

## Electron-Transfer Reactions in Organic Chemistry

Sebastião J. Formosinho<sup>a</sup> and Luís G. Arnaut<sup>a</sup>

Chemistry Department, University of Coimbra, 3049 Coimbra, Codex, Portugal

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The Intersecting-State Model (ISM) is used to calculate the absolute rate constants of self-exchange electron-transfer reactions (ET) of organic species. The systems studied include aromatic hydrocarbons, quinones, nitrobenzene, aromatic nitriles, tetracyanoethylene, aromatic amines, and alkylhydrazines. All of the calculated rates are within one order of magnitude of the experimental ones, and the correlation coefficient between the two sets is 0.96. An electron-tunneling model has been developed to calculate distance-dependent nonadiabatic factors of intramolecular ET. This model can be used with ISM to calculate intramolecular ET rates. The system biphenyl-spacer-naphthyl in tetrahydrofuran, whose distance-dependent intramolecular rates were measured by Closs and Miller, was used to test our calculations, because its ET rates can be calculated without adjustable parameters. Our absolute rate calculations are in an order-of-magnitude agreement with the experimental ones.

Electron-transfer processes (ET) are an important class of reactions in organic chemistry. Their relevance is particularly evident when one of the reactants is an organic radical or an electronically excited molecule. Then, the transfer of an electron to another organic molecule may be the dominant reaction path. Some practical applications of such processes have been summarized elsewhere.<sup>1,2</sup> ET reactions are also remarkable for their theoretical simplicity. The exchange of electrons between identical reactants and products is the simplest chemical reaction in a homogeneous solution, because it does not involve any bond-breaking or bond-forming process, and  $\Delta G^\circ = 0$ . Such electron self-exchanges can be represented by



with  $Z_2 - Z_1 = 1$ , where  $Z_1$  is the charge in a reduced form of species A and  $Z_2$  the charge in an oxidized form of the same species; the asterisk is used only to distinguish between the otherwise identical species. Although electron self-exchanges have few practical consequences, they are of paramount importance in the development of theoretical models for ET reactions, and have been extensively studied, both theoretically and experimentally.

The electron-exchange rate between a radical ion and its parent molecule in a homogeneous solution is most conveniently measured using magnetic-resonance techniques. Depending on the nature of the solvent and counterions, the radicals may exist as free ions, ion pairs, or a mixture of both. This is particularly evident when sodium or potassium are used as reducing agents to form a radical ion from the neutral molecule. The electron-exchange rates are 2–3 orders of magnitude slower in tight ion pairs formed with alkali metal ions as counter ions, than in loose ion pairs.<sup>3–20</sup> An ion pair

involving an alkali metal counterion,



is a more complex system than that represented by mechanism (1). We are primarily interested in testing models that may lead to absolute estimates of the electron-exchange rates for the simplest organic systems. Thus, we focus attention on systems where tight ion pairs are not important. Experimentally, this normally corresponds to systems where the radical ions are produced by electrolysis in the presence of tetrabutylammonium iodide, tetrabutyl- or tetraethylammonium perchlorate as supporting electrolytes, or from dissociation in a solution of stable butylammonium or perchlorate salts. To have a consistent set of experimental electron-exchange rate constants to test the validity of theoretical models aimed at absolute rate-constant calculations, Table 1 was prepared with rates measured either in *N,N*-dimethylformamide (DMF) or in acetonitrile, because the static dielectric constants of these solvents are high and similar ( $\epsilon_s = 36.7$  and  $\epsilon_s = 35.94$ , respectively). Furthermore, this table does not include data for systems involving simple changes of substituents, since at room temperature these lead to rate variations of less than a factor of 4;<sup>21</sup> i.e., benzonitrile is included, but methyl-substituted benzonitriles are omitted.<sup>21</sup>

Most theoretical approaches to the kinetics of the reactions illustrated by (1) are based on a theory developed by Marcus for inorganic ET reactions.<sup>2–11</sup> According to this theory, such reactions follow an outer-sphere mechanism, which postulates weakly bonded reactants (i.e. the electronic interaction between the reactants is less than  $5 \text{ kJ mol}^{-1}$ ), and their rates can be calculated from

$$k_{\text{ET}} = \kappa_{\text{el}} Z \exp(-\Delta G^\ddagger/RT), \quad (3)$$

where  $\kappa_{\text{el}}$  is a nonadiabatic factor ( $\kappa_{\text{el}} < 1$  for nonadiabatic

<sup>a</sup> Also Universidade Católica Portuguesa, 3500 Viseu, Portugal.

# Electron Transfer Reactions In Organic Chemistry

**Shasha Hu**



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