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A reactive BGK-type model: influence of elastic collisions and chemical interactions / Monaco, Roberto; Pandolfi, Miriam; A. J., Soares. - (2005), pp. 70-75. ( 24th International Symposium on Rarefied Gas Dynamics Bari (ITA) 10-16 July 2004) [10.1063/1.1941516].

*Availability:*

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DOI:10.1063/1.1941516

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Citation: *AIP Conf. Proc.* **762**, 70 (2005); doi: 10.1063/1.1941516

View online: <http://dx.doi.org/10.1063/1.1941516>

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# A reactive BGK-type model: influence of elastic collisions and chemical interactions

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**Abstract.** A BGK-type model for a reactive multicomponent gas undergoing chemical bimolecular reactions is here presented. The mathematical and physical consistency of the model is stated in detail. The relaxation process towards local Maxwellians, depending on mass and numerical densities of each species, as well as on common mean velocity and temperature, is investigated with respect to chemical equilibrium. Such a trend is numerically tested within the hydrogen-air reaction mechanism.

## INTRODUCTION

A large piece of research works has been addressed to simplified kinetic models of the Boltzmann equation, stimulated by the mathematical complexity of the true collision operator. A wide literature evidences the relevance of BGK-type models and their reliability also for computing gas transport properties far from equilibrium, assuming relaxation of the distribution function towards either a local Maxwellian or an anisotropic Gaussian [1, 2]. Extensions of BGK-type models to multicomponent systems can be found in [3], and more recently in [4] where a model satisfying the main properties of the true Boltzmann collision operator is presented. On this line, it seems to be a new interesting topic to deal with a BGK approximation of the extended Boltzmann equation for chemically reacting gases. The general structure of chemistry source terms, conservation and equilibrium properties in kinetic equations have been widely focused in [5]. More in detail considering bimolecular reactions, the exact kinetic equations which will be referred to in this work are those derived in paper [6].

A first attempt to build a BGK-type model for a mixture of four gas species with bimolecular chemical reaction of type



has been performed in paper [7], where the kinetic equations have been written in the form

$$\frac{\partial f_i}{\partial t} + \mathbf{v} \cdot \nabla f_i = \nu_i [\tilde{f}_i(\mathbf{v}) - f_i(\mathbf{v})] + \tilde{\mathcal{R}}_i[\underline{f}, \underline{f}](\mathbf{v}), \quad i = 1, \dots, 4. \quad (2)$$

In Equation (2)  $\tilde{f}_i$  denotes the local Maxwellian distribution of species  $i$  depending on the number densities  $n_i$  of each  $i$ -species, common mean velocity  $\mathbf{u}$  and temperature  $T$ , i. e.

$$\tilde{f}_i(\mathbf{v}) = n_i \left( \frac{m_i}{2\pi k_B T} \right)^{\frac{3}{2}} \exp \left( -\frac{m_i(\mathbf{v} - \mathbf{u})^2}{2k_B T} \right). \quad (3)$$

The term  $\tilde{\mathcal{R}}_i$  approximates the true reactive operator  $\mathcal{R}_i$ , introduced in Ref. [6], which includes the effects of the inelastic chemical process. In  $\tilde{\mathcal{R}}_i$  the chemical gain term involves mechanical equilibrium and chemical disequilibrium in such a way that the model verifies the indifferenciability principle and conservation of mass, momentum and total energy (kinetic plus internal chemical bond energy). Moreover the  $H$ -theorem holds true under a suitable hypothesis. In the last part of the paper the behavior of the model is numerically tested with respect to its trend to equilibrium for different initial conditions, in order to show the influence of elastic collisions and reactive interactions. The numerical experiments are performed for the elementary reaction occurring in the Hydrogen-Air reaction mechanism, namely  $H_2O + H \rightleftharpoons OH + H_2$  which is typical in Hydrogen combustion applications.

## KINETIC MODEL

With reference to Eq. (2), the microscopic state of a mixture of four gas species, say  $A_i$ ,  $i = 1, \dots, 4$ , is defined by the one-particle distribution function  $f_i = f_i(t, \mathbf{x}, \mathbf{v})$ ,  $t \in \mathbf{R}_+$ ,  $\mathbf{x} \in \mathbf{R}^3$ ,  $\mathbf{v} \in \mathbf{R}^3$  for each species  $A_i$  with molecular mass  $m_i$  and internal energy  $E_i$  such that  $m_1 + m_2 = m_3 + m_4$ . In addition, setting  $\Delta E = E_3 + E_4 - E_1 - E_2 > 0$  the forward reaction turns out to be the endothermic one. The time and space dependence, if useless, will be omitted in the sequel.

*Exact equations.* The exact kinetic equations for the reactive gas mixture are given by

$$\frac{\partial f_i}{\partial t} + \mathbf{v} \cdot \nabla f_i = J_i[\underline{f}](\mathbf{v}) + \mathcal{R}_i[\underline{f}](\mathbf{v}), \quad \underline{f} = \{f_1, \dots, f_4\}, \quad (4)$$

$$J_i[\underline{f}](\mathbf{v}) = G_i[\underline{f}](\mathbf{v}) - L_i[\underline{f}](\mathbf{v}), \quad \mathcal{R}_i[\underline{f}](\mathbf{v}) = \mathcal{G}_i[\underline{f}](\mathbf{v}) - \mathcal{L}_i[\underline{f}](\mathbf{v}). \quad (5)$$

The gain and loss terms  $G_i, L_i$  due to elastic collisions, and  $\mathcal{G}_i, \mathcal{L}_i$  due to chemical reactions, are not reported here for brevity, but can be recovered in paper [6]. They satisfy the following properties

$$\int_{\mathbf{R}^3} J_i[\underline{f}](\mathbf{v}) d\mathbf{v} = 0, \quad i = 1, \dots, 4 \quad (6)$$

$$\int_{\mathbf{R}^3} \mathcal{R}_1[\underline{f}](\mathbf{v}) d\mathbf{v} = \int_{\mathbf{R}^3} \mathcal{R}_2[\underline{f}](\mathbf{v}) d\mathbf{v} = - \int_{\mathbf{R}^3} \mathcal{R}_3[\underline{f}](\mathbf{v}) d\mathbf{v} = - \int_{\mathbf{R}^3} \mathcal{R}_4[\underline{f}](\mathbf{v}) d\mathbf{v}. \quad (7)$$

The former is typical of the elastic collision operator and accounts for conservation of the particle numbers of each species, the latter is due to the fact that the evolution of each species is predicted by chemical exchanges according to the bimolecular reaction (1). For what concerns mechanical equilibrium, each  $J_i[\underline{f}](\mathbf{v})$  vanishes when  $\underline{f}$  is a Maxwellian given by (3). Conversely, for what concerns chemical equilibrium, each  $\mathcal{R}_i[\underline{f}](\mathbf{v})$  vanishes if the distribution functions are Maxwellian and, in addition, the following condition holds

$$(m_3 m_4)^3 \tilde{f}_1(\mathbf{v}) \tilde{f}_2(\mathbf{w}) = (m_1 m_2)^3 \tilde{f}_3(\mathbf{v}_1) \tilde{f}_4(\mathbf{w}_1). \quad (8)$$

In Eq. (8),  $\mathbf{v}, \mathbf{w}$  and  $\mathbf{v}_1, \mathbf{w}_1$  are the pre and post-collisional velocities, respectively.

The post-collisional velocities,  $\Omega'$  being the unit vector of the relative post collisional velocity, are given by

$$\begin{aligned} \mathbf{v}_1 &= r_1 \mathbf{v} + r_2 \mathbf{w} - r_4 V \Omega', & \mathbf{w}_1 &= r_1 \mathbf{v} + r_2 \mathbf{w} + r_3 V \Omega', \\ r_i &= \frac{m_i}{m_1 + m_2}, & V &= \sqrt{\frac{(\mathbf{w} - \mathbf{v})^2}{\mu} - \frac{2\Delta E}{r_1 m_2}}, & \mu &= \frac{m_3 m_4}{m_1 m_2}, \end{aligned} \quad (9)$$

Condition (8), as it can be easily seen, implies the mass-action-law of chemical equilibrium

$$\frac{n_1 n_2}{n_3 n_4} = \mu^{-\frac{3}{2}} \exp\left(\frac{\Delta E}{k_B T}\right). \quad (10)$$

*Approximated equations.* Following the procedure adopted in paper [7], the BGK-type approximation of the exact equations (4) consists in inserting in both elastic and inelastic gain terms Maxwellian distributions with parameters  $n_i$  which do not satisfy condition (10) and, thus, do not imply chemical equilibrium. Such a procedure is justified since, in general [8], the relaxation time of elastic collision is of some order of magnitude smaller than the one of chemical interactions. The BGK equations then read

$$\frac{\partial f_i}{\partial t} + \mathbf{v} \cdot \nabla f_i = \tilde{J}_i[\underline{f}, \tilde{\underline{f}}](\mathbf{v}) + \tilde{\mathcal{R}}_i[\underline{f}, \tilde{\underline{f}}](\mathbf{v}), \quad i = 1, \dots, 4, \quad (11)$$

where  $\tilde{J}_i[\underline{f}, \tilde{\underline{f}}](\mathbf{v})$  and  $\tilde{\mathcal{R}}_i[\underline{f}, \tilde{\underline{f}}](\mathbf{v})$  approximate the true collision operators  $J_i$  and  $\mathcal{R}_i$ , according to the above said conjecture and assuming that the distribution functions satisfy conditions

$$\int_{\mathbf{R}^3} \varphi_i(\mathbf{v}) f_i(\mathbf{v}) d\mathbf{v} = \int_{\mathbf{R}^3} \varphi_i(\mathbf{v}) \tilde{f}_i(\mathbf{v}) d\mathbf{v}, \quad (12)$$

with  $\varphi_i$  alternatively equal to  $m_i$ ,  $m_i \mathbf{v}$  or  $\frac{1}{2}m_i \mathbf{v}^2 + E_i$ . Conditions (12) imply that the distributions  $f_i$  and  $\tilde{f}_i$ , which possess the same moments  $n, \rho \mathbf{u}, T$  ( $\rho$  being the total density), will be different only for what deals with the computation of higher moments.

The explicit expressions of  $\tilde{J}_i$  and  $\tilde{\mathcal{R}}_i$  will be here reported from paper [7], avoiding the details of their deduction. Adopting constant cross sections of Maxwell molecules,  $\alpha_{ij}$ , the BGK-type approximation of the elastic collision gain and loss terms, leads to the expression

$$\tilde{J}_i[\underline{f}, \underline{f}](\mathbf{v}) = v_i [\tilde{f}_i(\mathbf{v}) - f_i(\mathbf{v})], \quad v_i = 4\pi \sum_{j=1}^4 \alpha_{ij} n_j. \quad (13)$$

Conversely, the inelastic collision gain and loss terms have the expressions

$$\begin{aligned} \tilde{\mathcal{R}}_1[\underline{f}, \underline{f}](\mathbf{v}) &= \gamma(T) n_4 \tilde{f}_3 - \eta(T) n_2 f_1, & \tilde{\mathcal{R}}_2[\underline{f}, \underline{f}](\mathbf{v}) &= \gamma(T) n_3 \tilde{f}_4 - \eta(T) n_1 f_2 \\ \tilde{\mathcal{R}}_3[\underline{f}, \underline{f}](\mathbf{v}) &= \eta(T) n_2 \tilde{f}_1 - \gamma(T) n_4 f_3, & \tilde{\mathcal{R}}_4[\underline{f}, \underline{f}](\mathbf{v}) &= \eta(T) n_1 \tilde{f}_2 - \gamma(T) n_3 f_4 \end{aligned} \quad (14)$$

where

$$\begin{aligned} \gamma(T) &= \frac{4\pi\beta \theta S(T)}{k_B T}, & \eta(T) &= \gamma(T) (\mu)^{\frac{3}{2}} \exp\left[-\frac{\Delta E}{k_B T}\right], & \theta &= \frac{m_3 m_4}{m_1 + m_2}, \\ S(T) &= \frac{\xi}{\pi} \sqrt{\frac{2\pi k_B T}{\theta}} \exp\left[-\frac{\theta \xi^2}{2k_B T}\right] + \left[\frac{k_B T}{\theta} - \xi^2\right] \left[1 - \operatorname{erf}\left(\sqrt{\frac{\theta}{2k_B T}} \xi\right)\right] \end{aligned} \quad (15)$$

$\xi$  being the exothermic threshold velocity,  $\beta$  a scalar factor and  $k_B$  the Boltzmann constant. Expressions (14) have been derived using the proper cross sections, related to exothermic and endothermic reactions, proposed in paper [9].

## CONSISTENCY OF THE MODEL

On the line of standard procedures and notations of classical kinetic theory [10], and by using assumption (12), the following properties can be proven.

**Property 1** *The approximated elastic collision term  $\tilde{J}_i$  is such that*

$$\int_{\mathbf{R}^3} \tilde{J}_i[\underline{f}, \underline{f}](\mathbf{v}) d\mathbf{v} = 4\pi \sum_{j=1}^4 \alpha_{ij} n_j \int_{\mathbf{R}^3} [\tilde{f}_i(\mathbf{v}) - f_i(\mathbf{v})] d\mathbf{v} = 0, \quad i = 1, \dots, 4 \quad (16)$$

This property means that elastic collisions only, when modeled by the BGK equations (2), do not modify the species of the incoming particles. In fact, from condition (16), it results

$$\int_{\mathbf{R}^3} \left( \frac{\partial f_i}{\partial t} + \mathbf{v} \cdot \nabla f_i \right)_{\text{elast}} d\mathbf{v} = 0 \implies \left( \frac{dn_i}{dt} \right)_{\text{elast}} = 0. \quad (17)$$

**Property 2** *The approximated reactive collision terms  $\tilde{\mathcal{R}}_i$ ,  $i = 1, \dots, 4$ , are such that*

$$\int_{\mathbf{R}^3} \tilde{\mathcal{R}}_1[\underline{f}, \underline{f}](\mathbf{v}) d\mathbf{v} = \int_{\mathbf{R}^3} \tilde{\mathcal{R}}_2[\underline{f}, \underline{f}](\mathbf{v}) d\mathbf{v} = - \int_{\mathbf{R}^3} \tilde{\mathcal{R}}_3[\underline{f}, \underline{f}](\mathbf{v}) d\mathbf{v} = - \int_{\mathbf{R}^3} \tilde{\mathcal{R}}_4[\underline{f}, \underline{f}](\mathbf{v}) d\mathbf{v}. \quad (18)$$

This property implies

$$\left( \frac{dn_1}{dt} \right)_{\text{inelast}} = \left( \frac{dn_2}{dt} \right)_{\text{inelast}} = - \left( \frac{dn_3}{dt} \right)_{\text{inelast}} = - \left( \frac{dn_4}{dt} \right)_{\text{inelast}} \quad (19)$$

which agrees with the fact that through inelastic collisions with chemical reaction (1), if one particle of  $A_1$ -species is lost then also an  $A_2$ -particle vanishes with creation of two particles of  $A_3$  and  $A_4$ -species, and viceversa. Moreover, Eqs. (19) assures that the BGK approximated model reproduces the laws of chemical kinetics.

*Conservation laws.* Conservation of mass, momentum and total energy can be stated through the following

**Property 3** For every choice of collision invariants  $\varphi_i(\mathbf{v})$ , namely  $m_i$ ,  $m_i\mathbf{v}$  or  $\frac{1}{2}m_i\mathbf{v}^2 + E_i$ , the BGK-type collision operator verifies the following equality

$$\sum_i \int_{\mathbf{R}^3} \left( \tilde{J}_i[\underline{f}, \tilde{f}](\mathbf{v}) + \tilde{\mathcal{R}}_i[\underline{f}, \tilde{f}](\mathbf{v}) \right) \varphi_i(\mathbf{v}) d\mathbf{v} = 0 \quad (20)$$

*Entropy inequality.* Elastic collisions and chemical reactions contribute to increase the entropy of the system, according to the next

**Property 4** Let  $H$  and  $\mathbf{H}$  be the functionals defined by

$$H(x, t) = \sum_i \int_{\mathbf{R}^3} f_i \log \left( \frac{f_i}{m_i^3} \right) d\mathbf{v}, \quad \mathbf{H}(x, t) = \sum_i \int_{\mathbf{R}^3} f_i \log \left( \frac{f_i}{m_i^3} \right) \mathbf{v} d\mathbf{v}. \quad (21)$$

Then

$$\frac{\partial H}{\partial t}(x, t) + \operatorname{div} \mathbf{H}(x, t) \leq 0, \quad (22)$$

provided that

$$\sum_{i=2}^4 \int_{\mathbf{R}^3} \log \left( \frac{f_i}{\tilde{f}_i} \right) \tilde{\mathcal{R}}_i[\underline{f}, \tilde{f}](\mathbf{v}) d\mathbf{v} = 0. \quad (23)$$

Moreover one has  $\frac{\partial H}{\partial t}(x, t) + \operatorname{div} \mathbf{H}(x, t) = 0$  if and only if

$$\tilde{f}_i = f_i \quad \text{and} \quad (m_3 m_4)^3 \tilde{f}_1(\mathbf{v}) \tilde{f}_2(\mathbf{w}) = (m_1 m_2)^3 \tilde{f}_3(\mathbf{v}_1) \tilde{f}_4(\mathbf{w}_1). \quad (24)$$

The constraint (23) is purely mathematical. Nevertheless in the numerical experiments of the next Section this constraint has been checked finding a relative error whose order is  $10^{-4}$  at most. It obviously converges to zero approaching to equilibrium.

*Indifferentiability principle.* When all gas species are assumed to have same mass  $m$  and frequency  $\nu$ , it is straightforward to show that the total distribution  $f = \sum_i f_i$  verifies the single species BGK equation.

## NUMERICAL EXPERIMENTS

In this section some numerical tests for the proposed model, in the spatial homogeneous case and for the *Hydrogen-Air* reversible reaction, are presented in order to evaluate the trend to thermodynamical equilibrium and the influence of elastic collisions towards inelastic interactions.

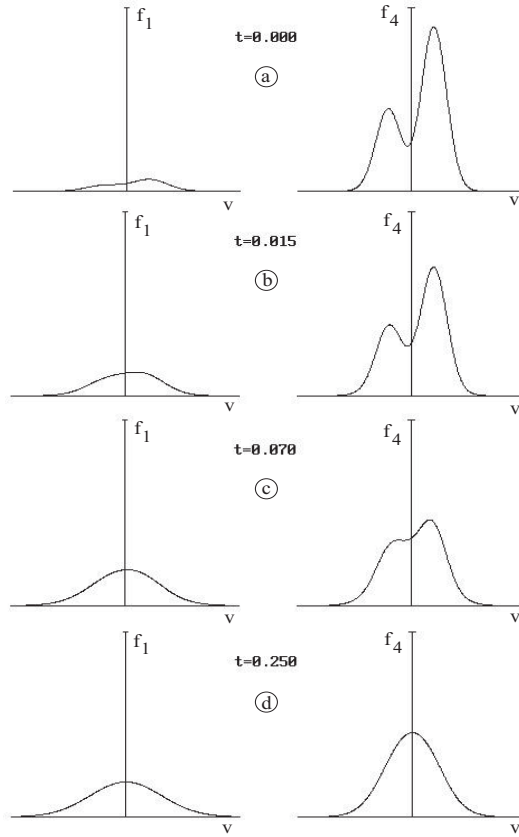
With reference to Fig.1, non-symmetric bimodal distributions are assumed as initial data for  $f_1, \dots, f_4$ . The corresponding macroscopic quantities (in *mole/l* for number densities and Kelvin degrees for temperature) are

$$n_1 = 0.0375, \quad n_2 = 0.0075, \quad n_3 = 0.225, \quad n_4 = 0.3375, \quad \mathbf{u} = 0, \quad T = 1200$$

We set  $\alpha_{ij} = \alpha = 1$  and  $\beta = 15$ ; for such values the ratio of the elastic and inelastic collision frequencies, that is  $\omega(T) = 4\pi\alpha/\gamma(T)$ , ranges between 50 and 75.

In Figs. 1a-1d the distributions  $f_1$  and  $f_4$  ( $f_2$  and  $f_3$  behave analogously) are plotted versus  $\mathbf{v}$  at different successive times. Since the reaction is prevalently exothermic  $f_1$  and  $f_4$  correspond, respectively, to distribution of product and reactant of the chemical process. It can be observed that, due to the assumptions on the BGK-type mechanism of collisions, the product distribution assumes a ‘‘Maxwellian’’ shape rather quickly, whereas the reactant distribution converges slower to such a shape.

In the last picture (d) both product and reactant have reached the equilibrium configuration which prescribes, with respect to initial data, a loss of the reactant  $H_2$  and a gain of the product  $H_2O$ .



**FIGURE 1.** Distributions  $f_1(v)$  and  $f_4(v)$  at different times.

In Fig.2, the influence of elastic collisions towards chemical interactions is shown, by plotting  $n_1, \dots, n_4$  versus time for the same initial data of Fig.1. In particular in Fig.2a  $\alpha = 1$  and in Fig.2b  $\alpha = 5$  (so that  $\omega$  now ranges approximately between 250 and 400). It can be noted that the increase of  $\alpha$  accelerates the trend to equilibrium which, in the pictures, is measured by the critical time  $t_c$ , defined as the time when  $n_2$  reaches the 90% of its asymptotic equilibrium value ( $t_c = 0.12$  for  $\alpha = 1$  and  $t_c = 0.0585$  for  $\alpha = 5$ ). Moreover the equilibrium values of the number densities are affected by the value of  $\alpha$ , since elastic collisions change significantly the temperature of the mixture, influencing the mass action law (10).

In Fig.3, the time evolution of the densities computed by the BGK model are compared to those obtained by the reactive Euler equations in the spatial homogeneous case, derived in paper [9], i.e.

$$\begin{aligned} \frac{dn_i}{dt} &= \lambda_i \hat{S}, \quad i = 1, \dots, 4, \quad \lambda_1 = \lambda_2 = 1, \quad \lambda_3 = \lambda_4 = -1 \\ \frac{dT}{dt} &= \frac{2}{3} \frac{\Delta E \hat{S}}{k_B n}, \quad \hat{S}(T) = \gamma(T) \left[ n_3 n_4 - \frac{\eta(T)}{\gamma(T)} n_1 n_2 \right], \quad n = \sum_{i=1}^4 n_i. \end{aligned} \quad (25)$$

Such equations give the time evolution towards chemical equilibrium of the physical state of a mixture which, at the microscopic scale, is in mechanical equilibrium (no elastic influence). < To make the comparison reasonable, the initial distributions of the BGK model have been chosen close to Maxwellians (symmetric bimodal distributions). They provide the values  $n_1 = 0.05$ ,  $n_2 = 0.1$ ,  $n_3 = 0.3$ ,  $n_4 = 0.45$ ,  $\mathbf{u} = 0$  and  $T = 1600$ , which are chosen as initial data for the Euler equations, as well. In such conditions, it can be underlined that the trend to equilibrium of the two models is quite similar, with an equilibrium reached a bit faster for the Euler model ( $t_c = 0.065$  against  $t_c = 0.0795$ ) and asymptotic values only slightly different. Finally in Fig.3a the entropy production profile  $\sigma$  (in arbitrary scale) is also provided, showing its monotone and concave shape.

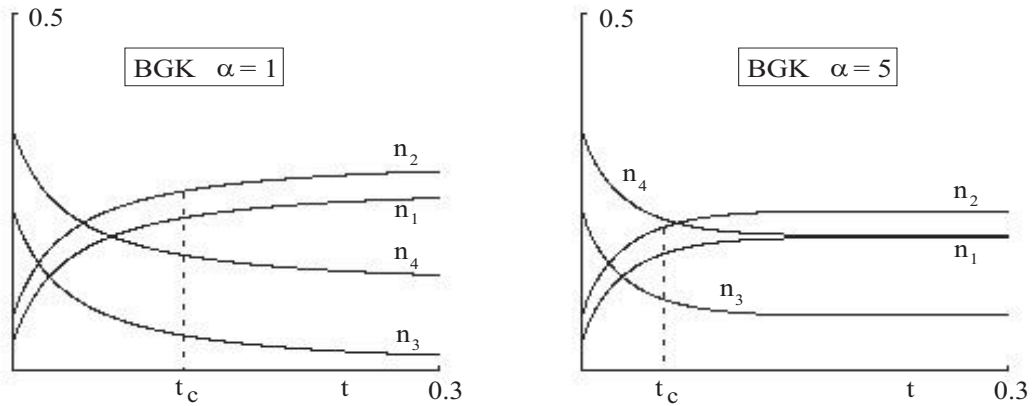


FIGURE 2. Time-evolution of number densities for two different values of  $\alpha$ .

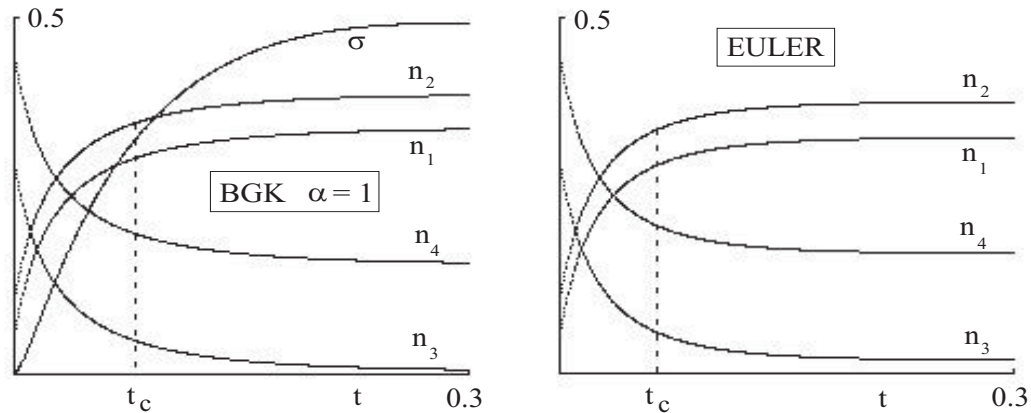


FIGURE 3. Entropy profile  $\sigma$  and comparison of trend to equilibrium for BGK and Euler equations.

## ACKNOWLEDGMENTS

The paper is partially supported by the National Research Project COFIN 2003 “Non linear mathematical problems of wave propagation and stability in models of continuous media” (Prof. T. Ruggeri), and Minho University Mathematics Centre (Financiamento plurianual, CMAT-FCT).

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