CATALYTICFOAM AND ISAT FOR THE EFFICIENT SIMULATION OF FIXED BED CATALYTIC REACTORS

MAURO BRACCONI[1], ALBERTO CUOCI[2], MATTEO MAESTRI[1]
[1]Dipartimento di Energia, Politecnico di Milano, via la Masa 34, 20156 Milano, Italy, mauro.bracconi@polimit.it
[2]Dipartimento di Chimica e Ingegneria Chimica, Politecnico di Milano, piazza Leonardo da Vinci, 32, 20133 Milano, Italy, alberto.cuoci@polimit.it
[1]Dipartimento di Energia, Politecnico di Milano, via la Masa 34, 20156 Milano, Italy, matteo.maestri@polimit.it

Keywords: *CFD, operator-splitting, ISAT, fixed bed reactor, heterogeneous catalysis*

Introduction

The worldwide demand for more efficient and sustainable exploitation of energy and material resources makes catalysis as one of the most relevant topics in the research agenda. Moreover, the tight targets on conversion, selectivity and stability under very broad operating conditions requires the development of new catalytic processes based on a deep understanding of the reacting system. The macroscopic behaviour of a catalytic reactor is intrinsically a multi-scale phenomenon, where the chemical events occurring at the catalytic surface depends on the local conditions of temperature pressure and composition, dictated by the transport phenomena of mass, energy and momentum. In this view, computational fluid dynamics (CFD), coupled with microkinetic descriptions of the surface reactivity, has demonstrated an unprecedented potential in the quest for the fundamental understanding and analysis of the chemical catalytic devices.

Recently, the catalyticFOAM [1] numerical framework has been proposed to efficiently solve the Navier-Stokes equations in general geometries with the accurate description of the surface reactivity by means of detailed microkinetic mechanisms. Furthermore, the framework has been extended to account for the intraphase transport within the solid catalyst, by splitting the computational domain in different regions, which are solved separately by considering the corresponding governing equations [2]. The numerical framework is based on the operator-splitting technique, which accounts, in separate fractional time steps, for the transport phenomena and the chemistry. The chemical sub-step is represented by a set of ordinary differential equations (ODEs), which represents the evolution of gas species, site densities and temperature. The system consists usually in 20-50 species involved in 80-100 reactions and might be coupled with homogeneous kinetic mechanism, which typically involves up to 200 species and thousands of reactions. The high non-linear and stiff ODE system has to be solved in each computational cell and every time step, strongly increasing the required computational effort.

A wide application of the envisioned approach is still hampered by the very high computational effort associated with the solution of the chemical sub-step. Indeed, most of the computational time is spent for the solution of ODE system associated to the description of the chemical sub-step. An efficient treatment of this sub-step is expected to strongly improve the performances of the envisioned approach. In this respect, we propose the application of in-situ adaptive tabulation (ISAT) [3], widely and successfully exploited in homogeneous reactive flow simulations, for the efficient solution of the chemical sub-step in the context of transient simulations of heterogeneous catalytic systems. The ISAT algorithm is a storage and retrieval method which allows the accurate approximation of multi-dimensional computational expensive functions by recording and reusing information obtained in previous evaluations of the function.

Results

We developed a numerical library, implementing the main features of the ISAT algorithm [3,4] and specifically conceived to efficiently deal with transient problems. In particular, a procedure to periodically reshape the binary tree and to remove infrequently used leaves from the storage table has been devoted to keep the search procedure efficient. The library has been developed in order to be easily interfaced with any existing operator-splitting numerical tool. The library has been successfully interfaced with the existent catalyticFOAM framework to efficiently solve the chemical sub-step. The accuracy of the numerical strategy has been investigated by comparing the results of simulations carried out with and without ISAT treatment of the chemical sub-step in a simple reactor geometry, in a wide range of operating conditions. The library ensures a very good error control as evident from the deviation between profiles obtained with and without ISAT, which is in most cases below the respective tolerance value.

In this work, we exploited the ISAT-based solver to investigate the computational efficiency in the context of large scale simulations of cases of industrial relevance. The adiabatic methane steam reforming carried out in a fixed-bed catalytic reactor has been adopted to analyse the computational performances of the modified solver dealing with complex geometries and detailed kinetic schemes. The reactor consists of 25 spherical pellets randomly inserted a square-base parallelepiped domain. The domain has a side length of 10.1 mm and the packed bed has a height of 14 mm, while the

spheres have a diameter of 3 mm. The generation of the numerical mesh has been carried out by means of snappyHexMesh [6]. Contact points among spheres have been meshed with the so called “bridge method” [7].
By comparing the ratio between the computational time required to solve a single chemical sub-step with and without ISAT, the capability of the library to reduce the chemical computational time up to ~500 times, depending on the dimension and stiffness of the system, has been proved. Considering the ratio between the time spent to carry out the solution of a single time-step with and without ISAT a speed-up factor of ~10-15 has been obtained, as shown in Figure 1.

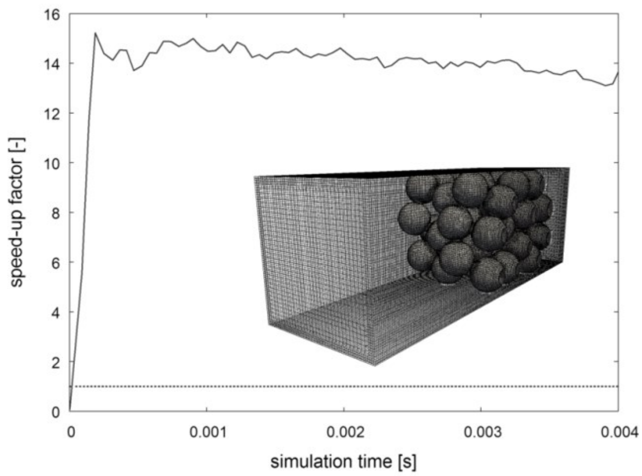

Figure 1. Speed-up factor against simulation time for methane steam reforming. The computational domain and the adopted mesh are also reported.

Conclusions
In this work we faced the problem of the reduction of the computational effort required in the solution of the chemical sub-step, in the context of heterogeneous reactive flow simulation. As a possible solution, we propose to exploit the in-situ adaptive tabulation algorithm. In this view, a library implementing the methodology has been developed and has been interfaced in the existent catalyticFOAM framework to efficiently solve the chemical sub-step.
The ISAT-based solver has been exploited to simulate cases of industrial relevance reducing the required computational effort. In particular, the ISAT treatment of the reactions sub-step allowed for a speed-up in the solution of the chemistry up to ~500 times, resulting in an overall simulation speed-up at most of ~10-15 times. In this context, ISAT can be considered as a helpful and handy tool to be interfaced in CFD framework to make possible the advanced design of catalytic devices of industrial relevance.

Acknowledgements
Computational time at CINECA (Bologna, Italy) is gratefully acknowledged.

References
[1] M. Maestri and A. Cuoci, "Coupling CFD with detailed microkinetic modeling in heterogeneous catalysis", Chemical Engineering Science, vol. 96, pp. 106-117, 2013.
[2] T. Maffei, G. Gentile, S. Rebughini, M. Bracconi, F. Manelli, S. Lipp, A. Cuoci and M. Maestri, "A multiregion operator-splitting CFD approach for coupling microkinetic modeling with internal porous transport in heterogeneous catalytic reactors", Chemical Engineering Journal, vol. 283, pp. 1392-1404, 2016.
[3] S. Pope, "Computationally efficient implementation of combustion chemistry using in situ adaptive tabulation", Combustion Theory and Modelling, vol. 1, no. 1, pp. 41-63, 1997
[4] L. Lu and S. Pope, "An improved algorithm for in situ adaptive tabulation", Journal of Computational Physics, vol. 228, no. 2, pp. 361-386, 2009.
[5] M. Maestri, D. Vlachos, A. Beretta, G. Groppi and E. Tronconi, "A C1 microkinetic model for methane conversion to syngas on Rh/Al_2O_3", AIChE Journal, vol. 55, no. 4, pp. 993-1008, 2009.
[6] OpenCFD, OpenFOAM: The Open Source CFD Toolbox. User Guide Version 1.4, OpenCFD Limited, Reading UK, Apr. 2007
[7] S. Rebughini, A. Cuoci and M. Maestri, "Handling contact points in reactive CFD simulations of heterogeneous catalytic fixed bed reactors", Chemical Engineering Science, vol. 141, pp. 240-249, 2016.