行政院國家科學委員會專題研究計畫 成果報告

球正對圓盤面電泳的理論分析(3/3)
研究成果報告(完整版)

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中 華 民 國 97 年 06 月 27 日

球正對圓盤面電泳的理論分析(3/3)
Electrophoresis of a Charge-Regulated Sphere Normal to a Large Disk (3/3)

計畫編號: NSC96-2221-E-002-079
執行期限: 96/08/01~97/07/31
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一、中文摘要

本研究在低表面電位及均勻若電場下探討環形粒子朝向一無窮大圓盤(或平板)的電泳行為並進行數值模擬。本研究在此系統下成功模擬細菌 DNA、質體 DNA 以及藻類等多種生物膠體在接近平面時之電泳動行為。所考慮環形粒子的表面荷電性質介於傳統固定電位與固定電荷密度模式之間，即所謂電荷可調整模式。文中並探討環形粒子之大小、環形粒子與無窮平板間距離、環形粒子與平板之表面電荷情況以及電雙層厚度等變因對環形粒子電泳動遷移率之影響。

關鍵詞: 電泳；邊界效應；電荷可調節粒子

二、計畫緣由與目的

For instance, bacterial DNA, plasmid DNA from within eukaryotic cells, and most viral DNA, all can be isolated as a single intact chromosome, is of toroid shape. DNA condensation in vitro has attracted much attention in gene therapy.[1] Within living cells, DNA is highly condensed in toroidal arrays as compared with free DNA in solution, especially in sperm cells and viruses where gene transcription is inactive.[2-4] Kong et al.[5] determined the structure of the beta subunit in E. coli and found that it was also a toroid which contained a hole big enough to encircle double-stranded DNA. They proposed that the beta subunit acts as a sliding clamp to hold the polymerase III. The toroid shape appearing in organisms, for example of a condensed DNA, is proposed to be related to the electrostatic charge density and water activity of the immediate microenvironment of the double-helix in DNA helix.[1] Anabaenopsis is another example of toroidal biocolloids.

The boundary effect on electrophoresis has been studied intensively in the literature. Various types of geometry have been considered and analytical,[6,7] semi-analytical,[8] and numerical results reported.[9-15] Due to the difficulty involved in solving the governing equations, analytical results are available mainly under drastic assumptions such as simple geometry, low surface potential, and infinitely thin or infinitely thick double layers. As pointed out by Ninham and Parsegian,[16] these are idealized, extreme conditions, and the actual situation is somewhere between the two. They proposed using a charge-regulate model where the surface of a particle carries ionizable groups and the dissociation/association of them yields fixed charge. Several attempts have been made to simulate the behavior of a system containing charge-regulated entities.[16-25] In this study, a toroid is of charge-regulated nature, which mimics bio-colloids such as cells, microorganisms, and DNA, and particles covered by a membrane layer, and the disk may be charged, that is, the effect of EOF can be significant. We focus on the classic electrophoresis problem, that is, a particle is driven solely by an applied electric field. Other possible driving forces, such as the concentration gradient near electrode surface and related gradient of electric field strength arising, for example, from surface reactions or deposition of particles,[26] are not within the scope of our analysis.

三、理論

Let us consider the electrophoresis of a rigid, nonconducting, toroid (doughnut-shaped) of inner radius (b-a) and outer radius (b+a) normal to an infinite, conducting disk illustrated in Figure 1. Let h be the center-to-center distance between the toroid and the disk. The cylindrical coordinates (r,θ,z) with its origin located at the center of the disk are adopted. The space between the toroid and the disk is filled with an incompressible Newtonian fluid of constant physical properties containing electrolytes. A uniform electric field $\mathbf{E_0}$ of strength E_0 is applied in

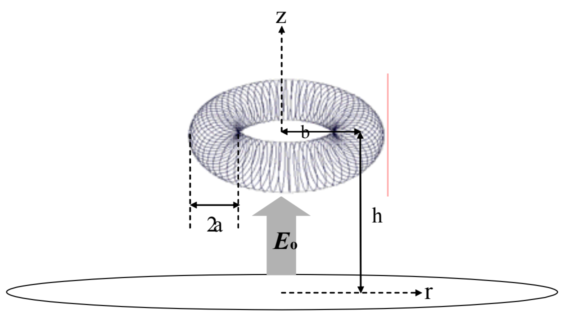

Figure 1. Systematic representation of the problem

the z-direction. Because of the axisymmetric nature of the present problem only the (r,z) domain needs be considered. We assume that the toroid moves in a completely symmetric fashion without rotation. For convenience, the electrical potential Ψ is partitioned into the equilibrium potential Ψ_1 arising from the charge on the toroid and the disk surface, and a perturbed potential Ψ_2 arising from $\mathbf{E_0}$. If $\mathbf{E_0}$ is weak and the surface potential is low, then it can be shown that Ψ_1 and Ψ_2 satisfy

$$\nabla^2\Psi_1 = \kappa^2\Psi_1 \quad (2)$$

$$\nabla^2\Psi_2 = 0 \quad (3)$$

where ∇^2 is the Laplace operator, κ^{-1} is the Debye length, ε is the permittivity, e is the elementary charge, n_{j0} is the bulk number concentration of jth ionic species, k is the Boltzmann constant, and T is the absolute temperature. We assume the following boundary conditions:

$$\mathbf{n}\cdot\nabla\Psi_1 = -\frac{\sigma_P}{\varepsilon} \text{ and } \mathbf{n}\cdot\nabla\boldsymbol{\Psi}_2 = 0 \text{ on surface} \quad (4)$$

$$\Psi_1 = \zeta_w \text{ and } \Psi_2 = \text{constant at } z=0 \quad (5)$$

$$\Psi_1 = 0 \text{ and } \nabla\Psi_2 = -E_0\mathbf{e}_z \text{ as } z\to\infty \quad (6)$$

$$\mathbf{n}\cdot\nabla\Psi_1 = 0 \text{ and } \mathbf{n}\cdot\nabla\Psi_2 = 0 \text{ as } r\to\infty,\ z>0 \quad (7)$$

Here, σ_P is the surface charge density of a toroid, ζ_w the surface potential of the disk, $\mathbf{n}$ the unit normal vector directed into the liquid phase, and $\mathbf{e_z}$ the unit vector in the z-direction.

Suppose that the surface of a toroid contains both acidic and basic functional groups, and the dissociation of them can be expressed by

$$AH \leftrightarrow A^- + H^+ \quad (8)$$

$$QH^+ \leftrightarrow Q + H^+ \quad (9)$$

Let K_a and K_b be the dissociation constants of these reactions. We have

$$K_a = \frac{[\mathrm{A}^-]_\mathrm{S}[\mathrm{H}^+]_\mathrm{S}}{[\mathrm{AH}]_\mathrm{S}} \quad (10)$$

$$K_b = \frac{[\mathrm{Q}]_\mathrm{S}[\mathrm{H}^+]_\mathrm{S}}{[\mathrm{QH}^+]_\mathrm{S}} \quad (11)$$

where subscript S denotes surface property. If we let N_s and N'_s be the total number of the acidic and the basic functional groups per unit area, then

$$N_s=[\mathrm{A}^-]_s+[\mathrm{AH}]_s \quad (12)$$

$$N'_s=[\mathrm{QH}^-]_s+[\mathrm{Q}]_s \quad (13)$$

We assume

$$[\mathrm{H}^+]_s = C^0_{H^+}\exp(-\frac{e\zeta_P}{kT}) \quad (14)$$

where $C^0_{H^+}$ is the bulk concentration of H^+. Combining eqns. 11, 13-14 yields

$$[\mathrm{QH}^+]_S = \frac{N'_S\frac{C^0_{H^+}}{K_a}\exp(-\frac{e\zeta_P}{kT})}{1+\frac{C^0_{H^+}}{K_a}\exp(-\frac{e\zeta_P}{kT})} \quad (15)$$

Therefore, the charge density of the associated basic groups on toroid surface, $\sigma_{P,b}=e[\mathrm{QH}^+]_s$, is

$$\sigma_{P,b} = \frac{eN'_S\frac{C^0_{H^+}}{K_a}\exp(-\frac{e\zeta_P}{kT})}{1+\frac{C^0_{H^+}}{K_a}\exp(-\frac{e\zeta_P}{kT})} \quad (16)$$

If ζ_P is low, this expression can be

$$\sigma_{P,b} = \frac{eN'_S\frac{C^0_{H^+}}{K_b}}{1+\frac{C^0_{H^+}}{K_b}} - \frac{e^2N'_S\frac{C^0_{H^+}}{K_bkT}}{(1+\frac{C^0_{H^+}}{K_b})^2}\zeta_P \quad (17)$$

It can be shown that the surface charge density of the dissociated acidic functional groups, $\sigma_{P,a}$, is [25]

$$\sigma_{P,a} = \frac{-eN_S}{1+\frac{C^0_{H^+}}{K_a}} - \frac{e^2N_S\frac{C^0_{H^+}}{K_bkT}}{(1+\frac{C^0_{H^+}}{K_a})^2}\zeta_P \quad (18)$$

Substituting eqns. 17 and 18 into eqn. 4 yields

$$\mathbf{n}\cdot\nabla\Psi_1 = -\frac{\sigma_{P,b}+\sigma_{P,a}}{\varepsilon} = -\frac{eN'_S\frac{C^0_{H^+}}{\varepsilon K_b}}{1+\frac{C^0_{H^+}}{K_b}} + \frac{e^2N'_S\frac{C^0_{H^+}}{\varepsilon K_bkT}}{(1+\frac{C^0_{H^+}}{K_b})^2}\Psi_1 + \frac{\frac{eN_S}{\varepsilon}}{1+\frac{C^0_{H^+}}{K_a}} + \frac{e^2N_S\frac{C^0_{H^+}}{\varepsilon K_akT}}{(1+\frac{C^0_{H^+}}{K_a})^2}\Psi_1 \quad (19)$$

or

$$\mathbf{n}\cdot\nabla^*\Psi_1^* = -\frac{\beta AB}{\alpha(1+\beta B)} + \frac{\beta AB}{\alpha(1+\beta B)^2}\Psi_1^* + \frac{A}{1+B} + \frac{AB}{(1+B)^2}\Psi_1^* \quad (20)$$

where $\nabla^* = a\nabla$, $\Psi_1^* = e\Psi_1/kT$, $A = e^2N_Sa/\varepsilon kT$, $B = C^o_{H^+}/K_a$, $\alpha = N_S/N'_S$ and $\beta = K_a/K_b$. The scaled charge density on toroid surface, σ_P^*, can be expressed as

$$\sigma_P^* = \sigma_{P,b}^* + \sigma_{P,a}^* = \frac{\beta AB}{\alpha(1+\beta B)} - \frac{\beta AB}{\alpha(1+\beta B)^2}\zeta_P^* - \frac{A}{1+B} - \frac{AB}{(1+B)^2}\zeta_P^* \quad (21)$$

where $\sigma_P^* = e\sigma_P a/\varepsilon kT = e(\sigma_{P,b}+\sigma_{P,a})a/\varepsilon kT$ and $\zeta_P^* = e\zeta_P/kT$.

If we let η, u, p, and U be respectively the viscosity and the velocity of the fluid, the hydrodynamic pressure, and the speed of the toroid in the z-direction, then the governing equations and the associated boundary conditions for the flow field can be described by

$$\nabla\cdot\mathbf{u} = 0 \quad (22)$$

$$\eta\nabla^2u - \nabla p = \rho\nabla\Psi \quad (23)$$

$$\mathbf{u} = U\mathbf{e}_z \text{ on toroid surface} \quad (24)$$

$\mathbf{u} = 0$ at z=0, $z \to \infty$, and $r \to \infty$ (25)

In our case, only the z-components of the relevant forces need be considered. These include the electrostatic force and the hydrodynamic force. The z-component of the former acting on a toroid can be calculated by integrating the Maxwell stress tensor $\boldsymbol{\sigma}^E = -\varepsilon \mathbf{EE} - (1/2)\varepsilon E^2 \mathbf{I}$ over the toroid surface S,

$$F_E = \iint_S (\boldsymbol{\sigma}^{\mathrm{E}} \cdot \mathbf{n}) \cdot \mathbf{e}_z dS \quad (26)$$

Here, $\mathbf{E} = -\nabla \Psi = \mathbf{n}(\partial \psi / \partial n) + \mathbf{t}(\partial \psi / \partial t)$ is the total electric field, **I** and **t** are respectively the unit tensor and the unit tangential vector on the toroid surface, n and t are, respectively, the magnitude of **n** and **t**, and E^2=**E•E.** For the present case, it can be shown that eqn. 26 leads to[27,28]

$$F_E = \iint_S \varepsilon [(\frac{\partial \Psi_1}{\partial n}\frac{\partial \Psi_2}{\partial z}) - (\frac{\partial \Psi_1}{\partial t}\frac{\partial \Psi_2}{\partial t}) n_Z] dS \quad (27)$$

The z-component of the hydrodynamic force acting on a toroid, F_D, can be evaluated by

$$F_D = \iint_S \eta \frac{\partial (\mathbf{u} \cdot \mathbf{t})}{\partial n} t_z dS + \iint_S - p n_z dS \quad (28)$$

where t_z and n_z are the z-component of t and that of n, respectively. At steady state, the total force acting on a toroid in the z-direction vanishes, that is,

$$F_E + F_D = 0 \quad (29)$$

For convenience, the problem under consideration is decomposed into two sub-problems. In this case, it experiences a hydrodynamic force $F_{D,1} = -UD$, D (>0) being the drag per unit velocity. In the second sub-problem, an external electric field is applied, but a toroid is held fixed. In this case, it experiences an electrostatic force F_E and an electric body force $F_{D,2}$. Both F_E and $F_{D,2}$ are functions of κa, (b/a), (h/a), A, B, α, and β; $F_{D,1}$ (or D) is a function of (b/a) and (h/a), but is independent of κa. Since $F_D = F_{D,1} + F_{D,2}$, eqn. (29) gives

$$U = \frac{F_E + F_{D,2}}{D} \quad (30)$$

The electrophoretic mobility of a toroid is evaluated based on the procedure adopted previously.[29]

四、結果與討論

For a more concise presentation, the scaled electrophoretic mobility $\omega = U / U_0$ is used in subsequent discussions, where $U_0 = \varepsilon (kT/e) E_0 / \eta$ is the electrophoretic velocity of a particle with a constant surface potential (kT/e) predicted by the Smoluchowski's theory when an electric field of strength E_0 is applied. Also, we define $F_E^* = F_E / F_{D,1,IS}$, $F_{D,2}^* = F_{D,2} / F_{D,1,IS}$, and $D^* = D/(F_{D,1,IS}/U_0)$, where $F_{D,1,IS} = 6\pi\mu a U_0$ is the conventional hydrodynamic force of an isolated sphere moving in an incompressible Newtonian fluid at steady state. Based on these scaled symbols, ω can be expressed as

$$\omega = \frac{F_E^* + F_{D,2}^*}{D^*} \quad (31)$$

Figure 2 shows the variations of the scaled electrophoretic mobility ω and the scaled forces F_E^*, and $F_{D,2}^*$ as a function of κa at various (b/a)'s for the case when A=1, B=1, α =4, β =4, and h/a=2.

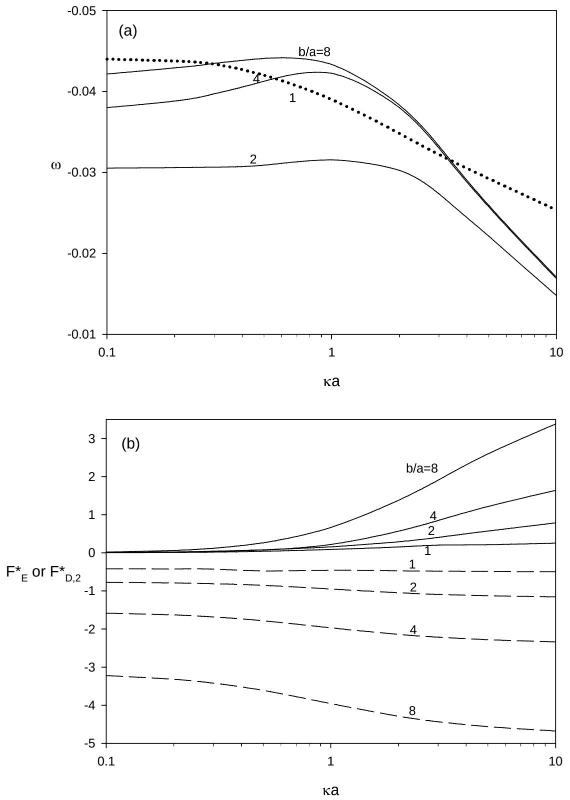

Figure 2. Variation of scaled electrophoretic mobility ω, (a), and F_E^* and $F_{D,2}^*$, (b), as a function of κa at various (b/a)'s .

Under these conditions, the amount of negative charge on the surface of a toroid is larger than that of positive charge, and the direction of its movement is opposite to that of the applied electric field. Figure 3 shows the influence of parameter A ($= e^2 N_S a / \varepsilon kT$) on the scaled electrophoretic mobility ω and the scaled forces F_E^*, and $F_{D,2}^*$ at various (b/a)'s for the case when B=0.3, α =2, β =0.5, κa =1, and h/a=2.

Note that an initially neutral toroid becomes charged as it approaches a charged disk, and an osmotic pressure field is established. These affect appreciably the behavior of the toroid. Figure 4 shows the variations of the scaled electrophoretic mobility ω and the scaled forces F_E^*, and $F_{D,2}^*$ as a function of κa at various (b/a) when A=1, B=1, α =4, β=4, and h/a=2.

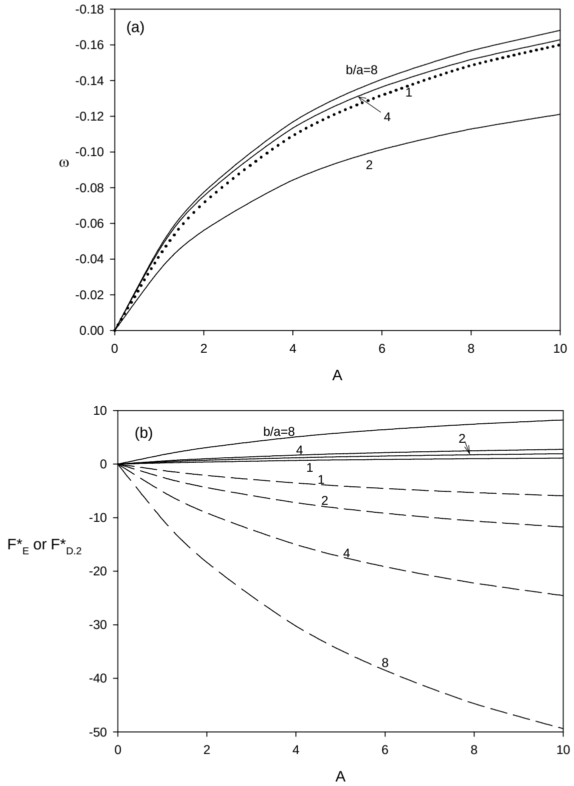

Figure 3. Variation of scaled electrophoretic mobility ω, (a), and F_E^* and $F_{D,2}^*$, (b) as a function of A at various (b/a)'s .

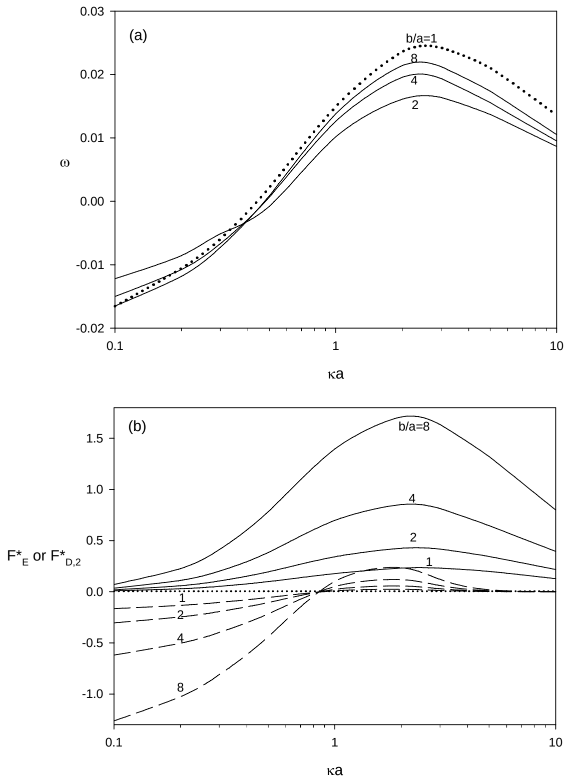

Figure 7. Variation of scaled electrophoretic mobility ω, (a), and F_E^* and $F_{D,2}^*$, (b), as a function of κa at various (b/a)'s

五、參考文獻

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行政院國家科學委員會補助國內專家學者出席國際學術會議報告

97年 6月 23日

附件三

報告人姓名	徐治平	服務機構及職稱	台大化工系　教授
會議 時間 地點	97.6.16 至 97.6.19 孔印布拉，葡萄牙	本會核定補助文號	NSC96-2221-E-002-079
會議名稱	(中文) 第7屆國際聚電解質研討會 (英文) 7th International Symposium on Polyelectrolytes (Polyelectrolytes 2008)		
發表論文題目	(中文) 聚電解在圓柱孔洞內之電泳 (英文) Electrophoresis of a Polyelectrolyte Entity in a Cylindrical Pore		

報告內容應包括下列各項：

一、參加會議經過
二、與會心得
三、考察參觀活動(無是項活動者省略)
四、建議
五、攜回資料名稱及內容
六、其他

一、參加會議經過

國際聚電解質研討會每二至三年舉辦一次，自 1995 於德國舉辦第一屆會議後，分別在日本(1998)、法國(2000)、瑞典(2002)、與美國(2004)、與德國(2006)舉辦，為國際間發表聚電解質相關研究成果的主要學術會議。本屆會議之主題包括：Biopolyelectrolytes、Polyelectrolyte complexes、Polysaccharides、Formulations and gels、Conformation and counterion binding、Self-assembly、Synthesis、Transport and rheology 等七大項。

國際聚電解質研討會之主旨在於提供工程師、科學家、研究人員、技術人員、與學生一個發表最新研究成果、交換意見、交流、建立合作研究管道、與其他學術功能的平台。由於其所牽涉到的領域相當廣泛，包括物理、化學、生物、醫學、與工程等，與聚電解質相關之基礎與應用已廣受重視。雖然此次會議並非大型的研討會，但是多數科技領先國家皆有代表與會；國內與大陸皆有數位學者參加。本會提供了一個理想的學術交流機會；一方面可以了解國際間關於聚電解質領域的重要發展與應用，另方面也達到了學術與文化外交的目的。本屆會議參與者包括全世界共 36 個國家共約 200 位的學者專家，主要的科技先後國家都有代表與會，是一個規模相當理想的國際學術會議。會

議期間除了關於聚電解質的各個主要領域課題之論文發表外，還有專題演講 6 篇、邀請演講 6 篇、口頭報告論文 71 篇、與海報論文 156 篇。整體而言，該研討會是一個精心籌劃、內容豐富、與執行成功的中小型國際學術會議。

筆者所發表的論文為『聚電解在圓柱孔洞內之電泳』(Electrophoresis of a Polyelectrolyte Entity in a Cylindrical Pore)。吾人以球在圓柱孔洞內之幾何架構，探討了邊界效應對電泳影響。主要的貢獻在於考慮了因邊界帶電所引發的電滲透流以及電泳粒子具多孔的特質，分析上具有相當的困難度，文獻中目前尚少見相關的探討。筆者於 6 月 14 日自台北啟程，於 15 日抵達孔印布拉(Coimbra)市，當日即完成報到註冊並熟析環境。會期中全程參與所有的學術及其它相關活動。會議結束後於 20 日啟程返國，於 6 月 21 日下午抵達台北。

二、與會心得

除了前兩屆之外，國際聚電解質研討會每二年舉辦一次，目前已是國際間發表聚電解質相關研究成果的主要學術會議之一。孔印布拉市是一個人口數萬人的大學城，其中學生即占了約半數。相較於其他國家，國人前往葡萄牙旅遊或進行學術或商業活動者相當少，有限的人次中又以前往首都里斯本者為多，鮮有前往孔印布拉市者。此次會議雖由孔印布拉大學(University of Coimbra)化學系主辦，但是孔印布拉市政府亦給予相當程度的支持與配合，例如提供活動場地。化學系內的投入也相當熱心，包括人力與行政資源的配合。整體而言，從規劃到執行，會議舉辦得算是相當成功，有許多地方值得我們學習。不過，也有一些小缺點，例如孔印布拉市與位於里斯本的國際機場有相當的距離，但是大會對於交通相關資訊的提供較不理想。另外，活動內容過於緊湊，較缺乏一些關於孔印布拉市的導覽與介紹，也有些遺憾。孔印布拉大學創立於 13 世紀，是歐洲，也是全世界歷史悠久的大學之一；校地雖然不算大，但是校園建築極具特色，特別是對於老舊建築的維修別具巧思。主辦單位化學系的設備與研究資源比不上國內大學的平均水準，但是以有限的人力與物力可以舉辦 Polyelectrolytes 這類國際會議，相當不容易。

葡萄牙在科技與工業上雖然並不突出，但是其文化藝術氣息濃厚，與歐盟其他歷史悠久的國家相較，並不遜色。特別是自然地呈現在生活與環境中，值得我們學習。孔印布拉市的都市建設及其環境較國際或我國同類型的城市為佳，建築雖非極具特色，但是整體感覺很好，不像國內因缺乏整體規劃而顯得不協調。英語雖非其國語，但是市民對於其尚稱熟悉，較我國為優。市民對於觀光客相當友善，也不吝於提供協助，市內治安甚佳，非我們所能及。孔印布拉市對於文化傳統的維護相當算是用心，對於市民的生活品質亦十分重視；街道與建築

雖然古舊，但是整潔而不髒亂。與台北市相較，孔印布拉市的生活水準與物價較低；整體而言，市民的日常生活簡單而不奢華；多數商家在晚間八時即已打烊，市民亦已就寢。這次是筆者第一次訪問葡萄牙及孔印布拉，相較於歐洲其他國家及地方，這裡是有趣且值得造訪的地方。

三、考察參觀活動(無是項活動者省略)

四、建議

歐盟對於推動國際學術合作與交流不遺餘力,許多成員國之政府與學術單位皆積極的支持與鼓勵國人參與，投入可觀的資源。這使得許多工作的推動達到事半功倍的效果。相較之下，我國在國際學術合作與交流之推動上，待努力的地方尚多，迄今亦尚缺乏舉辦如國際聚電解質研討會規模之國際會議的經驗與能力。第八屆國際聚電解質研討會已訂於 2010 年在上海舉辦,顯示大陸在爭取學術會議舉辦方面的努力已見成效，其學術水準與相關之配套措施的能力受到國際肯定。事實上，在整體華人世界中，大陸在許多領域中已處於領先的地位；而相對地，我國過去許多的優勢已不再具有，甚至已處於落後的地位，值得國內各界重視並一同努力，迎頭趕上。

五、攜回資料名稱及內容

會議之詳細內容及摘要合訂本。

六、其他