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Reaction–diffusion processes of electrically charged species

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1. Introduction

The investigation of nonlinear reaction–diffusion systems of the form

$$\frac{\partial u}{\partial t} - \nabla \cdot D(u)\nabla u = f(u, \nabla u) \qquad (1.1)$$

has received much attention in recent years [Am]. Here, $u = (u_1, ..., u_n)$ represents the concentrations of n species, $D = (D_1, ..., D_n)$ is the corresponding set of diffusion coefficients and f describes external sources and reactions. If transport includes drift in addition to diffusion, the flow $J = -D\nabla u$ has to be replaced by $J = -D\nabla u + \mathbf{v}$, where $\mathbf{v}$ is the convection flow. If the species are electrically charged, a drift is selfconsistently caused by the electrical field $\mathbf{v} = -c\nabla v_0$, where v_0 is the electrostatic potential satisfying Poisson's equation with the charge density as right-hand side. (The notation v_0 for the electrostatic potential will be convenient later on.) Drift–diffusion processes of charged species play an important rôle in many branches of modern natural sciences and technology. Especially in microelectronics, drift–diffusion models have fundamental significance for process simulation as well as for device simulation (see [Se]).

In this paper we state some basic facts about reaction–diffusion equations for charged species. Unlike the usual approach in device simulation, including only electrons and holes, we admit an arbitrary number of charged species. Our main aim is to show, that regardless of its complexity, the system of partial differential equations governing drift, diffusion and reaction of charged species has a convenient mathematical structure. Thus, following the ideas of S.G. Michlin [Mi], variational methods can be applied for proving global existence and uniqueness results. The key is a convex functional which can be interpreted from the viewpoint of thermodynamics as *free energy*. In particular, this functional turns out to be a Lyapunov function of the system and ensures exponential decay of arbitrary perturbations of thermal equilibria. We have to admit, however, that our existence results are based on additional assumptions restricting the growth of the source terms caused by chemical reactions.

The plan of the paper is following: First we discus some physical models for the drift–diffusion approach. Section 3 is devoted to the precise statement of the mathematical problem. The Lyapunov function is introduced in Section 4 and some a priori estimates are derived. An existence result is stated and a proof is sketched in Section 5. Section 6 deals with the question of uniqueness. Finally, in Section 7, we study the stationary problem and the asymptotic behaviour of transient solutions.

2. Mathematical modeling

The now classical drift–diffusion model of charged carrier transport in semiconductors was established by van Roosbroeck [vR] 1950. It consists of a Poisson equation for the electrostatic potential v_0 and continuity equations for the densities u_1, u_2 of electrons and

holes, respectively:

$$-\nabla \cdot (\varepsilon \nabla v_0) = f + \sum_{i=1}^{2} q_i u_i,$$

$$\frac{\partial u_i}{\partial t} + \nabla \cdot J_i + R = 0, \quad i = 1, 2.$$

Here ε is the dielectric permittivity, q_1, q_2 are the charges of electrons and holes, respectively, f denotes the net concentration of electrical active dopants, and R is the reaction (recombination/generation) term. The first equation expresses the Gauss law, the latter two local carrier conservation. Van Roosbroeck's equations are completed by current relations. It turns out to be useful both from the physical and from the mathematical point of view to introduce the electrochemical potentials (quasi Fermi potentials) ζ_1, ζ_2 of electrons and holes, respectively, by

$$\zeta_i = q_i v_0 + v_i, \quad i = 1, 2, \tag{2.1}$$

where v_1, v_2 are the chemical potentials (which are known functions of the densities u_1, u_2, cf. (2.3), (2.4) below). According to Ohm's law, the gradient of the electrochemical potential is postulated to be the driving force for the flow

$$J_i = -\mu_i \, u_i \nabla \zeta_i; \tag{2.2}$$

here μ_i is the mobility. The notation drift–diffusion model becomes clear by inserting (2.1) into (2.2). If a magnetic field B is present, in (2.2) a term due to the Lorentz power has to be added:

$$J_i - \beta_i \times J_i = -\mu_i \, u_i \nabla \zeta_i,$$

where $\beta_i = \mu_i r_i B$ (r_i is the so called Hall factor). Although the drift–diffusion model has proved to be of fundamental significance for the analysis and the numerical simulation of carrier transport in semiconductors, there are serious physical restrictions. First of all, the temperature is treated as a constant parameter. In order to model thermal effects the system has to be extended by an energy balance equation. Moreover, the trend to miniaturization forces modeling to become more microscopic and to take into account kinetic and even quantum mechanical transport effects. Some recent versions of the drift–diffusion model try to incorporate such effects via a careful and consistent choice of the physical model parameters. This approach is based on the fact that the drift–diffusion equations can be derived rigorously from kinetic models (Vlasov–Poisson–Boltzmann system) [P].

The drift–diffusion model describes electrons and holes in one semiconductor material (e.g. silicon) reacting via recombination and generation processes. In many situations different substances have to be taken into account and ionization as well as other chemical reactions occur. In process modeling, for instance, silicon as semiconductor and boron and phosphorus as dopants may be involved and may react according to different mechanisms (e.g. Frank–Turnbull, kick out [GGH, HS]).

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In what follows we shall admit n species with densities u_i and specific charges q_i. We discuss the physical model parameters from a more or less mathematical point of view. That means, we look for mathematically reasonable relations expressing carrier densities, mobilities and reaction rates in terms of the potentials to be determined as solutions of the equations. Fortunately, there is a correspondence between the mathematical and the physical point of view. As to the physical background we refer to [Se, SF].

Carrier densities

The introduction of a discrete number of charged species in a semiconductor is based on the energy band model of solid state physics and the effective mass approximation. Nonequilibrium situations are described by Fermi levels associated to the discrete energy bands. This means in particular that intraband relaxation processes are assumed to be much faster then interband ones. The standard drift–diffusion model distinguishes only two species, electrons and holes, associated to the conduction and valence band, respectively.

Frequently Boltzmann statistics is used for calculating the carrier distribution with respect to energy. Accordingly, carrier densities u_i and chemical potentials v_i are related by

$$u_i = u_i^* \, \exp\left(v_i\right), \tag{2.3}$$

where u_i^* is a reference density that generally depends on position because of doping or heterogeneous materials. Note that the chemical potential v_i is assumed to be scaled due to the fact that only isothermal processes are considered. In some situations (degeneration), e.g. at high doping levels, Boltzmann statistics has to be replaced by Fermi–Dirac statistics leading to

$$u_i = u_i^* \, \mathcal{F}_{1/2}(v_i), \quad \mathcal{F}_{1/2}(s) = \frac{2}{\sqrt{\pi}} \int_0^\infty \frac{\sqrt{t}\, dt}{1 + \exp(t - s)}. \tag{2.4}$$

We shall cover both options by assuming

$$u_i = u^* e_i(v_i) \tag{2.5}$$

with functions e_i satisfying mild conditions stated in the next section.

Mobilities

Mobility models have to account for different scattering mechanisms and high field effects. In particular, carrier–impurity scattering leads to an explicit space dependency of the mobilities. Carrier–carrier scattering can be modeled via a dependency of μ_i on the chemical potentials v_i.

The drift–diffusion model is mainly based on the linearized Boltzmann equation. At higher electric fields the carriers are able to accelerate and to heat up. Thus, linearization is no longer justified and a dependency of the mobilities on the gradients of the quasi Fermi potentials has to be admitted. In order to include the physical effects just mentioned and, possibly, a magnetic field, we assume that

$$J_i = -d_i(\cdot, v_i, \nabla \zeta_i), \tag{2.6}$$

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where the properties of the functions d_i are stated in the next section.
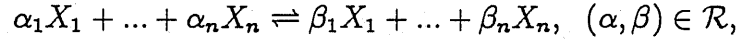
Reactions

Reactions between charged species are often recombination and generation processes. Recombination can happen e.g. via deep levels in the gap between conduction and valence band or as band–to–band transition. Which mechanism dominates depends on material properties and operation conditions. The most important recombination/generation models for electrons and holes are due to Shockley-Read-Hall and Auger:

$$R_{srh} = \frac{\exp\left(\zeta_1 + \zeta_2\right) - 1}{\tau_2(u_1 + r_1) + \tau_1(u_2 + r_2)},$$
$$R_{aug} = (\exp\left(\zeta_1 + \zeta_2\right) - 1)(a_1 u_1 + a_2 u_2),$$

where the parameters τ_i, r_i and a_i may depend on position x.

Generalizing these recombination models, we consider mass action type reactions of the form:

$$\alpha_1 X_1 + ... + \alpha_n X_n \rightleftharpoons \beta_1 X_1 + ... + \beta_n X_n, \quad (\alpha, \beta) \in \mathcal{R},$$

where $X_1, \ldots, X_n$ denote the species with the concentrations $u_1, \ldots, u_n$, respectively, and (α, β) is a pair of vectors $(\alpha_1, \ldots, \alpha_n)$, $(\beta_1, \ldots, \beta_n)$ of stoichiometric coefficients characterizing the reaction leading from $\sum_{i=1}^n \alpha_i X_i$ to $\sum_{i=1}^n \beta_i X_i$ and its converse reaction. We assume that the rates of these reactions are of the form $r_{\alpha\beta}(\cdot, v, \alpha \cdot \zeta)$ and $r_{\alpha\beta}(\cdot, v, \beta \cdot \zeta)$, respectively, where $v := (v_0, \ldots, v_n)$ and

$$\zeta := (\zeta_1, \ldots, \zeta_n), \quad \zeta_i := q_i v_0 + v_i, \quad i = 1, \ldots, n, \tag{2.7}$$

is the vector of electrochemical potentials ζ_i consisting of the electrostatic part $q_i v_0$ (q_i the specific charge of X_i) and the chemical part v_i ($i = 1, \ldots, n$). The assumption with respect to $r_{\alpha\beta}$ reflects the fact that the scalar product $(\alpha - \beta) \cdot \zeta$ is supposed to be driving force for the reactions. Correspondingly,

$$R_i = \sum_{(\alpha,\beta)\in\mathcal{R}} (r_{\alpha\beta}(\cdot, v, \alpha \cdot \zeta) - r_{\alpha\beta}(\cdot, v, \beta \cdot \zeta))(\alpha_i - \beta_i). \tag{2.8}$$

The finite set $\mathcal{R}$ denotes the set of reactions actually taking place in the volume Ω occupied by the species. Besides the reactions in the volume Ω further reactions may occur on the boundary Γ of Ω. In analogy to (2.8) we may assume that

$$R_i^\Gamma = \sum_{(\alpha,\beta)\in\mathcal{R}^\Gamma} (r_{\alpha\beta}^\Gamma(\cdot, v, \alpha \cdot \zeta) - r_{\alpha\beta}^\Gamma(\cdot, v, \beta \cdot \zeta))(\alpha_i - \beta_i) \tag{2.9}$$

is the contribution of these reactions to the balance of species X_i (which must be accounted for in the boundary conditions). Here $\mathcal{R}^\Gamma$ is a finite set of pairs of vectors of stoichiometric coefficients, and the functions $r_{\alpha\beta}^\Gamma$ model the surface reaction rates.

By specifying the coefficients of the reactions as

$$r_{\alpha\beta}(\cdot, v, s) = c_{\alpha\beta}(\cdot, v) \exp(s),$$

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it becomes clear, that standard mass action rates as well as the recombination models considered above are included as special cases.

Now we want to combine the physical models to get our final system of partial differential equations. To this end let Ω be a bounded domain in $\mathbb{R}^N$, $N \geq 2$, and $\partial \Omega = \Gamma$. We denote by $\nu(x_0)$ the outer unit normal at $x_0 \in \Gamma$. Then the initial boundary value problem we are interested in reads as follows:

$$\left. \begin{aligned} -\nabla \cdot (\varepsilon \nabla v_0) &= f + \sum_{i=1}^{n} q_i u_i \\ \frac{\partial u_i}{\partial t} + \nabla \cdot J_i + R_i &= 0, \ i = 1, \dots, n, \end{aligned} \right\} \text{ on } \mathbb{R}_+ \times \Omega, \qquad (2.10)$$

$$\nu \cdot (\varepsilon \nabla v_0) + \kappa v_0 = f^\Gamma, \quad \nu \cdot J_i + R_i^\Gamma = 0, \ i = 1, \dots, n \text{ on } \mathbb{R}_+ \times \Gamma, \qquad (2.11)$$

$$u_i(0, \cdot) = u_i^0, \ i = 1, \dots, n, \text{ on } \Omega. \qquad (2.12)$$

Here the densities u_i, the flows J_i, and the reaction terms R_i, R_i^Γ are given by (2.5) – (2.9). The functions f and f^Γ are fixed source terms (representing e.g. the charge density of dopants). The function κ represents a capacity of the boundary. The system (2.5) – (2.12) is to be regarded as an initial boundary value problem for the unknown vector $v = (v_0, v_1, \dots, v_n)$ of potentials and the corresponding vector $u = (u_0, \dots, u_n)$ of densities. Here $u_0 := \sum_{i=1}^{n} q_i u_i$ is the charge density caused by the mobile species.

Remark 2.1. An essential feature of the problem (2.5) – (2.11) is the fact that it allows so called thermal equilibria, i.e. steady states with vanishing driving forces. The results on steady states and asymptotic behaviour stated in Section 7 rest heavily on this property. All the other results remain true also in more general situations, for example, if Dirichlet conditions are posed on a part of the boundary Γ.

3. Precise statement of the problem

In this section we want to state precisely the problem discussed in the preceding section. We start with the formulation of basic hypotheses with respect to the data of the problem.

These hypotheses read as follows:

$$\Omega \text{ is a bounded Lipschitzian domain in } \mathbb{R}^N, \ N \geq 2, \text{ and } \Gamma := \partial \Omega, \qquad (3.1)$$

$$\varepsilon \in L^\infty(\Omega), \ \varepsilon \geq \varepsilon_0 > 0, \ \kappa \in L^\infty(\Gamma), \ \kappa \geq 0, \ \kappa \neq 0, \ f \in L^\infty(\Omega), \ f^\Gamma \in L^\infty(\Gamma), \qquad (3.2)$$

$$u_i^* \in L^\infty(\Omega), \ u_i^* \geq \delta > 0, \ u_i^0 \in L^\infty(\Omega), \ u_i^0 \geq \delta > 0, \ i = 1, \dots, n, \qquad (3.3)$$

$$q = (q_1, \dots, q_n) \in \mathbb{R}^n, \quad u_0^0 := \sum_{i=1}^{n} q_i u_i^0, \quad u^0 := (u_0^0, u_1^0; \dots, u_n^0), \qquad (3.4)$$

$$\left. \begin{aligned} &e_i \in C^1(\mathbb{R}) \text{ is strictly increasing}, \quad \lim_{y \to -\infty} e_i(y) = 0, \ \lim_{y \to \infty} e_i(y) = +\infty \\ &e_i' \leq e_i, \quad \int_{-\infty}^{0} e_i(y) dy < \infty, \quad i = 1, \dots, n, \end{aligned} \right\} \qquad (3.5)$$

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$$d_i : \Omega \times \mathbb{R} \times \mathbb{R}^N \longrightarrow \mathbb{R}^N \text{ satisfies the Carathéodory conditions,}$$

$$(d_i(x,y,\xi) - d_i(x,y,\eta)) \cdot (\xi - \eta) \geq \delta e_i'(y)|\xi - \eta|^2, \quad d_i(x,y,0) = 0,$$

$$|d_i(x,y,\xi) - d_i(x,y,\eta)| \leq \tfrac{1}{\delta} e_i(y)|\xi - \eta|$$

$$\text{for } x \in \Omega, \ y \in \mathbb{R}, \ \xi, \eta \in \mathbb{R}^N, \ i = 1, \dots, n, \text{ and some } \delta > 0, \tag{3.6}$$

$$\mathcal{R} \text{ and } \mathcal{R}^\Gamma \text{ are finite subsets of } \mathbb{Z}_+^n \times \mathbb{Z}_+^n,$$

$$\forall (\alpha, \beta) \in \mathcal{R}: \ r_{\alpha\beta} : \Omega \times \mathbb{R}^{n+1} \times \mathbb{R} \longrightarrow \mathbb{R}_+ \text{ satisfies the Carathéodory conditions,}$$

$$r_{\alpha\beta}(x,v,\cdot) \text{ is strictly increasing, } r_{\alpha\beta}(x,v,y) \leq c_0 \exp(y)$$

$$\text{for } x \in \Omega, \ y \in \mathbb{R}, \text{ and some constant } c_0,$$

$$\forall (\alpha, \beta) \in \mathcal{R}^\Gamma: \ r_{\alpha\beta}^\Gamma : \Gamma \times \mathbb{R}^{n+1} \times \mathbb{R} \longrightarrow \mathbb{R}_+ \text{ satisfies the Carathéodory conditions,}$$

$$r_{\alpha\beta}^\Gamma(x,v,\cdot) \text{ is strictly increasing, } r_{\alpha\beta}(x,v,y) \leq c_0 \exp(y)$$

$$\text{for } x \in \Gamma, \ y \in \mathbb{R}, \text{ and some constant } c_0, \tag{3.7}$$

$$\forall (\alpha, \beta) \in \mathcal{R} \cup \mathcal{R}^\Gamma: \ \alpha \cdot q = \beta \cdot q.$$

(As usual, $\mathbb{Z}_+ := \{m \in \mathbb{Z} : m \geq 0\}$ and $\mathbb{R}_+ := \{y \in \mathbb{R} : y \geq 0\}$.) The requirements $\alpha \cdot q = \beta \cdot q$ for $(\alpha, \beta) \in \mathcal{R} \cup \mathcal{R}^\Gamma$ express the fact that electrical charges are conserved during the reaction processes.

Throughout the paper we shall assume that $(3.1) - (3.7)$ are satisfied without mentioning this explicitly in our theorems. Further assumptions will be formulated later on in connection with existence and uniqueness results.

Let
$$V := H^1(\Omega; \mathbb{R}^{n+1}), \ H := L^2(\Omega; \mathbb{R}^{n+1}), \ W := V \cap L^\infty(\Omega; \mathbb{R}^{n+1}).$$

We define $E : W \longrightarrow V^*$ and $A : W \times V \longrightarrow V^*$ as follows:

$$\langle Ew, \bar{v} \rangle := \int_\Omega \left(\varepsilon \nabla w_0 \cdot \nabla \bar{v}_0 - f\bar{v}_0 + \sum_{i=1}^n u_i^* e_i(w_i)\bar{v}_i \right) dx + \int_\Gamma (\kappa w_0 - f^\Gamma)\bar{v}_0 d\Gamma,$$

$$\langle A(w,v), \bar{v} \rangle := \int_\Omega \left(\sum_{i=1}^n d_i(\cdot, w_i, \nabla\zeta_i) \cdot \nabla\bar{\zeta}_i \right.$$

$$\left. + \sum_{(\alpha,\beta)\in\mathcal{R}} (r_{\alpha\beta}(\cdot, w, \alpha \cdot \eta) - r_{\alpha\beta}(\cdot, w, \beta \cdot \eta))(\alpha - \beta) \cdot \bar{\zeta} \right) dx$$

$$+ \int_\Gamma \sum_{(\alpha,\beta)\in\mathcal{R}^\Gamma} (r_{\alpha\beta}^\Gamma(\cdot, w, \alpha \cdot \eta) - r_{\alpha\beta}^\Gamma(\cdot, w, \beta \cdot \eta))(\alpha - \beta) \cdot \bar{\zeta} d\Gamma,$$

where $w = (w_0, \dots, w_n) \in W$, $v = (v_0, \dots, v_n) \in V$, $\bar{v} = (\bar{v}_0, \dots, \bar{v}_n) \in V$,

$$\zeta := (\zeta_1, \dots, \zeta_n), \ \bar{\zeta} := (\bar{\zeta}_1, \dots, \bar{\zeta}_n), \ \eta := (\eta_1, \dots, \eta_n),$$

$$\zeta_i := q_i v_0 + v_i, \ \bar{\zeta}_i := q_i \bar{v}_0 + \bar{v}_i, \ \eta_i := q_i w_0 + w_i, \ i = 1, \dots, n.$$

Before we can formulate the initial boundary value problem to be solved we have to introduce some notation in connection with functions of time. Let Y be any Banach space and S any (bounded or unbounded) interval in $\mathbb{R}$. Then $L^p(S; Y)$ (resp. $L_{loc}^p(S; Y)$), $p \in [1, \infty]$, means the space of equivalence classes of Bochner measurable functions

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$u : S \longrightarrow Y$ such that $\|u(\cdot)\| \in L^p(S)$ (resp. $\|u(\cdot)\| \in L^p_{loc}(S)$). This space will be equipped with its standard norm (resp. the usual seminorms). $H^1(S;Y)$ is defined as the space of all $u \in L^2(S;Y)$ such that $u' \in L^2(S;Y)$, where u' denotes the derivative of u in the sense of Y–valued distributions. $H^1_{loc}(S;Y)$ is defined analogously.

Now the problem we want to solve can be stated as follows: Find (u,v) such that

$$
\left.
\begin{aligned}
& u \in H^1_{loc}(\mathbb{R}_+; V^*), \quad v \in L^2_{loc}(\mathbb{R}_+; V) \cap L^\infty(\mathbb{R}_+; L^\infty(\Omega; \mathbb{R}^{n+1})) \\
& u' + A(v,v) = 0, \quad u = Ev, \quad u(0) = u^0.
\end{aligned}
\right\}
\tag{P}
$$

Here and afterwards $A(v,v)$ and Ev mean the (equivalence classes of the) functions on $\mathbb{R}_+$ with the values $A(v(t),v(t))$ and $Ev(t)$, respectively.

Remark 3.1. Standard arguments show that a pair (u,v) of smooth functions solves (P) if and only if u and v satisfy the equations (2.5) – (2.12). In particular, by means of test functions of the form $(\bar{v}_0, -q_1\bar{v}_0, \ldots, -q_n\bar{v}_0)$ it is easy to check that for any solution (u,v) to (P) it holds $u_0 = \sum_{i=1}^{n} q_i u_i$.

Remark 3.2. Let (u,v) be a solution to Problem (P). As an element of $H^1_{loc}(\mathbb{R}_+; V^*)$ the function u is a continuous mapping from $\mathbb{R}_+$ into V^*. Using the boundedness of the functions v_i and the properties of the functions e_i, $i = 1, \ldots, n$, it is easy to show that $t \longmapsto u(t)$ is continuous from $\mathbb{R}_+$ to $L^\infty(\Omega; \mathbb{R}^{n+1})$, equipped with its weak* topology.

4. Physically motivated estimates and invariants

In this section we assume that we are given a solution to Problem (P). We shall show that physically motivated arguments lead to a priori estimates for the solution which are important for the proof of existence of solutions. In addition, we shall exhibit some invariants of the solution reflecting the "stoichiometric nature" of the reaction terms. These invariants will play a rôle in connection with the large time behaviour of the solution.

The boundary conditions introduced in Section 2 model a dissipative interaction of the physical system under consideration with its surrounding. Since we consider isothermic processes only we may expect that the *free energy* decreases along the solutions to (P). We are going to show that this is indeed the case. Moreover, giving an explicit expression for the free energy, we prove that this leads to interesting a priori estimates for the solutions to Problem (P).

In view of (3.5) it makes sense to define $\varphi_i \in C(\mathbb{R})$ and $\psi_i \in C(\mathbb{R}_+)$, $i = 1, \ldots, n$, by

$$
\varphi_i(v) := \int_0^v e_i(y)\,dy \text{ for } v \in \mathbb{R}, \quad \psi_i(u) := \int_{e_i(0)}^u e_i^{-1}(z)\,dz \text{ for } u \in \mathbb{R}_+, \ i = 1, \ldots, n. \tag{4.1}
$$

Next we introduce two convex functionals, namely

$$
\Phi(v) := \int_\Omega \left(\frac{1}{2}\varepsilon |\nabla v_0|^2 - f v_0 + \sum_{i=1}^{n} u_i^* \varphi_i(v_i) \right) dx + \int_\Gamma \left(\frac{1}{2}\kappa v_0^2 - f^\Gamma v_0 \right) d\Gamma, \ v \in V, \tag{4.2}
$$

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and its conjugate

$$\Psi(u) := \sup_{v \in V} \{\langle u, v \rangle - \Phi(v)\}, \ u \in V^*. \tag{4.3}$$

Note that the values $\Phi(v)$ and $\Psi(u)$ may be $+\infty$. Since $\Phi(0) = 0$ we have $\Psi(u) \geq 0$ for every $u \in V^*$. We refer to Ekeland–Temam [ET] for the basic notions and results from convex analysis.

The functional Φ is strictly convex. Hence for every $v \in V$ its subdifferential $\partial\Phi(v)$ contains at most one element. If $v \in W$ then, as is easily checked, $\partial\Phi(v) = \{Ev\}$.

A simple calculation shows that

$$\Psi(u) = \int_\Omega \left(\frac{1}{2}\varepsilon|\nabla v_0|^2 + \sum_{i=1}^n u_i^* \psi_i(u_i/u_i^*) \right) dx + \int_\Gamma \frac{1}{2}\kappa v_0^2 d\Gamma, \tag{4.4}$$

provided that $u \in V^*$, $u_i \in L^2(\Omega)$, $u_i \geq 0$, $i = 1, \dots, n$, and that v_0 is defined by

$$\forall \bar{v}_0 \in H^1(\Omega): \quad \int_\Omega (\varepsilon\nabla v_0 \cdot \nabla \bar{v}_0 - f\bar{v}_0)dx + \int_\Gamma (\kappa v_0 - f^\Gamma)\bar{v}_0 d\Gamma = \langle u_0, \bar{v}_0 \rangle. \tag{4.5}$$

The value $\Psi(u)$ is to be interpreted as the *free energy of the state u*. Therefore one is led to investigate the behaviour of this functional along solutions to Problem (P).

Theorem 4.1. *Let (u, v) be a solution to Problem (P) and let Ψ be the functional defined above. Then, for $0 \leq s \leq t$,*

$$\Psi(u(t)) \leq \Psi(u(s)) < \infty, \tag{4.6}$$

i.e., Ψ is decreasing along any solution to Problem (P). Moreover,

$$\|v_0\|_{L^\infty(\mathbb{R}_+;H^1(\Omega))} + \sum_{i=1}^n \|\psi_i(u_i/u_i^*)\|_{L^\infty(\mathbb{R}_+;L^1(\Omega))} \leq c, \tag{4.7}$$

and

$$
\begin{aligned}
&\sum_{i=1}^n \|e_i'(v_i)|\nabla\zeta_i|^2\|_{L^1(\mathbb{R}_+;L^1(\Omega))} \\
&+ \sum_{(\alpha,\beta)\in\mathcal{R}} \|(r_{\alpha\beta}(\cdot, v, \alpha \cdot \zeta) - r_{\alpha\beta}(\cdot, v, \beta \cdot \zeta))(\alpha - \beta) \cdot \zeta\|_{L^1(\mathbb{R}_+;L^1(\Omega))} \\
&+ \sum_{(\alpha,\beta)\in\mathcal{R}^\Gamma} \|(r_{\alpha\beta}^\Gamma(\cdot, v, \alpha \cdot \zeta) - r_{\alpha\beta}^\Gamma(\cdot, v, \beta \cdot \zeta))(\alpha - \beta) \cdot \zeta\|_{L^1(\mathbb{R}_+;L^1(\Gamma))} \leq c.
\end{aligned} \tag{4.8}
$$

where c is a constant depending on the data of the problem. As before, $\zeta = (\zeta_1, \dots, \zeta_n)$ is defined by $\zeta_i := q_i v_0 + v_i$, $i = 1, \dots, n$.

Proof.
1. First we mention that, for every $w \in W$,

$$
\begin{aligned}
\langle A(w, w), w \rangle \geq \ &\delta \sum_{i=1}^n \|e_i'(w_i)|\nabla\eta_i|^2\|_{L^1(\Omega)} \\
&+ \sum_{(\alpha,\beta)\in\mathcal{R}} \|(r_{\alpha\beta}(\cdot, w, \alpha \cdot \eta) - r_{\alpha\beta}(\cdot, w, \beta \cdot \eta))(\alpha - \beta) \cdot \eta\|_{L^1(\Omega)} \\
&+ \sum_{(\alpha,\beta)\in\mathcal{R}^\Gamma} \|(r_{\alpha\beta}^\Gamma(\cdot, w, \alpha \cdot \eta) - r_{\alpha\beta}^\Gamma(\cdot, w, \beta \cdot \eta))(\alpha - \beta) \cdot \eta\|_{L^1(\Gamma)},
\end{aligned} \tag{4.9}
$$

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where $\eta := (\eta_1, \ldots, \eta_n)$, , $\eta_i := q_i w_0 + w_i$, $i = 1, \ldots, n$. This is an immediate consequence of the definition of A (cf. (3.6), (3.7)).

2. Let (u, v) be a solution to (P). Then, for a.e. $t \in \mathbb{R}_+$,

$$u(t) = Ev(t) \in \partial \Phi(v(t)).$$

According to a standard result of convex analysis this implies that

$$v(t) \in \partial \Psi(u(t)), \quad \text{for a.e. } t \in \mathbb{R}_+.$$

Therefore, if $0 \leq s \leq t$, then (cf. Brézis [B], Lemma 3.3)

$$\Psi(u(t)) - \Psi(u(s)) = \int_s^t \langle u'(\tau), v(\tau) \rangle \, d\tau.$$

(Note that $\Psi(u(t))$ is finite for every $t \in \mathbb{R}_+$, cf. Remark 3.2 and (4.4).) Since (u, v) is a solution to (P) we obtain

$$\Psi(u(t)) - \Psi(u(s)) = \int_s^t \langle -A(v(\tau), v(\tau)), v(\tau) \rangle \, d\tau \leq 0. \qquad (4.10)$$

The last inequality follows from (4.9). Hence (4.6) holds.

3. The estimate (4.7) is an immediate consequence of (4.6) and (4.4). The assertion (4.8) follows from

$$\int_0^\infty \langle A(v(t), v(t)), v(t) \rangle \, dt \leq \Psi(u^0)$$

and the relation (4.9). $\square$

Remark 4.1.   The following theorem shows how to use (4.7) and (4.8) to get further information about a solution (u, v) to Problem (P).

Theorem 4.2. *Let (u, v) be a solution to Problem* (P). *Then*

$$u_i \log(u_i) \in L^\infty(\mathbb{R}_+; L^1(\Omega)), \ i = 1, \ldots, n. \qquad (4.11)$$

If $N = 2$ then $v_0 \in L^\infty(\mathbb{R}_+; L^\infty(\Omega))$. The same is true if

$$\limsup_{y \to \infty} e_i(y) y^{-1/(p_0-1)} < \infty \ \text{for some } p_0 > \frac{N}{2}, \ i = 1, \ldots, n. \qquad (4.12)$$

In that case we have also $u_i \in L^\infty(\mathbb{R}_+; L^{p_0}(\Omega))$, $i = 1, \ldots, n$.

Remark 4.2.   The condition (4.12) is satisfied for $N = 3$ with $p_0 = 5/3$ if the functions e_i are given according to the Fermi–Dirac statistics (cf. (2.4)).

Proof of Theorem 4.2.

1. The relation $e_i'(y) \leq e_i(y)$, $y \in \mathbb{R}$, implies that $e(y) \leq e(0) \exp(y)$ for $y \geq 0$. Consequently, for $u \geq e_i(0)$,

$$\psi_i(u) = \int_{e_i(0)}^u e_i^{-1}(z) dz \geq \int_{e_i(0)}^u \log(z/e_i(0)) dz \geq \frac{1}{2} u \log(u) - c. \qquad (4.13)$$

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An estimate of this form is true also if $0 < u < e_i(0)$. Therefore the assertion (4.11) follows from (4.7).

2. Let $N = 2$. The first part of the proof shows that for $u_0 = \sum\limits_{i=1}^{n} q_i u_i$ it holds $|u_0| \log(|u_0|) \in L^\infty(\mathbb{R}_+; L^1(\Omega))$. Since v_0 satisfies (4.5) the property $v_0 \in L^\infty(\mathbb{R}_+; L^\infty(\Omega))$ follows from the results in [G2].

3. Let $N > 2$, and let $p_0 > \dfrac{N}{2}$ be given such that (4.12) holds. Then $e_i(y) \le c_0 y^{1/(p_0-1)}$ for sufficiently large y and a suitable constant c_0. Consequently,

$$e_i^{-1}(z) \ge (z/c_0)^{p_0-1} \text{ for sufficiently large } z$$

and, for every $u \ge 0$,

$$\psi_i(u) = \int_{e_i(0)}^{u} e_i^{-1}(z)\,dz \ge c_1 u^{p_0} - c_2,$$

where $c_1 > 0$. Therefore (4.7) implies that $u_i \in L^\infty(\mathbb{R}_+; L^{p_0}(\Omega))$. The assertion with respect to v_0 now follows from a standard result on elliptic boundary value problems (see [LU]). $\square$

Next we shall discuss the invariants of the process mentioned in the beginning of this section. The space

$$\mathcal{S} := \text{span}\left\{\beta - \alpha : (\alpha, \beta) \in \mathcal{R} \cup \mathcal{R}^\Gamma\right\} \tag{4.14}$$

will be called the *stoichiometric subspace* of $\mathbb{R}^n$ associated to the system under consideration. By $\mathbf{1}$ we denote the function on Ω with the constant value 1. We define

$$U := \left\{u \in V^* : u_0 = \sum_{i=1}^{n} q_i u_i, \ (\langle u_1, \mathbf{1}\rangle, \ldots, \langle u_n, \mathbf{1}\rangle) \in \mathcal{S}.\right\} \tag{4.15}$$

(Note that $\langle u_i, \mathbf{1}\rangle = \int_\Omega u_i\,dx$ if $u \in V^* \cap L^1(\Omega; \mathbb{R}^{n+1})$.) The introduction of U is justified by the following

Theorem 4.3. *Let (u, v) be a solution to Problem* (P). *Then*

$$\forall t \in \mathbb{R}_+ : \quad u(t) \in U + u^0.$$

Proof. Let

$$U^\perp := \{v \in V : \nabla\zeta = 0, \ \zeta \in \mathcal{S}^\perp, \ \text{where } \zeta := (q_1 v_0 + v_1, \ldots, q_n v_0 + v_n)\}. \tag{4.16}$$

Here and afterwards $\mathcal{S}^\perp$ means the orthogonal complement of $\mathcal{S}$ in $\mathbb{R}^n$. From the definition of $U^\perp$ and that of the operator A it follows immediately that, for arbitrary $w \in W$ and $v \in V$, we have

$$\forall \bar{v} \in U^\perp : \quad \langle A(w, v), \bar{v}\rangle = 0.$$

In particular, if (u, v) is a solution to (P), then $\langle A(v(s), v(s)), \bar{v}\rangle = 0$ for every $\bar{v} \in U^\perp$. Hence

$$\forall \bar{v} \in U^\perp : \quad \left\langle u(t) - u^0, \bar{v}\right\rangle = \int_0^t \langle u'(s), \bar{v}\rangle\,ds = -\int_0^t \langle A(v(s), v(s)), \bar{v}\rangle\,ds = 0.$$

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It is easy to check that $U = \{u \in V^* : \langle u, v \rangle = 0 \text{ for every } v \in U^\perp\}$. Therefore, the preceding equality proves the assertion of Theorem 4.3. $\square$

Remark 4.3. It may well happen that the stoichiometric subspace $\mathcal{S}$ equals $\mathbb{R}^n$. In that case Theorem 4.3 reduces to the observation that $u_0 = \sum_{i=1}^n q_i u_i$ (cf. Remark 3.1).

5. Existence

As mentioned in the introduction we can prove existence only under additional restrictive hypotheses with respect to the reaction terms. We shall assume that

$$\left. \begin{aligned} &\forall v \in \mathbb{R}^{n+1},\ \forall(\alpha,\beta) \in \mathcal{R},\ \forall i \in \{1,\ldots,n\}:\\ &(r_{\alpha\beta}(\cdot, v, \alpha \cdot \zeta) - r_{\alpha\beta}(\cdot, v, \beta \cdot \zeta))(\beta - \alpha)_i \leq c(v_0) \sum_{j=1}^n |e_j(v_j)|^{1+\frac{2}{N}} + c(v_0), \end{aligned} \right\} \quad (5.1)$$

$$\left. \begin{aligned} &\forall v \in \mathbb{R}^{n+1},\ \forall(\alpha,\beta) \in \mathcal{R}^\Gamma,\ \forall i \in \{1,\ldots,n\}:\\ &(r_{\alpha\beta}^\Gamma(\cdot, v, \alpha \cdot \zeta) - r_{\alpha\beta}^\Gamma(\cdot, v, \beta \cdot \zeta))(\beta - \alpha)_i \leq c(v_0) \sum_{j=1}^n |e_j(v_j)|^{1+\frac{1}{N}} + c(v_0); \end{aligned} \right\} \quad (5.2)$$

here $\zeta_i := q_i v_0 + v_i$, $i = 1, \ldots, n$.

The conditions (5.1) and (5.2) impose restrictions only on the source terms whereas the sink terms may be large.

Theorem 5.1. *Suppose that* (5.1) *and* (5.2) *hold. Let* $N = 2$ *or let* (4.12) *and the following additional assumptions be satisfied:*

ε *is constant ,*

$d_i(x, y, \xi) = \sigma_i(y)\xi$ *for* $x \in \Omega$, $y \in \mathbb{R}$, $\xi \in \mathbb{R}^N$, *where* $\delta e_i' \leq \sigma_i \leq \frac{1}{\delta} e_i$ *for some* $\delta > 0$.

Then there exists a solution (u, v) *to Problem* (P).

We shall not give a complete proof of Theorem 5.1 but only sketch the main ideas.

The first idea is to investigate a "regularized" problem which arises from (P) by cutting off the nonlinearities in a suitable way at a certain level. Later one provides a priori estimates which are independent of that level. As a consequence a solution to the regularized problem will be a solution to the original problem (P) if only the cut off level is chosen sufficiently large.

Let $M > 0$ be a fixed number such that

$$u_i^* e_i(-M) \leq u_i^0 \leq u_i^* e_i(M), \quad i = 1, \ldots, n. \quad (5.3)$$

This number will play the rôle of the cut off level. By P_M we denote the convex projection from $\mathbb{R}$ onto $[-M, M]$, i.e., the mapping given by

$$P_M(y) := \begin{cases} M, & \text{if } y > M, \\ y, & \text{if } -M \leq y \leq M, \\ -M, & \text{if } y < -M. \end{cases}$$

We define $E_M : V \longrightarrow V^*$ and $A_M : V \times V \longrightarrow V^*$ by

$$\langle E_M v, \bar{v} \rangle := \int_\Omega \Big(\varepsilon \nabla v_0 \cdot \nabla \bar{v}_0 - f \bar{v}_0 + \sum_{i=1}^n u_i^* e_i(P_M v_i) \bar{v}_i \Big) dx + \int_\Gamma (\kappa v_0 - f^\Gamma) \bar{v}_0 d\Gamma$$

and (with the same notation as that used for the definition of A)

$$\begin{aligned}
\langle A_M(w, v), \bar{v} \rangle := \sum_{i=1}^n \Big\{ \int_\Omega \Big(& d_i(\cdot, P_M w_i, \nabla \zeta_i) \cdot \nabla \bar{\zeta}_i + (\zeta_i - P_M(q_i w_0) - P_M w_i) \bar{\zeta}_i \\
& + \lambda_M(w) \sum_{(\alpha,\beta) \in \mathcal{R}} (r_{\alpha\beta}(\cdot, v, \alpha \cdot \eta) - r_{\alpha\beta}(\cdot, v, \beta \cdot \eta))(\alpha - \beta) \cdot \bar{\zeta} \Big) dx \\
& + \int_\Gamma \lambda_M(w) \sum_{(\alpha,\beta) \in \mathcal{R}^\Gamma} (r_{\alpha\beta}^\Gamma(\cdot, v, \alpha \cdot \eta) - r_{\alpha\beta}^\Gamma(\cdot, v, \beta \cdot \eta))(\alpha - \beta) \cdot \bar{\zeta} d\Gamma \Big\},
\end{aligned}$$

where λ_M is a fixed function in $C(\mathbb{R}^{n+1}; [0, 1])$ such that

$$\lambda_M(\xi) := \begin{cases} 0, & \text{if } |\xi|_\infty \geq M, \\ 1, & \text{if } |\xi|_\infty \leq M/2 \end{cases}, \quad |\xi|_\infty := \max\{|\xi_0|, \ldots, |\xi_n|\}.$$

The definitions of the operators E_M and A_M are made in such a way that the essential properties of E and A are conserved. The regularized problem announced above reads as follows: Find (u, v) such that

$$\left. \begin{aligned}
& u \in H^1_{loc}(\mathbb{R}_+; V^*), \quad v \in L^2_{loc}(\mathbb{R}_+; V) \\
& u' + A_M(v, v) = 0, \quad u = E_M v, \quad u(0) = u^0.
\end{aligned} \right\} \tag{P_M}$$

Remark 5.1.   Let (u, v) be a solution to Problem (P_M). Then $u_i = u_i^* e_i(P_M v_i)$, $i = 1, \ldots, n$, and $u_0 = \sum_{i=1}^n q_i u_i$ (cf. Remark 3.1). If

$$\|v_i\|_{L^\infty(\mathbb{R}_+; L^\infty(\Omega))} \leq \frac{M}{2} \quad \text{and} \quad (1 + |q_i|) \|v_0\|_{L^\infty(\mathbb{R}_+; L^\infty(\Omega))} \leq \frac{M}{2}, \quad i = 1, \ldots, n,$$

then (u, v) is a solution to (P).

The solvability of Problem P_M can be proved by the investigation of systems which result from (P_M) by a discretization of time. To describe these systems we fix a sequence (τ_k) of time steps converging to 0. Let $S_k^j :=](j - 1)\tau_k, j\tau_k]$, $j \in \mathbb{N}$. If Y is any Banach space we denote by $C_k(\mathbb{R}_+; Y)$ the space of all functions $u : \mathbb{R}_+ \longrightarrow X$, which are constant

on each of the intervals S_k^j, $j \in \mathbb{N}$. We write u^j for the value of $u \in C_k(\mathbb{R}_+; X)$ on S_k^j. We define Δ_k as a mapping from $C_k(\mathbb{R}_+; H)$ into itself by

$$(\Delta_k u)^j := \frac{1}{\tau_k}(u^j - u^{j-1}), \quad j \in \mathbb{N}, \tag{5.4}$$

where u^0 is the initial value introduced in (3.4). The problem

$$\Delta_k u_k + A_M(v_k, v_k) = 0, \quad u_k = E_M v_k, \quad v_k \in C_k(\mathbb{R}_+; V), \tag{P_{Mk}}$$

which can be written more explicitly as

$$\frac{1}{\tau_k}(u_k^j - u_k^{j-1}) + A_M(v_k^j, v_k^j) = 0, \quad u_k^j = E_M v_k^j, \quad j \in \mathbb{N}, \quad u_k^0 = u^0, \tag{5.5}$$

is to be considered as the discrete version of (P_M) corresponding to the time step τ_k. One can prove that for every $k \in \mathbb{N}$ there exists a solution (u_k, v_k) to Problem (P_{Mk}) using a result on operators of variational type in the sense of Lions [L], Ch. 2, Sect. 2.5. Furthermore, one can find a priori estimates (depending on M) allowing to go to the limit as $k \longrightarrow \infty$. In this way one finds a solution to (P_M). We don't want to go into to details here. For similar considerations in a special case we refer to [GG2].

Next for a solution (u, v) to Problem (P_M) one has to derive a priori estimates which are independent of M. This is the most difficult part of the proof.

The operator E_M is defined in such a way that it is the Gâteaux derivative of a functional $\Phi_M : V \longrightarrow \mathbb{R}$, where $\Phi_M \le \Phi$ (for the definition of Φ cf. (4.2)). The conjugate functional Ψ_M to Φ_M satisfies $\Psi_M \ge \Psi$, where Ψ denotes again the conjugate of Φ. In the same way as the corresponding result for Ψ in Section 4 one can prove that Ψ_M decreases along the solutions to (P_M). Since by the choice of M the initial value $\Psi_M(u^0)$ is independent of M, this leads to a priori estimates independent of M for the following norms:

$$\left. \begin{array}{l} \|v_0\|_{L^\infty(\mathbb{R}_+; H^1(\Omega))}, \quad \|v_0\|_{L^\infty(\mathbb{R}_+; L^\infty(\Omega))}, \\ \sum_{i=1}^{n} \|u_i \log(u_i)\|_{L^\infty(\mathbb{R}_+; L^1(\Omega))} \text{ if } N = 2, \quad \sum_{i=1}^{n} \|u_i\|_{L^\infty(\mathbb{R}_+; L^{p_0}(\Omega))} \text{ if } N > 2. \end{array} \right\} \tag{5.6}$$

A priori bounds for the norms (5.6) are not sufficient for our purposes. What is needed are bounds for $\|v_i\|_{L^\infty(\mathbb{R}_+; L^\infty(\Omega))}$, $i = 1, \ldots, n$.

First we indicate how to obtain upper bounds for the densities u_i by means of a technique introduced by Moser [Mo]. Let $w := (0, w_1, \ldots, w_n)$, $w_i := \exp(pt)[e_i(P_M v_i)]^{p-1}$, $i = 1, \ldots, n$, where $p \ge 2$. Using w for the values $p = 2^k$, $k = 1, 2, \ldots$, as test functions for the equation $u' + A(v, v) = 0$ it is possible to derive successively bounds for the norms $\|u\|_{L^\infty(\mathbb{R}_+; L^{2^k}(\Omega; \mathbb{R}^{n+1}))}$ which are independent of M and of k. This implies that $e_i(P_M v_i) \le c$ or $P_M v_i \le e_i^{-1}(c)$, $i = 1, \ldots, n$. We omit the rather technical details. We mention only that it is this step which requires to distinguish the cases $N = 2$ and $N > 2$. For the case $N = 2$ the technique has been presented in [GG2] for a special case in some detail. From now on we assume that the choice of M is made in such a way that

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$M > e_i^{-1}(c)$, $i = 1, \ldots, n$. Then the estimate for $e_i(P_M v_i)$ implies that $v_i \leq e_i^{-1}(c)$, i.e. the components of the vector v of potentials are bounded from above independently of M.

To get lower bounds for the potentials one can use test functions of the form

$$w := (0, w_1, \ldots, w_n), \quad w_i := -\frac{p[(\log(e_i(P_M v_i)) + k)^-]^{p-1}}{e_i(P_M v_i)}, \quad i = 1, \ldots, n.$$

Here $p \geq 2$ and k is a sufficiently large parameter. The superscript "$-$" denotes the negative part of a function. This time one gets bounds for $\|(\log(e_i(P_M v_i)) + k)^-\|_{L^\infty(\mathbb{R}_+; L^p(\Omega))}$ independent of M and of p. Hence $(\log(e_i(P_M v_i)) + k)^- \leq c$ or $P_M v_i \geq e_i^{-1}(\exp(-c - k))$. If M is chosen such that $-M < e_i^{-1}(\exp(-c - k))$, $i = 1, \ldots, n$, then the components of v must be bounded from below by a constant independent of M.

Remark 5.2. Existence can be proved also without the assumption (4.12) if $q_i \geq 0$ or $q_i \leq 0$ for $i = 1, \ldots, n$ or if there are only two species taking part in the process $(n = 2)$.

6. Uniqueness

Due to the nonlinearities of the flow expressions (2.6), uniqueness of solutions to (P) cannot be proved by standard arguments without using additional regularity properties of the solutions. For example, boundedness of the gradients of the electrochemical potentials would imply uniqueness. But such strong regularity assumption excludes practically relevant geometries as well as heterogeneous structures.

In the case of Boltzmann statistics a quite satisfying uniqueness result has been proved in [GG1] for van Roosbroeck's system. This result rests on special properties of the exponential function and can be extended to (P) in case that $u_i = u_i^* \exp(v_i)$. However, for more general functions e_i, in particular for Fermi–Dirac statistics, there is still a gap between existence and uniqueness results, at least for $N \geq 3$. For two space dimensions uniqueness has been proved in [GR, Ga]. Thus the situation is quite similar to Navier–Stokes equations.

In this section we want to state a uniqueness result under a quite weak regularity hypothesis. To this purpose we assume the functions d_i from (2.6) to have the following special structure

$$d_i(\cdot, y, \xi) = e_i'(y)\gamma_i(\cdot, \xi). \tag{6.1}$$

Moreover, in addition to (3.5), we assume:

$$g_i := e_i' \circ e_i^{-1} :]0, \infty[\longrightarrow]0, \infty[, \quad i = 1, \ldots, n, \quad \text{is concave.} \tag{6.2}$$

Finally, we replace (3.6) by:

$$\left.\begin{array}{l}
\gamma_i : \Omega \times \mathbb{R}^N \longrightarrow \mathbb{R}^N \text{ satisfies the Carathéodory conditions, } \gamma_i(x, 0) = 0, \\
\gamma_i \text{ is strongly monotone and Lipschitzian:} \\
(\gamma_i(x, \xi_1) - \gamma_i(x, \xi_2)) \cdot (\xi_1 - \xi_2) \geq \delta|\xi_1 - \xi_2|^2, \quad |\gamma_i(x, \xi) - \gamma_i(x, \eta)| \leq \frac{1}{\delta}|\xi - \eta|, \\
\text{for } x \in \Omega, \; \xi, \eta \in \mathbb{R}^N, \; i = 1, \ldots, n, \text{ and some } \delta > 0.
\end{array}\right\} \tag{6.3}$$

Remark 6.1. In the case of Boltzmann statistics (cf. (2.3)) the condition (6.2) is trivially satisfied, since g_i is the identity map. It can be shown that for Fermi–Dirac statistics (cf. (2.4)) the function g_i is even *strictly* concave.

Now we are ready to state a mild regularity condition with respect to the electrostatic potential ensuring uniqueness.

Theorem 6.1. *Let the additional conditions (6.1) – (6.3) be satisfied. Then a solution (u, v) to problem (P) is unique if either $e_i = \exp$, $i = 1, \ldots, n$, (Boltzmann statistics) or*

$$\nabla v_0 \in L_{loc}^\infty(\mathbb{R}_+; L^p(\Omega; \mathbb{R}^N)) \text{ for some } p > N. \tag{6.4}$$

Proof. Suppose there are two solutions (u^j, v^j), $j = 1, 2$, to (P) satisfying (6.4). We set

$$(u, v) := (u^1 - u^2, v^1 - v^2), \quad \zeta := \zeta^1 - \zeta^2.$$

For the proof of uniqueness we may and we will restrict our considerations to a compact interval of time, say $S = [0, T]$. By means of the convex functional (4.4) we define a "distance"

$$\varrho(u^1(t), u^2(t)) := \Psi(u^1(t)) + \Psi(u^2(t)) - 2\Psi\left(\frac{u^1(t) + u^2(t)}{2}\right). \tag{6.5}$$

Since the functions ψ_i from (4.1) are locally uniformly convex, there exists a positive constant c_1 such that

$$\forall t \in S: \quad c_1\left(\|v_0(t)\|_{H^1(\Omega)}^2 + \sum_{i=1}^n \|u_i(t)\|_{L^2(\Omega)}^2\right) \leq \varrho(u^1(t), u^2(t)). \tag{6.6}$$

Hence, because of Gronwall's lemma, it suffices to show that

$$\varrho(u^1(t), u^2(t)) \leq c_2 \int_0^t \left(\|v_0(s)\|_{H^1(\Omega)}^2 + \sum_{i=1}^n \|u_i(s)\|_{L^2(\Omega)}^2\right) ds. \tag{6.7}$$

Now, setting

$$\bar{v}_0 = \frac{v_0^1 + v_0^2}{2}, \quad \bar{v}_i = e_i^{-1}\left(\frac{e_i(v_i^1) + e_i(v_i^2)}{2}\right), \quad \bar{\zeta}_i = q_i \bar{v}_0 + \bar{v}_i,$$

using the initial conditions along with (4.10) and the definition of the operator A, we obtain

$$\varrho(u^1(t), u^2(t)) = -\int_0^t \sum_{j=1}^2 \left\langle A(v^j(s), v^j(s)), v^j(s)) - \bar{v}(s)\right\rangle ds$$

$$= -\sum_{i=1}^n \int_0^t \sum_{j=1}^2 \left(\int_\Omega \left(e_i'(v_i^j)\gamma_i(x, \nabla\zeta_i^j) \cdot \nabla(\zeta_i^j - \bar{\zeta}_i) + R_i^j(\zeta_i^j - \bar{\zeta}_i)\right) dx \right.$$

$$\left. + \int_\Gamma R_i^{\Gamma j}(\zeta_i^j - \bar{\zeta}_i) \, d\Gamma\right) ds.$$

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(To simplify the notation we have omitted the time argument in the last expressions. This simplification will be used also in the following calculations.) To prove (6.7) we have to estimate the last expression from above. The only cumbersome term is the first one involving partial derivatives. Setting

$$\bar{g}_i := g_i\Big(\frac{e_i(v_i^1) + e_i(v_i^2)}{2}\Big), \quad G_i := 2\bar{g}_i - e_i'(v_i^1) - e_i'(v_i^2),$$

we can rewrite this term as follows

$$\sum_{j=1}^{2} e_i'(v_i^j)\gamma_i(\cdot, \nabla\zeta_i^j) \cdot \nabla(\zeta_i^j - \bar{\zeta}_i)$$

$$= \frac{1}{2\bar{g}_i}\sum_{j=1}^{2} e_i'(v_i^j)\gamma_i(\cdot, \nabla\zeta_i^j) \cdot (2\bar{g}_i\nabla(\zeta_i^j - q_i v_0^j) - e_i'(v_i^1)\nabla v_i^1 - e_i'(v_i^2)\nabla v_i^2)$$

$$= \frac{1}{2\bar{g}_i}\Big(e_i'(v_i^1)e_i'(v_i^2)(\gamma_i(\cdot, \nabla\zeta_i^1) - \gamma_i(\cdot, \nabla\zeta_i^2)) \cdot \nabla(\zeta_i^1 - \zeta_i^2 - q_i(v_0^1 - v_0^2))$$

$$+ G_i\sum_{j=1}^{2} e_i'(v_i^j)\gamma_i(\cdot, \nabla\zeta_i^j) \cdot \nabla(\zeta_i^j - q_i v_0^j)\Big).$$

Here, using the strong monotonicity and Lipschitz continuity of γ_i, the first term can be estimated easily. Since $G_i \geq 0$ by (6.2), it remains to estimate $\|\sqrt{G_i}|\nabla v_0^j|\|_{L^2(\Omega)}$. Under Boltzmann statistics G_i vanishes. Thus we can apply (6.4) and find by means of the inequalities of Hölder, Gagliardo-Nirenberg, and Young

$$\int_\Omega G_i|\nabla v_0^j|^2\, dx \ \leq\ \|G_i\|_{L^{p/(p-2)}(\Omega)}\||\nabla v_0^j|\|_{L^p(\Omega)}^2 \leq c\|v_i\|_{L^{2p/(p-2)}(\Omega)}^2\||\nabla v_0^j|\|_{L^p(\Omega)}^2$$

$$\leq\ c\|\nabla v_i\|_{L^2(\Omega;\mathbb{R}^N)}^{2N/p}\|v_i\|_{L^2(\Omega)}^{2-2N/p}\|\nabla v_0^j\|_{L^p(\Omega;\mathbb{R}^N)}^2$$

$$\leq\ \frac{\delta}{4}\|\nabla v_i\|_{L^2(\Omega;\mathbb{R}^N)}^2 + c\|v_i\|_{L^2(\Omega)}^2\|\nabla v_0^j\|_{L^p(\Omega;\mathbb{R}^N)}^{2p/(p-n)}$$

$$\leq\ \frac{\delta}{2}\|\nabla\zeta_i\|_{L^2(\Omega;\mathbb{R}^N)}^2 + c\|\nabla v_0^j\|_{L^2(\Omega;\mathbb{R}^N)}^2 + c\|u_i\|_{L^2(\Omega)}^2.$$

From this the theorem follows. □

Remark 6.2. The electrostatic potential v_0 satisfies the Poisson equation at any time $t > 0$. Hence, the condition (6.4) reduces to a standard question of regularity: Does the gradient of the solution to a linear elliptic boundary value problem with a right–hand side in $L^\infty(\Omega)$ belong to $L^p(\Omega)$ for some $p > N$? Since the answer is positive for a Lipschitzian domain Ω and $N = 2$ (see [G1]) the assumption (6.4) can be omitted if $N = 2$. For $N \geq 3$ sufficient conditions for (6.4) to hold can be found in [Sh]. As to smooth data, a positive answer is given by the classical regularity theory (cf. [LU]).

7. Equilibria and asymptotic behaviour

First we want to describe the set of all steady states of the system under consideration. By a steady state we mean a pair $(u,v) \in V^* \times W$ such that $A(v,v) = 0$, $u = Ev$, and $u_0 = \sum_{i=1}^{n} q_i u_i$.

We know already from Theorem 4.3 that, if (u,v) is a solution to Problem (P), then all values $u(t)$ remain in the affine subspace $U + u^0$ of V^* (cf. (4.15) for the definition of U). Thus, one might expect that there exists a steady state (u,v) such that u is in this subspace. The following theorem confirms this expectation.

Theorem 7.1. *For every $u^0 \in V^*$ such that $u_0^0 = \sum_{i=1}^{n} q_i u_i^0$ and $\langle u_i^0, 1 \rangle > 0$, $i = 1, \ldots, n$, there exists a unique $(u,v) \in V^* \times W$ such that $A(v,v) = 0$, $u = Ev$, and $u \in U + u^0$.*

Proof. Let $u^0 \in V^*$ be given such that $u_0^0 = \sum_{i=1}^{n} q_i u_i^0$ and $\langle u_i^0, 1 \rangle > 0$, $i = 1, \ldots, n$.

1. Suppose that (u^j, v^j), $j = 1, 2$, are steady states satisfying $u^j \in U + u^0$. Then

$$\left\langle A(v^j, v^j), v^j \right\rangle = 0, \tag{7.1}$$

and this implies that $v^j \in U^\perp$ (note that (7.1) implies the right hand side of (4.9) to vanish [of course with η replaced by ζ^j]). Hence $u^1 - u^2 \in U$, $v^1 - v^2 \in U^\perp$, and

$$0 = \left\langle u^1 - u^2, v^1 - v^2 \right\rangle = \left\langle Ev^1 - Ev^2, v^1 - v^2 \right\rangle.$$

Since E is strictly monotone this is possible only if $v^1 = v^2$ and $u^1 = u^2$.

2. We define

$$\forall v \in V: \quad \Phi_0(v) := \Phi(v) + I_{U^\perp}(v) - \left\langle u^0, v \right\rangle; \tag{7.2}$$

here Φ is the functional introduced in (4.2) and $I_{U^\perp}$ is defined by

$$I_{U^\perp}(v) := \begin{cases} 0, & \text{if } v \in U^\perp, \\ +\infty, & \text{if } v \in V \setminus U^\perp. \end{cases}$$

It is easy to check that Φ_0 is bounded from below and that $\lim_{\|v\|_V \to \infty} \Phi_0(v) = +\infty$. Consequently, there exists $v \in V$ such that $\Phi_0(v) = \inf_{\bar{v} \in V} \Phi_0(\bar{v})$. Obviously, we have $v \in U^\perp$.

3. Let $\zeta_i := q_i v_0 + v_i$, $i = 1, \ldots, n$, where v is the minimal element of Φ_0. Since $v \in U^\perp$, the functions ζ_i are constant. Note that $\langle u^0, v \rangle = \sum_{i=1}^{n} \langle u_i^0, \zeta_i \rangle$. We define, for $w \in H^1(\Omega)$,

$$g(w) := \int_\Omega \left(\frac{\varepsilon}{2} |\nabla w|^2 - fw \right) dx + \int_\Gamma \left(\frac{\kappa}{2} w^2 - f^\Gamma w \right) d\Gamma + \sum_{i=1}^{n} \int_\Omega u_i^* \varphi_i(\zeta_i - q_i w) dx.$$

The definition of g is made in such a way that v_0 minimizes g. In particular, $\varphi_i(\zeta_i - q_i v_0) \in L^1(\Omega)$. Exploiting our assumptions with respect to the functions e_i we can show that, for $\bar{w} \in H^1(\Omega) \cap L^\infty(\Omega)$,

$$\langle g'(v_0), \bar{w} \rangle = \int_\Omega \left(\varepsilon \nabla v_0 \cdot \nabla \bar{w} - \Big(f + \sum_{i=1}^{n} q_i u_i^* e_i (\zeta_i - q_i v_0) \Big) \bar{w} \right) dx + \int_\Gamma (\kappa v_0 - f^\Gamma) \bar{w} \, d\Gamma. \tag{7.3}$$

Because v_0 minimizes g, we have $\langle g'(v_0), \bar{w} \rangle = 0$ for every $\bar{w} \in H^1(\Omega) \cap L^\infty(\Omega)$. Standard arguments show that the solution to the last equation is necessarily in $L^\infty(\Omega)$. Therefore the functions v_0 and $v_i = \zeta_i - q_i v_0$, $i = 1, \ldots, n$ are all in the space $L^\infty(\Omega)$. From $v \in U^\perp \cap L^\infty(\Omega; \mathbb{R}^{n+1})$ it follows that $A(v, v) = 0$.

4. Next we define

$$\forall \eta \in \mathcal{S}^\perp : \quad h(\eta) := \sum_{i=1}^n \left\{ \int_\Omega u_i^* \varphi_i (\eta_i - q_i v_0) dx - \left\langle u_i^0, \eta_i \right\rangle \right\}.$$

Since we know already that $v_0 \in L^\infty(\Omega)$ the value $h(\eta)$ is finite for every $\eta \in \mathcal{S}^\perp$. The definition of h implies that ζ minimizes h (ζ defined as above). Hence, for every $\bar{\zeta} \in \mathcal{S}^\perp$,

$$0 = h'(\zeta)\bar{\zeta} = \sum_{i=1}^n \left\{ \int_\Omega u_i^* e_i (\zeta_i - q_i v_0) dx \bar{\zeta}_i - \left\langle u_i^0, \bar{\zeta}_i \right\rangle \right\}.$$

Let $u := Ev$. Then $u_i = u_i^* e_i(v_i)$, $i = 1, \ldots, n$, and the last equation shows that $\left(\left\langle u_1 - u_1^0, 1 \right\rangle, \ldots, \left\langle u_n - u_n^0, 1 \right\rangle \right) \in \mathcal{S}$. From $\langle g'(v_0), \bar{w} \rangle = 0$ for $\bar{w} \in H^1(\Omega) \cap L^\infty(\Omega)$ and (7.3) it follows that $u_0 = \sum_{i=1}^n q_i u_i$ (cf. the definition of E). These facts show that $u \in U + u^0$. $\square$

In the remaining part of this section we are going to investigate the asymptotic behaviour of transient solutions as time tends to infinity.

In order to obtain satisfactory results we impose the following (rather mild) additional condition on the functions $r_{\alpha\beta}$ and $r^\Gamma \alpha\beta$ modeling the reactions:

$$\left. \begin{array}{ll} \forall x \in \Omega, \ \forall v \in \mathbb{R}^{n+1} : & r_{\alpha\beta}(x, v, y) - r_{\alpha\beta}(x, v, z) \geq m(z)(y - z), \\ \forall x \in \Gamma, \ \forall v \in \mathbb{R}^{n+1} : & r_{\alpha\beta}^\Gamma(x, v, y) - r_{\alpha\beta}^\Gamma(x, v, z) \geq m(z)(y - z), \\ \text{if } y, z \in \mathbb{R}, \ y > z, \ \text{where } m : \mathbb{R} \longrightarrow]0, \infty[\text{ is continuous.} \end{array} \right\} \quad (7.4)$$

Under this hypothesis we have the following

Theorem 7.2. *Let (u, v) be a solution to Problem (P). Then, for some $\mu > 0$,*

$$\|u(t) - \tilde{u}\|_{L^2(\Omega; \mathbb{R}^{n+1})}^2 + \|v_0(t) - \tilde{v}_0\|_{H^1(\Omega)}^2 \leq c \exp(-\mu t),$$

where $(\tilde{u}, \tilde{v})$ denotes the unique steady state in the affine space $U + u^0$ (cf. Theorem 7.1).

Proof. Let $(\tilde{u}, \tilde{v})$ be the steady state in $U + u^0$. We introduce $\zeta(t)$, $\bar{\zeta}(t)$, and $\tilde{\zeta}$ by

$$\zeta_i(t) := q_i v_0(t) + v_i(t), \quad \bar{\zeta}_i(t) := \int_\Omega \zeta_i(t) dx, \quad \tilde{\zeta}_i := q_i \tilde{v}_0 + \tilde{v}_i, \quad i = 1, \ldots, n.$$

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Then, taking into account that $u = Ev$ and $\tilde{u} = E\tilde{v}$, we obtain (using Poincaré's inequality)

$$
\left.
\begin{aligned}
&\|v_0(t) - \tilde{v}_0\|^2_{H^1(\Omega)} + \sum_{i=1}^n \|u_i(t) - \tilde{u}_i\|^2_{L^2(\Omega)} \\
&\leq c \Big\{ \langle u_0(t) - \tilde{u}_0, v_0(t) - \tilde{v}_0 \rangle + \sum_{i=1}^n \langle u_i(t) - \tilde{u}_i, v_i(t) - \tilde{v}_i \rangle \Big\} \\
&= c \sum_{i=1}^n \langle u_i(t) - \tilde{u}_i, \zeta_i(t) - \tilde{\zeta}_i \rangle = c \sum_{i=1}^n \langle u_i(t) - \tilde{u}_i, \zeta_i(t) \rangle \\
&\leq c \sum_{i=1}^n \Big(\|u_i(t) - \tilde{u}_i\|_{L^2(\Omega)} \|\nabla \zeta_i(t)\|_{L^2(\Omega;\mathbb{R}^N)} + \bar{\zeta}_i(t) \int_\Omega (u_i(t) - \tilde{u}_i)dx \Big).
\end{aligned}
\right\} \tag{7.5}
$$

Since $\left(\int_\Omega (u_1(t) - \tilde{u}_1)dx, \ldots, \int_\Omega (u_n(t) - \tilde{u}_n)dx \right) \in \mathcal{S}$ the estimate (7.5) proves that

$$
\|v_0(t) - \tilde{v}_0\|^2_{H^1(\Omega)} + \|u_i(t) - \tilde{u}_i\|^2_{L^2(\Omega)} \leq c \sum_{i=1}^n \|\nabla \zeta_i(t)\|^2_{L^2(\Omega;\mathbb{R}^N)} + c|P_{\mathcal{S}}\bar{\zeta}(t)|^2, \tag{7.6}
$$

where $P_{\mathcal{S}}\bar{\zeta}(t)$ denotes the orthogonal projection of $\bar{\zeta}(t)$ onto the subspace $\mathcal{S}$.

On the other hand starting from (4.9) and exploiting the hypothesis (7.4) one can easily show that, for some $\delta > 0$,

$$
\langle A(v(t), v(t)), v(t) \rangle \geq \delta \sum_{i=1}^n \|\nabla \zeta_i(t)\|^2_{L^2(\Omega;\mathbb{R}^N)} + \delta |P_{\mathcal{S}}\bar{\zeta}(t)|^2. \tag{7.7}
$$

Finally, we note that

$$
\Psi(u(t)) - \Psi(\tilde{u}) = \int_\Omega \frac{\varepsilon}{2} |\nabla(v_0(t) - \tilde{v}_0)|^2 dx + \int_\Gamma \frac{\kappa}{2} |v_0(t) - \tilde{v}_0|^2 d\Gamma
$$
$$
+ \sum_{i=1}^n \int_\Omega u_i^* \int_{\tilde{u}_i/u_i^*}^{u_i/u_i^*} (e_i^{-1}(y) - e_i^{-1}(\tilde{u}_i/u_i^*)) dy.
$$

Combining the preceding relations we find that, for sufficiently small $\mu > 0$,

$$
\exp(\mu t) \Big(\|v_0(t) - \tilde{v}_0\|^2_{H^1(\Omega)} + \sum_{i=1}^n \|u_i(t) - \tilde{u}_i\|^2_{L^2(\Omega)} \Big)
$$
$$
\leq c \exp(\mu t)(\Psi(u(t)) - \Psi(\tilde{u}))
$$
$$
= c \Big(\Psi(u^0) - \Psi(\tilde{u}) \Big) + c \int_0^t \exp(\mu s) \Big(\mu(\Psi(u(s)) - \Psi(\tilde{u})) - \langle A(v(s), v(s)), v(s) \rangle \Big) ds
$$
$$
\leq c \Big(\Psi(u^0) - \Psi(\tilde{u}) \Big).
$$

This proves the desired asymptotic behaviour of (u, v). $\square$

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