6 different systems using the same strategy.
- Given the impossibility of theoretical verification of N_{st}=min, we proposed to verify it by discrete computer simulations. In 1994, discrete simulation was started by proposing the so-called pseudo-particle model (9, 10). That is, the fluid was treated as pseudo-particles, each with a diameter much smaller than that of real particles so that the interaction between fluid and particles was transformed into the interaction between real particles and pseudo-particles, making it possible to generate macro-phenomena by micro-analysis and to calculate the value of N_{st} directly. At the beginning of verifying N_{st}=min, the progress was slow due to limited computer capacity. Some preliminary results were obtained first (11), indicating that the heterogeneous structure was formed only when two dominant mechanisms compromise with one another. Though these results were only qualitative, it gave us confidence and encouragement. Since 2002, computer capacity and computational capability have improved significantly at our lab, facilitating the verification of N_{st}=min. In 2004, N_{st}=min was successfully verified (12, 13), removing the confusion with regard to the EMMS concept, encouraging its extension to other complex systems, and even facilitating explorations of its generality. At the same time, powerful computational capability was established to study micro-mechanisms and to solve industrial problems.
- Application of the EMMS model was extended. First, the drag coefficient in heterogeneous structure was analyzed to tackle the difficulties in calculating the drag coefficient in CFD. It was concluded that the drag coefficient in the dense phase is several orders larger than in the dilute phase, and that the average

treatment of the structure leads to overestimation of drag coefficient. These results were published in 1993 (2, 14), but their relevance to CFD calculations was realized only recently due to the complexity of solving the EMMS model, as observed by some of our international colleagues. To facilitate the application of the EMMS model, we made simplifications (8), and the solver is now available online (http://pevrc.ipe.ac.cn/emms/emmsmodel.php3). These efforts promoted its application in CFD and in solving industrial problems, as discussed below.

4. **Theoretical validation of N_{st}=min is not possible due to the lack of a single and general variational criterion for non-linear and non-equilibrium systems. However, discrete simulation has paved the road to verify N_{st}=min, leading to the conclusion that N_{st}=min is valid for a volume of sufficient size but not valid for a point, due to the compromise between the movement tendency of the fluid (W_{st}=min) and the particles (ε=min):**

Preliminary results (11) showed the critical role of the compromise between two dominant mechanisms in the formation of multi-scale structures, but was not sufficient to verify N_{st}=min since the value of N_{st} could not be calculated at that time. The progress of discrete simulation and the increase of computation capability made the quantification of N_{st} possible (12, 13), leading to the confirmation that N_{st}=min and the understanding of its mechanism.

In complex systems dominated by two mechanisms, each mechanism tends to realize its own movement tendency. However, due to the constraint between the two, their extremum tendencies cannot be simultaneously realized at a point, and they have to compromise with each other with respect not only to time, but also to space. That is, any point in space shows an alternance of the two states dominated exclusively by one of the two, showing time compromise, whereas throughout space at large, these two states can co-exist, but only at different points, showing spatial compromise. It is such compromises that lead to spatio-temporal multi-scale structures in complex systems for which stability exists only for a large enough volume, but not for a point. For instance, in a gas-solid system as simulated, $N_{st}=W_{st}/[(1-\varepsilon)/\rho_p]$ (W_{st}=min featuring the movement tendency of gas, and ε_{min} that of the particle). At a local point, W_{st}=min and ε_{min} cannot be realized simultaneously, so that, N_{st} cannot be a minimum at this point, but instead shows fluctuations, as indicated in Figure 2. However for a volume, due to the compromise in space between W_{st}=min and ε=min at different points, it is possible that N_{st}=min. With increasing size of the volume from Region D to Region G, the extremum tendency of N_{st} becomes more and more obvious. Therefore, we confirmed the correctness of N_{st}=min for a volume and identified its non-existence at a point. This represents very important progress not only for the development of the EMMS model, but also for understanding the stability criteria of other complex systems.

5. **The analysis of the compromise between the extremum tendency of gas and that of particles in gas-solid system was next applied to 5 other systems, leading to the extension of the EMMS model to a general analytical multi-scale methodology following the strategy of defining, first of all, the stability condition of complex systems by analyzing the compromise between dominant mechanisms so that phenomena of different scales can be correlated. This methodology was conceptually formulated into a multi-objective variational problem:**

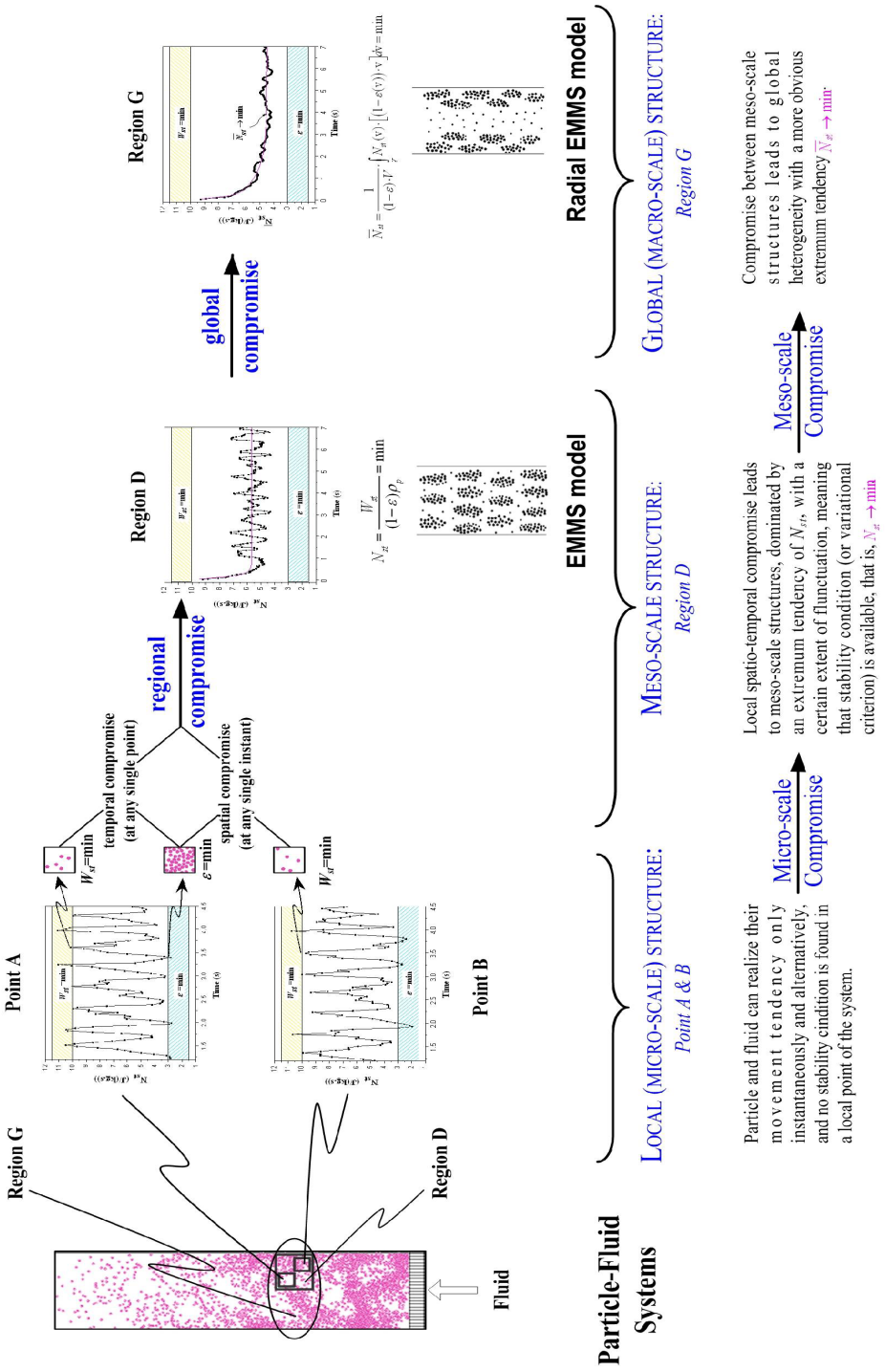


Fig. 2 Spatio-temporal compromise between dominant mechanisms (15).

The first extension of the EMMS model was to turbulent pipe flow. The results indicated that the radial distribution of fluid velocity in a pipe is subject to the compromise between viscosity and inertia (16). In combination with the simulation results on two interacting discrete granular media, indicating that a heterogeneous structure can appear only when these two media compromise with each other (17, 18), we recognized that stability conditions are specific to different complex systems due to difference in dominant mechanisms. However, there is indeed a common feature that stability conditions are dominated by a compromise between dominant mechanisms, leading to spatio-temporal multi-scale structures. Variational criteria exist to characterize such compromises and to express the stability conditions.

Therefore, we decided to explore this common feature in more complex systems. Along with the physical analysis of the compromise between different dominant mechanisms in these systems, we carried out simulations to reveal the importance of the compromise and to explore the generality of the existence of variational criteria in these systems. The results are summarized in Figure 3, indicating that these six systems are all dominated by two different mechanisms which compromise with one another. They show a common feature that there is no stability conditions at a point, but system stability for a volume with a sufficient size indeed exist. Therefore, the EMMS model for gas-solid systems was conceptually extended to an analytical multi-scale methodology for complex systems. The analytical multi-scale methodology in Fig. 4c is summarized in comparison with other multi-scale methods, descriptive in Fig. 4a and correlative in Fig. 4b.

The closures of the description of different scales for three types of multi-scale methods follow different strategies. That is, the descriptive method pays no attention to the correlation between scales. The correlative method makes the closure by inputting the lower-scale analysis into the higher-scale formulation without considering the stability of the structure. The analytical method resulted in closed descriptions of multi-scale structures by establishing corresponding stability conditions. This methodology can be formulated into a multi-objective variational problem (19):

For complex systems defined by n variables $X(r,t)=\{x_1, x_2, \cdots, x_n\}$ *and dominated by k mechanisms, assume that the rate equations can be formulated by a set of m functions of* $X(r,t)$*, that is,*

$$F_i(X)=0,\quad i=1,2,\cdots,m \qquad m<n$$

and the extremum tendencies of these dominant mechanisms are expressed as

$$E_j(X)=\min,\quad j=1,2,\cdots,k$$

Then the steady state of the system could be formulated, conceptually as:

$$\min \begin{pmatrix} E_1(X) \\ \vdots \\ E_k(X) \end{pmatrix}$$

$$s.t.\ F_i(X)=0,\ i=1,2,\cdots\cdots,m$$

This is a multi-objective variational problem challenging even mathematicians. As a conceptual model to guide our analysis on complex systems, its detailed formulation and solution require further efforts. It is evident that the EMMS model is

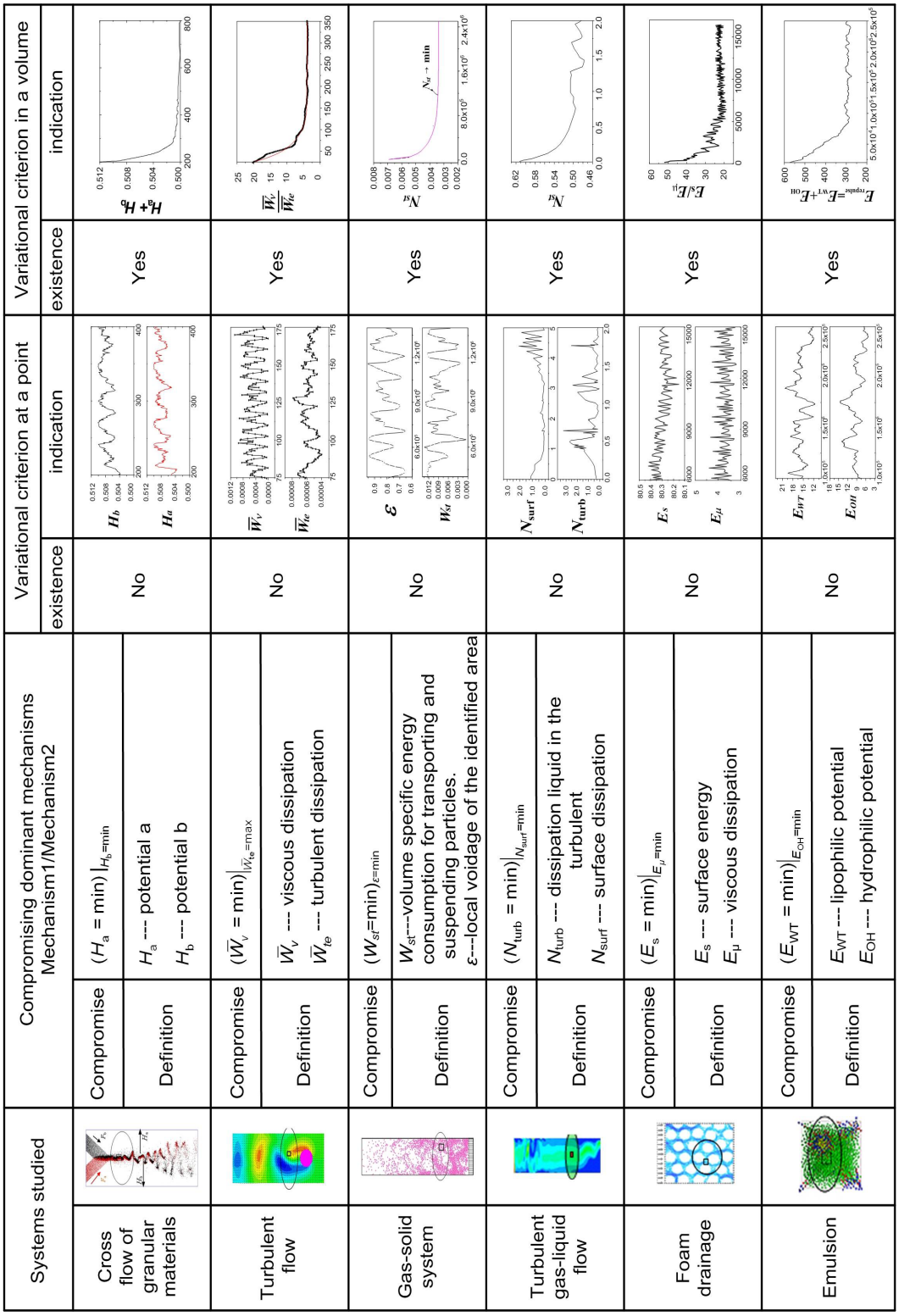


Fig. 3 Compromise between the dominant mechanisms and indications for the existence of stability conditions for 6 different systems(the horizontal axes represent time steps, modified from the work of Ge et al. (20))

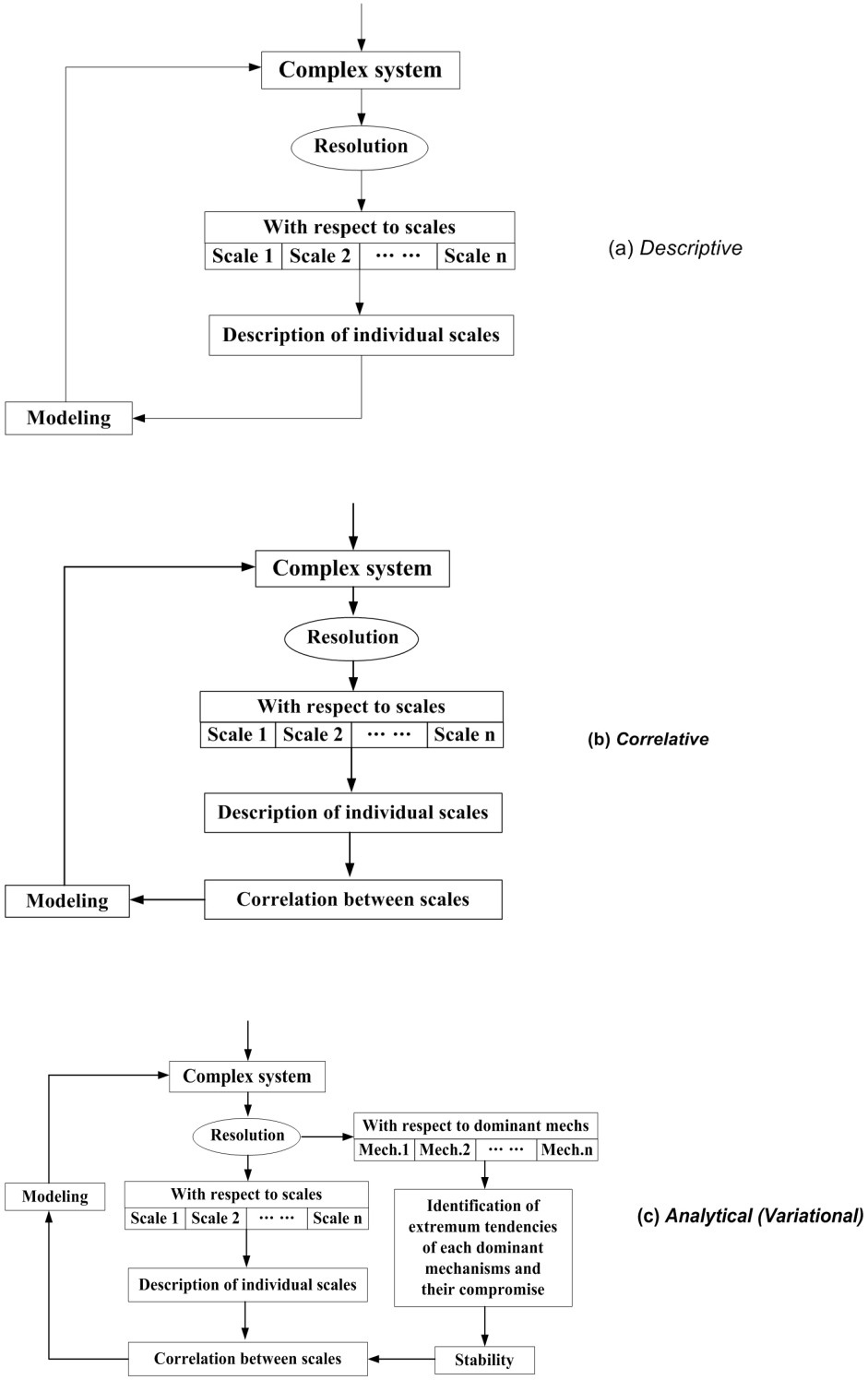


Fig. 4 Three different types of multi-scale methodology (21)

a simplified case, of the multi-objective variational problem, with two dominant mechanisms and without considering changes over time.

6. **Understanding spatio-temporal multi-scale structures provides an effective tool for calculating the drag coefficient in such heterogeneous structures, and facilitates CFD simulations for industry:**

To describe the local deviation from global behavior of fluidization, the original EMMS model was extended to include an acceleration term (8, 22), making it possible to couple the time variation and the space variation. Recent efforts even extended the EMMS framework to the sub-grid level (23). With the global searching scheme of the EMMS model (8), the drag coefficient in a CFD control volume can be approximately calculated. This can be used to replace the C_D correlation used in current commercial codes, such as Fluent and CFX as shown in Figure 5. Mass transfer has also been studied with the multi-scale strategy (24), which is a step forward to integrate a multi-scale mass transfer model into CFD.

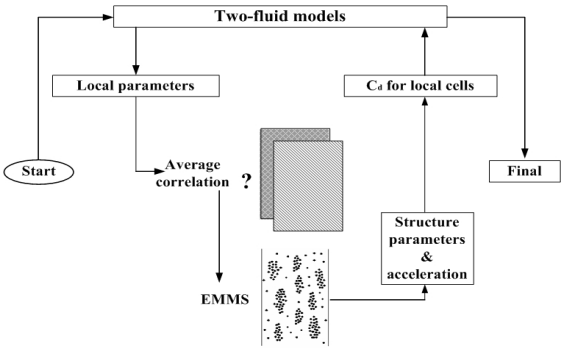


Fig. 5 Incorporating the EMMS model into two-fluid models

The integration of the EMMS model into CFX and Fluent enabled the computation to capture meso-scale structures as shown in Figure 6 (22, 23), and to reproduce choking and the co-existence of the top dilute and the bottom dense regions, as shown in Figure 7 (25). The choking region in Figure 7(a), the bell-shaped area, is hard to be clearly defined, as experimental data is scarce. It should also be noted that the experimental data beyond the abscissa limit of 14 is omitted, to better compare the results.

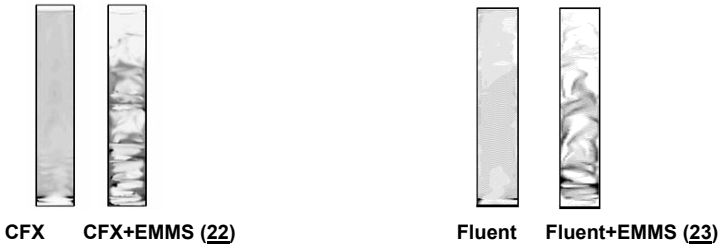


Fig.6 Comparison between snapshots from simulations of multiphase flows with commercial softwares alone and with incorporation of the EMMS model.

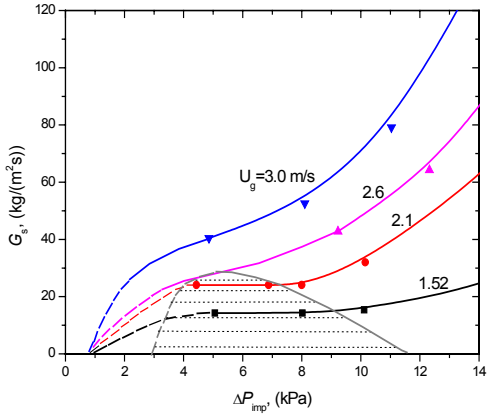
The prospect of industrial application for the stand-alone EMMS model and its integration with CFD software has been demonstrated by some successful attempts. The EMMS model has been successfully applied in the design of a novel FCC process for maximizing iso-paraffins (MIP), developed by Research Institute of Petroleum

Processing (RIPP), SINOPEC. The MIP process is capable of reducing olefin content and increasing the iso-paraffin content of gasoline to meet the new specification of gasoline with olefin content of less than 35 vol.%. The MIP reactor is a combination of a conventional FCC riser and a dense circulating fluidized bed, acting as two different reaction zones. The flow pattern inside the MIP reactor needs to be known before process design and there is no experience in designing such a reactor. At the request of SINOPEC, the flow patterns including axial and radial voidage profiles, pressure drop profile, and gas and solid velocities were calculated for given operating conditions (U_g, G_s), properties of gas and solid, and the reactor configuration. Optimal operating conditions and geometrical dimensions were determined from calculations. The commercial MIP process for a reactor of 50 m tall and 0.9-3.2m inner diameter was scaled up successfully by SINOPEC directly from lab-scale to industrial scale based on the parameters specified by the EMMS method. The ability to identify the chocking point through the EMMS model is the key to the successful design of this process. To some extent the EMMS simulation remedied the lack of experience in designing such a novel reactor with its unique geometry and facilitated the identification of the optimal operating conditions.

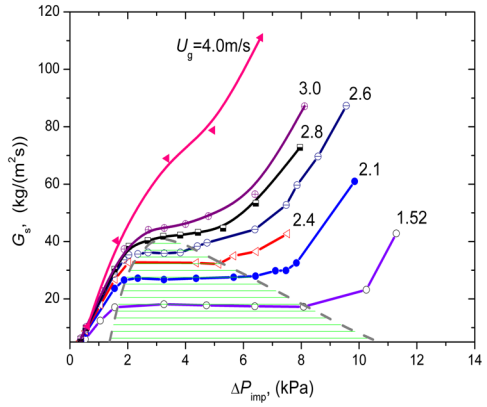
The successful application of the EMMS model in assisting the design of the MIP reactor has boosted the cooperation between IPE and RIPP. In a recent program for troubleshooting the MIP process, the CFD software (Fluent 6.2) incorporating the EMMS, was used to investigate the effects of operating conditions and geometrical configuration, (e.g. inlet/outlet and distributor geometry), on the flow behavior inside an industrial MIP riser. Simulation results indicated that the geometrical configuration had little effect on the density distribution of catalyst particles. It is the operating conditions, i.e. gas velocity and solids flux, that determine the flow behavior, including the onset of choking transition.

The EMMS model has also been extended to simulating the flow structure in a slurry bed loop reactor, developed by PetroChina for hydrogenation of residual oil. Without detailed knowledge of flow structure, it is difficult for engineers to choose the parameters in designing such reactor. The EMMS model has been adapted to predicting the equivalent bubble diameter, a key parameter for CFD simulation of two-fluid models. Then, the combined models were employed to predict gas-liquid flow for reactors of various sizes and with different internals, and therefore to provide useful guidelines for the optimization of reactor structures, such as how to avoid dead zones at the bottom of the reactor. The proposal about the shape of the bottom of the reactor based on our simulation results has been adopted by the engineers designing this reactor.

Although the integration of EMMS and CFD codes has significantly improved the calculation accuracy, the prediction of cluster diameters remains an unsolved problem. At present, we found that the original model is applicable to Geldart A particles, but not suitable for B, C and D particles. We have been making efforts to solve this problem. It is independent of the whole EMMS model, but closely related to the correlation of cluster diameter. The EMMS model should be, in principle, also applicable to dense babbling beds, which will be another area for future work.



(a)



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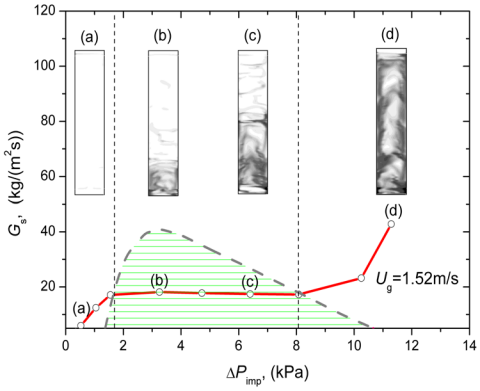

(c)

Fig. 7 Relationship between gas flow rate（U_g）, solids flux（G_s） and solids inventory（I) for different operating modes in an air-FCC particle CFB system (25). The abscissa variable ΔP_{imp} refers to the total imposed pressure drop across the riser. (a): experimental data after Li & Kwauk (2), the solids flux G_s is redrawn as a function of ΔP_{imp}; (b): simulation results; (c): simulated choking transition and representative snapshots of density distribution for different flow regimes.

7. **Our 20-year efforts in following the analytical multi-scale strategy have developed from a simple idea for gas-solid fluidization into the EMMS model and further to a more general framework for studying different complex systems —— the analytical multi-scale methodology. We are encouraged by its application to the solutions of industrial problems and by its potential to analyze multi-scale systems. As for the future, we feel continually challenged by unsolved problems and unanticipated difficulties. We are convinced, however, that the multi-scale methodology will play a more and more important role in dealing with routine problems as well as many spatio-temporal multi-scale structures in the future of chemical engineering:**

Progress in understanding multi-scale structures will be of substantial significance for chemical engineering science, presenting, at the same time, a challenge and an opportunity for us and calling for joint efforts from different disciplines. We believe that spatio-temporal multi-scale structure is a core problem for chemical engineering science in the 21st century, which may well be the essence of the so-called third paradigm of chemical engineering, as well as a focus for complexity science at large. Multi-scale structures present challenges everywhere from various nano-phenomena to mega ecology and from traditional scale-up of reactors to high-tech production processes. Therefore, while complexity science is claimed to be the science for the 21st century, multi-scale science could at least be the challenge of the 21st century! Transdisciplinarity is the best way to transform a challenge into an opportunity. Computation, which is effective in tackling various complex processes, should be structure-oriented. The advantage of increasing computer capacity could be made use of only when we focus correctly on the physical mechanisms of structure. We expect even more significant progress in this new era of science and technology since we have much more powerful computation facilities.

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