Hyperfine Interact (2008) 181:127–130
DOI 10.1007/s10751-008-9703-z

Fitting PAC spectra with a hybrid algorithm

M. A. Alves · A. W. Carbonari

Published online: 6 August 2008
© Springer Science + Business Media B.V. 2008

Abstract A hybrid algorithm (HA) that blends features of genetic algorithms (GA) and simulated annealing (SA) was implemented for simultaneous fits of perturbed angular correlation (PAC) spectra. The main characteristic of the HA is the incorporation of a selection criterion based on SA into the basic structure of GA. The results obtained with the HA compare favorably with fits performed with conventional methods.

Keywords Genetic algorithms · Simulated annealing · Data fitting · Perturbed angular correlation

PACS 23.20.En · 07.05.Kf

1 Introduction

Several procedures can be used in the fitting of perturbed angular correlation (PAC) spectra; the most common ones being based on gradient methods. Although these methods can be very precise and fast, they have limitations since they depend on the correct choice of starting parameters and can get trapped in local minima of the parameter space. These difficulties are compounded when several spectra need to be

M. A. Alves
Instituto de Aeronáutica e Espaço, São José dos Campos, SP, Brazil

M. A. Alves
Faculdade de Engenharia de Guaratinguetá, Guaratinguetá, SP, Brazil
e-mail: mauro@sepn.org

A. W. Carbonari (✉)
Instituto de Pesquisas Energéticas e Nucleares, São Paulo, SP, Brazil
e-mail: carbonar@ipen.br

fitted simultaneously to the same model. A number of algorithms can be used to solve complex optimization problems, such as data fitting. Genetic algorithms (GA) [1] are one of the best methods due to their parallel search capability, lack of dependence on initial values and insensitivity to local minima. Simulated annealing (SA) [2] is another method that employs a random search on the parameter space and also avoids becoming trapped in local minima. Here, a hybrid algorithm (HA) [3] that combines the search properties of GA and SA was implemented to fit simultaneously PAC spectra. Fitting data with the HA results in considerable savings in analysis time, and allows the determination of good fits without well-defined initial values.

2 The hybrid algorithm

Genetic algorithms are optimization tools based on the principles of the Darwinian evolution. In GA, members of a population (solutions to a problem) compete with each other to survive and reproduce (generate new solutions). Each member of the population (a chromosome) is a concatenation of encoded parameter values in the form of a string of digits. A selection operator chooses among the fittest members of the population from the current generation which one will survive and be present in the next generation. A crossover operator creates new chromosomes (children) by mixing segments of two paired chromosomes (parents). Digits in the chromosome strings are randomly changed by a mutation operator. A fixed-size initial population is randomly generated and new populations result from the application of the selection, crossover and mutation operators. This process is repeated until a termination criterion, such as the average fitness of the population, is satisfied.

Simulated annealing is another technique that exploits an analogy with a natural system. Annealing is the process in which a heated metal is cooled very slowly until it reaches a minimum-energy state. The SA algorithm searches for the optimum solution moving randomly in the solution space seeking for a solution that minimizes the value of an objective function. The state of a system in SA is defined by the values of the parameters that characterize the solution. State transitions take place by changes in these parameters according to a probability distribution function (PDF) given by $p = exp(-dE/T)$, where dE is the change in energy of the system and T is a control parameter (temperature). In SA, changes that lead to lower energy are always accepted while an increase in energy is accepted according to the PDF. The system initially is at a high temperature that is reduced gradually until the system solidifies at a low energy solution.

SA and GA are very useful in many applications but they have some disadvantages. For instance, GA does not offer any guarantee that an optimal solution will be reached, and SA may be computationally intensive. To improve the search ability of the GA, candidate solutions from the population pool of a generation are selected based on the PDF. Assuming that f' is the fitness of the fittest member of a population, if another member has a fitness f'' such that $f'' > f'$, then the new member is selected, if not the new member is selected according to $p = exp(-(f' - f'')/T)$. The final state is reached when T approaches zero, which corresponds to the optimal solution. As the temperature decreases, it is less likely that a “bad” solution is chosen and a “good” solution is discarded. Thus, it is not necessary to generate a new and complete population of solutions every generation. In the HA, the probability of the fittest members of a population being selected is higher than in a conventional

Table 1 Dynamical parameters derived from the fitting of 0.1 and 0.2 at.% yttria-doped zirconia PAC spectra with the HA

Sample	E_h (eV) w_0 (rad/s)	E_d (eV) w_{d0} (rad/s)	E_t (eV) w_{t0} (rad/s)
0.1 at%	0.89(8)	1.54(5)	1.01(7)
	$15.0(5) \times 10^{13}$	$14.1(7) \times 10^{13}$	$2.3(8) \times 10^{11}$
0.2 at%	0.86(7)	1.49(7)	0.95(8)
	$11.6(3) \times 10^{13}$	$14.1(6) \times 10^{13}$	$3.9(7) \times 10^{11}$

GA. Since the fittest member in the GA population influences the evolution of the entire population, the HA has better convergence.

The parameters that define the best fit to a model that characterizes the experimental data are found by the method of least squares. The goodness of the fit is described by χ^2, which in its reduced form is defined as

$$\chi^2 = \frac{1}{n - n_p} \sum_{i}^{N} \frac{(y_i - y(x_i, a_i))^2}{\sigma_i^2} \tag{1}$$

where y_i are the experimental data, $y(x_i, a_i)$ is the function chosen as model, a_i are the model parameters, σ_i the variance , n the number of data points, and n_p the number of free parameters used in the fitting. The fitness of the solution, i.e., the quality of a given set of parameters used in the fit is defined as the inverse of χ^2. The HA is used to find the best combination of parameters in the solution space such that the inverse of χ^2 is minimized.

3 An application example of the HA

As an example, the HA was used to fit PAC data from two samples of yttria-doped zirconia (0.1 and 0.2 at%). Fifteen PAC spectra were collected for each sample in the temperature range of 1,000 to 1,300°C [4]. At these temperatures, zirconia ceramic is an excellent ionic conductor due to the high mobility of oxygen vacancies. A vacancy can be trapped by a PAC probe nucleus; while trapped, the vacancy can hop between equivalent sites around the probe nucleus, and it will eventually detrap. The effects of the fluctuating electric field produced by the moving vacancy on the perturbation function $G_{22}(t)$ (the PAC spectrum) can be represented by a stochastic model adapted in order to take into account the effects of static defects [5, 6]. With this model it is possible to find the values of physical quantities related to the dynamics of the oxygen vacancies. The model assumes that the fluctuating electric fields in the material are caused by thermal processes, and that the hopping, trapping and detrapping rates follow an Arrhenius behavior given by $w = w_0 e^{-E_h/kT}$, $w_d = w_{d0} e^{-E_d/kT}$ and $w_t = w_{t0} e^{-E_t/kT}$, where E_h, E_t and E_d are the hopping barrier between equivalent trap sites, the trapping and detrapping energy barriers, respectively; w_0, w_{d0} and w_{t0} are prefactors that depend on physical properties of the material being analyzed.

The HA was used to fit simultaneously the two sets of data to extract the values of the prefactors and of E_h, E_t and E_d. Table 1 lists the dynamical parameters determined by the simultaneous fits with the HA, and Fig. 1 shows an example of a fit.

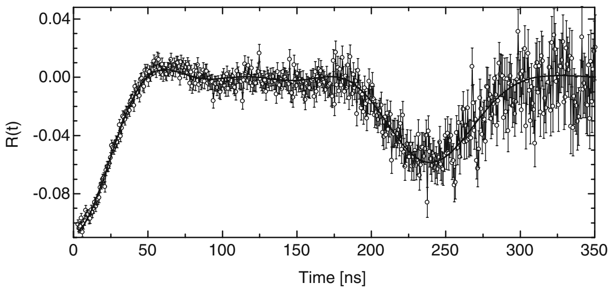

Fig. 1 PAC time spectra for 0.2 at% yttria-doped tetragonal zirconia at 1,250°C and HA fit

All the spectra were also fitted individually using the values of the parameters obtained with the simultaneous fits. The values of χ^2 for the individual fits are in the interval $1.08 < \chi_{\mu}^2 < 1.25$, showing that the overall quality of the fits is satisfactory. The comparison of the values listed in Table 1 with those found in [6], and references therein, shows a good agreement between them.

4 Discussion and conclusions

The simultaneous fitting of sets of data using conventional methods can be a time-consuming task. The use of the HA is advantageous since it eliminates most of the repetitive work, needs no intervention during its execution or "correct guesses" of initial values. Also, intermediate results obtained with the HA can serve to initialize fitting programs based on other methods, thus eliminating the trial and error process to determine the ideal starting parameters. The HA can be seen as an important tool for fitting PAC spectra, it is rather simple to implement and provides quality solutions with less effort.

References

1. Goldberg D.E.: Genetic Algorithms in Search, Optimization and Machine Learning. Addison Wesley, New York (1989)
2. Kirkpatrick, S., Gellat, C., Vecchi, M.P.: Optimization by simulated annealing. Science **220**, 671–680 (1983)
3. Pakhira, M.K.: A hybrid genetic algorithm using probabilistic selection. J. Inst. Eng. **84**, 23–30 (2003)
4. Lee, T.A.: Dependence of oxygen vacancy concentration in zirconia on processing parameters and reducing conditions. Dissertation, Oregon State University (2000)
5. Evenson, W.E., Lu, J., Winz, M.W., Gardner, J.A., Zacate, M.O., Lee, T.A., Mommer, N.: Stochastic PAC models for vacancy motions with trapping and detrapping. Hyperfine Interact. **120/121**, 427–431 (1999)
6. Alves, M.A.: The dynamics of oxygen vacancies in zirconia: an analysis of PAC data. Dissertation, Oregon State University (2003)