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ASBO BASED COMPOSITIONAL OPTIMIZATION IN COMBINATORIAL CATALYST

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Abstract. The application of different engineering fields in the discovery and development of new materials, especially of new catalyst, is changing the conventional research methodology in materials science.For Heterogeneous catalysts, their catalytic activity and selectivity are dependant on chemical composition, micro structure and reaction conditions. Therefore, it is worth to do the research over the composition of the catalyst and the reaction conditions that will boost its performance.This paper proposes a computational intelligence approach based on adaptive social behavior optimization (ASBO) for catalyst composition optimization to enhance the resulting yield or achieving objective maximal.The proposed approach is especially useful in the combinatorial catalysis optimization wherein the fitness function is unknown, in result cost and time can be drastically reduced with intelligent search method instead of applying real time chemical reaction.Challenge of handling higher dimensionality and achieving a global solution can be fulfilled by ASBO which is based on human behavior under social structure which makes human as a most successful species in nature.Two different mathematical models of the catalyst composition problem, which contains the optimal complexity and represents practical scenarios have taken to explore the quality of solution. Particle swarm optimization (PSO) which is considered as a successful heuristic method among others has also been applied to get the comparative performance analysis in detail.

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I. Introduction

The pace at which major technological changes take place is often dictated by the rate at which new materials are discovered, and the timely arrival of new materials has always played a key role in bringing advances to our society. It is no wonder that the so-called combinatorial or high-throughput strategy has been embraced by practitioners of materials science in virtually every field. High throughput experimentation allows simultaneous synthesis and screening of large arrays of different materials. Pioneered by the pharmaceutical industry, the combinatorial method is now widely considered to be a watershed in accelerating the discovery and optimization of new materials.

The field of catalysis is poised for further major progress and significant successes in the 21st century, with some innovations occurring in an evolutionary manner and many more likely to result from technological discontinuities or movement to a new learning curve. During the last 20 years, catalysis has been increasingly recognized as a multidisciplinary science, setting the stage for substantial changes.Catalysis in the 21st century will be trans-disciplinary, with continuous synergistic interactions and collaborations between mathematicians, chemists, physicists, biochemists, chemical, mechanical and electronic engineers, and many others. The basis for this change will be: the rational design of catalysts underpinned by the fundamental understanding of their action and of reaction mechanisms; novel routes to catalyst discovery using combinatorial methods and high throughput experimentation; the need for cheaper and sustainable catalytic processes in all chemical sectors, with increased selectivity and lifetime (due to environmental, societal, and economic considerations) and necessarily an increased collaboration between academic and industrial scientists having a common interest in catalysis and its applications.Sectors that are particularly open to innovation are, in order of increasing maturity and interest, pharmaceuticals, power generation, fine chemicals, natural gas conversion,and transportation. Other sectors we shouldn't overlook include the food industry, petrochemicals, and petroleum refining. These last three sectors are often considered as technologically matured, with most of research in these fields being explanatory. However, we believe that there is still ample room for "extrapolatory" research and new developments. Nevertheless, it is the former five sectors where exploratory research and emerging technologies are likely to have the greatest impact.

The use of Genetic algorithm based solution for the search for optimal catalytic materials has been applied in [1]. They have applied the concept of surrogate models in integration of neural network to handle the issues. A methodical basis of the evolutionary method for selection and optimization of heterogeneous catalytic materials has been developed and presented in [2]. They have defined a mathematical model for catalyst performance in terms of yield. Authors in [3] tried to decrease the number of necessary experiments by proposing an optimization algorithm based on a genetic algorithm. By combining a trained neural network with the genetic algorithm software virtually computational experiments were aimed at adjusting the control parameters of the optimization algorithm to the special requirement of catalyst development. Authors in [4] presented issues of heterogeneous catalyst optimization in the framework of high throughput iterative screening. This work highlights the most important features of the evolutionary strategies (ES) that lead to successful optimizations. The authors in [5] have presented a two-stage design framework approach to the discovery and optimization of heterogeneous catalysts by complete liquid phase technology for direct synthesis DME from syngas. In this proposed approach, a trained SVR model is constructed for correlating process data comprising values of the input variables of catalyst compositional, operating conditions and output variables of performance of catalyst. Next, the trained SVR model is employed as an approximate model in calculating on fitness function values of multi-objective CPSO architecture. A soft computing technique based on the combination of Artificial Neural Networks (ANNs) and a Genetic Algorithm (GA) has been developed for the discovery and optimization of new materials when exploring a high-dimensional space has been presented in [6]. This technique allows the experimental design in the search of new solid materials with high catalytic performance when exploring simultaneously a large number of variables such as elemental composition, manufacture procedure variables, etc. An approach to constrained mixed optimization based on formulating a separate linearly-constrained continuous optimization task for each combination of values of the discrete variables has been proposed in [7]. Then, discrete optimization on the set of nonempty polyhedra describing the feasible solutions of those tasks is performed, followed by solving those tasks for each individual of the resulting population of polyhedra.The relationship between the chemical reaction controlled (shrinking core) model and cellular automata, to study the dissolution of particles, is derived in [8]. Use of computational methods to design new catalysts has been reviewed in [9]. Examples include screening of catalysts

with increased activity and catalysts with improved selectivity. Authors in [10] hybridize a classic genetic algorithm with a knowledge discovery system which extracts information from a database containing known observations allowing building a model replacing the fitness function. They have used the k nearest neighbour's algorithm to solve such a problem sat in heterogeneous catalysis, a division of chemical science where a compound shall be optimized to favour a reaction. Detailed discussion related to PSO has presented by the authors in [11] and they have given mathematical analysis about the convergence requirement of PSO. Concept related to ASBO has presented in[12].Surrogate modelling based approach to the optimization of objective functions evaluated via measurements has presented in [13] and purpose to decrease the time and costs of evaluation of the objective function.In [14] authors have tried to find the best catalyst, the best combination of compounds, in order to optimize a chemical reaction.Variant of evolutionary optimization algorithm, defined as meta modeling presented .They have combined a statistical learning algorithm with the optimization process. The approach has applied in the combinatorial catalysis optimization where the fitness function is unknown and the labelled individual is obtained by the real chemical reaction. Optimization of catalytic materials though evolutionary algorithms have presented in [15]. They have described the various kinds of encountering constraints, and explain how to handle constraints.A discussion about Combinatorial Heterogeneous Catalysis has presented in [16]. State of the art discussion for predictive computational organometallic chemistry in reference to the different stages of catalyst development by considering characterization, mechanistic studies, fine-tuning/optimization, and evaluation of novel designs has presented in [17]. The computational enzyme design has tremendous potential for a wide range of important applications and to illuminate fundamental issues in catalysis has proposed in [18]. Most organic and organometallic catalysts have been discovered through serendipity or trial and error, rather than by rational design. Computational methods, however, are rapidly becoming a versatile tool for understanding and predicting the roles of such catalysts in asymmetric reactions. Such methods should now be regarded as a first line of attack in the design of catalysts [19].Review of the historical development of computational quantum chemistry, in the context of catalysis and a subjective selection of past and present research, as well as subjective views on future directions has presented in [20].

Catalysis is a ubiquitous process in the modern world, and there is a perennial need to improve and discover new catalysts. Heterogeneous catalysts are multifunctional materials composed of several active components, promoters, and a high-surface-area support material. The compositional parameter space associated with them is therefore very large.In addition, one also needs to consider a wide variety of parameters related to the method of preparation, as well as reaction conditions to which a given catalyst could be subjected, which increases the total number of possible experimental combinations even more. In the past decade, the field has experienced significant growth, with multimillion dollar investments by almost all major chemical and petrochemical companies.High-throughput studies of heterogeneous catalysts consist of three components – rapid catalyst synthesis, high throughput testing of catalyst materials, and appropriate data processing and information mining techniques – which feed back into the synthesis.For primary screening experiments, libraries are typically synthesized via radio frequency sputtering, pulsed laser deposition, molecular beam epitaxy, or chemical vapor deposition. However, it can be difficult to extrapolate the catalytic properties derived from such model thin-film libraries to much more realistic supported catalyst materials, where a typical synthesis requires phase chemistry and solutions of high-surface-area support materials. Therefore, most work in high-throughput catalysis is now performed on high-surface-area materials, which allow much easier scale up.To this end; liquid precursors can be dispensed with automated robots to synthesize libraries of more realistic catalytic materials. However, the synthesis of supported catalysts (powders) has a multitude of variables, including precursor materials, support materials, temperature, and drying and calcination parameters. All these variables must be screened and optimized for the particular system under study.

Catalytic processes can be divided in three main types: heterogeneous, homogeneous and enzymatic processes.In a heterogeneous reaction, the catalyst is in a different phase from the reactants. Normally, the catalyst is a solid and reactants are fluids (liquids or gases). It is characterized by the presence of “active sites” on the catalyst surface.In a homogeneous reaction, the catalyst is in the same phase as the reactants .The enzymatic catalysis (biocatalysis) has an intermediate character between homogeneous and heterogeneous processes, because although the enzymes and reactants are in the same phase (solution), they have “active sites”in their structures. Even with the development of sophisticated high-throughput techniques, it is frequently unfeasible

to collect data for all experimental combinations. Experimental design strategies can reduce the number of experiments required to sample the parameter space. Approaches include standard statistical design techniques, such as response surface methodology and D-optimal designs. Other approaches used to guide high-throughput experimentation include the use of artificial neural networks, a holographic strategy, and factor-based methods from chemometrics. Caruthers et al. have investigated the process of analyzing high-throughput data to extract chemical understanding using a process termed 'knowledge extraction'.

A. Present possibilites and hopes

(a) New approaches to discover potentially new catalytic materials, including innovative preparation methods and combinatorial catalysis;

(b) Increased molecular understanding of the mechanism of heterogeneous and homogeneous catalytic transforma tions based on in-situ investigations and extensive quantitative kinetic models and studies;

(c) Scientific computing making possible quantum chemistry,molecular modeling, thermodynamic and kinetic predictions, reaction and reactor modeling, and, last but not least, all the resources needed to enable quantitative high throughput evaluation of catalyst performance;

(d) New approaches to catalytic processes enabling product yields to exceed those predicted by thermodynamics;

(e) Combinatorial catalysis and high throughput catalyst testing, speeding up the discovery of novel catalysts and catalyst optimization.

II. Combinatorial catalysis: accelerating catalyst discovery and evaluation

If we consider that the periodic table contains approximately 75 useful and stable elements, the number of possible compounds which can be created is extremely large. The elements form about 5600 binary, 4×10^5 ternary, 3×10^7 quaternary and 10^{18} decanery compounds, without even considering stoichiometric and structural variations. The synthesis, not to mention the analysis, of such numbers of compounds would be prohibitively time consuming and expensive and a more selective approach is required. Instead of randomly synthesising new compounds the search for new material designs begins with the synthesis of materials similar to already well-known compounds. The results of the initial process are used to obtain trends and patterns which are then

used to select optimal compositional ranges for further exploration, and the synthesis recommences. Integration of rapid chemical synthesis and high-throughput screening with large-scale data analysis methods that constitutes the essence of combinatorial material science. By utilising the power of these automated techniques, the time required to converge upon new materials can be reduced.

In about five years, combinatorial methods have gone from being a curiosity generating with lot of skepticism, to occupying center stage in catalysis research. Major players in this arena are Symyx Technologies (USA), HTE (Germany), and Avantium (The Netherlands). Several R&D companies also elected to develop their own in-house capabilities, e.g., General Electric, DuPont, UOP, ICI, Johnson Matthey, to cite but a few.The term "combinatorial catalysis," which harks back to its original use in the rapid screening of drug candidates, is now broadly applied to any miniaturized, high-throughput R&D system, in particular those used for the accelerated discovery and development of new catalysts and materials. Fig.1 illustrates the various stages involved in the discovery and development of catalytic processes, including their attrition over the carrier of 12 year period usually necessary to reach commercialization. Combinatorial catalysis, via high-throughput screening and testing of potential catalytic materials, aim to collapse the time necessary to discover or optimize, and define new catalytic formulations, i.e., compressing it from 4–5 years to 6–12 months.The approach is still maturing and involves a number of steps. The design and construction of catalyst libraries, which may comprise thousands of candidate materials, involves theory, modeling, careful screening of the journal and patent literature, chemical intuition and continuous feedback — this through various algorithms, of information generated by the primary screenings and secondary screenings (tests) that are progressing. Primary screening is usually highly sophisticated, involving highly miniaturized reactor systems and specific, sometimes homedesigned, analytical techniques. Secondary screening is performed with parallel multichannel microreactor systems equipped with more conventional analytical tools. A third screening stage at the conventional microreactor scale is still likely to be needed in order to generate data necessary to reach the pilot stage. Many challenges are met by this novel approach to accelerated catalyst discovery and optimization. Considering the large number of potential catalytic materials which will have to be generated, the tremendous amount of data (operating conditions and catalytic system performance) to be stored and mined, and the need for state-of-the

art automation at all levels with scientific computing will play a decisive role. This will be even more so when the potential contributions of theoretical methods, such as quantum chemistry and molecular modeling/simulations, are taken into account. Another major challenge is the preparation of catalysts for the first and second screening levels by methods that can be realistically and reproducibly scaled up for their conventional microreactor testing if they become the lead compositions.

The catalyst sector has clearly become the next leading proponent of combinatorial chemistry after the pharmaceutical and agrochemical sectors. Combinatorial catalysis enable us to look with a new eye at what, unfortunately, has been many times characterized as a mature and old science or industry.There is no longer any doubt that the development of combinatorial, high throughput technology for catalyst design, evaluation, and testing is a new and proven tool for catalyst discovery. It is certainly very target-oriented. Industry and academia have complimentary objectives and novel ways must be found to enhance the synergies between these two communities.High throughput catalyst design and evaluation is definitely not a roulette game. The combinatorial approach relies on scientific knowledge and statistical methods, and benefits from the input of many other disciplines, including among others solid state and organometallic chemistry, automation, scientific computing, etc.

Combinatorial catalysis is a methodology where a large number of new materials are prepared and tested in a parallel fashion. The global search/optimisation strategy is the main difference with the traditional catalyst research and allows reduction of the number of experiments needed to find an optimal catalyst composition. Combinatorial catalysis involves the co-ordination of : high-throughput systems for preparation, characterisation and catalytic test; large information data management; and rapid optimisation techniques. This promising approach requires therefore the development and optimisation of the following items: (i) high-throughput equipment, which allows the reliable preparation and characterisation/testing preferentially under realistic conditions of large quantities of materials (ii) optimisation techniques, adapting their structure and parameters by implementing the chemical knowledge/experience of the experts. With this, it would be possible to increase the number of variables to study and this would result in a potentially rather more powerful final catalyst and shorter search times. Indeed, if this methodology is properly followed it can be very helpful in the scientific understanding of catalysis.

An important issue in combinatorial catalysis is how to design the experiments in order to explore and optimize the high-dimensional solution space while minimizing the number of trials to achieve a solution. The approaches employed for experimental design can be distinguished in three groups: (i) statistics procedures like factorial designs, (ii) deterministic optimization procedures like simplex, holographic search or split & pool and (iii) stochastic procedures like simulated annealing or Genetic Algorithms (GAs).

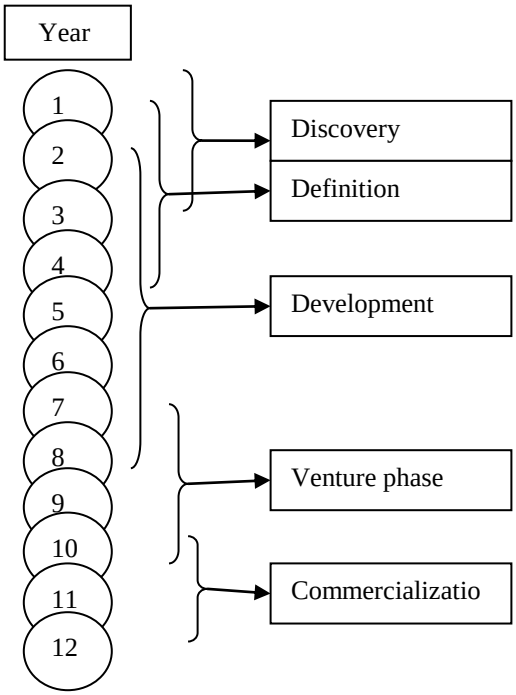

Fig.1 Project stages from discovery to commercial application and attrition

Stochastic procedures inspired by natural computing are procedures well-suited for the optimization of multi-dimensional problems, being especially useful for application in the field of combinatorial catalysis, since (i) they use a population of points to conduct the search, making this approach appropriate for high-throughput (HT) tools,(ii) the objective is to find an approximate global maximum and (iii) they tolerate noisy experimental data

In chemical engineering, much effort is devoted to increasing the performance of industrially important chemical processes, i.e., to achieve a higher yield of the desired reaction products without higher material or energy costs. Over 90% of the processes use a catalyst to speed up the reaction or to improve its selectivity to the desired products. Catalysts are materials that decrease

the energy needed to activate a chemical reaction without being themselves consumed in it. Catalytic materials typically consist of several components with a different purpose to increase their functionality. The components typically can be selected from among many substances. Chemical properties of those substances usually constrain the possible ratios of their proportions, but since the proportions are continuously-valued, they still allow for an infinite number of catalyst compositions. Moreover, the catalyst can usually be prepared from the individual components in a number of ways, and the preparation method also influences the performance of the chemical process.Consequently, the search for catalysts leading to optimal performance entails a complex optimization task with the following features:

(a) high dimensionality (30-50 variables are not an exception);

(b) mixture of continuous and discrete variables;

(c) constraints;

(d) objective function cannot be explicitly described and its values must be obtained empirically.

III. Test bench

At this step of the research, to show the efficiency of the modeling approach, we examine the optimization of virtual catalysts according to a theoretical response surface. Considering the evolution control step, this means that instead of synthesizing and testing the catalyst, a theoretical fitness value will be attributed. It is calculated according to a mathematical function, which reflects the behavior of the catalyst in real conditions.In this research, two benchmark functions have been employed for experimental studies carried out with the PSO and ASBO individually and comparative performances analysed in details. The first test bench describes the dependence on the catalyst composition of the final catalytic performance in the oxidative dehydrogenation of propane, when prepared by incipient wetness.The catalyst variables considered in this model are the content of eight different elements: V, Mg,B, Mo, La, Mn, Fe and Ga. The objective function to be maximized is the propylene yield (Y, %) which is the product of selectivity S and the conversion X and is described as follows in (1) :

$$Y = \begin{cases} S_1 \times X_1 \rightarrow If \quad Incipient\ Wetness \\ S_2 \times X_2 \rightarrow If \quad Coprecipitation \\ 0 \qquad \rightarrow If \quad (x_B \vee x_{La}) > 0 \end{cases} \tag{1}$$

Where

$$S_1 = 66x_V x_{Mg}\left(1 - x_v - x_{Mg}\right) + 2.0x_{Mo} - 0.1x_{Mn} - 0.1x_{Fe}$$

$$X_1 = 66x_V x_{Mg}\left(1 - x_V - x_{Mg}\right) - 0.1x_{Mo} + 1.5x_{Mn} + 1.5x_{Fe}$$

$$S_2 = 60x_V x_{Mg}\left(1 - 1.3x_v - x_{Mg}\right)$$

$$X_2 = 60x_V x_{Mg}\left(1 - 1.3x_V - x_{Mg}\right)$$

The non-continuous function in (1) reveals two maxima (one is global and other is local) corresponding to different molor functions of components and preparation method.A global maximum occurs if incipient wetness is applied while a local minimum occurs if coprecipitation method is applied for catalyst preparation.

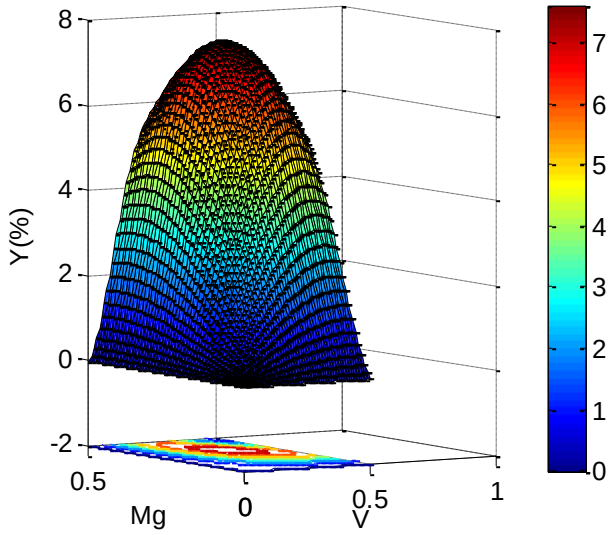

Fig.2 Yield variation with variation in Mg and V in range of [0 0.5] with constant value of B[0], Mo[0.3496], La[0], Mn[0.0064], Fe[0.0031], Ga[0].

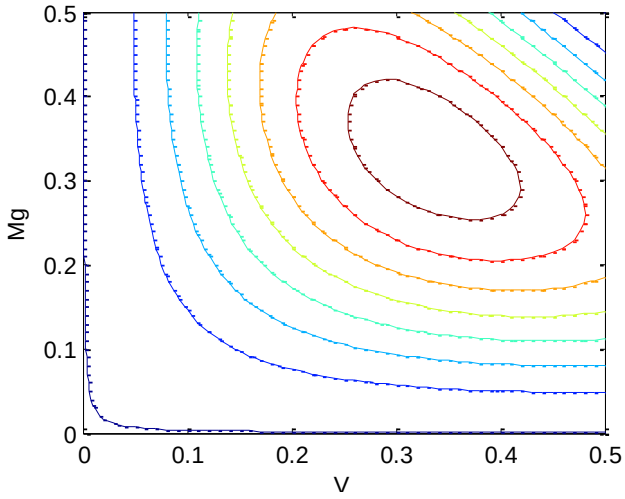

Fig.3 Yield contour with Mg and V with other parameters constant.

The second test bench has a much more complex topology with several local maximums. As can be seen in Fig.4, this function presents three high-activity areas while presenting some periodicity, this behavior being common for heterogeneous catalysts, when varying their composition and synthesis conditions. The function is defined as follows in (2):

$$Y(x_1,x_2,x_3,x_4,x_5)=Z_i(x_1,x_2)+Z_j(x_2,x_3)Z_k(x_3,x_4,x_5) \quad (2)$$

Where

$$\sum_{i=1}^{5} x_i = 1, \quad \forall\, i \geq 0\,,$$

(x_i represents the molar percentage of the catalyst material)

$$Z_i(u,v)=0.6g(100u-35,100v-35)+0.75g(100u-10,100v-10)+g(100u-35,100v-10)$$

$$Z_j(u,v)=0.4g(100u-10,100v-30)\,;$$

$$Z_k(u,v,w)=5+25(1-(1+(u-0.3)^2+(v-0.15)^2+(w-0.1)^2)^{0.5})$$

$$g(u,v)=100-\left(u^2+v^2\right)^{0.5}+50\frac{\left(\sin\left(\left(u^2+v^2\right)^{0.5}\right)\right)}{\left(u^2+v^2+0.001\right)^{0.5}}$$

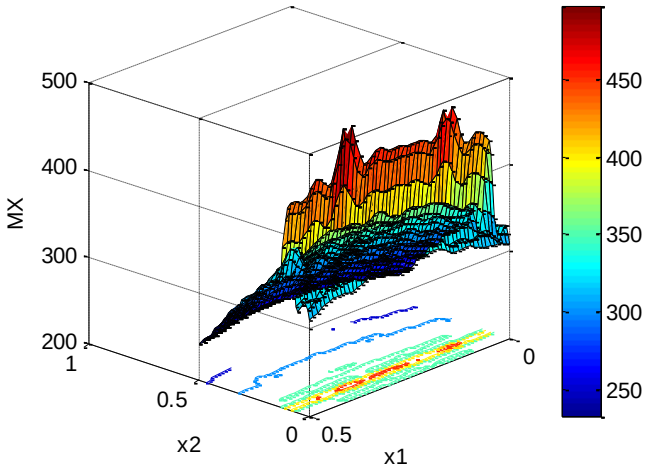

Fig.4 Yield variation with variation in X1 and X2 in range of [0 0.5]While other parameters are constant x3[0.1026], x4[0.2469], x5[0.1969]

IV. Proposed solution

Classical linear programming and traditional non-linear optimization techniques such as Lagrange's Multiplier, Bellman's principle and Pontyagrin's principle were prevalent until this century. Unfortunately, these derivative based optimization techniques can no longer be used to determine the optima on rough non-linear surfaces. The aim of optimization is to determine the best-suited solution to a problem under a given set of constraints. Several researchers over the decades have come up with different solutions to linear and non-linear optimization problems. Mathematically an optimization problem involves a fitness function describing the problem, under a set of constraints representing the solution space for the problem. Unfortunately, most of the traditional optimization techniques are centered around evaluating the first derivatives to locate the optima on a given constrained surface. Because of the difficulties in evaluating the first derivatives, to locate the optima for many rough and discontinuous optimization surfaces, in recent times, several derivative free optimization algorithms have emerged. The optimization problem, now-a-days, is represented as an intelligent search problem, where one or more agents are employed to determine the optima on a search landscape, representing the constrained surface for the optimization problem. Suppose the global optimum of a n-dimensional function is to be located. The function may be mathematically represented as:

$$f(x_1, x\ , x\ , \ldots . x_n) = f(X)$$

Where $\boldsymbol{X}$ is the search-variable vector, which actually represents the set of independent variables of the given function. The task is to find out such a $\boldsymbol{X}$,that the function value $f(\mathrm{X})$is either a minimum or a maximum denoted by f^* in the search range. If the components of $\boldsymbol{X}$ assume real values then the task is to locate a particular point in the n-dimensional hyperspace which is a continuum of such points.

A. Particle Swarm Optimization (PSO)

PSO is a multi-agent parallel search technique. Particles are conceptual entities, which fly through the multi-dimensional search space. At any particular instant, each particle has a position and a velocity. The position vector of a particle with respect to the origin of the search space represents a trial solution of the search problem. At the beginning, a population of particles is initialized with random positions marked by vectors $\vec{x}_i$ and random velocities $\vec{v}_i$. The population of such particles is called a "swarm" S. A neighborhood relation N is defined in the swarm. N determines for any two particles Pi and Pj whether they are neighbors or not. Thus for any particle P, a neighborhood can be assigned as N(P), containing all the neighbors of that particle.Different neighborhood topologies and their effect on the swarm performance will be discussed later. However, a popular version of PSO uses N = S for each particle. In this case, any particle has all the remaining particles in the swarm in its neighborhood. Each particle P has two state variables viz., its current position $\vec{x}(t)$ and its current velocity $\vec{v}(t)$It is also equipped with a small memory comprising its previous best position (one yielding the highest value of the fitness function found so far) $\vec{p}(t)$i.e., personal best experience and the best $\vec{p}(t)$of all $p \in N(P): \vec{g}(t)$, i.e., the best position found so far in the neighborhood of the particle. When we set $N(P) = S, \vec{g}(t)$ is referred to as the globally best particle in the entire swarm. Once the particles are all initialized, an iterative optimization process begins where the positions and velocities of all the particles are altered by (3) and (4).

$$V_{id}(t+1) = K * \begin{bmatrix} W * V_{id}(t) \\ + C_1 r_1 \left(P_{id}(t) - X_{id}(t)\right) \\ + C_2 r_2 \left(P_{gd}(t) - X_{id}(t)\right) \end{bmatrix} \qquad (3)$$

$$X_{id}(t+1) = V_{id}(t+1) + X_{id}(t) \qquad (4)$$

The main parameters of the canonical PSO model are W, C1, C2, K and the swarm size S. The settings of these parameters determine how it optimizes the search-space. The inertia weight ω

controls the momentum of the particle: If ω << 1, only little momentum is preserved from the previous time-step; thus quick changes of direction are possible with this setting. The concept of velocity is completely lost if ω = 0, and the particle then moves in each step without knowledge of the past velocity. On the other hand, if ω is high (>1) we observe the same effect as when C1 and C2 are low: Particles can hardly change their direction and turn around, which of course implies a larger area of exploration as well as a reluctance against convergence towards optimum. K Constriction coefficient results in the quick convergence of the particles over time. That is the amplitude of a particle's oscillation decreases as it focuses on the local and neighborhood previous best points. Though the particle converges to a point over time, the constriction coefficient also prevents collapse if the right social conditions are in place. The particle will oscillate around the weighted mean of p_{id} and p_{gd}, if the previous best position and the neighborhood best position are near each other the particle will perform a local search. If the previous best position and the neighborhood best position are far apart from each other, the particle will perform a more exploratory search (global search). During the search, the neighborhood best position and the previous best position will change and the particle will shift from local search back to global search. The constriction coefficient method therefore balances the need for local and global search depending on what social conditions are in place.

B. ADAPTIVE SOCIAL BEHAVIOR OPTIMIZATION (ASBO)

ASBO is a very new heuristic and stochastic search method inspired by human social behavior to obtain the global solution.Social interactions enable individuals to adapt and improve faster than biological evolution based on the genetic inheritance alone. This is the driving concept behind the optimization algorithm that makes use of the competition and influence available within a formal society. Particle swarm optimization and ant colony algorithms are two very successful and established computing models already justifying the importance of the above statements. These two computing models are having the bias reference of social life activities either with respect to a very few species likes bird flocking or fish schooling or species like ants.

In ASBO optimization process, behavior of entity to inspire automatically by various social elements are taken as fundamental operators to optimize the solution iteratively. It is a well known

fact that every action of human is the result of influences. The nature and characteristics of influence may be different from person to person and time to time. In ASBO, three macro social influence operators namely: inspiration by leader, inspiration by neighbours and self inspiration have taken. Level of influences is defined by corresponding adaptive constants. These constants play very important in changing the status of the individual; because the influence is dynamic with time variable hence it can not be fixed for all life periods. The Adaptive characteristics of these constants are defined by self adaptive mutation strategy. Mathematical modelling for ASBO is given below.

There are two sets of populations; a solution population and the other is influenced factor population. Each member of solution population represents the solution in a phenotype format (direct form i.e. not in coded form) and influence factor population contains the same number of members as solution population has with three parameters each corresponding to leader, neighbours and self influence. With respect to the problem at hand using the fitness function a fitness value for each and every member defined. An Individual having the maximum value of fitness treated as a leader at the present time. A group of individuals having next nearest higher value of fitness will be treated as neighbours for a particular individual. The change in existing status because of influence is innovated by each and every member of the population using (5) and the next location of status given by (6).

$$\Delta X(i+1) = [\quad C_g R_g (G_{bi} - X_i) \quad + C_s R_s (S_{bi} - X_i) \quad + C_n R_n (N_{ci} - X_i) \quad] \qquad (5)$$

$$X(i+1) = X_i + \Delta X(i+1) \qquad (6)$$

Where $\Delta X(i+1)$ represents the new change in i'th dimension of an individual element. Cg, Cs, Cn are adaptive progress constants ≥ 0; R_g , R_s , R_n are uniformly distributed random number in range [0 1], G_b , global best individual at present population's, S_b is the self best for an individual till present and Nc is the center position of a group formed by an individual and its neighbours in present population, For a D-dimensional problem, Gb, Sb, & Nc represent vectors of D-dimension.

$G_b = [G_{b1}, G_{b2}, G_{b3}, G_{b4} \ldots \ldots \ldots G_{bD}];$

$S_b = [S_{b1}, S_{b2}, S_{b3}, S_{b4} \ldots \ldots \ldots \ldots S_{bD}];$

$N_b = [N_{b1}, N_{b2}, N_{b3}, N_{b4} \ldots \ldots \ldots N_{bD}];$

A. Evolution of New Set of Progress Constant

A population of N initial random solution initialized. Each solution is taken as a pair of real valued vector called progress constant vector (p_i) and strategy parameter vector (σ_i), with each vector there are three dimensions corresponding to the number of adaptive progress constant. The initial components of each $p_i, i \in \{1..... N\},$, were selected in accordance with a uniform distribution ranging over a presumed constant space. The values of σi , $i \in \{1..... N\}$, were initially set to some smaller value. A new solution $(p_i^{'}, \sigma_i^{'})$ generated from each previous solution (p_i, σ_i) by (7) and corresponding strategy parameters upgraded by using (8).

$$p_i^{'}(j) = p_i(j) + \sigma_i(j)N(0,1) \qquad (7)$$

$$\sigma_i^{'}(j) = \sigma_i(j)e^{(\tau' N(0,1) + \tau N_j(0,1))} \qquad (8)$$

$$\forall\ j \in \{1,2,3\}$$

Where $p_i(j), p_i^{'}(j), \sigma_i(j), \sigma_i^{'}(j)$ denote the jth component of the vectors $p_i, p_i^{'}, \sigma_i, \sigma_i^{'}$ respectively and N(0,1) is a random number from Gaussian distribution. $N_j(0,1)$ is a new random number sampled for each value of the counter j using Gaussian distribution.τ' and τ are constants.

There are two phases under which the whole process to get the global solution. (i) A PF number of different populations having same population size (PZ) initially are taken and ASBO method is applied independently up to a fixed number of iterations say P. At the end, values of fitness and all progress constants are stored for each and every member from each final population. This phase will help to maintain the diversity and in result there is better exploration to localize the region of the solution. (ii) From all final population, depending upon the fitness, members who are having best PZ number of fitness values are selected to form new population and their progress constants are also taken to form the second stage single population. Over this newly generated population ASBO is applied to get the final solution. This phase will help to get the optimal solution in a faster manner.

V. Experimental setup

For both test problems we have applied PSO and ASBO under two different environment.First we have given a comparative environment for both methods with maintaing balance in terms of population size and number of iterations.Total fifty different trails have given with population size equal to 100 for both algorithms while number of allowed iteration for PSO has given equal to

2000 while for ASBO it is 100 in first stage while 1000 in second stage so that overall there are effectively 2000 iterations available.

In another case both algorithm has given a chance to a very high number of generations (5000) to explore the capability in delivering the global solution under different population size say 25, 50, 75 and 100. Experiment was repeated for 20 independent trails and best, worst, mean and standard deviation and confidence interval (CI) for 95% and 99% are estimated for comparative purpose.Distribution of results for all trails are also plotted in terms of histogram to understand the precision capability in a better manner.We have applied an annealing scheme for the ω-setting, where ω decreases linearly from ω = 1.2 to ω = 0.1 over the whole run under PSO.

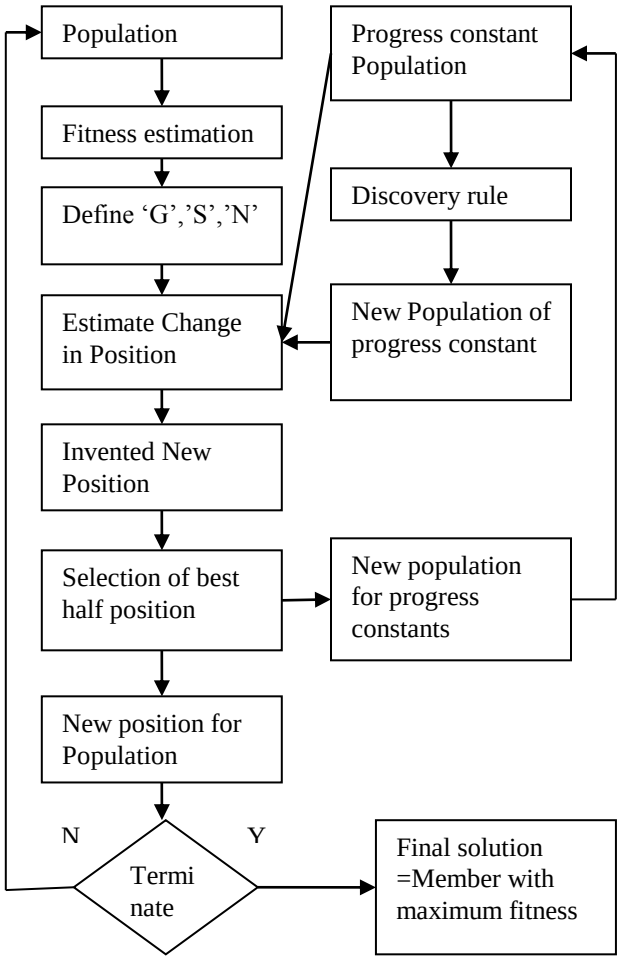

Fig.5 working flow of ASBO

A. TEST BENCH1: Under First test environmeont

Performance obtained with both algorithms are obtained graphically and numerically.Journey of parameters using PSO has shown in Fig.6 while obtained corresponding yield values are represented in Fig.7 and in Fig.8.in Fig.7 at the beginning large negative appeared because of constraint violation there is a penalty value associated which makes their fitness lower.After certain number of iterations there are feasible solutions and their details behavior is appeared in Fig.8.In Fig.9 yield values obtained in first phase of ASBO for all 10 different population each with 100 iterations have shown.Best solution fitness in second stage of ASBO has shown in Fig.10.Comparative performance between PSO and ASBO has shown in Table I and it is clear that ASBO has achieved global solution even in worst result case.ASBO has delivered better results compare to PSO in meeting similar computational complexity.Individual component final fractional values are also shown in Table II.

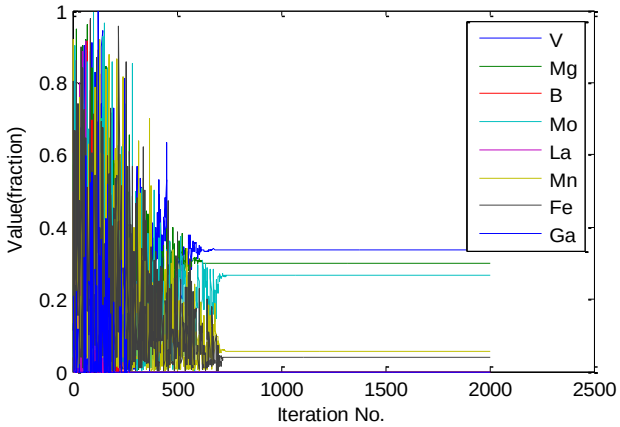

Fig.6.PSO catalyst component estimation by PSO with population size 100 for 1st test problem.

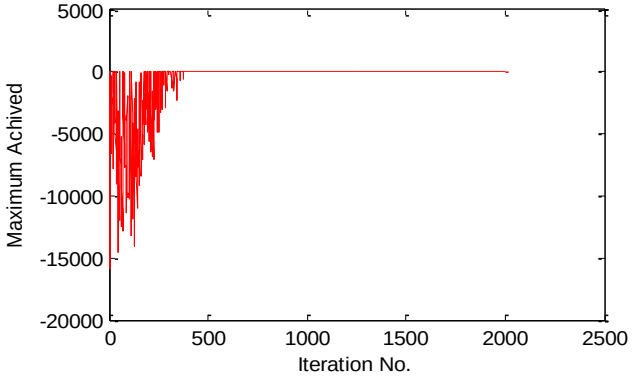

Fig.7 Yield achived by PSO in 1st test problem

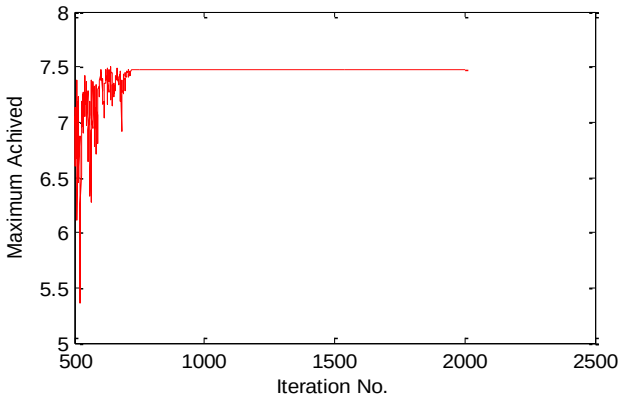

Fig.8 Yield achived by PSO in later generation for 1st test problem.

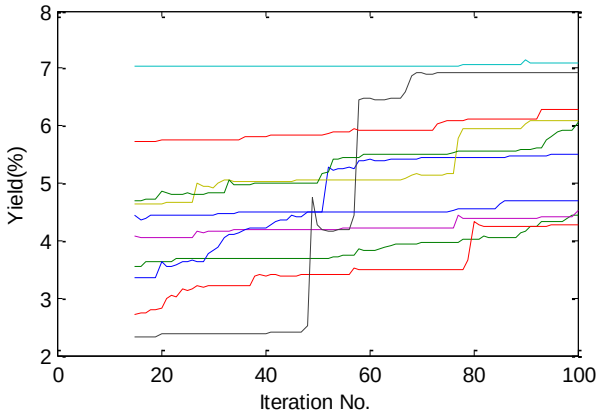

Fig.9 Yield achived by ASBO in first stage for 1st Test problem with all 10 different population

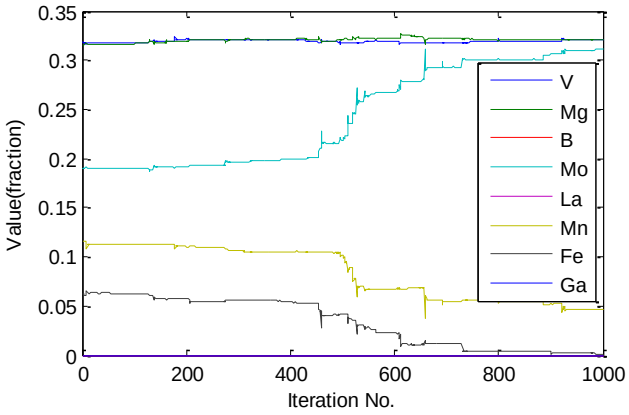

Fig.10 PSO catalyst component estimation by ASBO for 1st test problem in second stage.

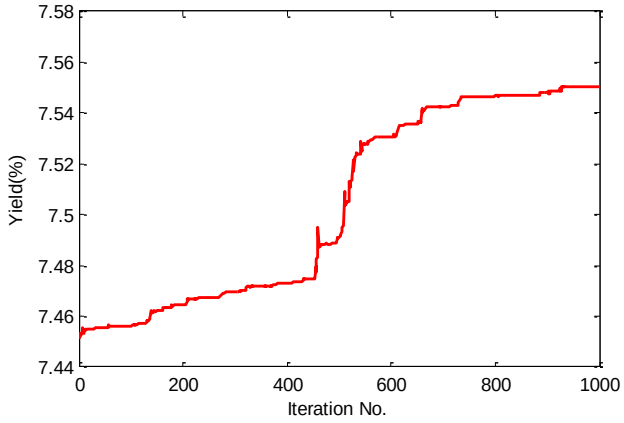

Fig.11 Yield achived by ASBO in 2nd stage for 1st test problem

TABLE I. comparative performances for 1st test problem under first environment for 50 independent trails

Yield(%)	PSO	ASBO
Best	7.5495	7.5550
Worst	7.3761	7.5111
Mean	7.5032	7.5409
Std.Dev	0.0304	0.0127
C.I (95%)	∓0.0086	∓0.0036
C.I (99%)	∓0.0115	∓0.0048

TABLE II. Best solution result for 1st test problem under first environment

Catalyst	PSO	ASBO
V	0.3221	0.3201
Mg	0.3159	0.3208
B	0	0
Mo	0.3156	0.3333
La	0	0
Mn	0.0339	0.0094
Fe	0.0122	0.0160
Ga	0	0

B. TEST BENCH 1:EXPERIMENT RESULT UNDER SECOND ENVIRONMENT

To understand the effect of population size over the exploration capabilities of algorithms ,different population size like 25,50,75 and 100 have applied for PSO and ASBO and performances have obtained.From Table III it is clear that PSO could reach close to global solution in nearly half trails but could reach close to ASBO results even in a single time with population size equal to 25 while ASBO almost in all trails deliver the global solution with high precision or very with same size of population. This can be more clearly understand with histogram representation for best value obtained in all trials have as shown in Fig.12 and in Fig.13.Fitness on average for all trails and best solution among trails for ASBO have shown in Fig.14 and it is obtained that they very close to each other on all periods of exploration.Experimental results for all other population sizes have shown in Fig.14 toFig.22 and performances have tabulated in Table IV to Table X.

TABLE III. COMPARATIVE PERFORMANCES FOR 1st TEST PROBLEM UNDER SECOND ENVIRONMENT FOR 20 INDEPENDENT TRAILS WITH POPULATION SIZE 25.

Performance	PSO	ASBO
Best	7.4982	7.5534
Worst	5.9252	7.5140
Mean	7.1642	7.5387
Std.Dev	0.4199	0.0110
C.I (95%)	∓0.1959	∓0.0051
C.I (99%)	∓0.2671	∓0.0070

TABLE IV. BEST SOLUTION RESULT FOR 1st TEST PROBLEM UNDER SECOND ENVIRONMENT WITH POPULATION SIZE 25

Catalyst	PSO	ASBO
V	0.3154	0.3199
Mg	0.3215	0.3210
B	0	0
Mo	0.2295	0.3212
La	0	0
Mn	0.0519	0.0349
Fe	0.0817	0.0029
Ga	0	0

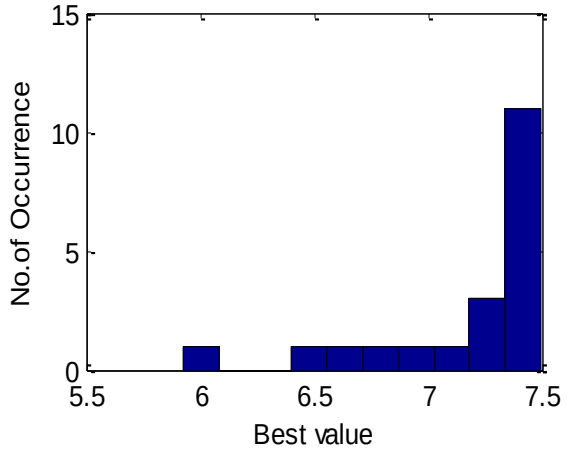

Fig.12 Distribution of best value over 20 trails in PSO with population size 25 for 1st test problem.

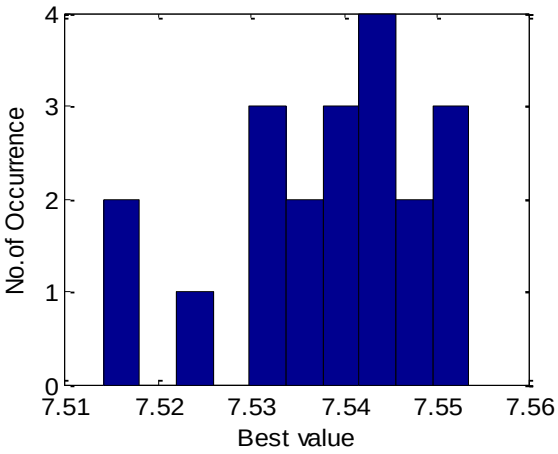

Fig.13 Distribution of best value over 20 trails in ASBO with population size 25 for 1st test problem.

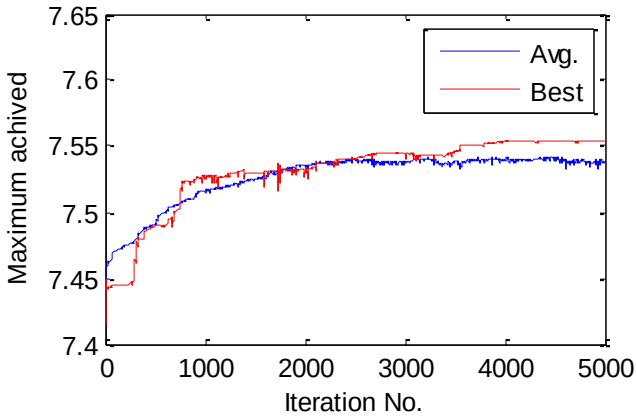

Fig.14 Average and best solution performance over 1st test problem w.r.t iterations in second phase of ASBO with population size equal to 25.

TABLE V. COMPARATIVE PERFORMANCES FOR 1st TEST PROBLEM UNDER SECOND ENVIRONMENT FOR 20 INDEPENDENT TRAILS WITH POPULATION SIZE 50.

Performance	PSO	ASBO
Best	7.5523	7.5576
Worst	7.2076	7.5473
Mean	7.4609	7.5534
Std.Dev	0.0881	0.0031
C.I (95%)	$\mp$0.0411	$\mp$0.0014
C.I (99%)	$\mp$0.0560	$\mp$0.0020

TABLE VI. BEST SOLUTION RESULT FOR 1st TEST PROBLEM UNDER SECOND ENVIRONMENT WITH POPULATION SIZE 50

Catalyst	PSO	ASBO
V	0.2494	0.3202
Mg	0.3029	0.3196
B	0	0
Mo	0.2551	0.2745
La	0	0
Mn	0.1302	0.0323
Fe	0.0624	0.0533
Ga	0	0

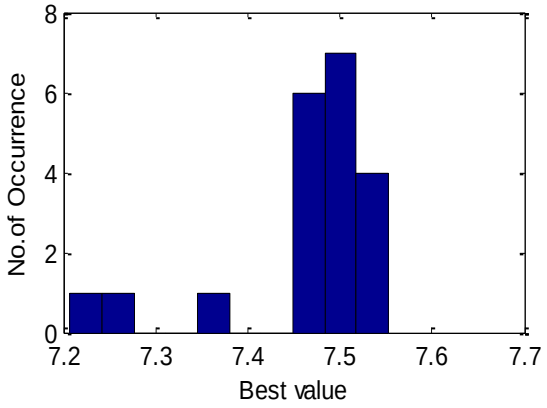

Fig.15(a) Distribution of best value over 20 trails in PSO with population size 50 for 1st test problem.

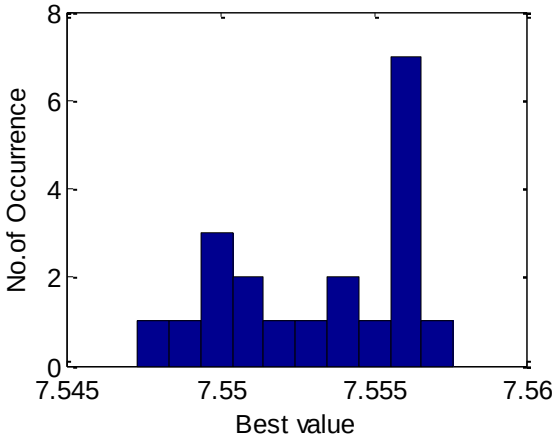

Fig.15(b) Distribution of best value over 20 trails in ASBO with population size 50 for 1st test problem

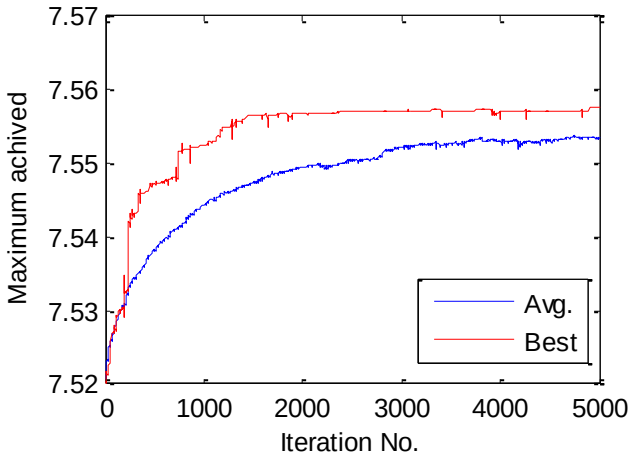

Fig.16 Average and best solution performance over 1st test problem w.r.t iterations in second phase of ASBO with population size equal to 50.

TABLE VII. COMPARATIVE PERFORMANCES FOR 1st TEST PROBLEM UNDER SECOND ENVIRONMENT FOR 20 INDEPENDENT TRAILS WITH POPULATION SIZE 75.

Performance	PSO	ASBO
Best	7.5333	7.5577
Worst	7.3388	7.5544
Mean	7.4907	7.5564
Std.Dev	0.0440	0.0011
C.I (95%)	$\mp$0.0205	$\mp$0.0005
C.I (99%)	$\mp$0.0280	$\mp$0.0007

TABLE VIII. BEST SOLUTION RESULT FOR 1st TEST PROBLEM UNDER SECOND ENVIRONMENT WITH POPULATION SIZE 75

Catalyst	PSO	ASBO
V	0.3137	0.3203
Mg	0.3273	0.3204
B	0	0
Mo	0.2832	0.3482
La	0	0
Mn	0.0190	0.0030
Fe	0.0567	0.0081
Ga	0	0

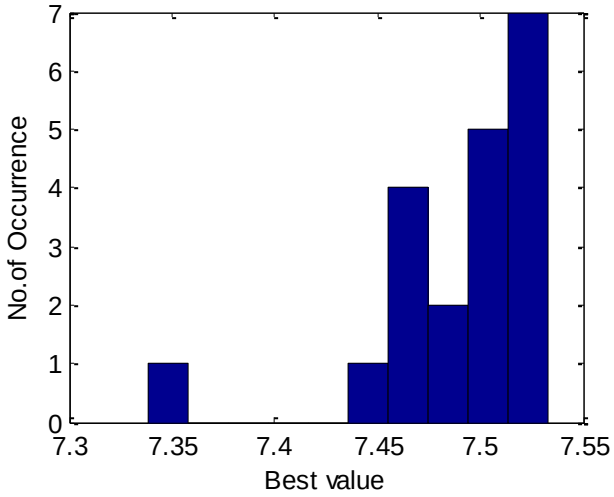

Fig.17 Distribution of best value over 20 trails in PSO with population size 75 for 1st test problem.

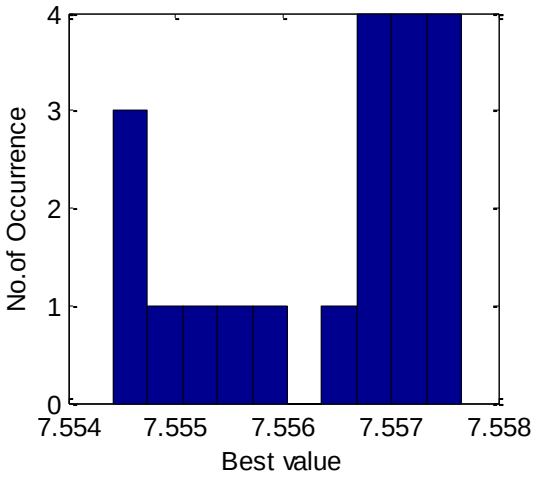

Fig.18 Distribution of best value over 20 trails in ASBO with population size 75 for 1st test problem.

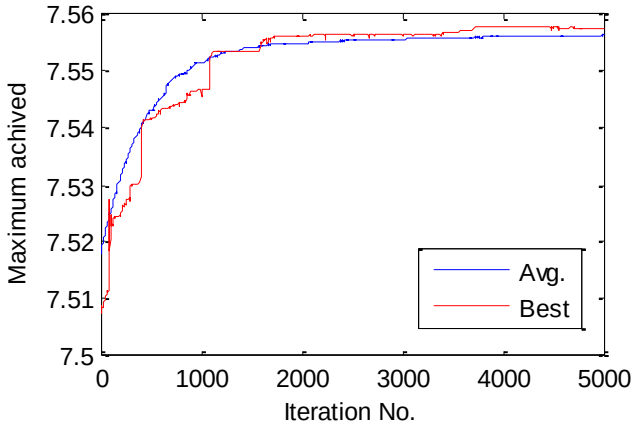

Fig.19 Average and best solution performance over 1st test problem w.r.t iterations in second phase of ASBO with population size equal to 75.

TABLE IX. COMPARATIVE PERFORMANCES FOR 1st TEST PROBLEM UNDER SECOND ENVIRONMENT FOR 20 INDEPENDENT TRAILS WITH POPULATION SIZE 100.

Performance	PSO	ASBO
Best	7.5404	7.5578
Worst	7.4689	7.5558
Mean	7.5079	7.5572
Std.Dev	0.0215	0.0005
C.I (95%)	$\mp$0.0100	$\mp$0.0002
C.I (99%)	$\mp$0.0137	$\mp$0.0003

TABLE X. BEST SOLUTION RESULT FOR 1st TEST PROBLEM UNDER SECOND ENVIRONMENT WITH POPULATION SIZE 100

Catalyst	PSO	ASBO
V	0.3191	0.3203
Mg	0.3184	0.3206
B	0	0
Mo	0.2895	0.3496
La	0	0
Mn	0.0261	0.0064
Fe	0.0470	0.0031
Ga	0	0

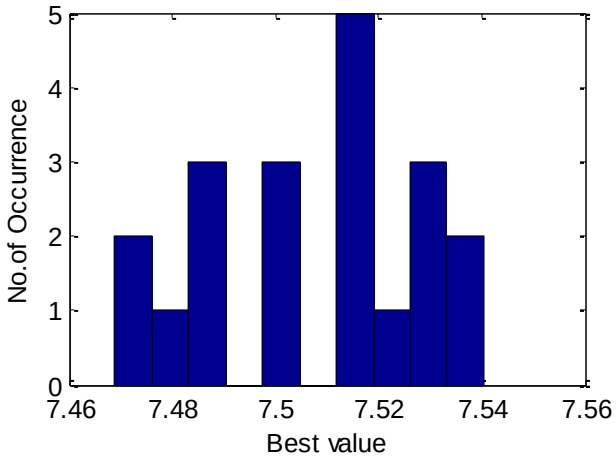

Fig.20 Distribution of best value over 20 trails in PSO with population size 100 for 1st test problem.

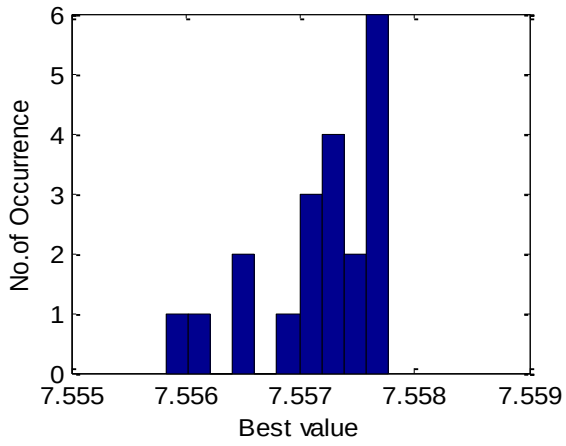

Fig.21 Distribution of best value over 20 trails in ASBO with population size 100 for 1st test problem.

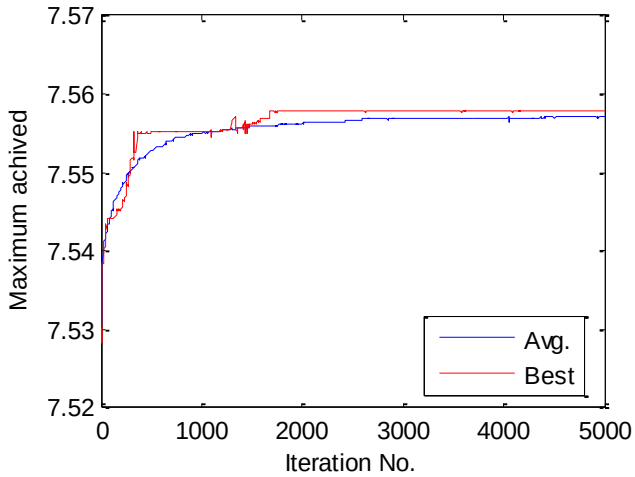

Fig.22 Average and best solution performance over 1st test problem w.r.t iterations in second phase of ASBO with population size equal to 100.

C. TEST BENCH 2: Under First test environmeont

In second problem again for 50 independent trials have given for both algorithms and results have analysed graphically and numerically and represented in Fig.23 to Fig.28.Eventhough PSO manage to deliver the global solution but the success rate is very low whereas ASBO has delivered the global solution in all time as shown in Table XI and component best values have shown in Table XII.

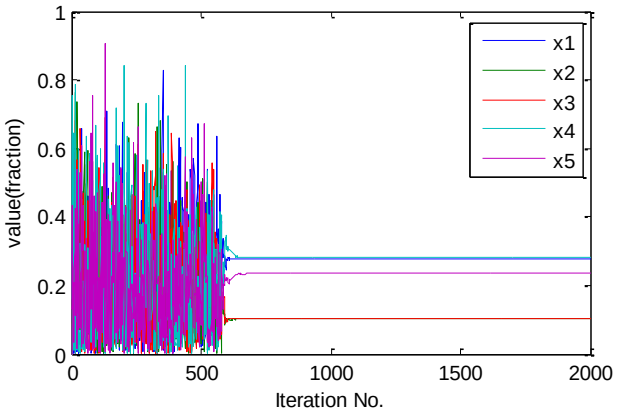

Fig.23 PSO catalyst component estimation by PSO with population size 100 for 2nd test problem.

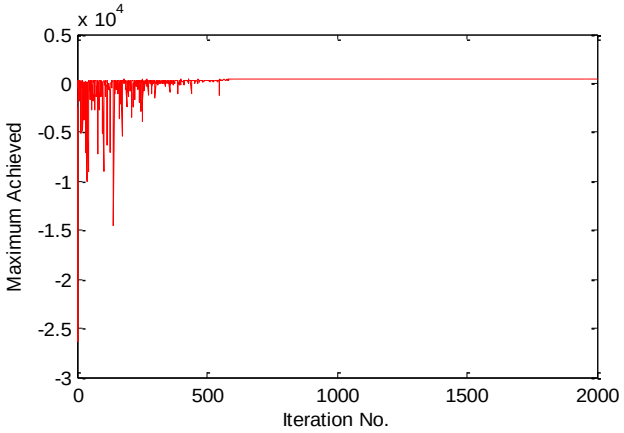

Fig.24 Yield achived by PSO in 2nd test problem

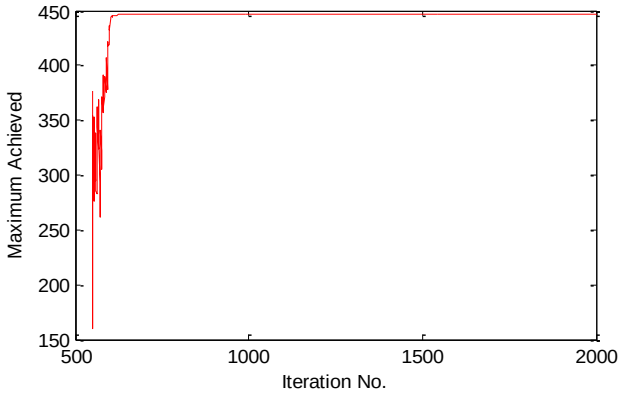

Fig.25 Yield achived by PSO in later generation for 2nd test problem.

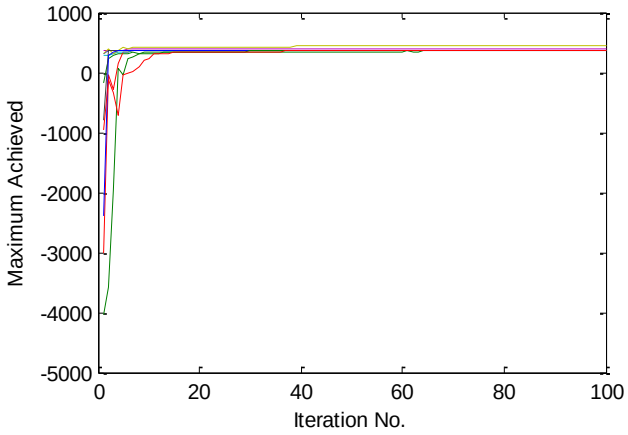

Fig.26 Yield achived by ASBO in first stage for 2nd test problem with all 10 different population

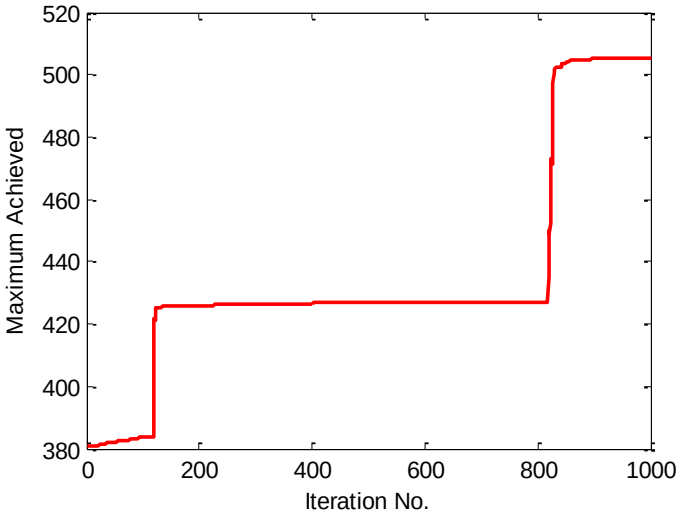

Fig.27 Yield achived by ASBO in 2nd stage for 2nd test problem

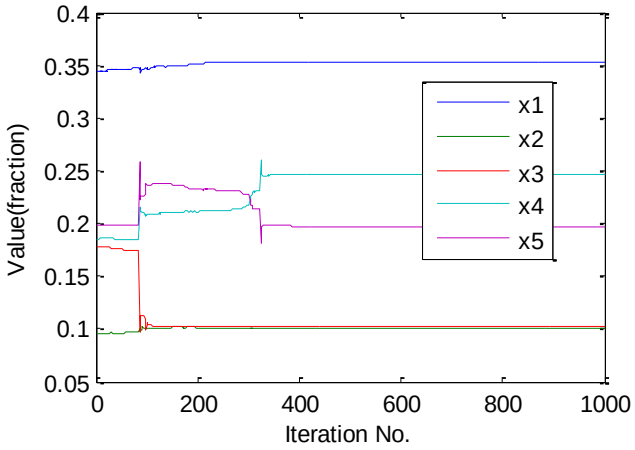

Fig.28 Yield achived by ASBO in first stage for 2nd Test problem with all 10 different population

TABLE XI. COMPARATIVE PERFORMANCES FOR 2nd TEST PROBLEM UNDER FIRST ENVIRONMENT FOR 50 INDEPENDENT TRAILS WITH POPULATION SIZE EQUAL TO 100

Performance	PSO	ASBO
Best	505.09487	505.0948
Worst	375.7112	505.0948
Mean	497.0352	505.0948
Std.Dev	26.10707	7.97380e-013
C.I (95%)	∓ 7.4174	∓2.2655e-013
C.I (99%)	∓ 9.8874	∓3.0199e-013

TABLE XII. BEST SOLUTION RESULT FOR 2nd TEST PROBLEM UNDER FIRST ENVIRONMENT

Catalyst	PSO	ASBO
X1	0.3525	0.3525
X2	0.1003	0.1003
X3	0.1025	0.1025
X4	0.2469	0.2469
X5	0.1969	0.1969

D. TEST CASE 2: EXPERIMENT RESULT UNDER SECOND ENVIRONMENT

As in case of the first test problem, experiment has done with a second test problem with different population size for 20 independent trials and results are shown in Fig. 29 to Fig.37.With population size 25, PSO could deliver the optimal value while ASBO has not only deliver the optimal value but also its precision is very high as shown in Table XIII.

TABLE XIII. COMPARATIVE PERFORMANCES FOR 2nd TEST PROBLEM UNDER SECOND ENVIRONMENT FOR 20 INDEPENDENT TRAILS WITH POPULATION SIZE 25

Performance	PSO	ASBO
Best	502.5570	505.0949
Worst	270.6913	505.0616
Mean	380.2910	505.0917
Std.Dev	55.0546	0.0084
C.I (95%)	∓25.6799	∓0.0039
C.I (99%)	∓35.0236	∓0.0053

TABLE XIV. BEST SOLUTION RESULT FOR 2nd TEST PROBLEM UNDER SECOND ENVIRONMENT WITH POPULATION SIZE 25

Catalyst	PSO	ASBO
X1	0.3525	0.3525
X2	0.1001	0.1003
X3	0.1027	0.1026
X4	0.2051	0.2469
X5	0.2385	0.1969

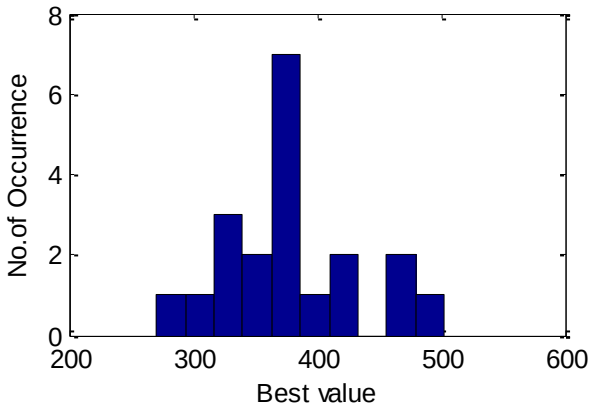

Fig.29 Distribution of best value over 20 trails in PSO with population size 25 for 2nd test problem

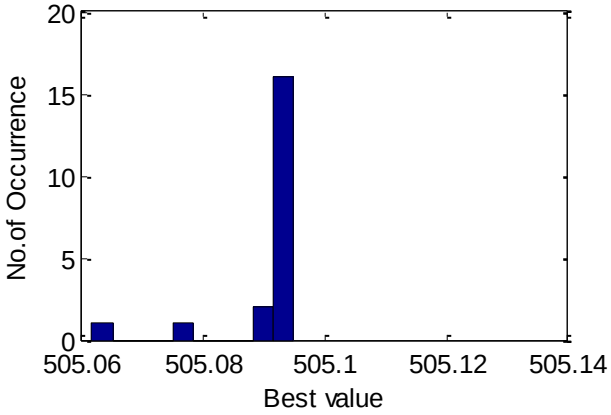

Fig.30 Distribution of best value over 20 trails in ASBO with population size 25 for 2nd test problem.

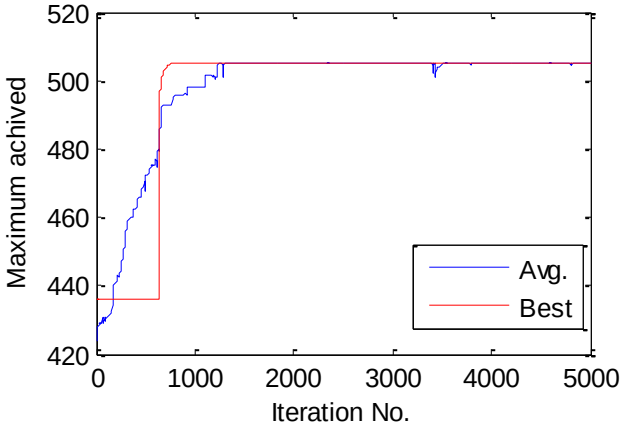

Fig.31 Average and best solution performance over 2nd test problem w.r.t iterations in second phase of ASBO with population size equal to 25.

TABLE XV. COMPARATIVE PERFORMANCES FOR 2nd TEST PROBLEM UNDER SECOND ENVIRONMENT FOR 20 INDEPENDENT TRAILS WITH POPULATION SIZE 50

Performance	PSO	ASBO
Best	505.0949	505.0949
Worst	354.6355	505.0949
Mean	436.2163	505.0949
Std.Dev	54.6224	0.0000
C.I (95%)	∓25.4783	∓0.0
C.I (99%)	∓34.7487	∓0.0

TABLE XVI. BEST SOLUTION RESULT FOR 2nd TEST PROBLEM UNDER SECOND ENVIRONMENT WITH POPULATION SIZE 50

Catalyst	PSO	ASBO
X1	0.3525	0.3525
X2	0.1003	0.1003
X3	0.1026	0.1026
X4	0.2469	0.2469
X5	0.1969	0.1969

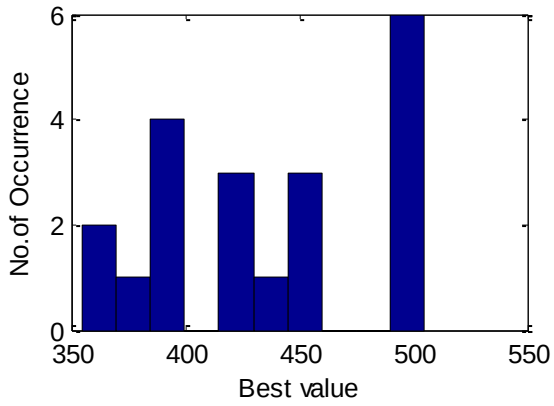

Fig.32 Distribution of best value over 20 trails in PSO with population size 50 for 2nd test problem

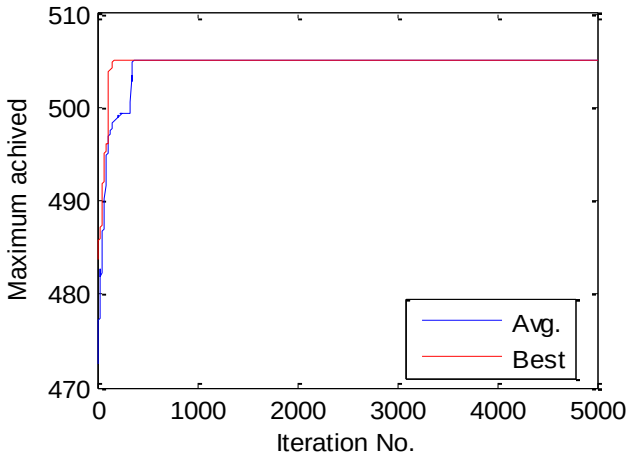

Fig.33 Average and best solution performance over 2nd test problem w.r.t iterations in second phase of ASBO with population size equal to 50.

TABLE XVII. COMPARATIVE PERFORMANCES FOR 2nd TEST PROBLEM UNDER SECOND ENVIRONMENT FOR 20 INDEPENDENT TRAILS WITH POPULATION SIZE 75

Performance	PSO	ASBO
Best	505.0949	505.0949
Worst	367.6223	505.0949
Mean	454.3058	505.0949
Std.Dev	56.0447	0.0000
C.I (95%)	∓26.1417	∓0.0
C.I (99%)	∓35.6535	∓0.0

TABLE XVIII. BEST SOLUTION RESULT FOR 2nd TEST PROBLEM UNDER SECOND ENVIRONMENT WITH POPULATION SIZE 75

Catalyst	PSO	ASBO
X1	0.3525	0.3525
X2	0.1003	0.1003
X3	0.1026	0.1026
X4	0.2469	0.2469
X5	0.1969	0.1969

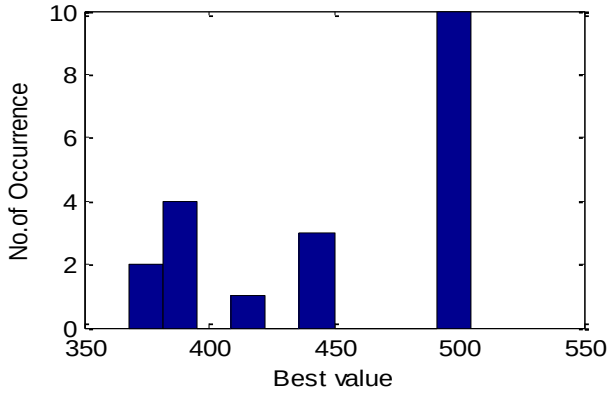

Fig.34 Distribution of best value over 20 trails in PSO with population size 75 for 2nd test problem

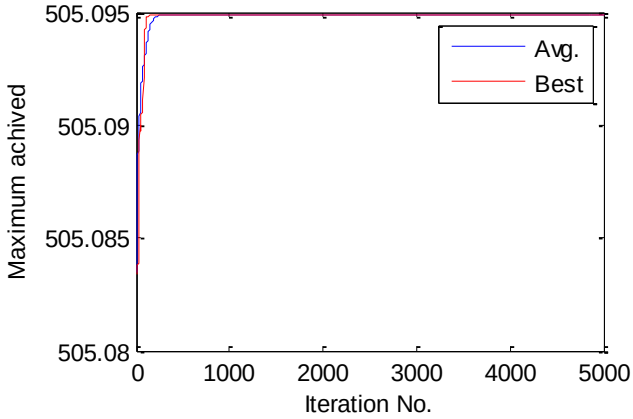

Fig.35 Average and best solution performance over 2nd test problem w.r.t iterations in second phase of ASBO with population size equal to 75.

TABLE XIX. COMPARATIVE PERFORMANCES FOR 2nd TEST PROBLEM UNDER SECOND ENVIRONMENT FOR 20 INDEPENDENT TRAILS WITH POPULATION SIZE 100

Performance	PSO	ASBO
Best	505.0949	505.0949
Worst	366.9733	505.0949
Mean	456.5000	505.0949
Std.Dev	49.0142	0.0000
C.I (95%)	∓22.8624	∓0.0
C.I (99%)	∓31.1809	∓0.0

TABLE XX. BEST SOLUTION RESULT FOR 2nd TEST PROBLEM UNDER SECOND ENVIRONMENT WITH POPULATION SIZE 100

Catalyst	PSO	ASBO
X1	0.3525	0.3525
X2	0.1003	0.1003
X3	0.1026	0.1026
X4	0.2469	0.2469
X5	0.1969	0.1969

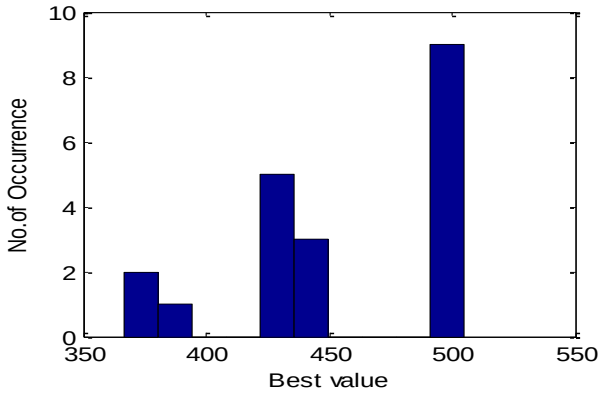

Fig.36 Distribution of best value over 20 trails in PSO with population size 100 for 2nd test problem

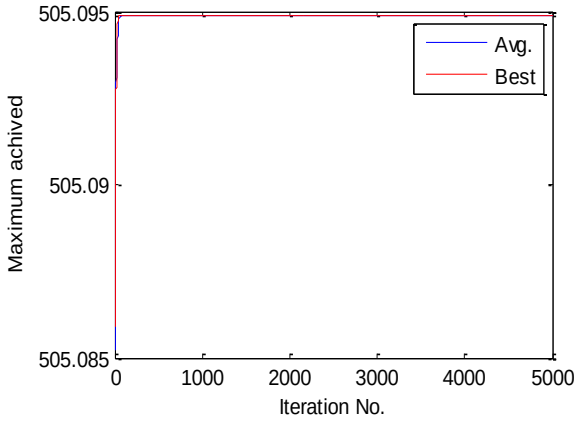

Fig.37 Average and best solution performance over 2nd test problem w.r.t iterations in second phase of ASBO with population size equal to 100.

VI. Conclusion

We have applied a newly developed global optimization method ASBO to a heterogeneous catalysis problem whose fitness function is unknown. Its objective is to estimate the value of a potential solution to a problem which is not defined by a mathematical expression but by a set of observations, each of the high monetary cost. We compare the results obtained by PSO in different aspects like capability of delivering global solution and effects of size of population in the exploration of solutions. We show that the use of ASBO improves the robustness compared to PSO in all aspects. The catalyst sector has clearly become the next leading proponent of combinatorial chemistry after the pharmaceutical and agrochemical sectors. Combinatorial catalysis enable us to look with a new eye at what, unfortunately, has been many times characterized as a mature and old science or industry.

Conflict of Interests

The authors declare that there is no conflict of interests.

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