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科　　　別：工程學

作 品 名 稱：電容超音波膠體金粒子電位調控系統研發

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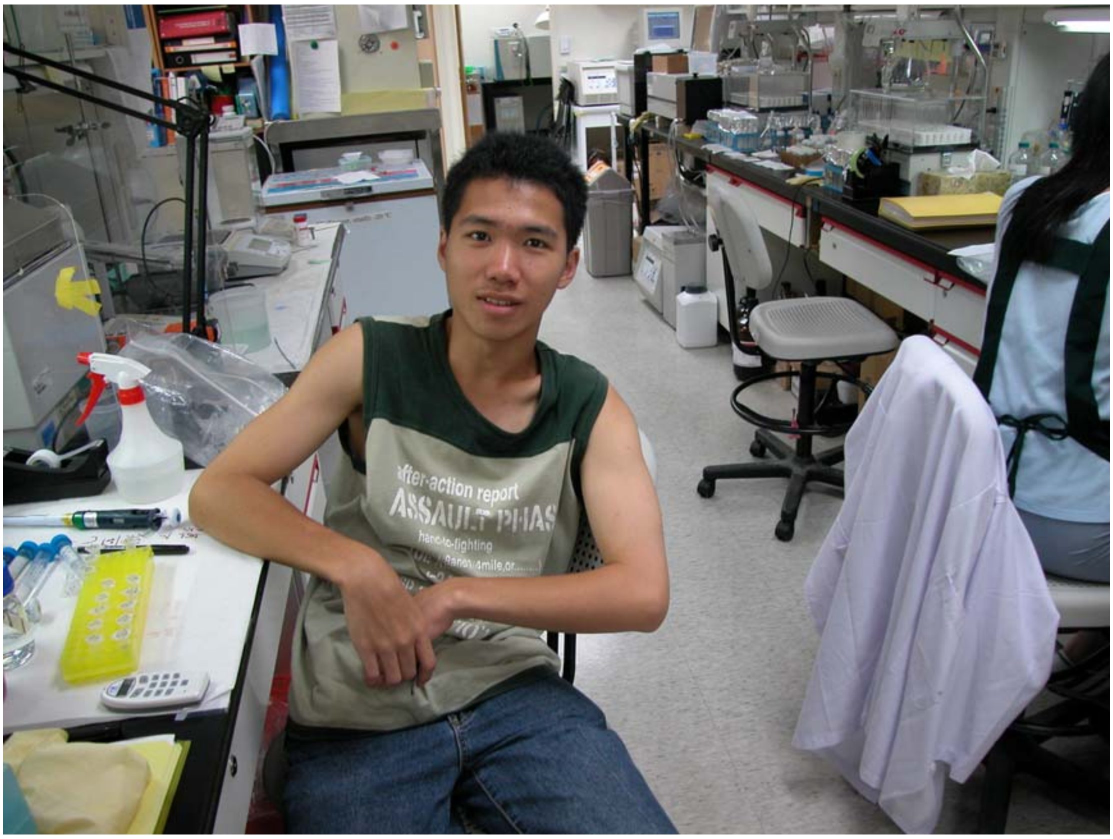
作者簡介

鍾介恆



我從小學三年級就開始做科展，算算已經七年了，參加國際科展則是第四年。這四年當中，科展讓我的生活比其他的同學多了一塊……挑戰？經驗？不管如何，我很高興我的生活中有這一部份，讓我有機會在台大和中研院做實驗，接受教授們的指導。最後，我在這裡感謝這四年來所有幫助過我的人，讓我有支持下去的毅力和決心。

作者簡介

簡佑軒

我是簡佑軒，於民國 79 年出生於台北，目前就讀麗山高中二年級，從小對各種領域都有涉獵，尤其對語文和自然科學。

由於學校致力教育一批具有科學實做能力的青年，因此特別開放其他高中沒有的科學領域專題課程，而我在高一時就對物理有濃厚的興趣，所以高一就跟著老師做科展。我曾研究過位能井不同梯度下所擁有的向心力、水滴模擬布朗運動，以及篩選奈米金顆粒大小以運用在各領域。

我相信自己想成就什麼，並不是由天賦決定，而是自己的選擇，因此我盡全力做每件事，並享受可以成長的機會。

題目：**電容超音波膠體金粒子電位調控系統研發**

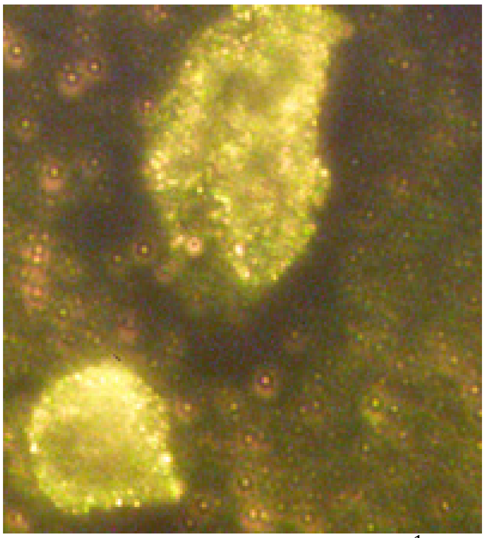
摘 要

費曼曾說：There is plenty of room at the bottom。喬治亞理工大學的 Mostafa El-Sayed 教授發表的癌細胞辨識、與科學月刊報導『台大抗煞一號』引發我們對膠體金粒子的興趣。膠體的性質主要是由界達電位 (zeta potential)決定。參考台科大、成大、中山…等超音波應用研究，提出改良篩選物理法製造之膠體金粒子的儀器設計與製作。

經沉降過濾可達平均粒徑 100 nm；而離心式篩選機與超音波管式篩選機可達平均粒徑 30 nm。篩選後的膠體粒子以電容原理調控膠體金粒子之界達電位 (zeta potential)，成功地從-30 mV 提升至-59 mV，並發展成**電容超音波界達電位控制儀(**Capacitor Ultrasonic Zeta Potential Controller)。

以膠體金粒子與蛋白質鍵結量來測試調控界達電位的效果，發現蛋白質鍵結量之增加曲線與界達電位的增加曲線的增加趨勢相似；此功能的發現對於生物科技方面的應用應會有很大的幫助。

透過**界達電位控制系統**，本研究達到費曼先生所期望的「**在原子或分子的尺度上來加工材料和製造設備**」。



以金膠體粒子辨識癌細胞[1]
Source: Mostafa El-Sayed

[1] Georgia Tech News Room, Gold Nanoparticles May Simplify Cancer Cells.

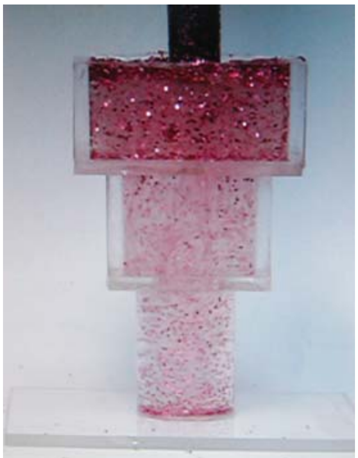
Abstract

"There is plenty room at the bottom." The words of Mr. Feynman are the beginning of nano technology. Mostafa El-Sayed, a professor of Georgia Institute Technology, identified cancer cells through nano gold-antibody complex. So, our study focuses on the zeta potential of colloidal gold particles.

At first, the filtering method and equipments were developed. The theories were based on the ultrasonic studies of universities such as National Taiwan University of Science and Technology. Then the colloidal gold's sizes were filtered to100 nm through settling. At last, by using Continual-Filtering Centrifuge (CoCe.) and Tube Well Mass (TW-MS), the mean particles sizes can be filtered to 30 nm.

The most important results are: Zeta potential of the gold colloid was controlled with Capacitor Ultrasonic Zeta Potential Controller. The zeta potential can be raised from -30 mV up to -59 mV, which is -20 mV higher than the conventional pH-changing way. The function of zeta potential to protein binding quantity was tested. The increasing curves of zeta potential and protein binding quantity were similar. This property would be a significance of biotechnology.

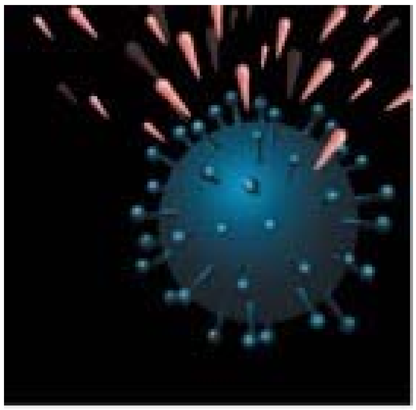
Thourgh Capacitor Ultrasonic Zeta Potential Control system, the zeta potential's limitation of gold colliod, which is produced by SANSS (Submerged Arc Nanoparticles Synthesis System), can be controled in a wilder range. The study which is focused on nano-scale, like the wish of Mr. Feynman – "To manufacture material and produce equipment in atom and molecular scale".



超音波振動水模模擬

Theory of of ultrasonic

Source: T.S. Lei



Mechanism of『NTU Anti-SARS No.1』

Source: C.K. Lee

目　錄

表圖索引

壹、前 言

一、研究動機

1965 年的諾貝爾物理學獎得主費曼(Richard F. Feynman)的名言「**底下空間還很大**(There is plenty of room at the bottom.」象徵奈米時代的開始[1]。目前當紅的奈米科技，正是在研究如何達到費曼**(見照片 2)**先生提到的「**在原子或分子的尺度上來加工材料和製造設備**」

在 SARS 流行期間由台灣大學結合多個學院及台大醫院，研發出一種藉由相反電性吸引崩解 SARS 病毒蛋白質外殼的「台大奈米生科一號」，亦稱為『抗煞一號』**(見照片 1。)**[2]

2005 年，喬治亞理工大學的教授 Mostafa A. El-Sayed，藉由接有 EFGR(Epidermal Growth Factor Receptor, 乳癌細胞的蛋白質)之抗體的奈米金膠體粒子來辨識癌細胞**(見照片 3)**，有非常顯著的效果。[3]

不論是金顆粒與抗體的鍵結還是崩解病毒外殼，都和膠體粒子的一個重要特性有關—界達電位(zeta potential)，傳統調整界達電位的方法是利用 pH 值[4]，但界達電位在 0 mV 的時候不穩定，且極端的酸鹼度對於應用(如蛋白質的活性)可能會有不良的影響，本研究是在以電容法(物理法)控制並提高金膠體的表面電位，並提出儀器設計、構想與製作。

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1 微觀世界─高中生奈米科技營研習手冊,教育部,北區奈米科技 K-12 教育發展中心

2 李世光, 林世明, 李世元, 蘇慶琅, 歐芳序, 抗煞一號背後的奈米生物科技, 科學月刊, 2003, 34(8), 713-717.

3 I.H. El-Sayed; X.H Huang; M.A. El-Sayed, Nano Letts ,Surface Plasmon Resonance Scattering and Absorptin of Anti-EGFR antibody Conjugated Gold Nanoparticles in Cancer Diagnostics: Applications in Oral Cancer,,2005, 5 (5), 829-834.

4 D.J. Shaw, D.H. Everett, S. Ross, I.D. Morrison, J. Lyklema,Zeta Potential An Introduction in 30 minutes, Malvern Instruments Ltd,

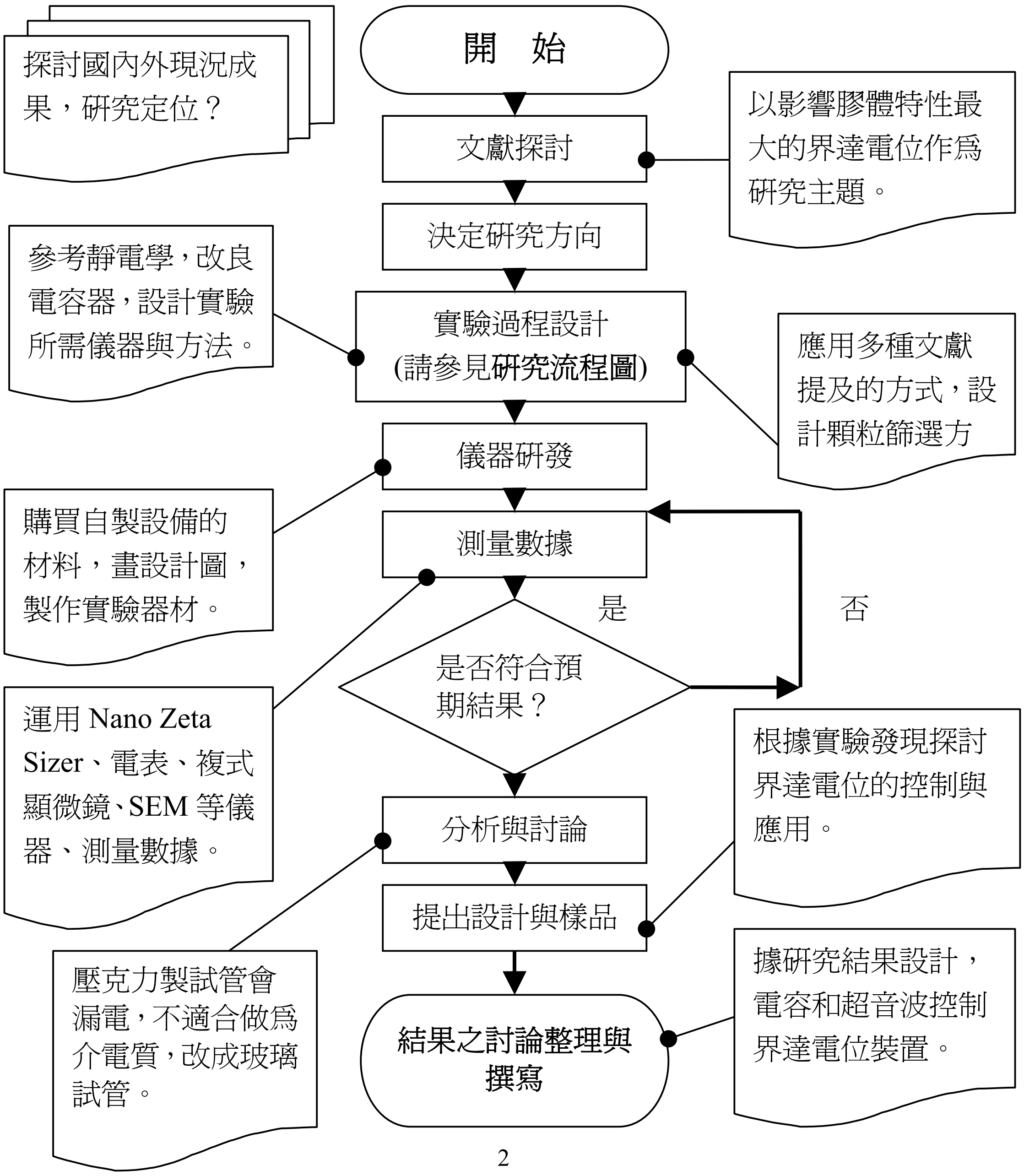
二、研究目的

(一) 以電容法控制膠體金粒子之界達電位(zeta potential)，並研發界達電位調控儀。

(二) 探討改變界達電位的電容法(物理法)和傳統 pH 值調整法(化學法)比較。

(三) 評估不同的膠體金粒子篩選方式，提出以超音波**(見照片 4, 5)**改良膠體金粒子分散篩選機設計構想與試製。

三、研究流程



貳、研究方法與過程

一、材料

由參考資料提供的應用物理、化學原理與工程知識分析後，確定以下表列的主要材，並以產業研發新產品之物料管控 BOM 表(Bill of Materials)表示：

表一、沉降篩選之 BOM

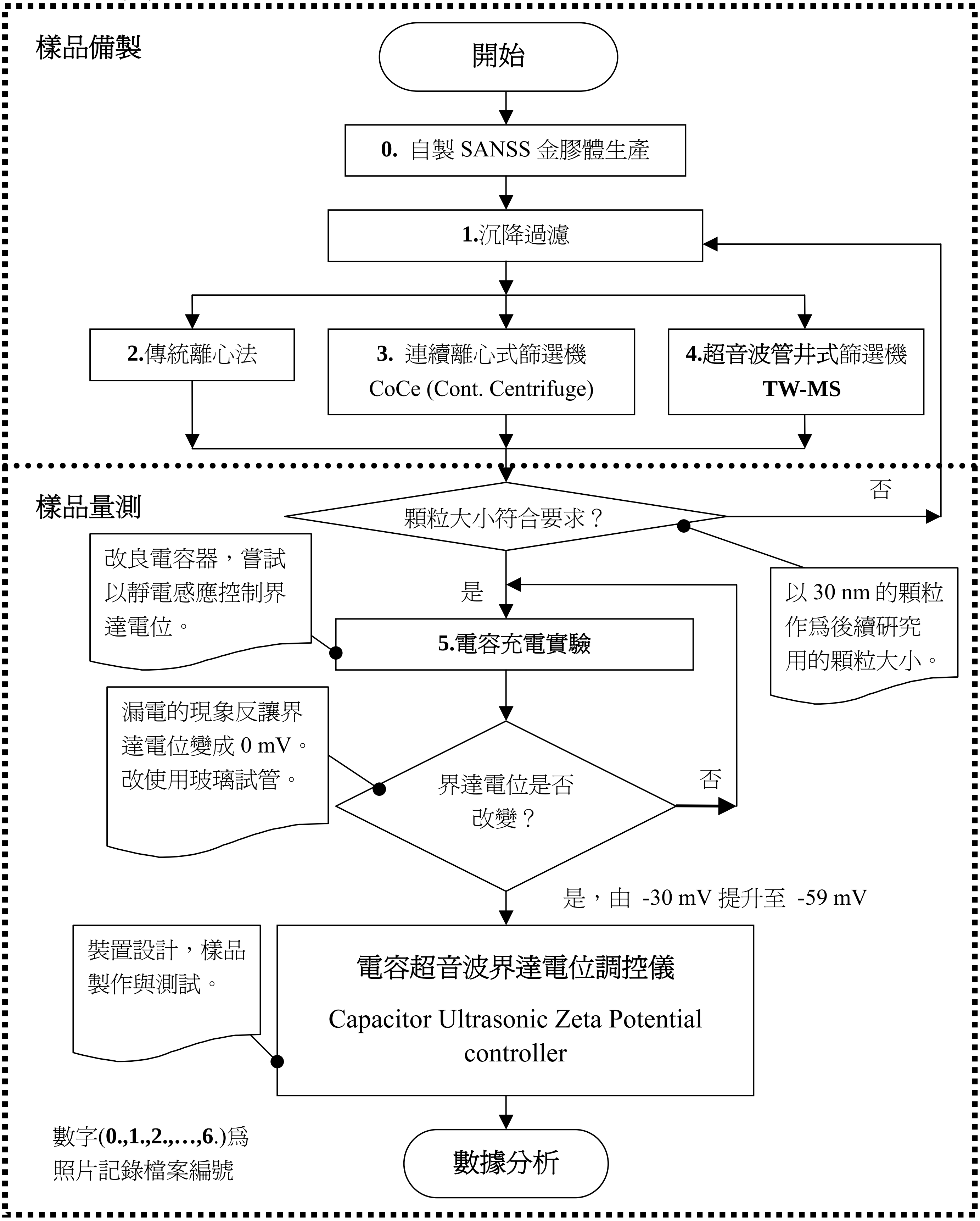
	name	Specification	Item No	company	Address & Tel.
1.	試管				
2.	T-Joint				
3.	Silicon-tube				
4.	Fittings	1/8”消音末(R)	2 只 NT$25/只	聯成空油壓五金行 盧春玉 翁佩雯	北市華陰街 119 號 Tel:02-25550988(O) 02-25550698(O) 02-25550068(H) Fax:02-255581680
5.	Fittings	Connector	3 只 No PC401 NT$40/只	聯成空油壓五金行 盧春玉 翁佩雯	北市華陰街 119 號 Tel:02-25550988(O) 02-25550698(O) 02-25550068(H) Fax:02-255581680
6.	Clamps	H36	3 只 NT$35/只	聯成空油壓五金行 (Gold Seal) 盧春玉 翁佩雯	北市華陰街 119 號 Tel:02-25550988(O) 02-25550698(O) 02-25550068(H) Fax:02-255581680

表二、電容超音波界達電位調控儀之 BOM

	name	Specification	Item No	Company	Address & Tel.
1.	solenoid	AC110V 50-60Hz 1.5kg 15mm	TAS-10 cont	Shih Hsing electric co.,Ltd.	北市錦西街 114 號 Tel:02-25573539(O) 02-25535228(O)
2.	T-Joint				
3.	Silicon-tube				
4.	Fittings			聯成空油壓五金行 盧春玉 翁佩雯	北市華陰街 119 號 Tel:02-25550988(O) 02-25550698(O)
5.	壓克力塊			橋達壓克力	
6.	壓克力管			橋達壓克力	

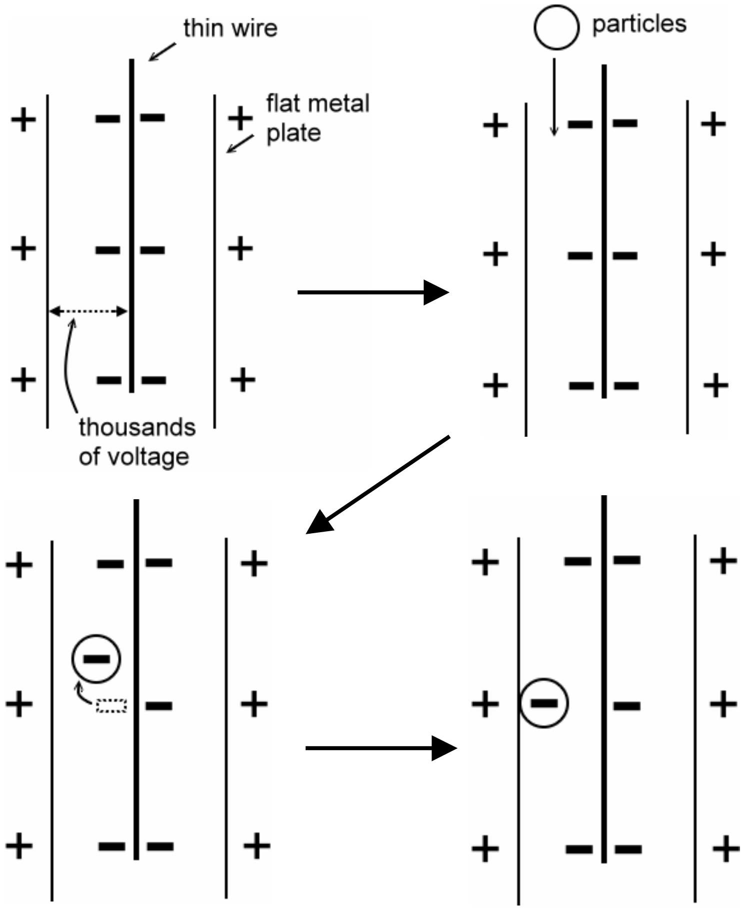
二、方法與步驟

(一)研究流程圖



(二) 原理及概念探討

美國國家科學研究院院士 Frederick G. Cottrell (1877-1948)在任教於柏克萊大學時，發明了**靜電除塵機**[5] **(electrostatic precipitator)**，原理如下圖所示：

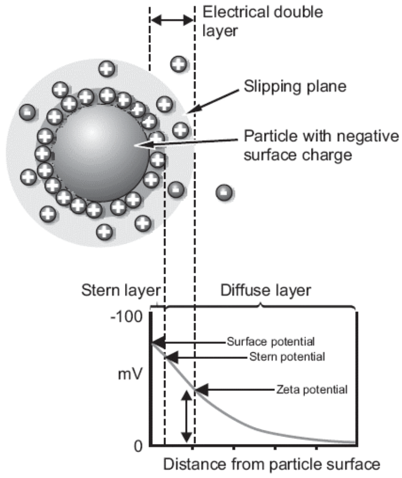


靜電除塵機的基本構造是由兩金屬板與一條導線組成，幾千伏特的電壓在兩者間產生，當電壓足夠時，導線周圍的負電荷會飄向金屬板的正極，若電荷碰到了氣體中的微塵（µm)，會使灰塵產生靜電，再藉由靜電將其吸附在帶正電的金屬板上。

本研究將此原理推廣至奈米尺度，並對儀器構造加以改良，來調控影響膠體粒子性質最重要的關鍵—界達電位。

[5] Wikipedia English version

界達電位 (zeta potential)是指膠體粒子上累積的離子造成的靜電壓，由固定層(Stern layer)和擴散層(diffuse layer)構成。可藉由 Henry function 導出電泳的移動率(Ue)，進而求出其界達電位的值[7]。



界達電位的固定層(Stern layer)和擴散層(diffuse layer)[6]

$$U_e = \frac{2\varepsilon\zeta f(ka)}{3\eta} \quad \text{------------------------------ (1)}$$

U_e 是電泳移動率，ζ 是界達電位，$f(ka)$ 是 Henry's function，ε 是樣品的介電常數，η 是黏滯系數。

界達電位有對膠體粒子的懸浮、分散、聚集、維持穩定及應用都有極大的影響。Zeta-Meter inc.是一間位於紐約的奈米科技公司，其創辦人 Thomas M. Riddick 曾在其著作中提到[9]：「界達電位是自然界的基本定律，且在植物和動物生命中都是非常重要的角色。對控制膠體的穩定性正是需要透過界達電位。」


Nano ZetaSizer，本研究測量界達電位與粒徑大小之儀器。[8]

目前界達電位的調控大多借由 pH 值的不同來改變，pH 值對界達電位範圍(±40 mV)的影響請參見 Malvern 公司所提供 Nano Zeta Sizer 儀器之使用說明的附圖**(見附件圖 25)**，使用 pH 調整界達電位之缺點有二：

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[6] D.J. Shaw, D.H. Everett, S. Ross, I.D. Morrison, J. Lyklema,Zeta Potential An Introduction in 30 minutes, Malvern Instruments Ltd..
[7] Wikipedia English version
[8] http://www.nbtc.cornell.edu/facilities/tools/Nano-ZS%20image.bmp
[9] T. M. Riddick,Control of colloid stability through zeta potential;: With a closing chapter on its relationship to cardiovascular disease, Livingston Pub. co, 1968

1、膠體金粒子在有高濃度離子的環境下會聚集沉澱[10]。

2、pH 值對後續應用會有不良的影響，如些微的 pH 值改變就會迫壞蛋白質的結構。

因此，以物理法調整界達電位對於奈米科技的發展是非常重要的。

膠體(colloid)是一種介於溶液與懸浮液間的混合物[11]，如表三所列，其組成爲大小在奈米等級的固體懸浮在液體中[12]。懸浮在膠體中的固體顆粒被稱爲**膠體粒子**，屬於奈米顆粒的一種。其表面帶有界達電位(zeta potential)。

表三、不同混合物顆粒大小的比較[13]

名稱	大小
溶液(均勻混合物)	< 10^{-9} nm (原子等級)
膠體	10^{-9} ~ 10^{-6} nm (奈米等級)
懸浮液(不均勻混合物)	>10^{-6} nm (大於微米等級)

以膠體金粒子爲本研究主題的原因有三：

1、金的化學性質穩定，不易起化學反應。目前也沒有對人體會有重大傷害的報告。對於後續應用較無顧慮。

2、較多人研究，目前的背景知識及可參考之資料較其它奈米粒子研究充足。

3、可應用範圍較廣，例如：醫學、電子、化學、日用品等。

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10 A.D. Mcfarland, C.L. Haynes, C.A.Mirkin, R.P. Van Duyne, H.A. Godwin, Color My Nanoworld, Journal of Chemical Education, Vol. 81, No. 4, April 2004, p544A~544BS

11 Wikipedia http://en.wikipedia.org/wiki/Colloid

12 Francis S. Key, George maass, Ions, Atoms and Charged Particles, Colloid Science laboratory Inc., 2001

13 微觀世界─高中生奈米科技營研習手冊,教育部,北區奈米科技 K-12 教育發展中心,1996,p25

(三)國內外相關研究和論文

從國內外研究膠體金粒子的文獻探討，如表四與五所示，爲國內外研究重點與論文摘要；可知膠體金粒子在各方面的應用越來越重要；膠體的性質主要是由聚集程度、粒子大小與表面電位決定；應用在生化方面的研究很多；大部分的膠體金粒子以化學法製造，強調物理法製造之膠體金粒子的研究很少。

表四、國外歷年的膠體金粒子研究與論文

序號	年份	作　　者	單　　位	研究〔論文〕名稱	成　果
1	2002	Langmuir, Rajendra Bhat	North Carolina State University, US	Gold nanoparticles make the grade	nanoparticles can form a gradient of decreasing concentration along a surface.
2	2002	Jorge Gardea-Torresdey, et al.	University of Texas-El Paso, US	Alfalfa plants harvest gold nanoparticles	Using alfalfa is a cost-effective and environmentally friendly method of producing gold nanoparticles.
3	2003		Hebrew University of Jerusalem, Israel, and Brookhaven Nat. Lab., US	Gold nanoparticles could boost biosensors	were used to attach glucose-oxidizing enzymes to electrodes which could ultimately be used as miniaturized biosensors inside the body that measure blood glucose.
4	2003	Murali Sastry, et al.	Nat.Chemical Lab. and the Armed Forces Medical College, India,	Micro-organism synthesizes gold nanoparticles	Using a micro-organism to synthesize gold nanoparticles which was much more uniform than the particles formed using other biological methods.
5	2003	Brahim Lounis, et al.	University of Bordeaux in France	Gold lights up biological cells	Gold nanoparticles was developed a new way of visualizing proteins in cells by labelling them.
6	2004	Chad Mirkin, et al.	Northwestern University, US	Gold nanoparticles and bio-bar codes bring sensitive DNA detection	“bio-bar-code-based” DNA detection technique has a sensitivity similar to that of the commonly used polymerase chain reaction (PCR) method.
7	2004	Crego-Calama, Holger Schönherr, et al.	University of Twente in the Netherlands	Self-assembly wins with gold rosette	Using the self-assembly of hydrogen-bonded rosettes to create nanostructures containing gold. The technique could have applications in the fabrication of nanowires.
8	2004	Jeff Brinker, et al.	University of New Mexico & Davidson College, US	Gold nanocrystals self-assemble to form 3D arrays	
9	2004	Swiss-US team	Swiss Federal Institute of Technology and the University of California Berkeley,USA,	Laser sintering strikes gold with nanoparticle ink	A laser technique for fabricating gold microstructures could provide a new powerful way to create miniature resistors or conductive tracks for flexible electronics.
10	2005	Zare , et al.	Stanford University, U.S.	Gold nanoparticles monitor protein folding	Gold nanoparticles could monitor changes in the folding of a yeast protein by attaching gold nanoparticles.
11	2005	Uzi Landman, et al.	Georgia Tech., US, and Technical University of Munich, Germany	Nanocatalysts charge into action	gold nanoclusters on a ceramic surface gain an electrical charge while they act as a catalyst for the low-temperature oxidation of carbon monoxide.
12	2005	Chad Mirkin, et al.	Northwestern University and Rush University ,USA	Nanoparticle technique detects Alzheimer's-related proteins	based bio-barcode has been assayed to measure the concentration of amyloid-β-diffusible ligands (ADDLs) in cerebrospinal fluid (CSF).
13	2006	Richardson	Ohio University, US	Gold nanoparticles enhance laser heating	Gold nanoparticles was used as site-directed nano heaters that can be actuated with optical light

表五、國內歷年的膠體金粒子研究與論文

序號	年份	作者	單　位	研究﹝論文﹞名稱	成　果
1	1999	張德有	國立中山大學化學系研究所	融合微粒電噴灑游離質譜法的發展與應用	先以微滴產生器（超音波霧化器）將其霧化成極小微滴（aerosol），再以一股氮氣流將微滴經由一鐵氟龍管吹送到游離源中，正進行電噴灑之毛細管前端。
2	2001	李尙謙	國立中山大學材料科學研究所	以超音波霧化法製備之銻及鉭摻雜氧化錫薄膜之光電特性	超音波鍍膜系統。以 $SnCl_4.5H_2O$, $SbCl_3$, and $TaCl_4$ 爲溶質，乙醇爲溶劑。配置不同 Sn^{4+}濃度的溶液、不同 Sb/Sn 原子比的溶液、不同 Ta/Sn 原子比的溶液。藉由震盪頻率 1.65MHz 之超音波裝置的強烈震盪以產生霧氣，霧氣則藉由氮氣的流動，攜帶至已加熱的基材（康寧 7059 玻璃 ）上，因此霧氣可被熱分解。由於此熱解反應，SnO_{2-x} 薄膜則就被沈積在基材上了。
3	2003	陳正雍	國立中央大學物理研究所	奈米金粉粒的原子結構及吸收光譜與粒徑關係探討	將 3 種不同的奈米金粉粒的結構利用有限尺度散射強度進行模擬，得知當奈米粉粒的粒徑越小，其繞射譜圖與塊材越不相同，由模擬結果得知，當粒徑小於 2.7nm，結構發生扭曲，但無法確定原子結構是否有改變。
4	2003	林宏聲	中原大學化學工程研究所	蛋白質於金奈米粒子表面吸附之探討	利用不同鏈長的硫氫化合物(mercapto-aliphatic acids)來修飾奈米金粒子表面，探討人血清蛋白(Human serum albumin)及澱粉分解脢(Bacillus amyloliquefaciens α-amylase)於不同表面之吸附行爲，以及蛋白吸附後構形的變化。
5	2004	賴建宇	國立成功大學機械工程學系博士班	高強度超音波與氣泡空蝕場應用於奈米粉體製備與養分萃取	利用超音波產生的氣泡空蝕場，在氣泡空蝕場崩裂時所產生的微噴流以及震波來衝擊粉粒表面，以達到粉粒細化的效果。
6	2005	謝倩芳	中原大學化學研究所	製備零維與一維之金奈米材料及奈米金粒子在多孔二氧化鈦上特性之研究	(1)利用化學溶液的方式，將塊材金合成出奈米金粒子。(2)藉由模板法與電化學沉積法製備出一維的金奈米材料。(3)將奈米金粒子負載於中孔洞的二氧化鈦上，並探討其光催化特性與研究。

表六、國內歷年的超音波研究與論文

序號	年　份	作　者	單　位	研究﹝論文﹞名稱	成　果
1	1999	Der-Yeou Chang	Department of Chemistry National Sun Yat-Sen University	Application and Development of Fused-Droplet Electrospray Mass Spectrometry	The technique makes use of both gold nanoparticles attached to “bar code” DNA and magnetic microparticles.
2	2001	Shang-Chien Li	Institute of Materials Science and Engineering ,National Sun Yat-Sen University	Electro-optical properties of Sb and Ta doped SnO_2 thin films derived from an ultrasonic atomization process	using ultrasonic nebulization was adopted in this study. a solution by the agitation of an ultrasonic device operating at about 1.65MHz.
3	2003	Cheng-Yung Chen	Department of Physics, National Central University, Taiwan	Atomic structure and absorption of Au nanoparticle with different diameter	the structure of Au nanoparticle is distorted when the diameter is smaller than 2.9 nm. But Au nanoparticle’s structure is changed.
4	2003	Hong-Sheng Lin	Department of Chemistry, Chung Yuan Christian University, Taiwan R.O.C.	Investigation of Proteins Adsorption on Colloidal Gold Nanoparticles	to modify the surfaces of the colloidal gold nanoparticles by mercapto-aliphatic acids of various lengths (C2 to C16),
5	2004	Chien-Yu Lai	Dept. of Mechanical Eng. National Cheng Kung University, ROC	High Power Ultrasound and Bubble Cavitation for Fabricating Nanoparticles and Nutrition Extraction	using high-power ultrasound and cavitation field can result in particle-size reduction..
6	2005	Chain-Fang Shieh	Department of Chemistry, Chung Yuan Christian University, Taiwan R.O.C.	Synthesis of 0-D and 1-D Gold Nanomaterials and Properties of Gold Nanoparticles-deposited Mesoporous TiO_2	the synthesis of gold nanoparticles from bluk gold by chemical solution process,the synthesis of 1D gold nanorods/nanotubes via template growth and electrochemical deposition

(四) 實驗步驟

1、金膠體的製備

參考北區奈米科技 K-12 教育發展中心提供的高中生奈米科技營研習手冊，本組決定測試熔膠凝膠法和台北科技大學電能中心所使用的潛弧合成系統(Submerge Arc Nanoparticles Synthetic System, SANSS) **(見照片 6)**，分別屬化學法和物理法製程。

(1) 熔膠凝膠法：

以吸量管取 10ml 的過氯酸金，放入試管中。再以另一隻吸量管取 0.5ml 的檸檬酸溶液，同樣放入試管樣品瓶中。靜置約十五分鐘。

(2). 潛弧合成系統(Submerge Arc Nanoparticles Synthetic System, SANSS)

SANSS 的原理[14]是藉由金屬在常壓的環境下在水中進行相變化來製作膠體。此系統包含純金電極、電力供應系統、去離子水供應系統與加熱系統**(見圖 1 與照片 6)**。壓力一大氣壓和溫度維持在 80℃狀態，去離子水(介電液)維持穩定進出，兩個純金(99.9%)的電極置於去離子水裏，當電極互相介接近時，6000~10000℃的電弧產生高熱使金屬汽化，80℃之水碰到汽態金，亦迅速被汽態金的熱汽化，汽態金的熱被水吸收後，會迅速被冷凝固集結成奈米顆粒，而得研究所需之膠體金粒子。

[14] TiO2 Nanoparticle Suspension Preparation Using Ultrasonic Vibration-Assisted Arc-Submerged Nanoparticle Synthesis System(ASNSS) Materials Transactions, The Japan Institute of Metals, Vol.45 No.3 (2004).pp.806-811. (SCI)

2、膠體金粒子篩選

(1) 多種方法測試

請參考實驗流程圖，有三種篩選方式可得研究所需之粒子大小的膠體金粒子。整個過程是從 SANSS 產出粒子大小分佈很廣 (200000 nm 至 10 nm)之膠體金粒子，先以沉降系統**(見圖 2 與照片 7)**篩選過濾，得到大約 100 nm 至 10 nm 之膠體金粒子，再進入依測試不同的篩選方式：(詳細的比較請見研究結果與討論之表八)

左線：商用離心式篩選機，廠牌：Hettich，型號：MIKRO 22 R**(見圖 3 與照片 8)**；其特性是少量需求時使用。

中線：自行製作連續式離心篩選機**(見圖** 4, **5 與照片 9)** ；其特性是單一粒徑需求時使用，原理較複雜，技術性高。

右線：是自行製作超音波管井式篩選機(Tube Well Mass Spectrometer:TW-MS)**(見圖 6 與照片 10)**；其特性是構造簡單、具超音波攪拌、分散篩選又可加電場調變，最具發展潛力者，因此對本路線--超音波管式質譜分散篩選機--做較深入地研究，特別適合大量生產使用。

(2)超音波管井式篩選機 (TW-MS)

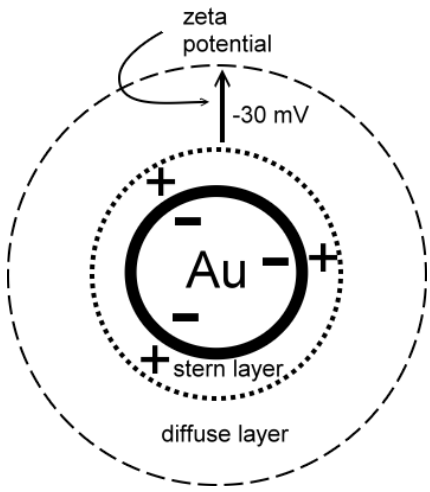
當收集並分析國內歷年的超音波霧化研究與論文，較爲精確發展狀況可參考的資訊如表六所列。由優缺點比較表**(表八)**，故採用此法，本研究將超音波霧化與攪拌**(見照片4, 5)**之功能拓展，利用水面共振的效果，穿過薄金屬箔將膠體金粒子霧化，並以鼓風機用定壓定量之空氣吹，再加電場調變，就具有類似質譜儀，而達到超音波管式質譜分散篩選機之功能。根據研究結果評估不同的膠體金粒子篩選方式，並提出改良的設計構想，與試製實驗機印證，而發展出更新更好之膠體金粒子分散篩選機。

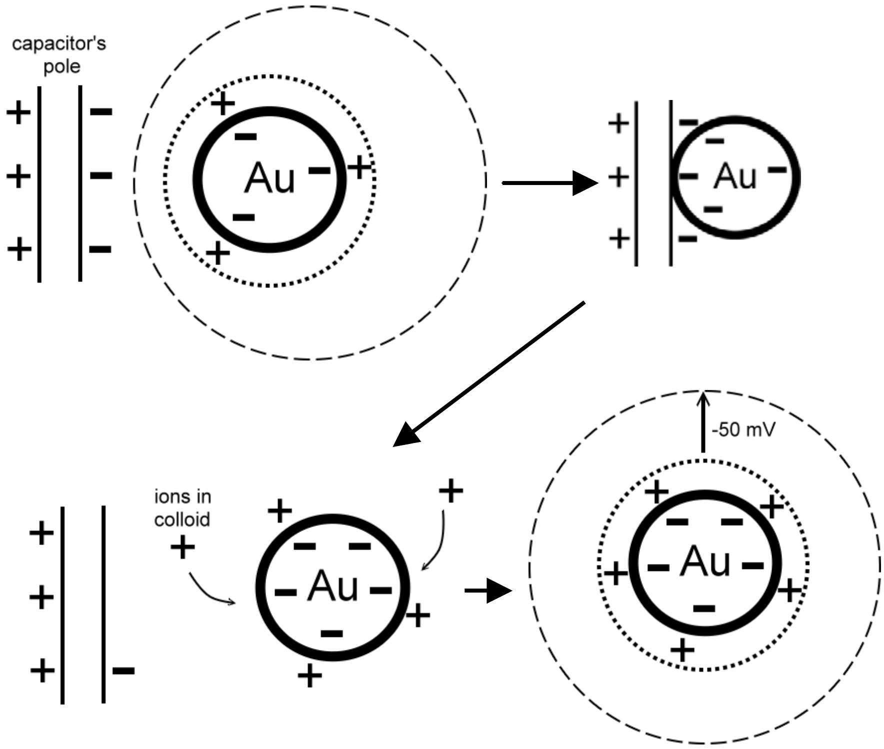
3、電容充電實驗

當膠體金粒子分散篩選之技術已相當掌握後，而且可以掌握自己需要之粒子大小分佈後，即進行以電容法控制膠體金粒子之界達電位，並研製膠體金粒子界達電位調控儀，如**圖** 7 與**照片** 11 所示。在試管內外各安裝一不銹鋼箔(50 μm 厚)電極，以試管之管壁玻璃璃(500 μm 厚)為介電材料，其構造即為一電容器。於試管內之不銹鋼箔(50 μm 厚)電極接到 1500 V 電源供應器之負極；於試管外之不銹鋼箔(50 μm 厚)電極接到 1500 V 電源供應器之正極。當兩電極接上電源時，其作用就如同電容器被充電。當電容器被充飽滿時，兩電極間之電位差為 1500 V。此時試管內膠體金粒子就有機會接觸 1500 V 之電極。當膠體金粒子接觸到 1500 V 之電極時，就被充電了。

利用超音波造成水面共振的效果，霧化金膠體達到強力攪伴之功，促進增加膠體金粒子接觸之機會，再以時間長短控制接觸之次數，接觸次數多寡得到控制，就可以達到調控膠體金粒子**界達電位 (zeta potential)**之功能。

當膠體金粒子分散篩選之技術已相當掌握後，而且可以掌握自己需要之粒子大小分佈後，即進行以電容法控制膠體金粒子之界達電位，並研製膠體金粒子界達電位調控儀，如**圖 7** 與**照片 14** 所示。





界達電位充電示意圖

參、研究結果與討論

一、本研究最主要的研究結果

(一) 電容超音波界達電位控制儀可把膠體金粒子之界達電位從原尖峰值-30 mV 可調控至 -59 mV，真實大小分佈曲線請見**圖 18~19**，大部分係以常態分佈的方式出現。從**圖 24** 可知，界達電位的調控量與充電時間成正比。

(二) 以本研究實驗所得的結果比較改變界達電位的電容法(物理法)和傳統 pH 值調整法(化學法)可知，電容法調控界達電位的量與充電時間成正比，請參見**圖 24**。傳統 pH 值調整法則是成 Z 型曲線。並發現用電容法比 pH 值調整法之極限 -40 mV，高出約 -10 mV 左右。請參見**圖 25**。

(三) 本研究結果提出超音波管井式篩選機 TW-MS、連續離心式篩選機 CoCe (Cont. Centrifuge)不同的膠體金粒子篩選方式，提出以超音波改良膠體金粒子分散篩選機設計構想，經理論探討完成設計構想，**如圖 6,8,9 與 10** 所示，並完成試驗之雛形機。所得測試結果如**圖 11** 以及**圖 26**。

(四) 經過電容超音波界達電位控制儀調整界達電位的膠體金粒子與蛋白質鍵結的效果見**圖 27** 蛋白質鍵結量的關係，可發現其蛋白質增量與**圖 24** 充電時間與界達電位(30 nm)的關係之增加趨勢相似。

二、金膠體的製備

目前製備膠體金粒子的方法有五種[15]。經初步的比較後，潛弧合成系統(Submerge Arc Nanoparticles Synthetic System, SANSS)[16]和熔膠凝膠法較適合本研究。後來發現，熔膠凝膠法的化學反應非常難控制，每次做出來的金膠體顏色都不一樣，很難做出顆粒大小均勻的紫色金膠體，常常都因用來穩定金顆粒的反應太快，變成顆粒過小的黑色金膠體。因此決定使用 **SANSS**，以物理法製造之膠體金粒子爲**研究奈米尺度的載體**。

表七 膠體金粒子製備方式之比較[17]

方法名稱 / 特點項目	**潛弧奈米合成系統 (SANSS)**	**熔膠凝膠法 (主流方式)**	電漿技術	物理粉碎法	氣相沉積法
製備原理	物理性	化學還原	物理性	物理性	化學性
產品純度	**高**	低	高	低	高
粒徑	分佈廣，經 **TW MS** 篩選可**依不同應用提供不同大小**	小	小	較大	---
顆粒均一度		佳；但顆粒大小不符合本研究之要求	佳	不佳	佳
操作難易度	容易	容易	尚可	容易	困難
優點	**成本低廉 完全無雜質 過程簡單 大量生產**	適合製備氧化物	適合製備金屬微粒	成本低廉	---
缺點	製作時間長	製程中引入的**離子不易完全去除**	設備要求高	雜質多	原料成本高昂

[15] 微觀世界─高中生奈米科技營研習手冊,教育部,北區奈米科技 K-12 教育發展中心
[16] TiO2 Nanoparticle Suspension Preparation Using Ultrasonic Vibration-Assisted Arc-Submerged Nanoparticle Synthesis System(ASNSS) Materials Transactions, The Japan Institute of Metals, Vol.45 No.3 (2004).pp.806-811. (SCI)
[17] 微觀世界─高中生奈米科技營研習手冊,教育部,北區奈米科技 K-12 教育發展中心,1996,p95

三、膠體金粒子篩選

表八 膠體金粒子篩選方式之比較

	傳統離心法 **(主流方式)**	連續式離心篩選機	管井式篩選機 (TW-MS)
製作難易度	無法自行製作 必須用購買的	尚可	簡單
篩選速度	尚可	慢	尚可
特點	1. 操作簡單	1. 可連續得到需要的顆粒大小。 2. 篩選量少，適合醫療研究之應用。	1. 可自製成本低 2. 取樣品方便 3. 大量篩選顆粒，適合量產。
缺點	1. 機器價格昂貴 2. 取樣品不便，使篩選的效果降低。	篩選速度不快	篩選時間長，每次必須大量生產。

三條路線篩選方式特性，如**表八**所列，可看出：左線少量、中線單一與右線多元的特性。再深入分析可得知：利用自製的連續離心機篩選顆粒大小，與普遍常用之離心機比較，效果不錯，但變數複雜，其分離顆粒原理：膠體金由上面的入口注入 PU 管內開始離心**(圖 4)**，顆粒即以負等加速度運動達到一個終端速度 **(圖 5)**，公式如下：

$$V_0 = \sqrt{\frac{\pi d(\delta - \rho)g}{6\psi\rho}}$$

-- (2)

d：顆粒直徑 δ：顆粒密度 ρ：介質密度 ψ：阻力係數 g：重力

當雷諾數 Re < 1 時，有史托克公式

$$V_0 = 54.8d^2 \frac{\delta - \rho}{\rho} \quad \text{-- (3)}$$

雷諾數 Re = 2~300 時

$$V_0 = 25.8d^3 \sqrt{\left(\frac{\delta - \rho}{\rho}\right)\frac{\rho}{\mu}} \quad \text{-- (4)}$$

當顆粒走的距離 S 大於管壁 L 時，將會緊貼著管壁而不會從出口流出，改變離心的時間得到不同顆粒大小的奈米金。審視目前的奈米顆粒製備方法，已有人用超音波製備奈米粒子，其原理是以震波震碎粒子，達到奈米等級，而化學法則是引發溶液的催化反應，產生奈米粒子，於是本研究採用類似質譜儀的原理，構想以超音波將金膠體霧化，再以風力將大顆粒及小顆粒分類，風力大小對於顆粒大小的篩選機制將是本題目重點之一。風力大小由鼓風機風壓決定，因此控制風壓就可將大顆粒及小顆粒分類，如**圖 8** 所示，就是風壓校正曲線。鼓風機風壓從 4.5 V 至 12 V 機乎是成直線比例上升，為了控制方便，研究中，皆設定電壓 12 V，並維持風壓在 5 mmAq 水柱高，確保實驗準確。

風量大小對於顆粒大小的篩選機亦是重要變因之一。風量大小由鼓風機出口的小圓孔(φ0.5 mm 直徑圓孔) 的個數控制，因此控制小圓孔的個數就可控制風量，風量大小就可控制膠體金粒子霧化後在管子理飄浮多遠，飄浮至多少節管子，其結果如**圖 11** 所示，圓孔個數與飄浮至多少節管子關係，從圖上可知以純水試驗，圓孔數為 8 時，霧化飄浮至 6 節管子；若以金膠體試驗，圓孔數為 8 時，霧化飄浮至 5 節管子； 推測應該是金膠體比重大於純水，因此超音波管井式篩選機究中，皆設定圓孔數為 8，膠體金粒子霧化飄

浮至 5 節管子，確保實驗之可重複性。

霧化量最大之超音波(頻率 1.65 MHz)霧化器振盪，造成水面(距霧化器底 65 ~ 70 mm 之處)共振，效果最佳，液滴之大小最佳可至 1 μm。在 μm 級之液滴中，推論有如**圖 10** 所示之分佈狀況。從實驗結果印證，如**圖 11** 所示,，變化趨勢有相當高相似性。不管在理論與實驗都值得繼續深入研究。

四、電容充電實驗

利用物理公式將電容器之各物理量精確地掌控

平板電容公式：

$$C = k\varepsilon_0 \frac{A}{d} \text{ -- (5)}$$ [18]

C：電容 (F) k：介電係數 ε_0：$8.85^{-12}\ {}^{F}\!/\!{}_{m}$ A：電極面積 d：電極距離

電容計算，

$$4.7 \times 8.85^{-12}\ {}^{F}\!/\!{}_{m} \times \frac{6\times10^{-4}}{5\times10^{-4}} = 49.914^{-12} F \approx 50^{-12} F = 50 pF$$

$$Q = CV \text{ -- (6)}$$ [19]

Q：電量 (coul) C：電容 (Farad) V：電壓 (volt)

電量計算

$$Q = 50 \times 1500 = 75000 pcoul = 7.5 \times 10^{-8} coul$$

$1.6\times10^{-19}\ {}^{coul}\!/\!{}_{electron}$ $\frac{7.5\times10^{-8}}{1.6\times10^{-19}} \approx 5\times10^{11}$ electrons

(8 mm/7.5 mm)

[18] 黃崢瑜，王耀常，賴俊豪，廖顯文，物理(下) 第六版，金華科技圖書股份有限公司, 2004, p26-5

[19] TiO2 Nanoparticle Suspension Preparation Using Ultrasonic Vibration-Assisted Arc-Submerged Nanoparticle Synthesis System(ASNSS) Materials Transactions, The Japan Institute of Metals, Vol.45 No.3 (2004).pp.806-811. (SCI)

$$C = k2\pi\varepsilon_0 \frac{L}{\ln(b/a)}$$ -- (7) [20]

C：電容 (F)　k：介電係數 ε_0：$8.85\ {}^{pF}\!/\!_{m}$ A：電極面積 d：電極距離

$$C = 4.7 * 2\pi * 8.85 \frac{30*10^{-3}}{\ln(8^{-3}/7.5^{-3})} \approx 261.35*1001.46 \approx 261731.85\ pF$$

$$Q = 261731.85*1500 = 392597775.5\ pC \approx 3.9*10^{-4}\ C$$

Electron quantity = $\dfrac{3.9*10^{-4}}{1.6*10^{-19}} \approx 2.44*10^{15}$

經由以上的計算電容器的電容及表面所帶的電荷量可以知道，當膠體金粒子的濃度確定後，即知道膠體金粒子的數量，藉由電荷量與金粒子數量，可推估需要多少充電時間，但是實際值仍須以實驗來得到正確的充電時間。

[20]黃崢瑜, 王耀常, 賴俊豪, 廖顯文, 物理(下) 第六版, 金華科技圖書股份有限公司, 2004, p26-5

五、電容超音波界達電位調控儀設計與製作

當確定膠體金粒子之界達電位(zeta potential)可以被電容法(物理法)調控時，即探討電容法(物理法)以靜電感應改變界達電位的效能並與傳統 pH 值調整法(化學法)之特性，**見圖** 24, 25。以所得的結果可知充電時間越長，平均界達電位(zeta potential)可提得越高，上升曲線漸趨遲緩**見圖 24**。傳統 pH 值調整法則是成 Z 型曲線，以 pH 值調整界達電位(zeta potential)，可從正值到負值，其極限在 ±40 mV，電容法(物理法)只有單向上升，差異甚大，此現象相當有趣值得深入研究。利用氣壓缸或電磁作動器，將類似電容器的試管上下帶動。利用定時器可以控制上下的時間。當試管往下至下死點時，試管底部正好處在水面共振處。利用 1.65 MHz 超音波將水面共振的原理，傳到試管內，試管內的金膠體亦產生共振，而將試管內的膠體金粒子劇烈攪拌，使膠體金粒子有充分接觸電容器電極，而達到將電荷從電極傳導至膠體金粒子上，因爲膠體金粒子處在去離子水中，離子水的電阻約在 1 MΩ至 26 MΩ之間，所以 1.65 MHz 超音波劇烈攪拌膠體金粒子，以提高膠體金粒子接觸電容器電極的機會，即增加電荷從電極傳導至膠體金粒子上的機會與電荷量。氣壓缸或電磁作動器將試管上下帶動的方式各有特色：氣壓缸帶動的方式上下運動平穩順暢，但週邊設備複雜；電磁線圈構造簡單，但是運動速度較快，容易引起震動，各有優缺點。從然如此，利用控制上下停滯的時間，就可設計出許多不同的充電方式，本研究就是控制充電時間與電壓高低，來調控金顆粒的界達電位，相信，本研究繼續以調控電壓與充電時間將可得到許多有趣的效果，若再配上膠體金粒子的大小和不同的製作參數，將有非常多的變化。

肆、結論與未來之應用潛力

一、結論

膠體的性質主要是由界達電位 (zeta potential)決定，可從鍵結蛋白質的測試中證明。

應用超音波霧化與攪伴之功能，利用水面共振的效果，穿過薄金屬箔將膠體金粒子成功地達到超音波管式質譜分散篩選。

不同的膠體金粒子篩選方式結果：沉降過濾可達平均粒徑 100 nm；離心式篩選機與**超音波管井式篩選機(TW-MS)**可達平均粒徑 30 nm。

膠體粒子以電容調控膠體金粒子之界達電位 (zeta potential)，可成功地從-30 mV 提升至-59 mV，超過 pH 調整法的極限 -40 mV。並成功地製作完成**電容超音波界達電位控制儀** (Capacitor Ultrasonic Zeta Potential Controller)、**超音波管井式篩選機** (TW-MS)、連續離心式篩選機 CoCe (Continue Centrifuge)、沉降過濾器(Settling tube)，組成**電容超音波界達電位控制儀系統(Capacitor Ultrasonic Zeta Potential System)**。

二、未來之應用潛力

奈米金應用範圍已被報導可應用在醫學、電子、化學、日用品……，奈米科技 K-12 教育發展中心的研習手冊中提到了十項主要應用：

表九、奈米金的多種應用[21]

奈米金的應用	與 Zeta potential 相關
1. 奈米金分子自組裝系統	○
2. 奈米金觸媒	
3. 驗孕試紙	○
4. I 型大腸桿菌偵測劑	○
5. DNA 檢測與基因治療	○
6. 藥物載體	○
7. 奈米金/C60 複合製造單電子電晶體	
8. 奈米金針測家用塗料的含鉛量	
9. 奈米金催熟縮短酒的成熟時間	

本研究在應用上選擇以藥物載體、醫學檢測上追蹤標記的材料、驗孕試紙、DNA 檢測與基因治療等與醫學較相關的應用上進行測試，這些應用大部分都源自於膠體金粒子與蛋白質的鍵結。

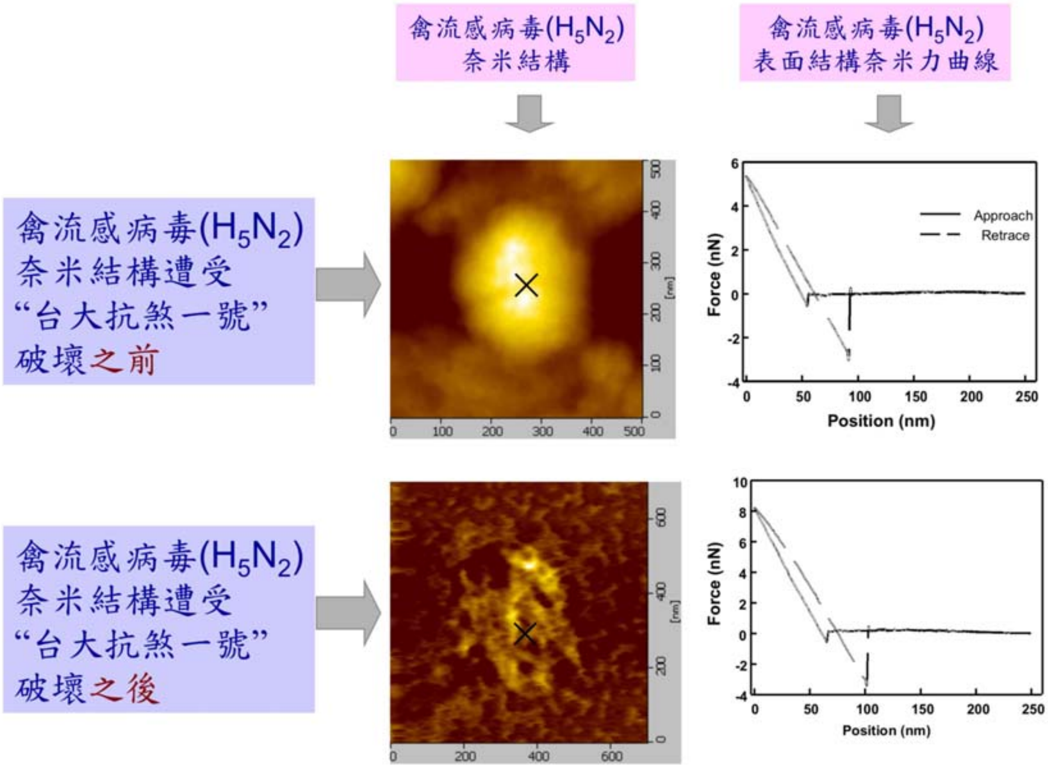
本研究參考荷蘭學者 Jan.W.Slot, (2002)[21]等人，University of Utrech 之研究成果的方法[22]，進行膠體金粒子的應用測試。並以 SDS-PAGE (Sodium Dodecylsulfate Polyacrylamide Gel Electrophoresis, 十二烷基硫酸鈉-聚丙烯醯胺凝膠電泳)分析經電容法調控界達電位之膠體金粒子，是否會增加其能鍵結蛋白質能力(Ovalbumin, 卵白蛋白)的量。本研究以結合卵白蛋白質之相對結合量來代表其結合能力。

[21] 微觀世界─高中生奈米科技營研習手冊,教育部,北區奈米科技 K-12 教育發展中心
[22] Jan W. Slot, Hans J. Geuze, A New Method of Preparing Gold Probes for Multiple-labeling Cytochemistry, European Journal of Cell Biology 38., p87~93, 1985

經初步的測試發現，調控界達電位後的膠體金粒子與蛋白質的鍵結量明顯的增加

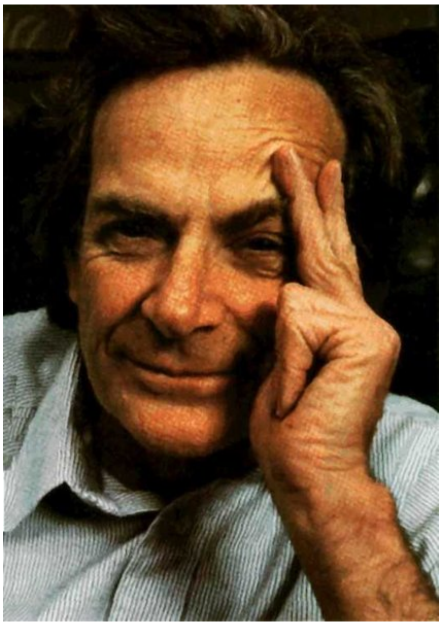
時間 Min	0	1	2	3	4	5	標準
量 μg	0.07	0.64	0.54	1.30	1.34	1.52	1 μg

由此可見，經過調控界達電位的膠體金粒子與蛋白質鍵結的效果有非常明顯的改變，將結果畫成圖後**(見圖 26)**，可發現其蛋白質增量趨勢與**圖 24** 相似。因此，藉由界達電位**電容超音波界達電位控制儀系統**，能對膠體金粒子的特性與效能做更嚴密的掌控，達成費曼先生所說的「**在原子或分子的尺度上來加工材料和製造設備**」。

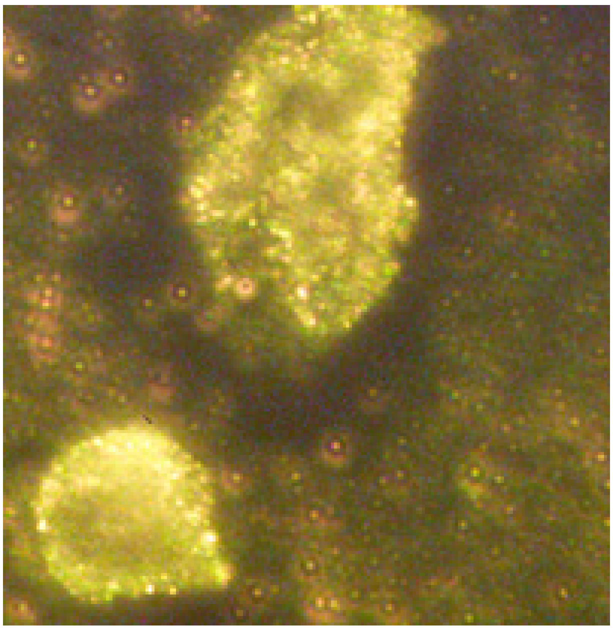


Source: httpwww.aicgroup.com.twNTU1-1.htm

照片 1 抗煞一號的理論(Picture 1 Theory of Anti-SARS No1)



照片 2 Richard F. Feyman



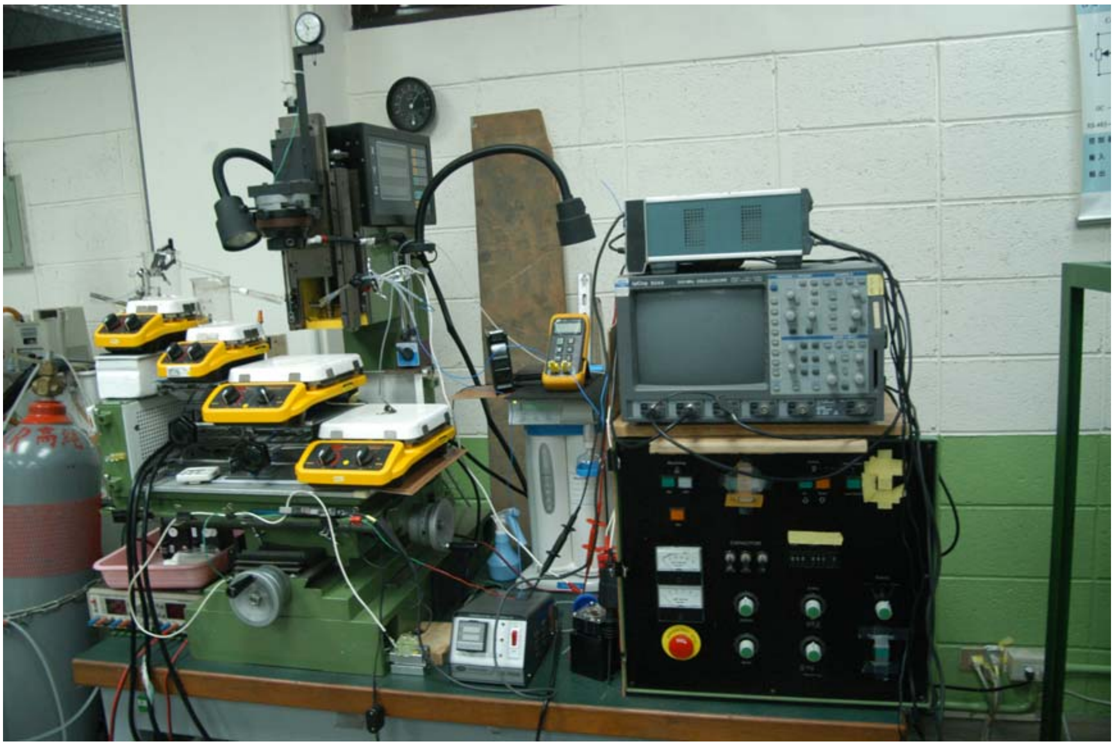
照片 3 膠體金粒子附著在癌細胞上閃閃發光

(Picture 3 Gold nanoparticles stick to cancer cells and make them shine.)

Source: Geprge Tech. Prof. David Terraso

Source: National Taiwan University of Technology Prof. Lei
照片 4 超音波作用原理(Picture 4 Theory of Ultrasonic)

照片 5 超音波霧化器(Picture 5 Fog is produced by Ultrasonic Atomizer
Source: Flower market Taipei)



照片 6 膠體金粒子潛弧合成系統

(Picture 6 Au-NPs is produced by SANSS)

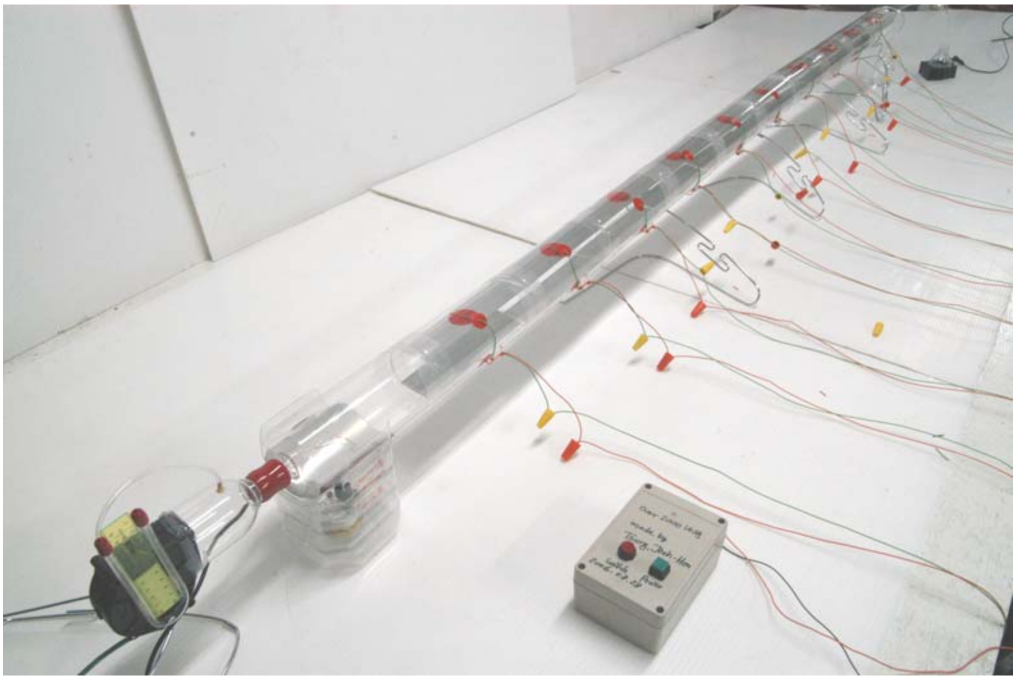
照片 7 沉澱過濾器

(Picture 7 沉降過濾器)

Source: Hettich MIKRO 22

照片 8 一般離心機(Picture 8 General Centrifuge)

照片 9 連續篩選式離心機(Picture 9 連續式離心式篩選機)



照片 10 超音波管井式篩選機(Picture 10 Tube Well Mass Spectrometer)

照片 11 電容超音波界達電位控制儀(Picture 11 界達電位 Zeta Potential 調控機)

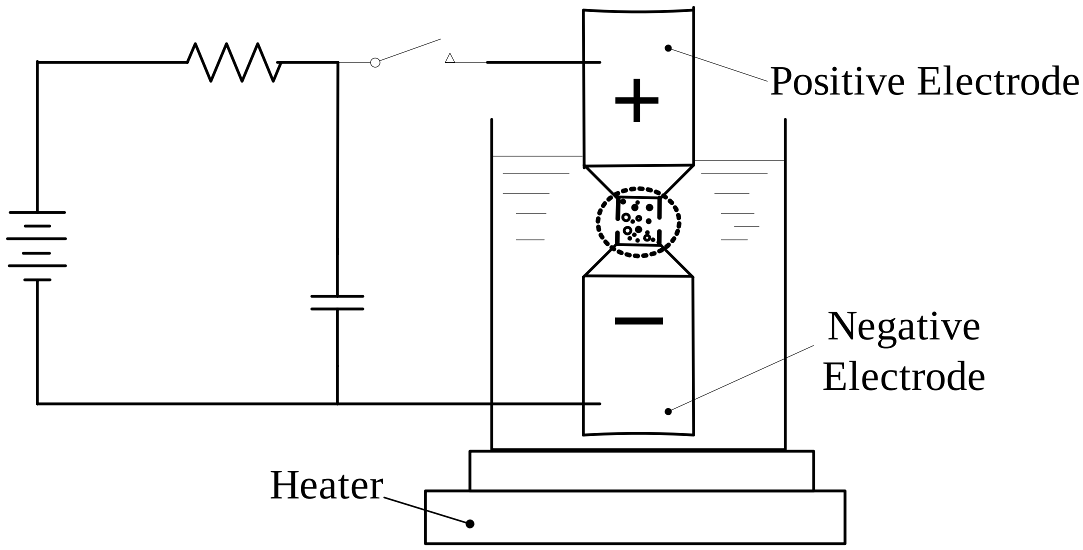


圖 1 奈米潛弧合成系統

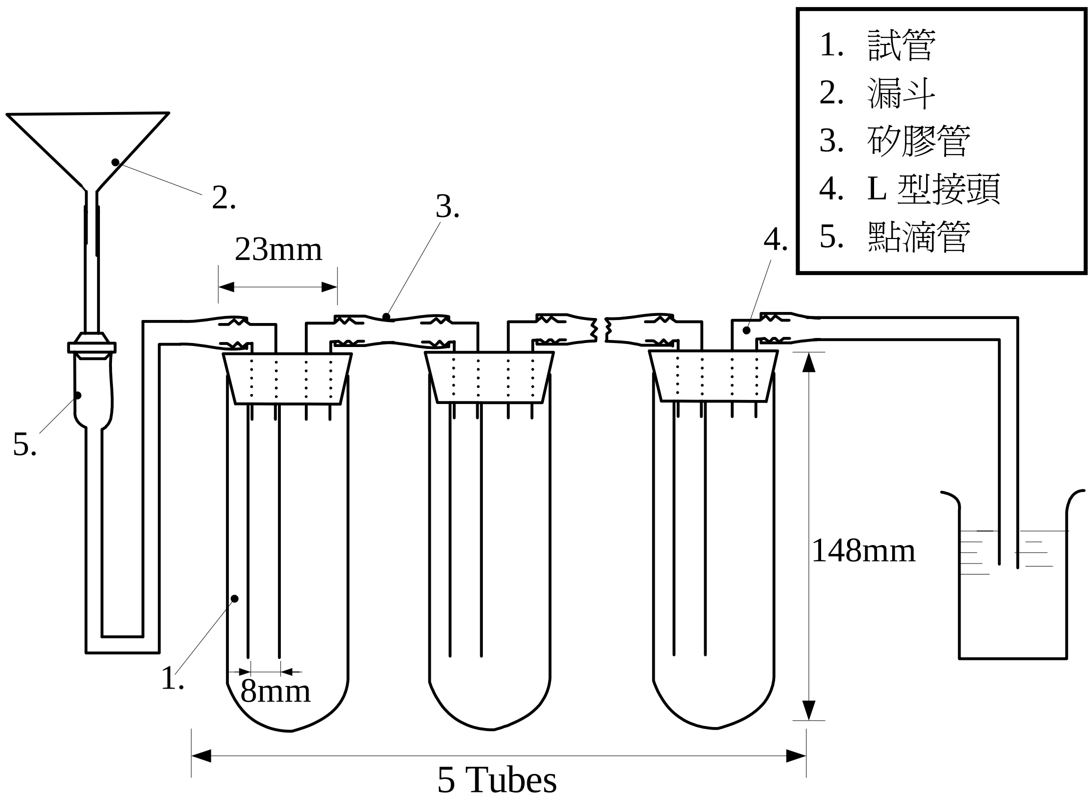


圖 2 沉澱過濾器

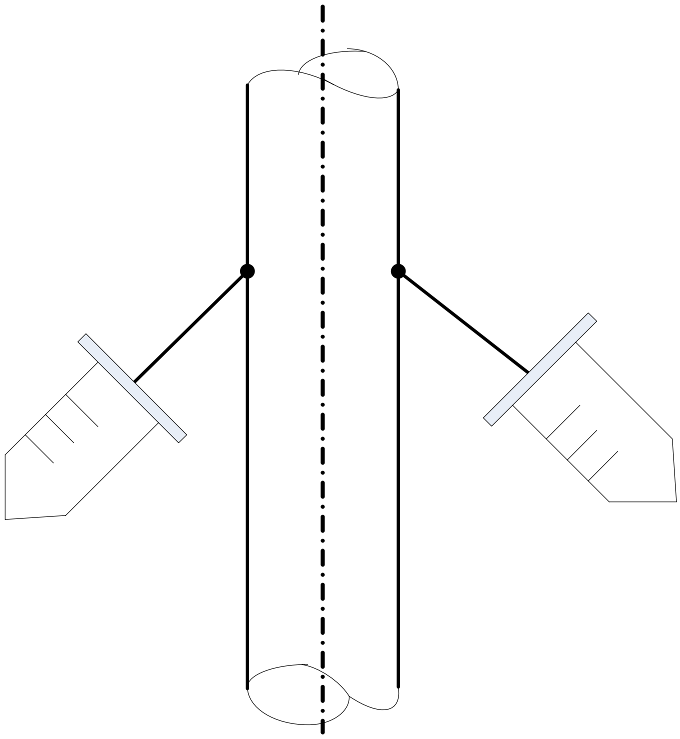


圖 3 一般離心機示意圖

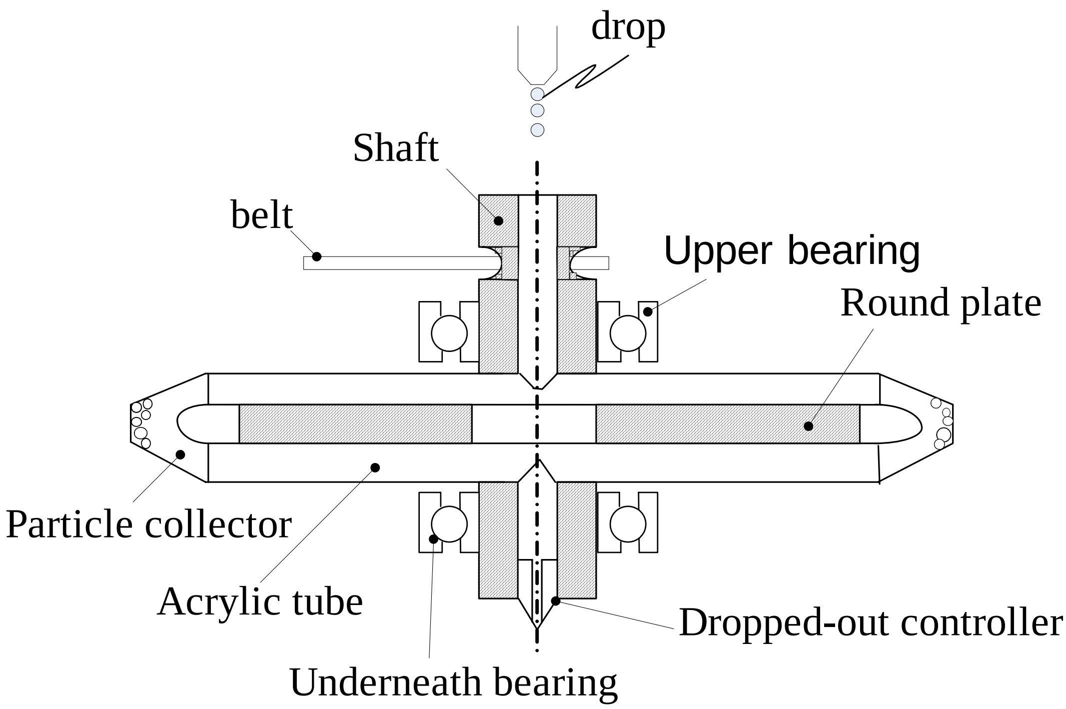
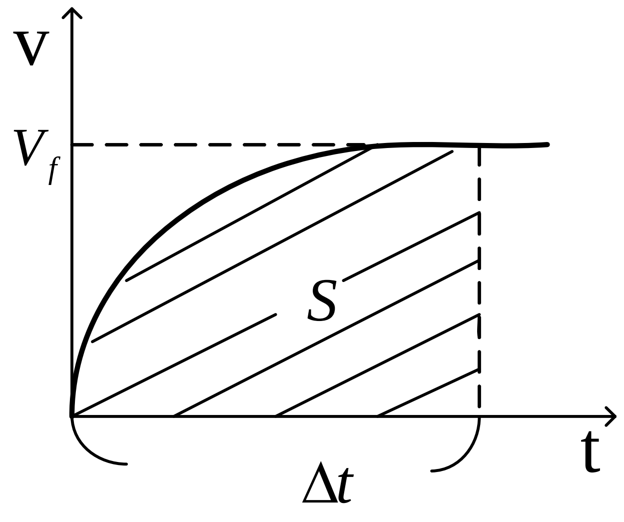


圖 4 連續篩選式離心機草圖



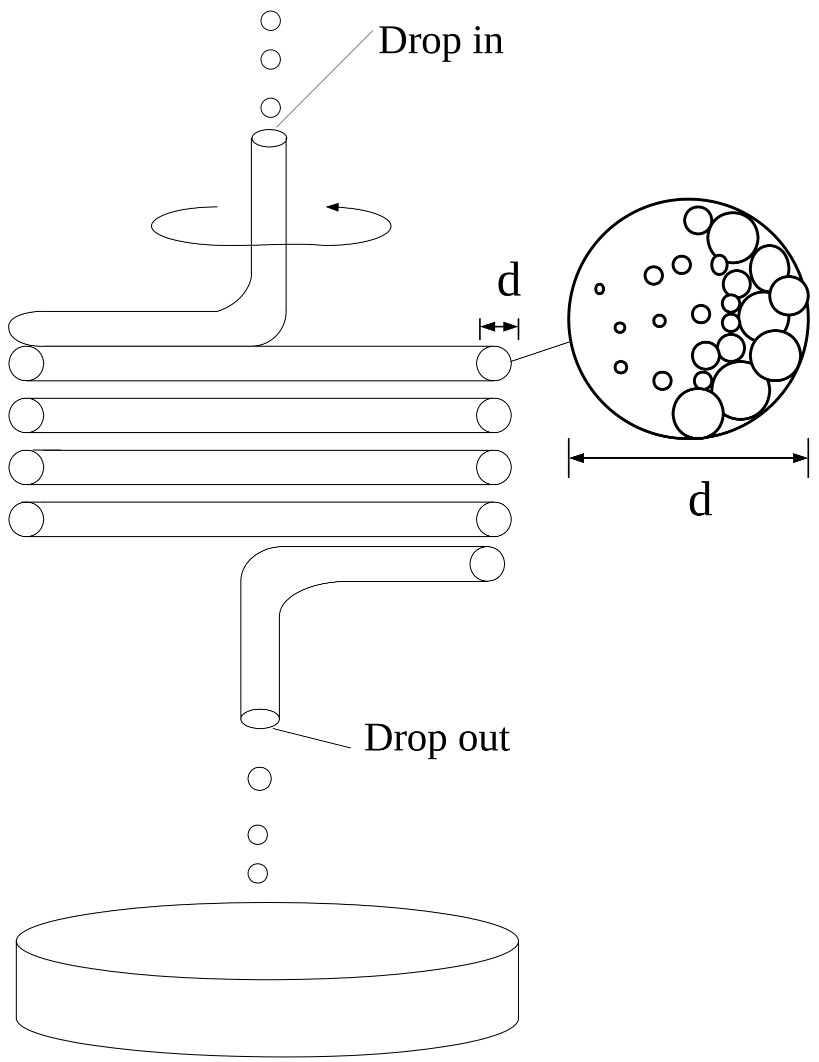


圖 5 連續篩選式離心機作用原理示意圖

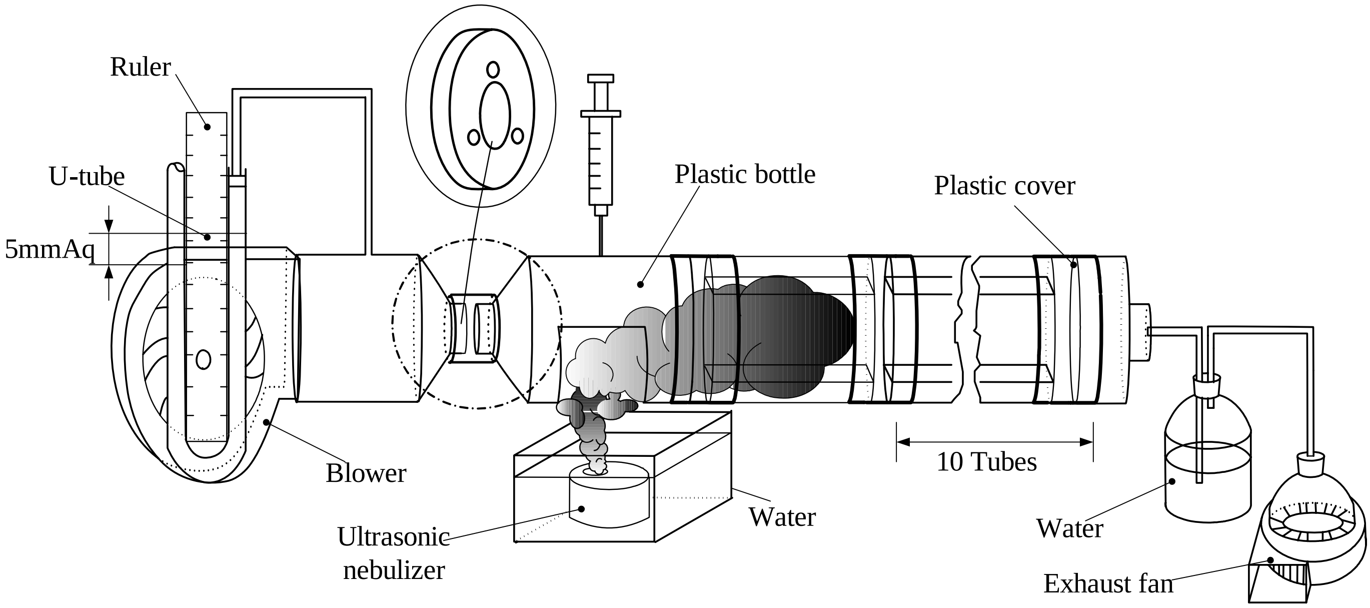


圖 6 超音波管井式篩選機示意圖(TWMS)

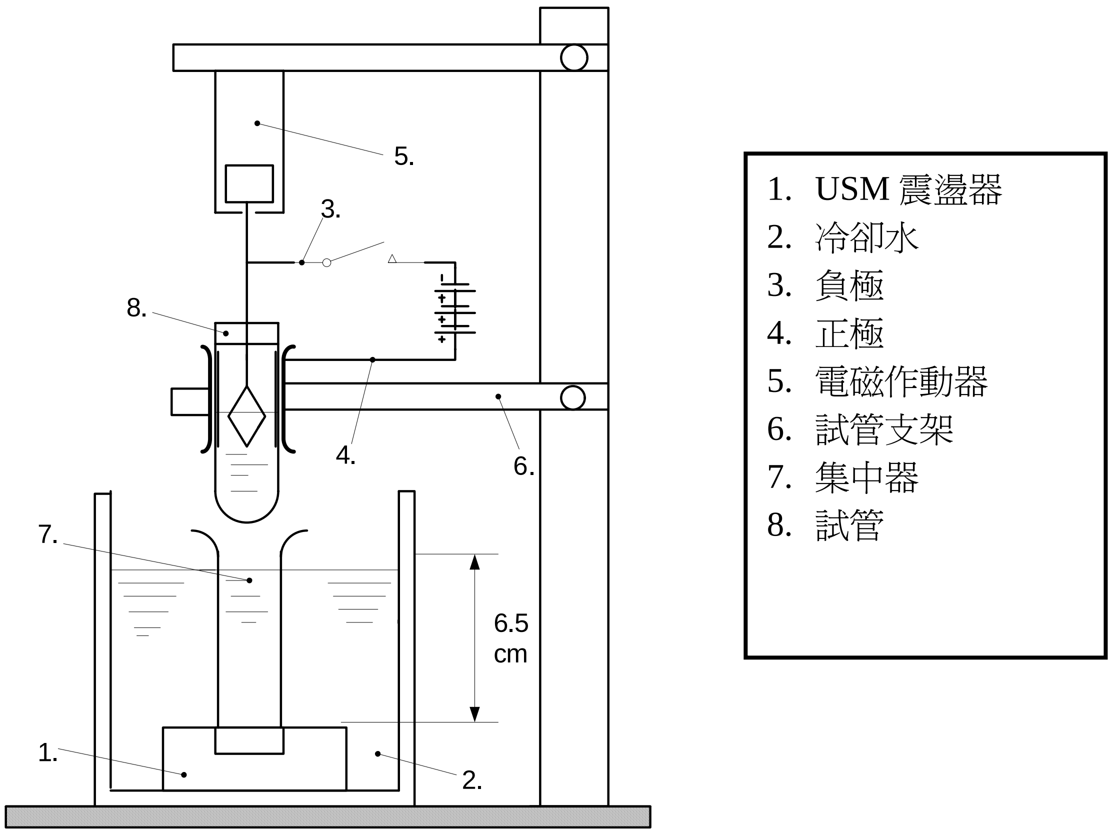


圖 7 電容超音波界達電位控制儀示意圖

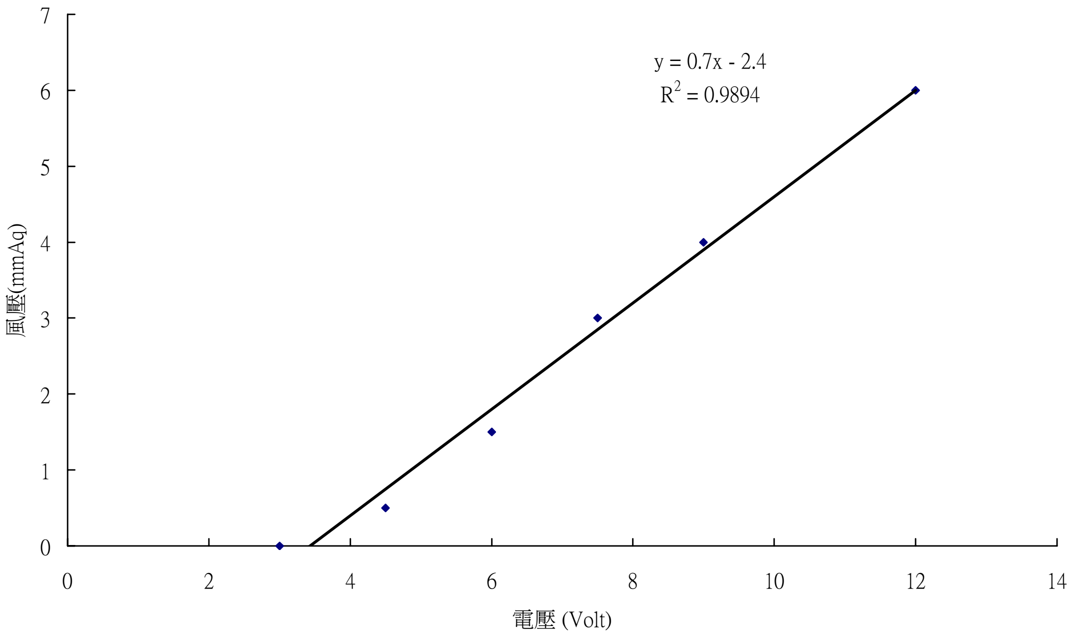


圖 8 超音波管井式篩選機風壓校正曲線

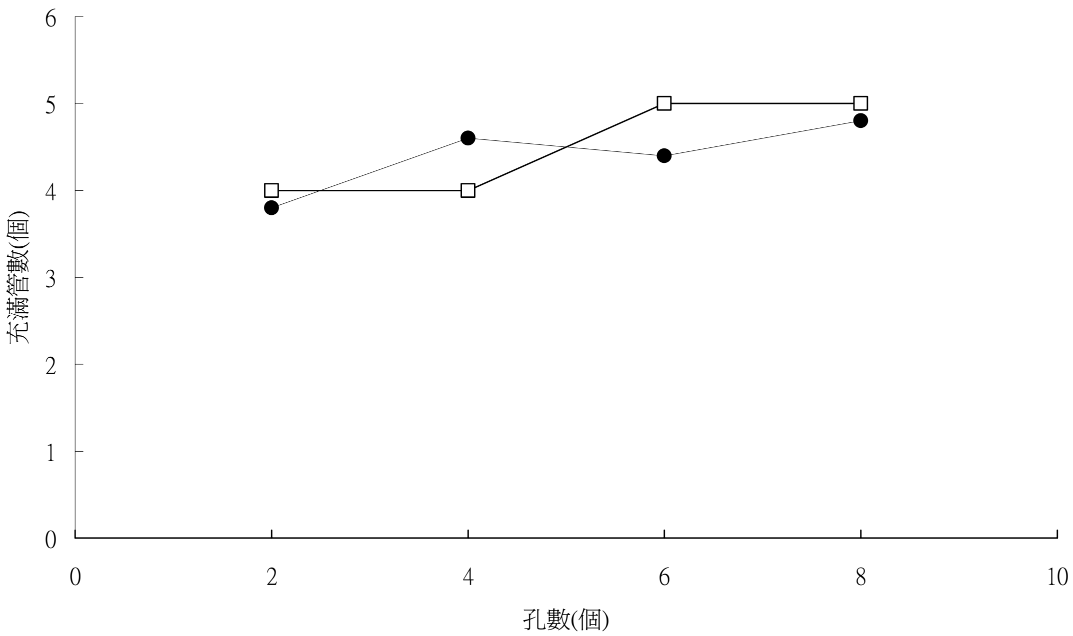


圖 9 超音波管井式篩選機孔數與霧氣充滿管數的關係

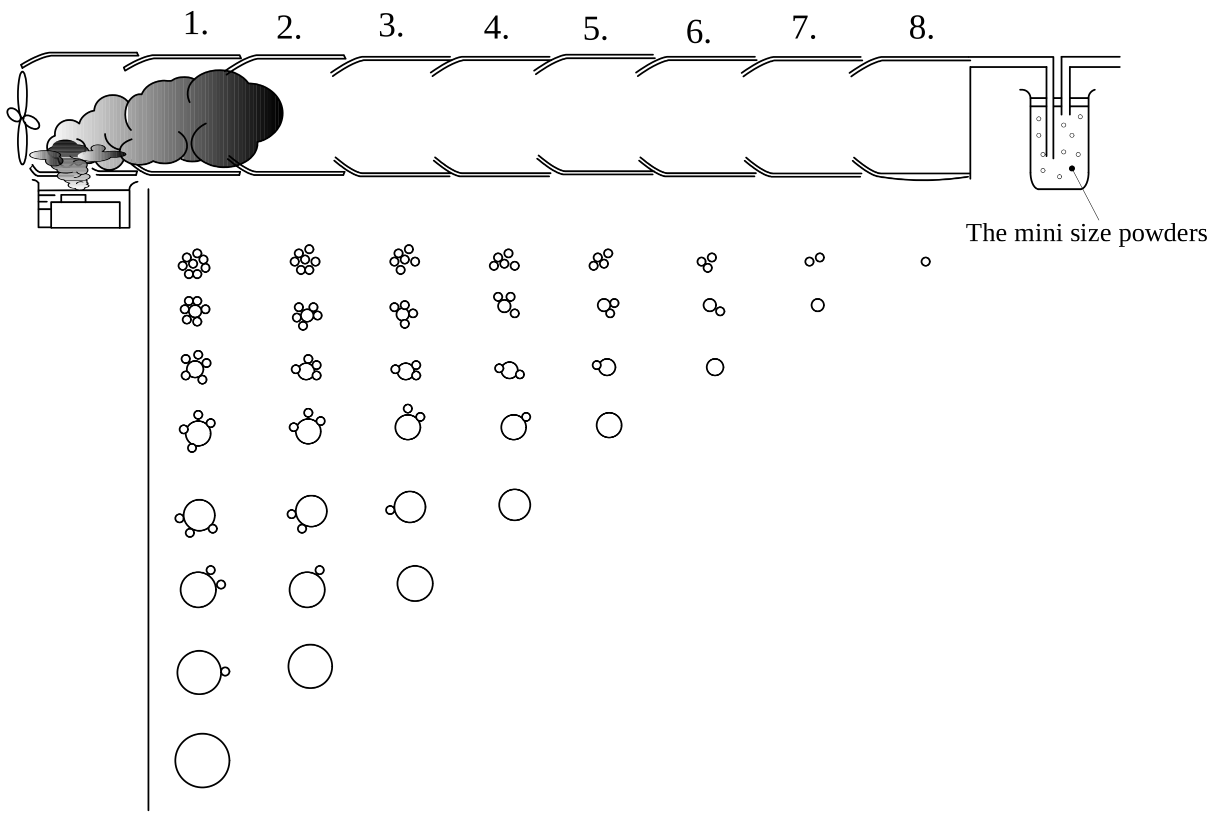


圖 10 膠體金粒子分在超音波管井式篩選機散理論分佈圖

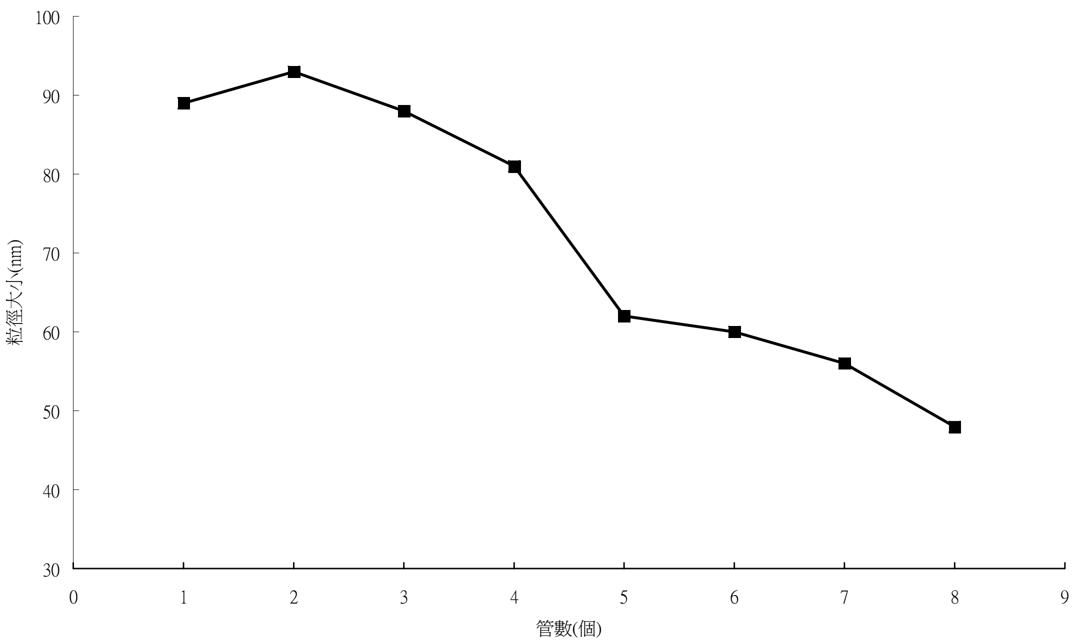


圖 11 管井中奈米顆粒的粒徑分佈實驗分佈圖

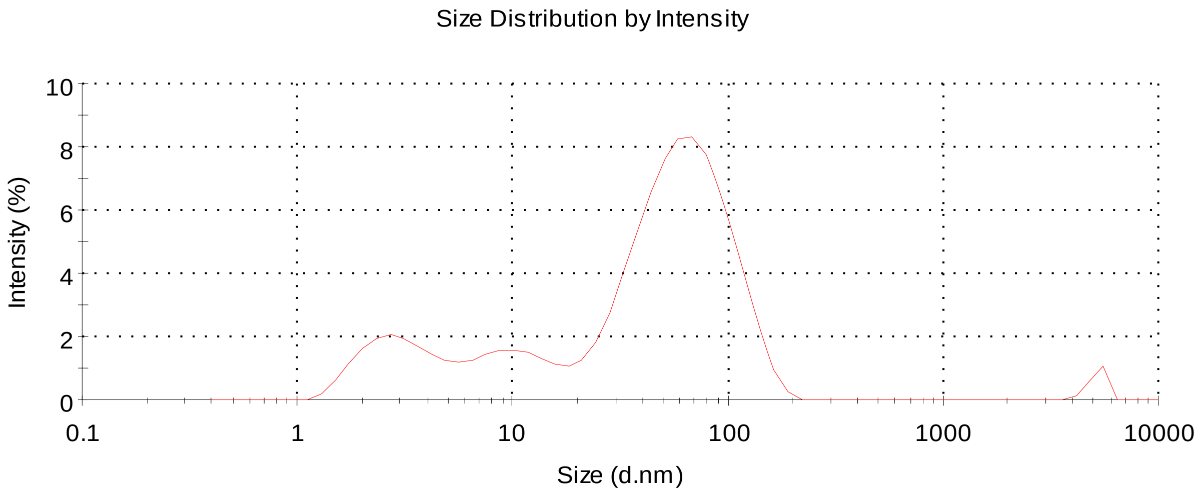


圖 12 峰值 80 nm 的膠體金粒子粒徑分佈曲線

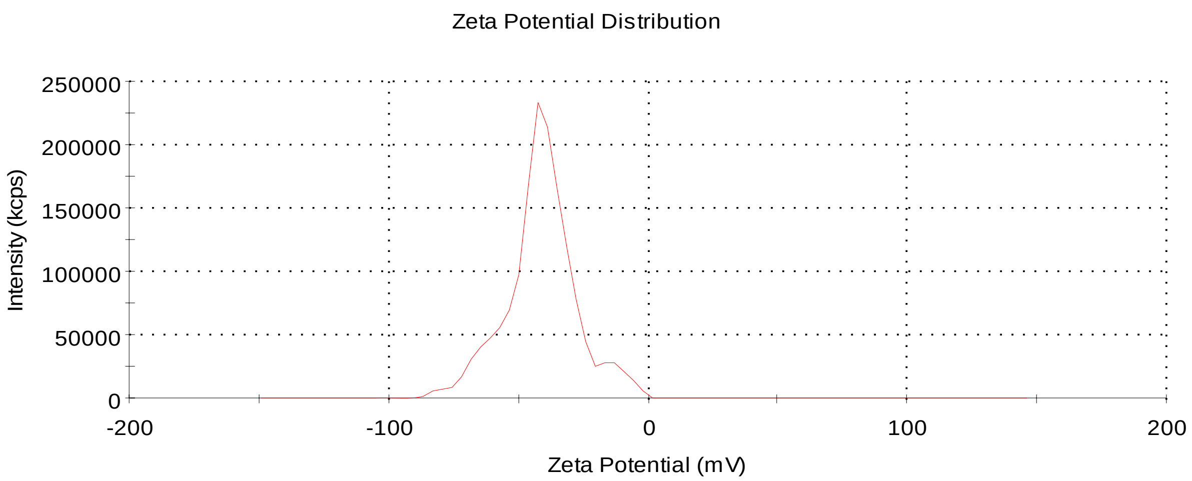


圖 13 峰值 80 nm 的膠體金粒子界達電位分佈曲線

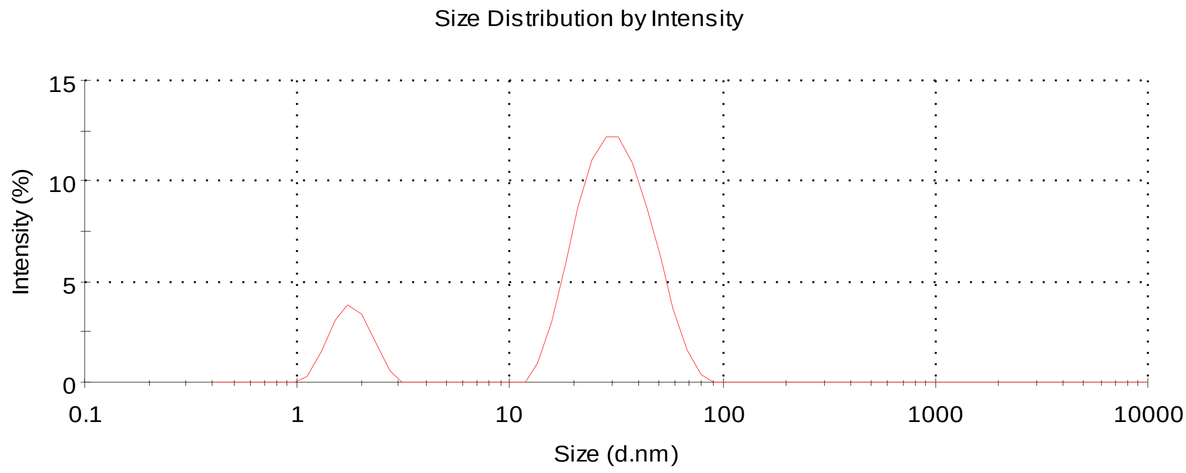


圖 14 峰值 30 nm 的膠體金粒子粒徑分佈曲線

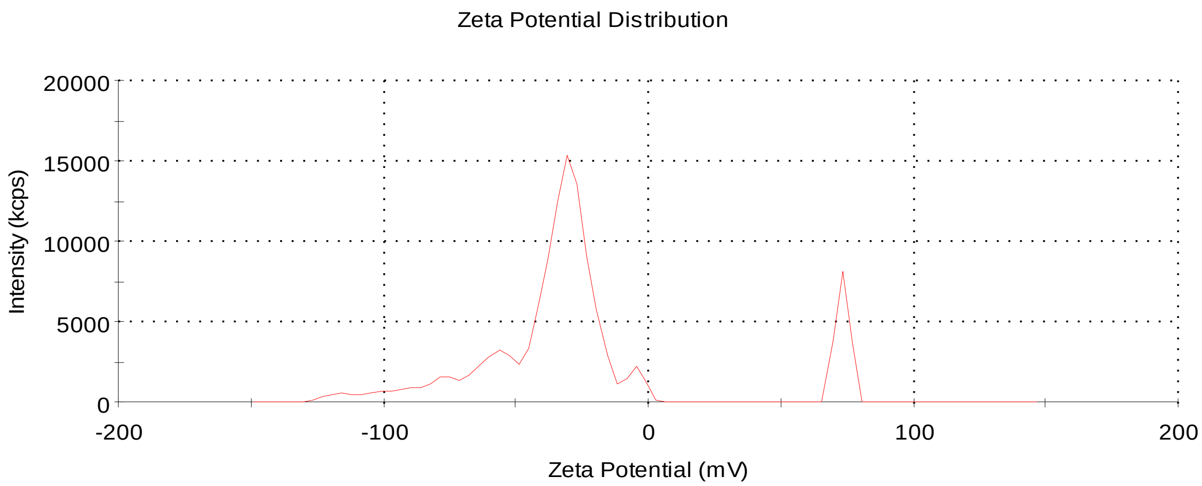


圖 15 峰值 30 nm 的膠體金粒子界達電位分佈曲線

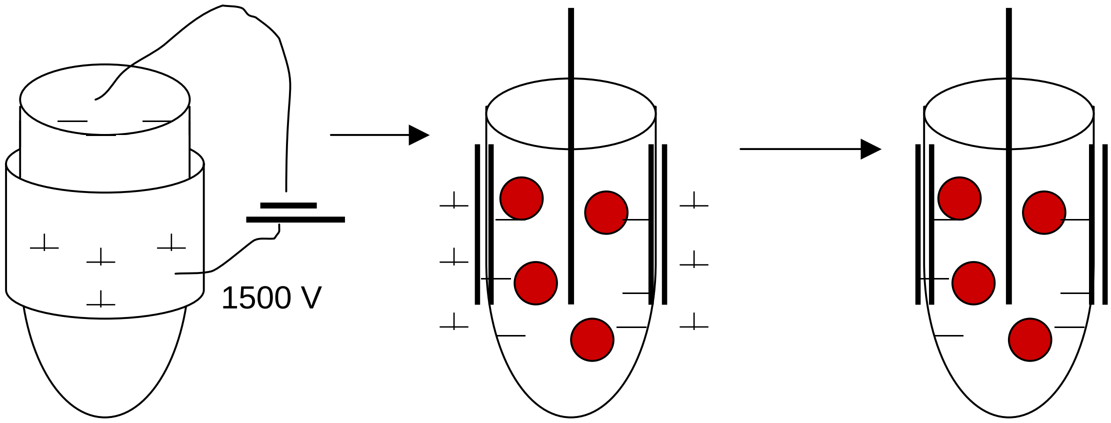


圖 16 界達電位(30 nm) 調控作用原理示意圖

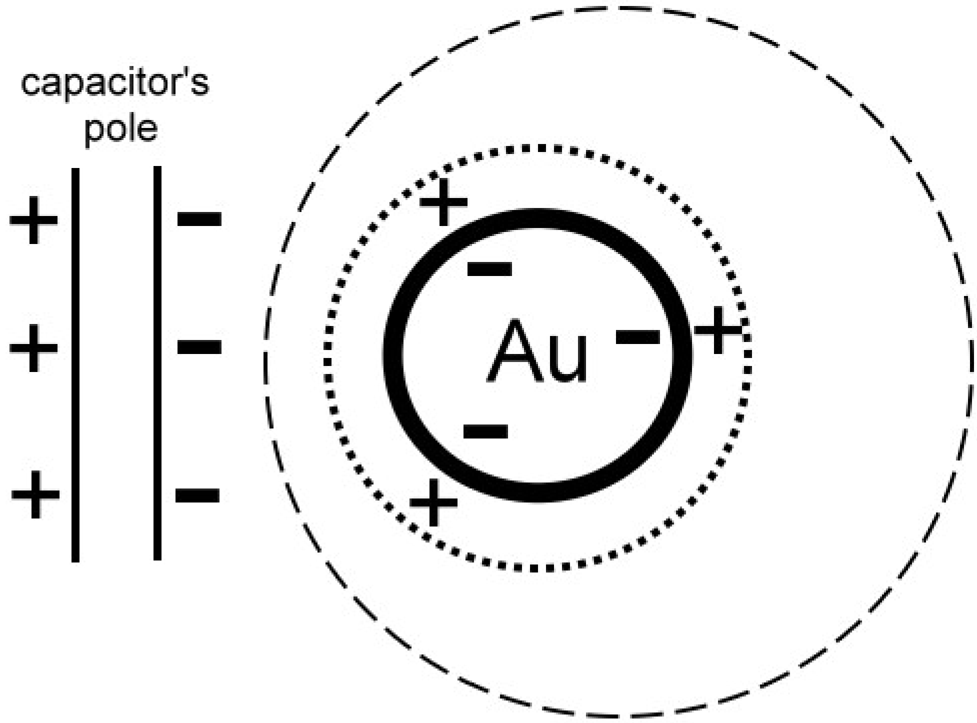


圖 17 電容器之電極與膠體金粒子作用原理示意圖

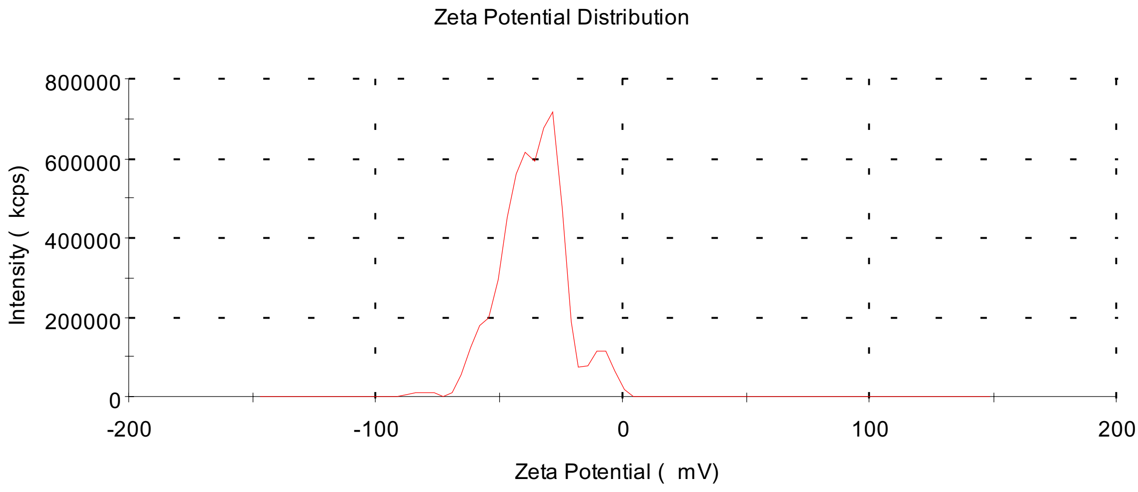


圖 18 未調控之界達電位(30 nm)分佈曲線

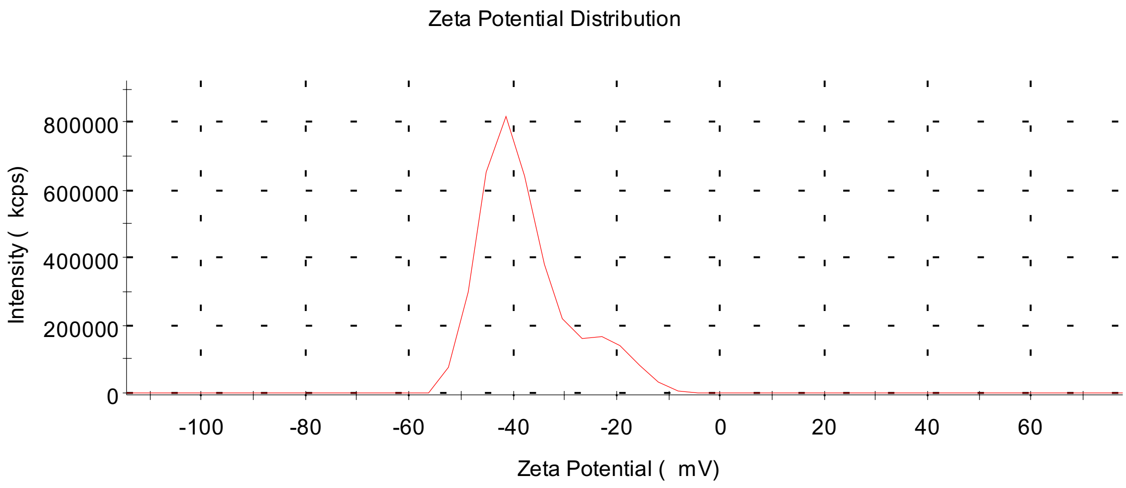


圖 19 調控 1 分鐘之界達電位(30 nm)分佈曲線

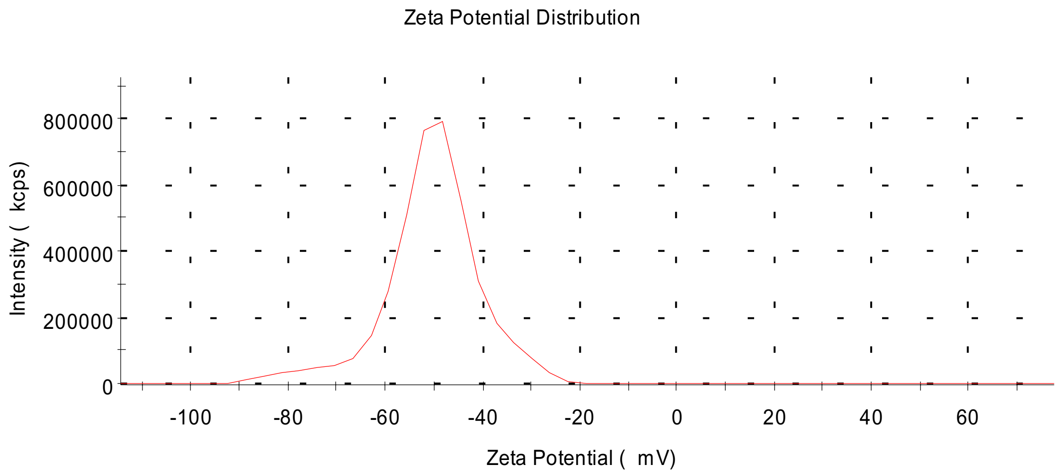


圖 20 調控 2 分鐘之界達電位(30 nm)分佈曲線

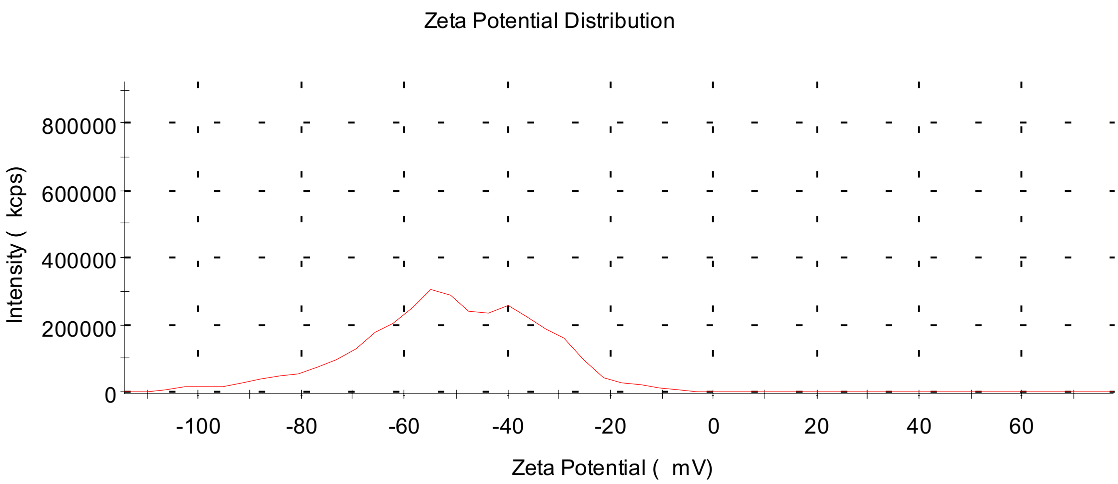


圖 21 調控 3 分鐘之界達電位(30 nm)分佈曲線

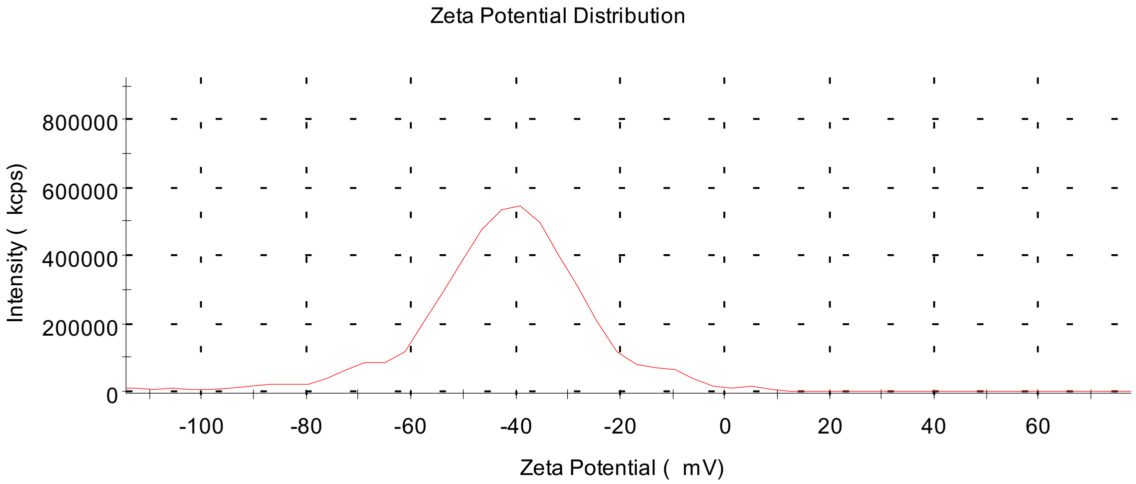


圖 22 調控 4 分鐘之界達電位(30 nm)分佈曲線

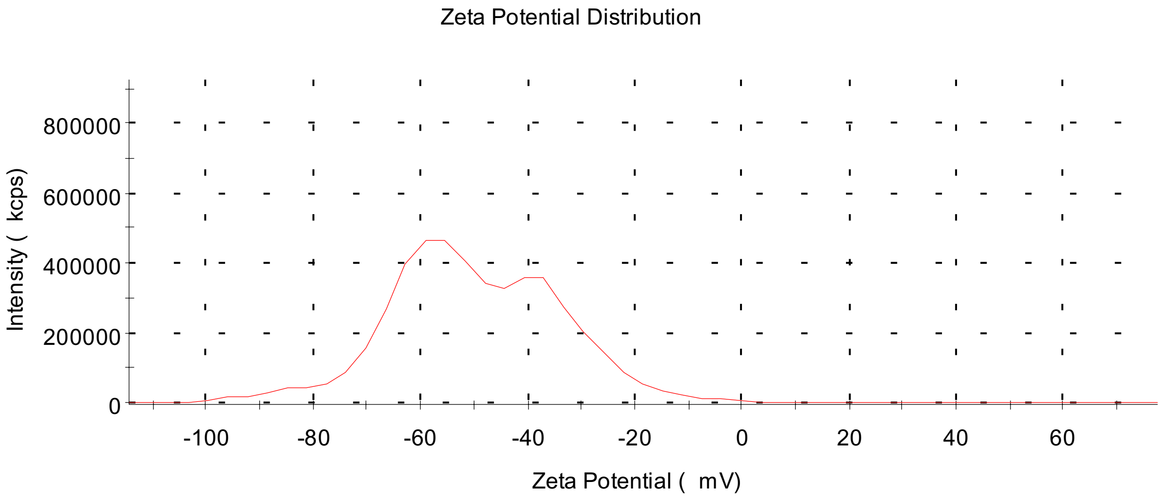


圖 23 調控 5 分鐘之界達電位(30 nm)分佈曲線

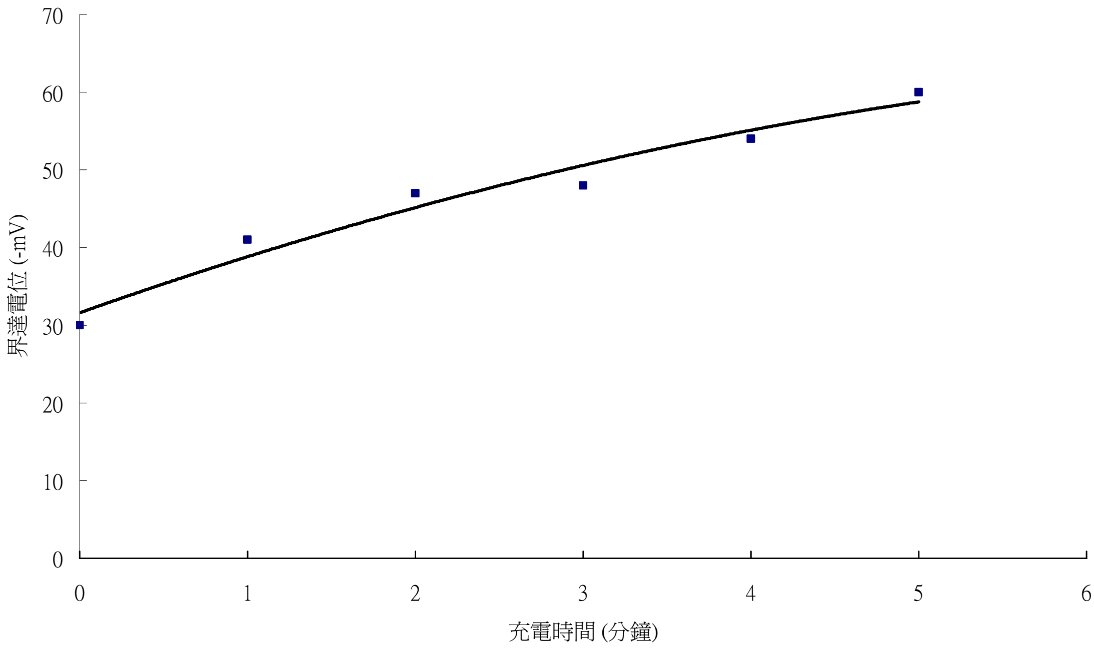
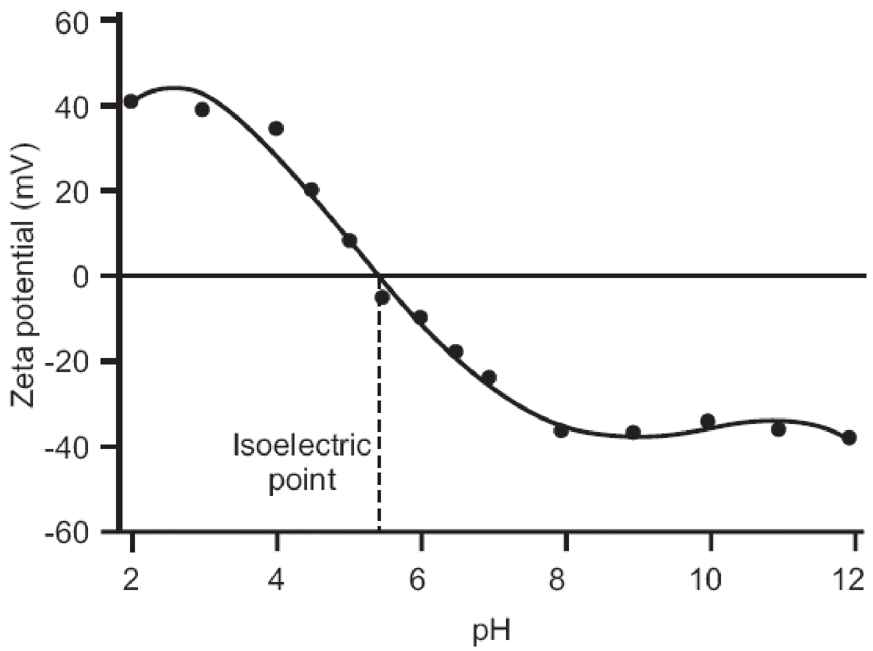


圖 24 充電時間與界達電位(30 nm)的關係



Source: Mavern

圖 25 界達電位與 pH 的曲線

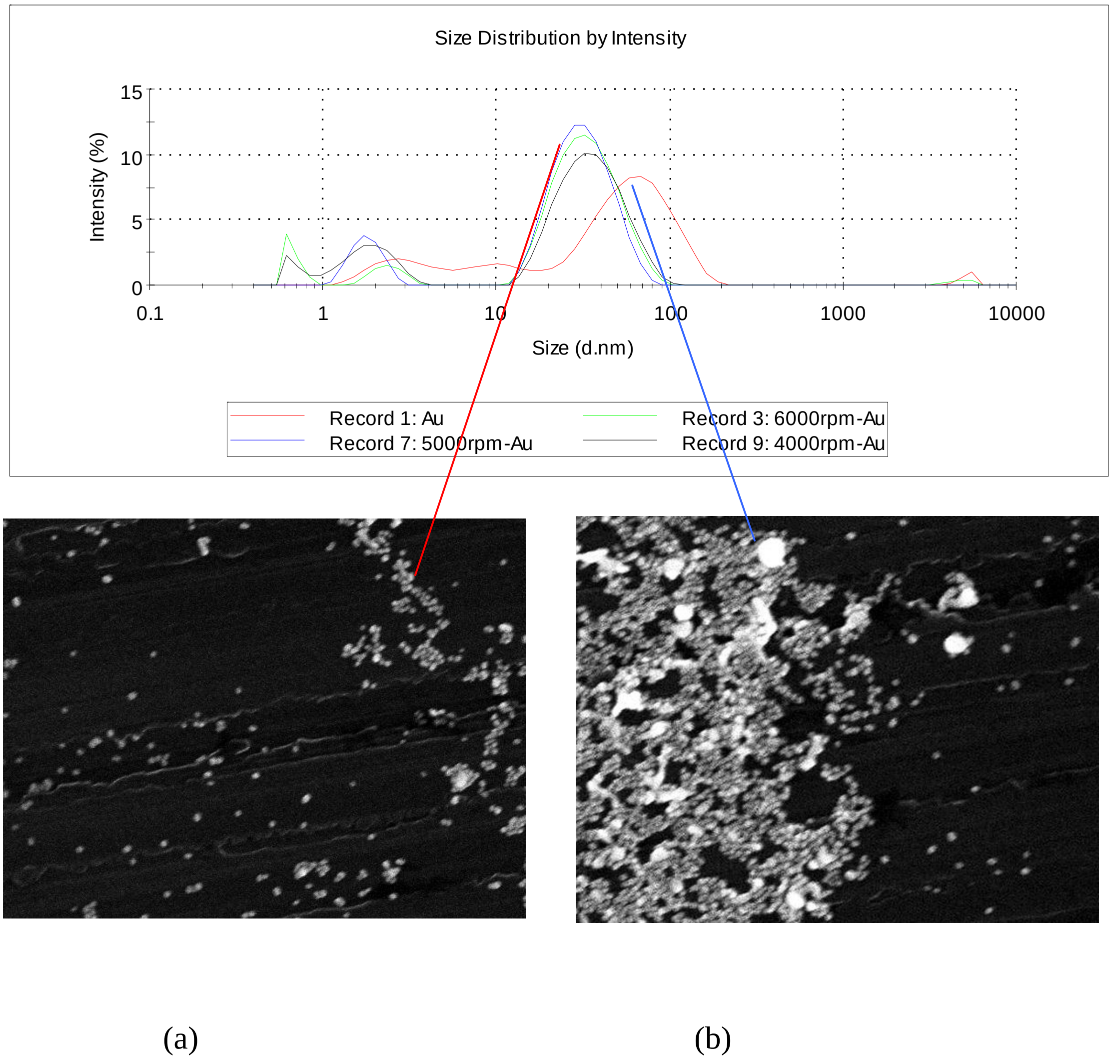


圖 26 利用電子穿透式顯微鏡分析離心作用對膠體金粒子的影響

(a) 離心篩選前之膠體金粒子

(b) TEM 照片

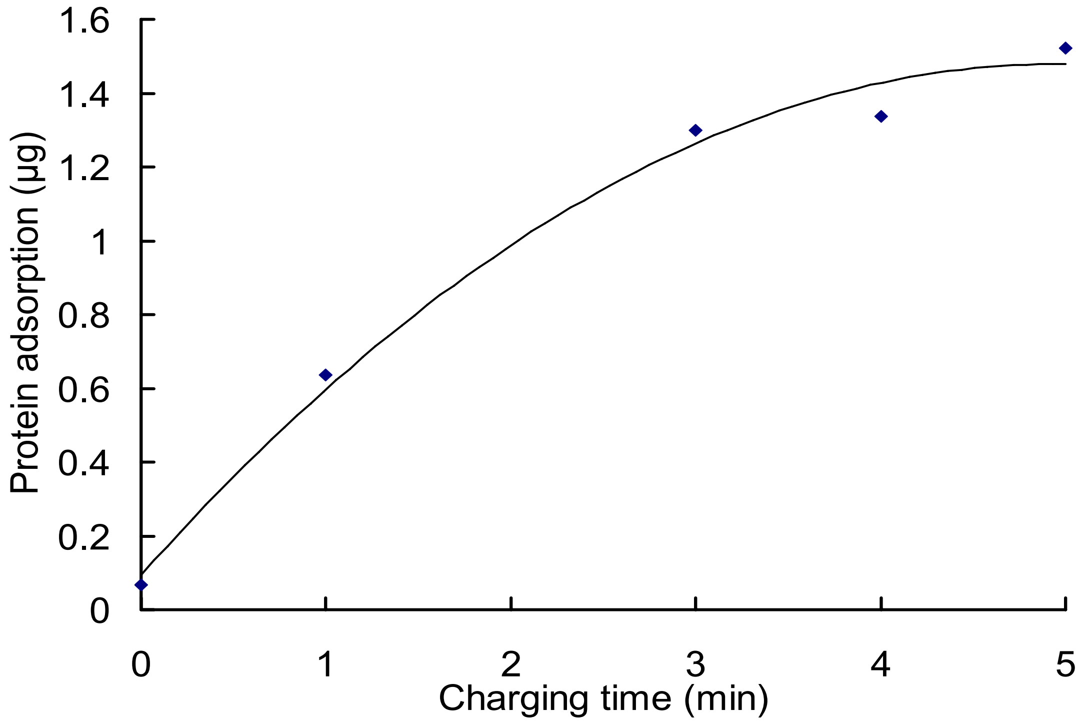


圖 27 蛋白質結合量與充電時間(min)的關係

伍、參考文獻

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區別：北

科別：Engineering (工程學科)

Capacitor Ultrasonic Zeta Potential Control System of Colloidal Gold

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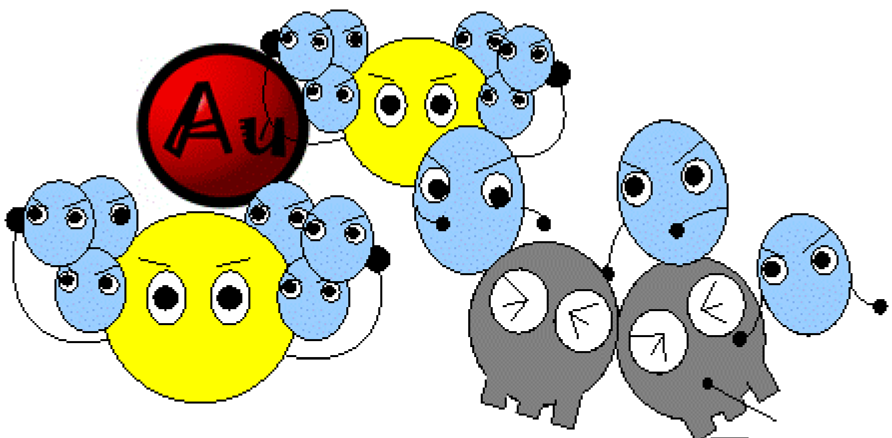
Keywords : Zeta potential 界達電位 、Colloidal gold 膠體金粒子 、Ultrasonic 超音波



Made in Taiwan Feb., 6, 2007 **LS**

No:

Abstract

"There is plenty room at the bottom." The words of Mr. Feynman are the beginning of nano technology. Mostafa El-Sayed, a professor of Georgia Institute Technology, identified cancer cells through nano gold-antibody complex. So, our study focuses on the zeta potential of colloidal gold particles.

Since that zeta potential is well known to strongly affect bio-material adhesion, we've designed a small experiment. We conjugated protein with colloidal gold. The result shows that when zeta potential increased, the protein conjugated quantity increased.

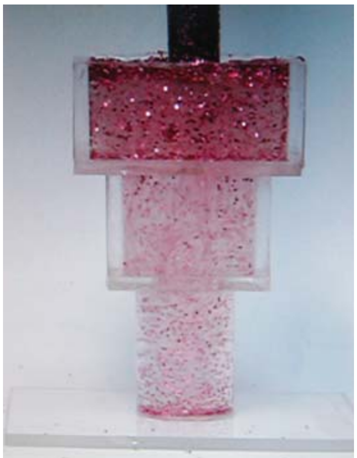
Nowadays, lots of academia institute have found many bio-engineering applications for colloidal gold. For instance, Prof. Angela Belcher of MIT found the Virus-Enable Nanowires. Prof. Mostafa El-sayed of Georgia tech is the inventor of colloidal gold cancer diagnosis. No mention to the well known anti-SARS No 1 of prof. C.K. Lee. Our zeta potential control technique is a part of these future industries.

At first, the filtering method and equipments were developed. The theories were based on the ultrasonic studies of universities such as National Taiwan University of Science and Technology. Then the colloidal gold's sizes were filtered to100 nm through settling. At last, by using Continual-Filtering Centrifuge (CoCe.) and Tube Well Mass (TW-MS), the mean particles sizes can be filtered to 30 nm.

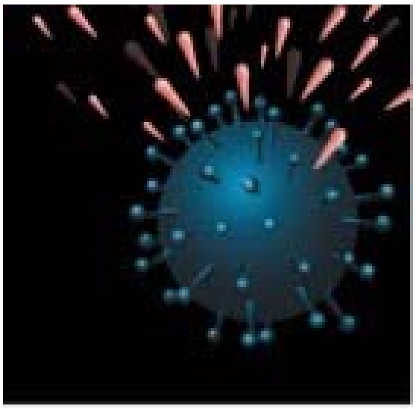
The most important results are: Zeta potential of the gold colloid was controlled with Capacitor Ultrasonic Zeta Potential Controller. The zeta potential can be raised from -30 mV up to -59 mV, which is -20 mV higher than the conventional pH-changing way. The function of zeta potential to protein

binding quantity was tested. The increasing curves of zeta potential and protein binding quantity were similar. This property would be a significance of biotechnology.

Thourgh Capacitor Ultrasonic Zeta Potential Control system, the zeta potential's limitation of gold colliod, which is produced by SANSS (Submerged Arc Nanoparticles Synthesis System), can be controled in a wilder range. The study which is focused on nano-scale, like the wish of Mr. Feynman – "To manufacture material and produce equipment in atom and molecular scale".



Theory of of ultrasonic
Source: Prof. T.S. Lei



Mechanism of『NTU Anti-SARS No.1』
Source: Prof. C.K. Lee

Introduction

In 1959, Mr. Feynman described nanothechnology as "There is *Plenty* of Room at the Bottom." Then Mostafa El-Sayed, a professor of Georgia Institute Technology, identified cancer cells through nano gold-antibody complex. It shows that's a big potential for identification of cancer, then a precise operation and therapy can be performed. And Colloidal Gold can be used in many anther fields, as the applications are list in this Table.This interesting finding on the characteristics of nano gold particles has drawn much attention from us in science and engineering in the world. So, our study focuses on the zeta potential of colloidal gold particles. Then in the past decades, more special characteristics of colloidal gold had been discovered. The widespread international researches have concentrated great efforts in this area. Naturally, nano-biotechnology will play an important role in the next decade.

"During the time of SARS, anti- SARS No.1 is invented by National Taiwan University." and "Binding gold nanoparticles to a specific antibody for cancer cells could make cancer detection much easier, suggests researcher Prof. Dr. El-Sayed at Georgia Tech." introduced us to study Binding gold of protein.

"Capacitor Ultrasonic Zeta Potential Control System of Colloidal Gold" the most important thing about a project is "what's new?"Our project has 2 news:The 1st one is: By using 1.65MHz Ultrasonic introduce water surface resonance, to disperse and filter Colloidal Gold. The 2nd one is: By using Capacitor's theory and Ultrasonic water resonance To control zeta potential of Colloidal Gold.

The chemical additive of in colloid is well known to strongly affect the zeta potential, which indicates colloid's stability and function. Our most important breakthroughs are: 1. The desired colloidal gold (35 nm) can be obtained using ultrasonic atomizer. 2. We demonstrate a physical method using electrostatic precipitator to control the zeta potential of colloidal gold between -28.5 mV and -59.1 mV. These physical approaches and methods can avoid the possible interferences from chemicals and improve the function or application of colloidal gold.

Normally, the colloidal gold is produced by a chemical method. The chemical colloidal gold has 4 weak points,

1. (the 1st is) not pure, there is chemical residual,

2. (the 2nd is) high cost, there are too many key factors for production, process is too complex, It's not so easy to reproduce the same colloidal gold. no mass but. Up to now the pure colloidal gold is high cost. Like this one, 25 cc, the cost is 2500 NT $.

3. (the 3 rd is) gathering. Gathering is always a big problem for nano-particles. Dispersion and filtration are key technologies for use of the colloidal gold.

4. (the 4th is)zeta potential of chemical colloidal gold is controlled by change the pH value. There is too much chemical additive in the chemical colloid.

Engineering goal

To avert the 4 weak points of chemical colloidal gold, We design a physical method to produce the colloidal gold. To reduce chemical additive influence and to improve the function of colloidal gold in bio-engineering.

The physical method has 3 parts:

1. (the 1st is) Production
2. (the 2nd is) Dispersion and filtration, and
3. (the 3 rd is) Control the zeta potential.

The first, Production of the colloidal gold is by using a physical method, the name is SANSS in Taipei University of Technology. The primary particle size is about 30 nm.

And then, Dispersion and filtration of the colloidal gold is by using this tow home made machines. (CoCe and TW-MS.) This one is through centrifugal force to filter the colloidal gold. And this one is by using 1.65MHz Ultrasonic introduce water surface resonance, to disperse and filter the Colloidal Gold, this can different kind of filtration techniques and propose a new filtration for colloidal gold.

At last, Control the zeta potential of the colloidal gold, we design this home made machine. By using Capacitor’s theory and 1.65MHz Ultrasonic introduce water resonance to control zeta potential of Colloidal Gold. To devise a physical zeta potential controlling technique and system, this can promote the function of colloidal gold on cancer diagnostics and drug delivering.

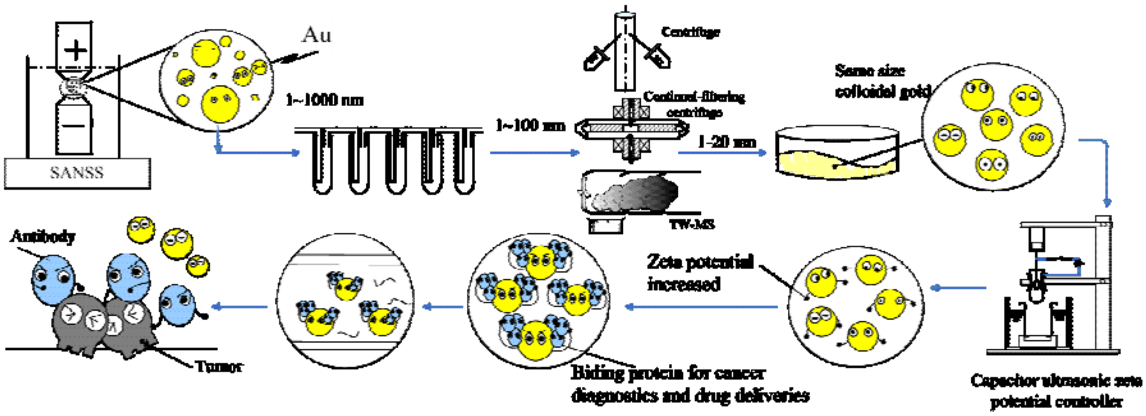


Fig. 1 Engineering goal for long term

Therefore, the devise which can be applied to filter colloidal gold, such as by using Continual-Filtering Centrifuge (CoCe.) and Tube Well Mass (TW-MS), are invented in this research and applied to develop the technology of control the zeta potential's limitation of gold colliod, which is produced by SANSS (Submerged Arc Nanoparticles Synthesis System).

Materials and Methods

A. Literature Review

Tabel 1. Notable work on colloidal gold and zeta potential

Year	Author(s)	institution	Title	Result
2004	Patrick Warren, , Nature 429, 822		Electrifying effects in colloids. (published on ***nature***)	Electro field influence the charge and potential of colloid.
2005	K. Rezwan, , et al.	ETH Zurich, Switzerland	Protein adsorption changed the Potential of Colloidal Particles.	Surface potential and charge strongly effect protein adsorption.
2005	M.A. El-Sayed, et al.	Georgia tech., U.S.A	Colloidal gold as Cancer Diagnostics.	Significant function
	D.J. Shaw, et al.	Malvern Instruments Ltd.	Zeta Potential An Introduction in 30 minutes	The theory review of zeta potential.
2005	Tsing-Tshih Tsung, et al , Jan W. Slot, Hans J. Geuze(1985), A New Method of Preparing Gold Probes for Multiple-labeling Cytochemistry, European Journal of Cell Biology 38., 87~93, 1985	Ni Nano-magnetic fluid prepared by submerged arc nano synthesis system (SANSS), JSME Inter national Journal. Series B, Fluids and Thermal Engineering, Vol. 48, No. 4, 750-755.【13】		
2004	Chad Mirkin, et al.	Northwestern University, US	Gold nanoparticles and bio-bar codes bring sensitive DNA detection	“bio-bar-code-based” DNA detection technique has a sensitivity similar to that of the commonly used polymerase chain reaction (PCR) method. The technique makes use of both gold nanoparticles attached to “bar code” DNA and magnetic microparticles.
2006	Richardson	Ohio University, US	Gold nanoparticles enhance laser heating	Gold nanoparticles was used as site-directed nano heaters that can be actuated with optical light
2002	Langmuir, Rajendra Bhat	North Carolina State University, US	Gold nanoparticles make the grade	First found that nanoparticles can form a gradient of decreasing concentration along a surface.
2003		Hebrew University of Jerusalem, Israel, and Brookhaven National	Gold nanoparticles could boost biosensors	Gold nanoparticles were used to attach glucose-oxidizing enzymes to electrodes

		Laboratory, US		which could ultimately be used as miniaturized biosensors inside the body that measure blood glucose.
2003	Murali Sastry, et al.	National Chemical Laboratory and the Armed Forces Medical College, India,	Micro-organism synthesizes gold nanoparticles	Using a micro-organism that normally grows on fig trees to synthesize gold nanoparticles which was much more uniform than the particles formed using other biological methods.
2003	Brahim Lounis, et al.	University of Bordeaux in France	Gold lights up biological cells	Gold nanoparticles was developed a new way of visualizing proteins in cells by labelling them.
2004	Crego-Calama, Holger Schönherr, et al.	University of Twente in the Netherlands	Self-assembly wins with gold rosette	Using the self-assembly of hydrogen-bonded rosettes to create nanostructures containing gold. The technique could have applications in the fabrication of nanowires.
2004	Jeff Brinker, et al.	University of New Mexico & Davidson College, US	Gold nanocrystals self-assemble to form 3D arrays	
2004	Swiss-US team	Swiss Federal Institute of Technology and the University of California Berkeley,USA,	Laser sintering strikes gold with nanoparticle ink	A laser technique for fabricating gold microstructures could provide a new powerful way to create miniature resistors or conductive tracks for flexible electronics.
2005	Uzi Landman, et al.	Georgia Institute of Technology, US, and Technical University of Munich, Germany	Nanocatalysts charge into action	It is found that gold nanoclusters on a ceramic surface gain an electrical charge while they act as a catalyst for the low-temperature oxidation of carbon monoxide.
2005	Chad Mirkin, et al.	Northwestern University and Rush University ,USA	Nanoparticle technique detects Alzheimer's-related proteins	Gold nanoparticle-based bio-barcode has been assayed to measure the concentration of amyloid-β-diffusible ligands (ADDLs) in cerebrospinal fluid (CSF).
2002	Jorge Gardea-Torresdey, et al.	University of Texas-El Paso, US	Alfalfa plants harvest gold nanoparticles	Using alfalfa is a cost-effective and environmentally friendly method of producing gold nanoparticles.

Tabel 3. Notable international work on Capacitor in lately 5 years

序號	年份	作　　者	單　　位	研究﹝論文﹞名稱	成　果
1.	2005	Zare , et al.	Stanford University, U.S.	Gold nanoparticles monitor protein folding	Gold nanoparticles could monitor changes in the folding of a yeast protein by attaching gold nanoparticles.
2.	2004	Chad Mirkin, et al.	Northwestern University, US	Gold nanoparticles and bio-bar codes bring sensitive DNA detection	"bio-bar-code-based" DNA detection technique has a sensitivity similar to that of the commonly used polymerase chain reaction (PCR) method. The technique makes use of both gold nanoparticles attached to "bar code" DNA and magnetic microparticles.
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5.	2003		Hebrew University of Jerusalem, Israel, and Brookhaven National Laboratory, US	Gold nanoparticles could boost biosensors	Gold nanoparticles were used to attach glucose-oxidizing enzymes to electrodes which could ultimately be used as miniaturized biosensors inside the body that measure blood glucose.

Tabel 3. Notable international work on gold nanoparticles in lately 5 years

序號	年份	作　　者	單　　位	研究﹝論文﹞名稱	成　果
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Tabel 3. Notable international work on colloidal gold in lately 5 years

序號	年份	作　　者	單　　位	研究〔論文〕名稱	成　果
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B. The Zeta potential Studied

Zeta potential calculation

$$\zeta = \frac{3U_e\eta}{2\varepsilon \cdot f(ka)} \quad \text{...(1)}$$

ζ Zeta potential

$f(\kappa a)$ Henry's Function

ε Dielectric constant

a

U_e Electrophoretic mobility

κ

η viscosity

A property of a colloid, Zeta potential was shown in the formula (1). The Zeta potential ζ in Eq. 1 was used to judge the precipitation tendency of the colloid. In Eq. 1, If ζ is zero of of the colloid, the Au-nanoparticles will gather together, and the Au-nanoparticles will precipitate.

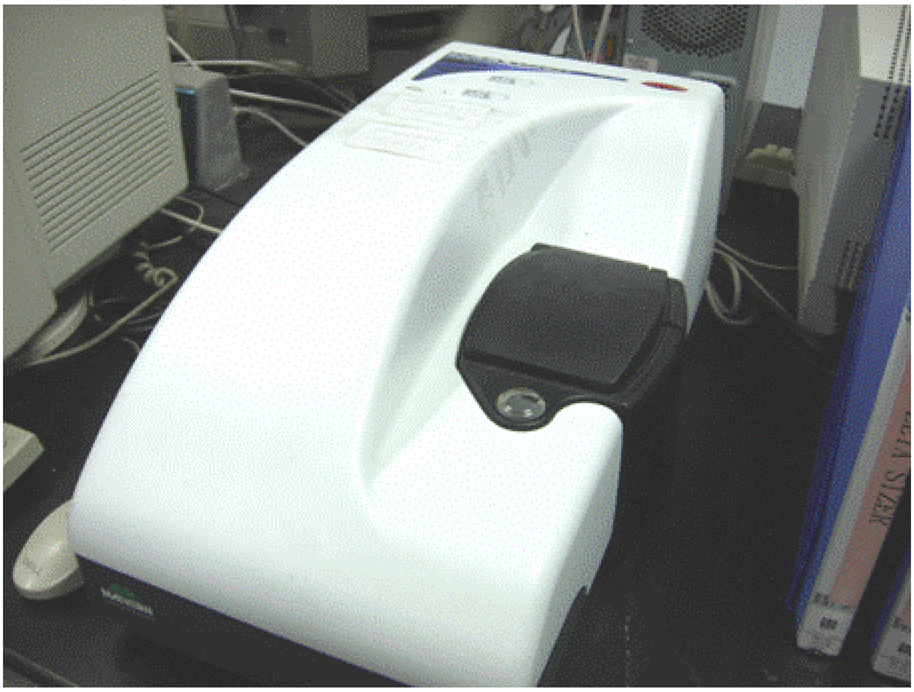

Fig. 2 Zetasizer nano ZS90 Zeta potential and particle size

C. Experimental Design

Why did we do the experiments?

Because zeta potential is so important. Zeta potential the overall charge a particle acquires in a medium. It was first discovered by Zsigmondy, Chemistry noble prize awarder in 1925. The magnitude indicates the stability of colloid. It’s also well known to strongly affect the adhesion with bio-materials.

How did we carry out the experiment?

We review many papers. Our idea comes from a paper in Journal, Nature, which indicated electrifying colloidal would influence the charge on particles. We reform the electrostatic precipitator, which invented by Dr. Frederick Cottrell, the Academician of National Academy of Sciences in USA.

Our theory was from a paper in Nature, which indicated electrifying colloidal would influence the charge on particles. We reform the electrostatic precipitator, which invented by Dr. Frederick Cottrell, the Academician of National Academy of Sciences in USA. Please look the “Capacitor Ultrasonic Zeta Potential controller”. This text tube was made into a capacitor. Inside is negative, outside is positive. 1500 V of electricity will be applied in addition to make negative charges. The test tube will be pull down with Actuator so as to separate positive and negative pole. The colloid will be atomized for making colloidal gold touch the pole. And the zeta potential will be increased by electrostatic induction.

This is the experimental setup for Zeta potential controller. Next, we’ll introduce you the the colloidal particles filtering section

D. Procedures

How did we carry out the experiment?

There are two sections in our project.

1. Zeta potential controller.

2. The colloidal particles filtering.

We'll introduce you the Zeta potential controller first.

Normally, the zeta potential of colloid is control through pH value. But there are two defaults. 1. Colloidal particles are not stable in high concentration ion medium. 2. Chemical additives may affect further applications. So a physical method is necessary.

The study can be summarized as following: Base on the theory of ultrasonic atomizer, the filtering method and device were developed. The sizes of colloidal gold, produced by SANSS (Submerged Arc Nanoparticles Synthesis System), were filtered to 100 nm through settling. By using Continual-Filtering Centrifuge (CoCe.) and Tube Well Mass (TW-MS), the particles sizes were filtered to 30 nm.

The most important result is: through "Capacitor Ultrasonic Zeta Potential Controller", the zeta potential of gold colloid can be controled in wilder range (-30 mV ~ -59 mV) .the saturation is -20 mV higher than the conventional pH-changing way.

The function of zeta potential to protein binding quantity was tested. The increasing curves of zeta potential and protein binding quantity were similar. This property would be a significance of biotechnology.

The study procedure in this research is as follows: At first, the filtering method and equipments were developed. The theories were based on the ultrasonic studies of universities such as National Taiwan University of Science and Technology. Then the colloidal gold's sizes were filtered to100 nm through settling. At last, by using Continual-Filtering Centrifuge (CoCe.) and Tube Well Mass (TW-MS), the mean particles sizes can be filtered to 30 nm.

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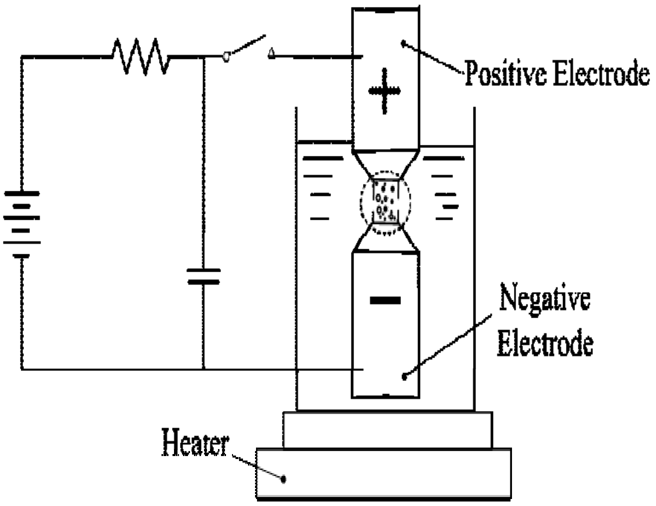
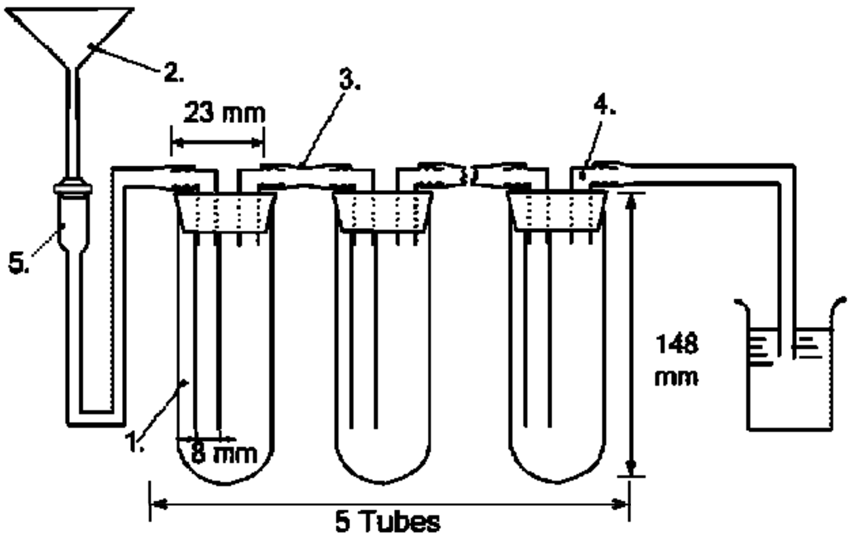
C. Instrument Design and Produce

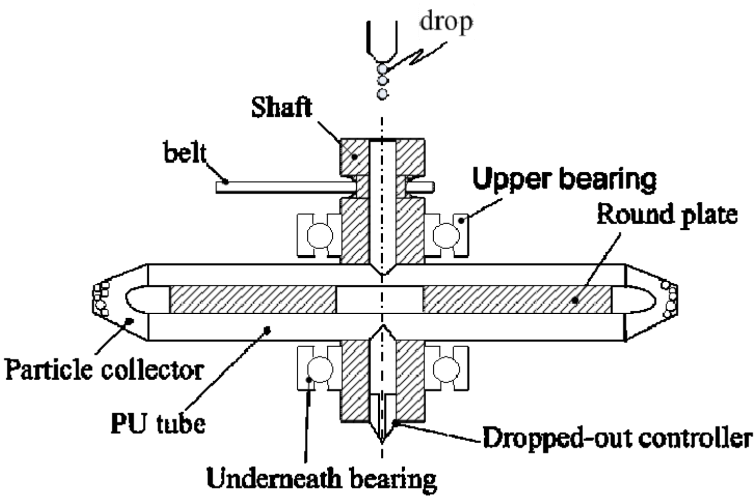
Fabrication and preparation of colloidal gold, the raw material, gold wire with size ψ1.0mm x 25 mm, purity 99.95% of Au-nanoparticles was used in this study. By SANSS, the Au-nanoparticles will be produced. Through setting tube, the mean size of the Au-nanoparticles is roughly about 100nm.

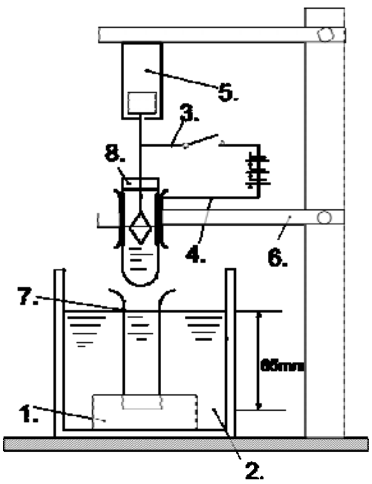


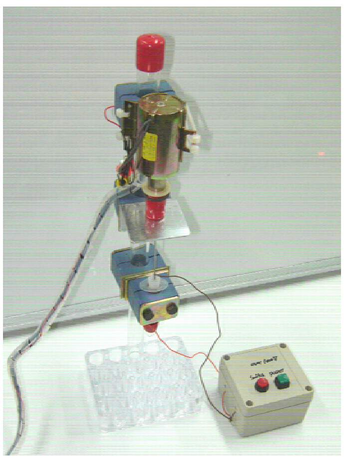
The colloidal gold were produced by SANSS. The diameter of particle size is about 1000 ± 10 nm. The deionized water was employed as a solution and its resistance is 17.9 MΩ.

The structure schematic diagram of the Settling System is shown in Fig.1 and the device is shown in Photo 1. It is composed of five test tubes and a dropper. The flow rate of solution is controlled at 60 ml/min by dropper. As gold colloid pass through the tubes, big particles will settle down at the bottom.





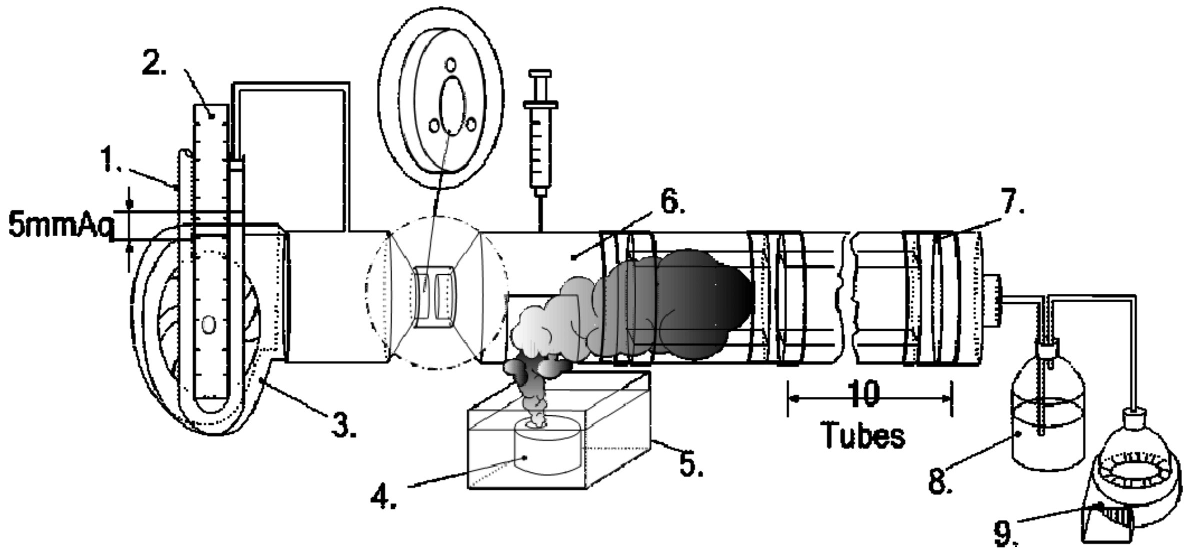


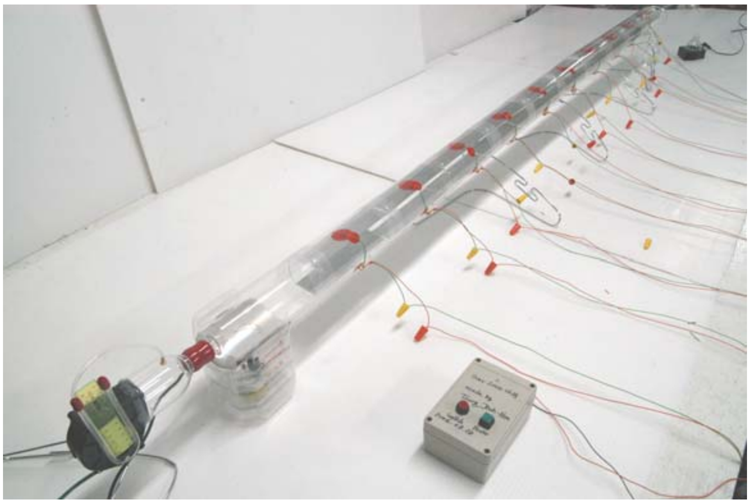


1. Ultrasonic atomizer
2. Cooling water
3. Negative electrode
4. Positive electrode
5. Actuator
6. Test tube holder
7. Silicon tube
8. Capacitor (Test tube)

Among the three kinds of methods we've tried, TW-MS is the best. The principle of TW-MS was from Mass spectrum. Colloidal gold is filtered by mass difference. When the 1.65MHz atomizer is working, water and metal slice from coke can will resonate. Then gold colloid will be nubilized. The gold colloid will be blow into the tube. The large particles will settle faster.

Theoretically, the more after the tube, the smaller particle size will be. But colloidal gold will distribute in TW-MS as figure 20. There are separated and aggregated particles in the same tube. This phenomenon may affect the filtering effect. As our lately result, the size of colloidal gold was successfully filtered to 30 nm with TW-MS.





1. U-tube
2. Ruler
3. Blower
4. Ultrasonic atomizer
5. Water
6. Plastic bottle
7. Plastic cover
8. Water
9. Exhaust fan

Results and Discussion

What is the significance of our results?

1.The peak number was appeared in ca. 35 nm, after filtering by Continual Centrifuge. It’s the 2nd particle size. There is Gathering hire and hire.(point the picture Fig. 10)

2. 35.6 nm of colloidal gold in tube No.8 was obtained using TW-MS. It’s the 2nd particle size too. There is Gathering too hire and hire. To reduce variable, the gold colloid was produced by SANSS, the physical way. As you can see, gold bar are the electrodes. When the electrodes are close enough, gold bar will be vaporized by arc. The vaporized gold will be quenched by water rapidly. Thus colloidal gold nucleated and formed. Because the range of size will be from 1 nm to 1000 nm, filtering methods are necessary.

The zeta potential of colloidal gold was controlled successfully through Capacitor Ultrasonic Zeta Potential Controller. The peak value shifted form -29.5 mV to -59 mV. Also, the longer the changing time, the higher zeta potential will be. The saturation of average zeta potential is about -50 mV, which is 10 mV higher than the conventional pH changing way.

In this device, A, are tubes. B, the 1.65MHz ultrasonic atomizer, which is to nebulize the gold colloid; C, the blower, which is to blow the colloidal gold into the tubes;

In order to observe the diameter of the gold colloid after filtering and before filtering, the reacted product were examined with the transmission electron microscope (TEM) and it were identified and analyzed by the particle size analyzer with the laser diffraction methods.

The structure schematic diagram of Continual-filtering Centrifuge (Coce.)

is shown in Fig. . According to the experience and theory analyze, the first generation has been developed. It is mainly composed of a rotor, an acrylic shaft and a motor. In order to make the filtering time longer for increasing the efficiency, the PU tube is coiled around the rotor so that the gold colloid can flow in it and be filtered by the centrifugal force. The shaft was driven by the motor and the highest rotational speed can be up to 6000rpm.

When colloidal gold was filtered by centrifugal force, the movement of particles is as fig . If the displacement of colloidal gold is bigger than the inside diameter of the tube, the particle will attach to the inner wall of it.

But due to the weight of the first generation is too heavy, it is difficult to assemble in the afterwards experiment. Thus, the second generation of Coce. was design and set up and is shown in Fig. . The body of the devise is composed of a slab of acrylic and rest on a base of vice. It can be easily disassembled to two parts and take with one. In addition, the rotational speed can be controlled with the step pulley.

It is found that in the afterward experiment, the shape of acrylic shaft were changed due to the overheating caused by the bearings. Therefore, the final generation was designed in Fig. . The bearings were changed to smaller ones so that the shaft will not be melt by the overheating caused by the bearings.

Nowadays, By using anyone Internet Search Engine to find the “http://” about Ultrasonic, capacitor or Zeta potential, Search Results are always over thousand thousands Web site, that come to us in ms. From this information, we can feel that this three nouns are so popular and important in our life and industry. What’s new can we do? This question is easy, but the job is difficult. If we can do what a little new is, it’s surely an innovation.

The little new is:1. Capacitor Zeta Potential Control System of colloidal gold, 2. Ultrasonic Atomizer for colloidal gold.

How important is Zeta Potential for a colloid? We can understand as follows:

1. Zeta Potential is a measure of the electrical force that exists between atoms, molecules, particles, suspensoids, cells, etc., in a fluid【1】.
2. Hundreds of industries work with Zeta Potential to control their chemical reactions【2】. — Tommy Cichanowski —
3. The Science of "Zeta Potential" is used by Hundreds of different industries. This knowledge is used to improve many things and processes; from concrete to beer, and much much more【3】 ... Dr. T.C. McDaniel
4. Zeta Potential represents a basic law of Nature, and it plays a vital role in all forms of plant and animal life. And Control of Colloid Stability through Zeta Potential. 【4】

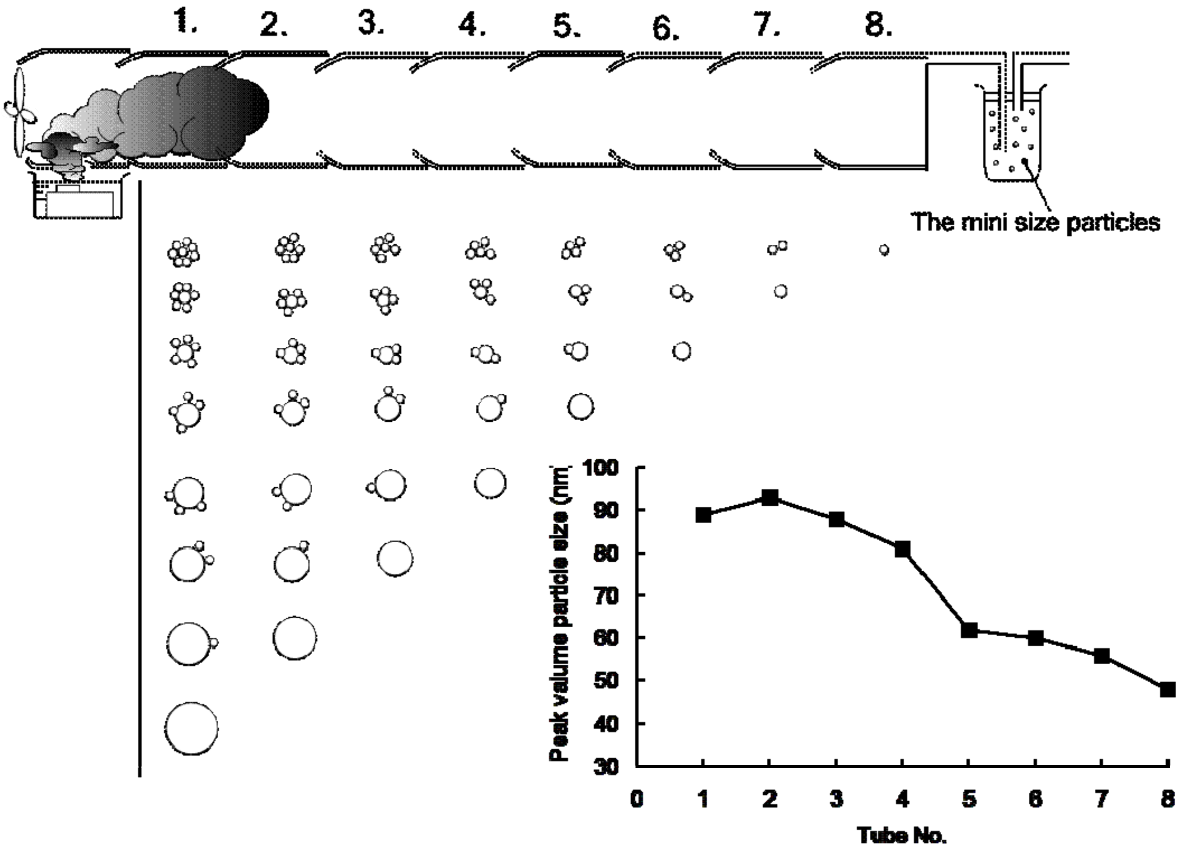
... Thomas M. Riddick

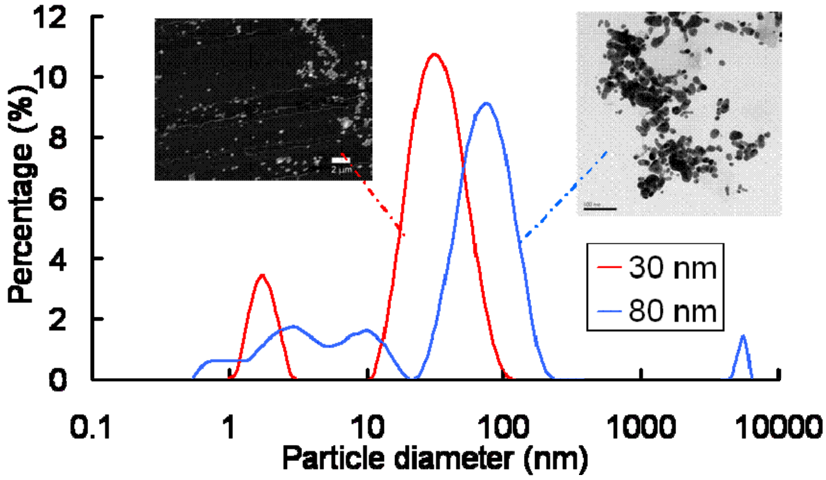
Practice work of industries with Zeta Potential is a natural law in following【5】:

1. The zeta potential is the overall charge a particle acquires in a specific medium.
2. The magnitude of the zeta potential gives an indication of the potential stability of a colloid.
3. If all the particles have a large negative or positive zeta potential they will repel each other and there is dispersion stability.
4. If the particles have low zeta potential values then there is no force to

prevent the particles coming together and there is dispersion instability

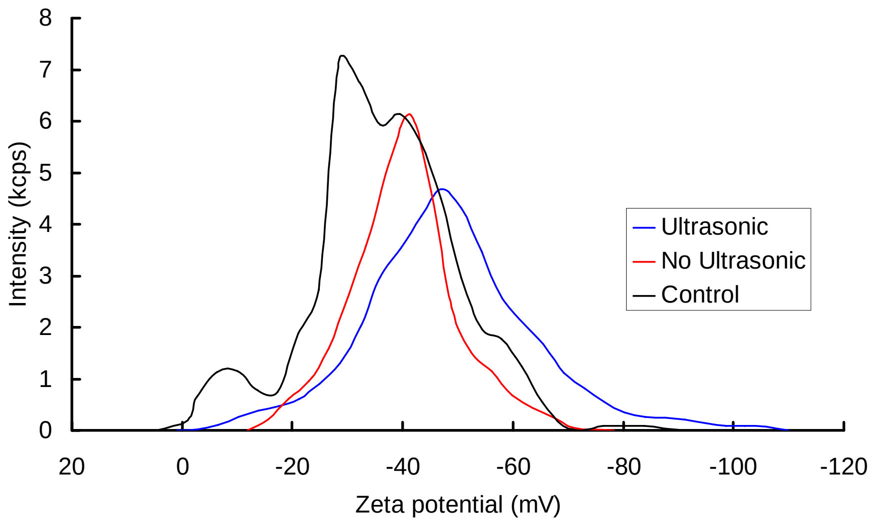
5. A dividing line between stable and unstable aqueous dispersions is generally taken at either +30 or -30mV
6. Particles with zeta potentials more positive than +30mV are normally considered stable.
7. Particles with zeta potentials more negative than -30mV are normally considered stable.

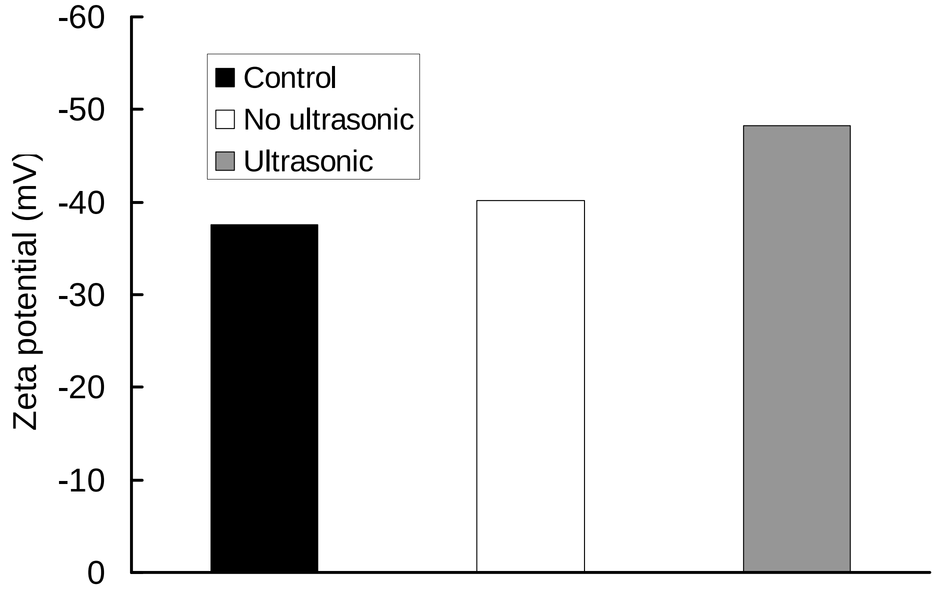




The distributing hypothesis and result of colloidal gold in TW-MS

It is assume that the of colloidal gold distributed as Fig. 18. So there were separated and aggregated particles in the same tube. The more after the tube, the smaller will be.





The mean zeta potential of colloidal gold with different treatments, the peak value increased after 1 min, 1500 V charging. The one atomized with ultrasonic increased more significantly. Comparing to no ultrasonic, the average zeta potential increased about 8 mV after atomizing with ultrasonic.

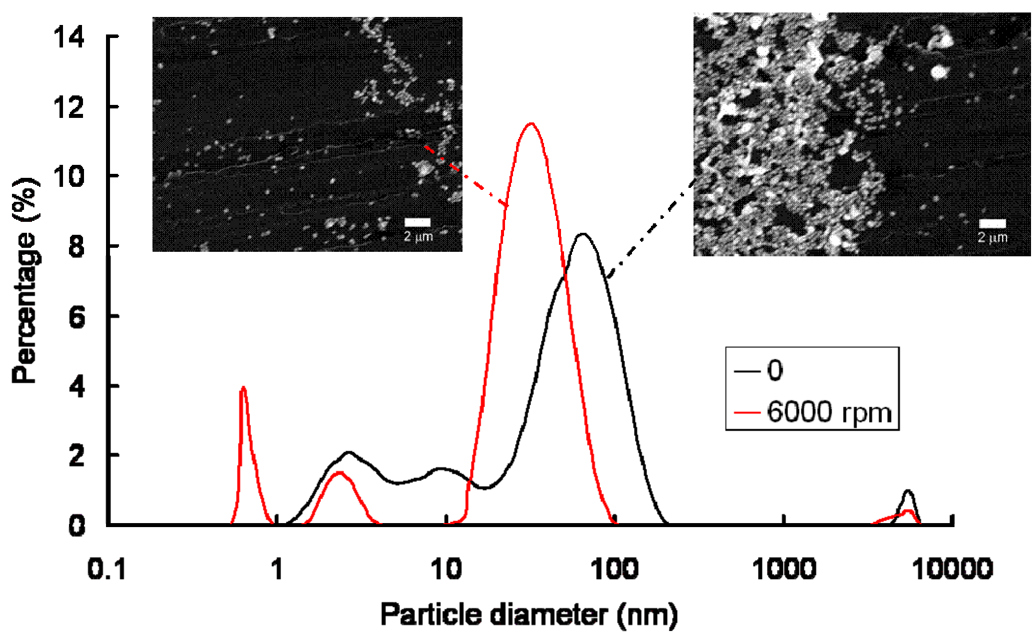
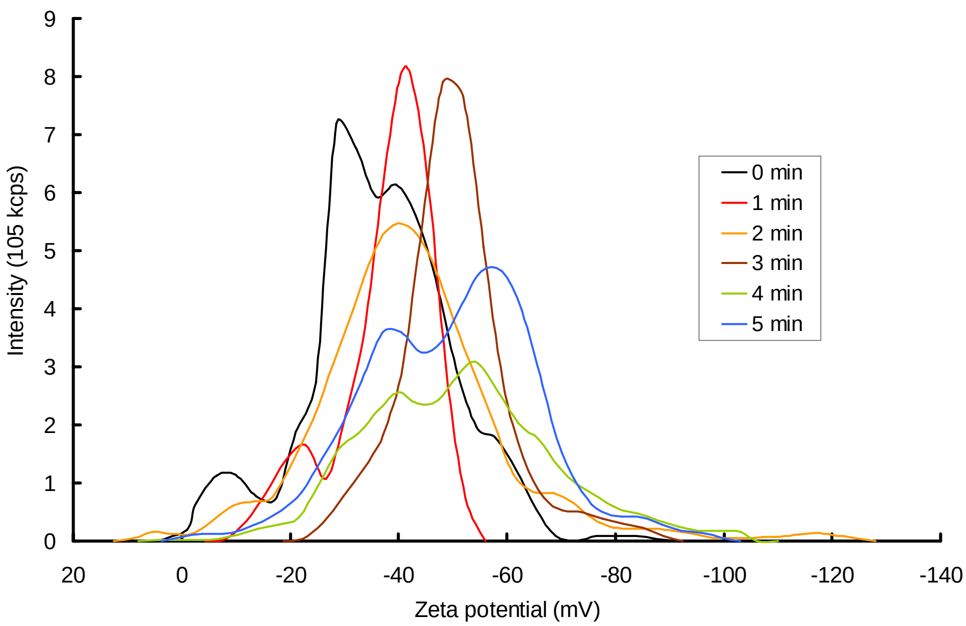
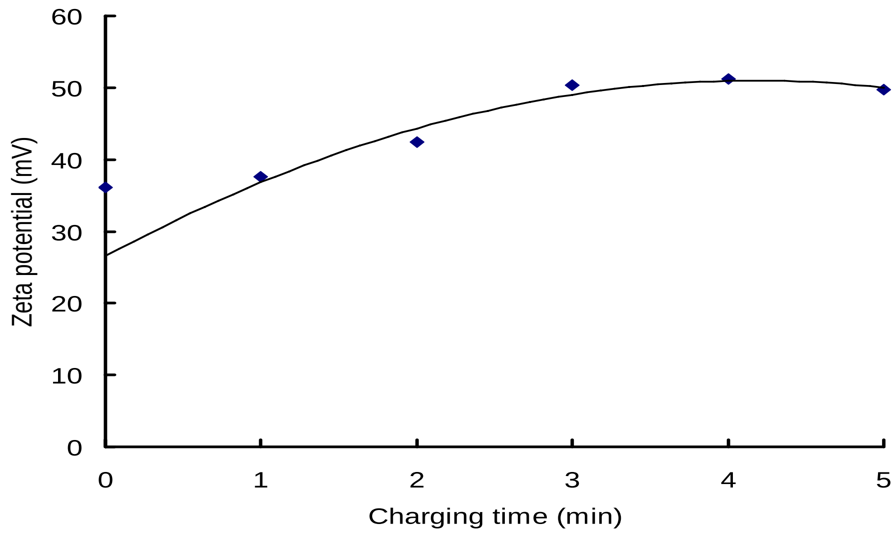


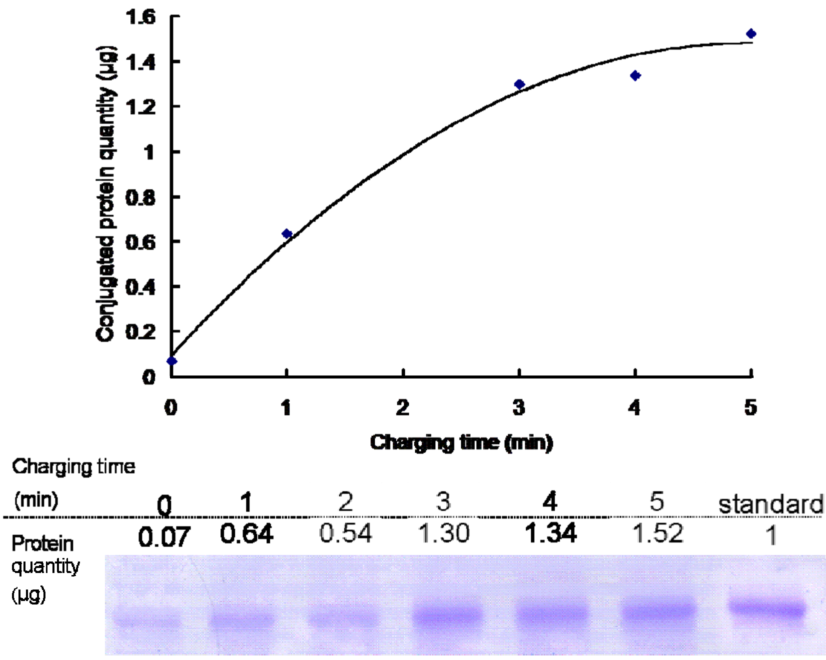
Fig. 15 shows that the colloidal gold being filtered by Coce.. In these two samples, the black curve is a sample, which wasn't being filtered. The peak number was appeared in ca. 80 nm. The red curve was filtered by Coce. at 6000 rpm. The peak number was shifted to left and was appeared in 30 nm. In order to observe and characterize the result of the filtered colloidal gold, the TEM images were displayed to correspond with the laser diffraction result. The quantity of aggregated colloidal gold, which was filtered by Coce. is fewer than without being filtered, is confirmed by TEM observation.

It means that the aggregated colloidal gold were filtered by Coce.. So, the laser diffraction result shows that the peak number of colloidal gold is larger than without being filtered due to the aggregated colloidal gold is in majority.

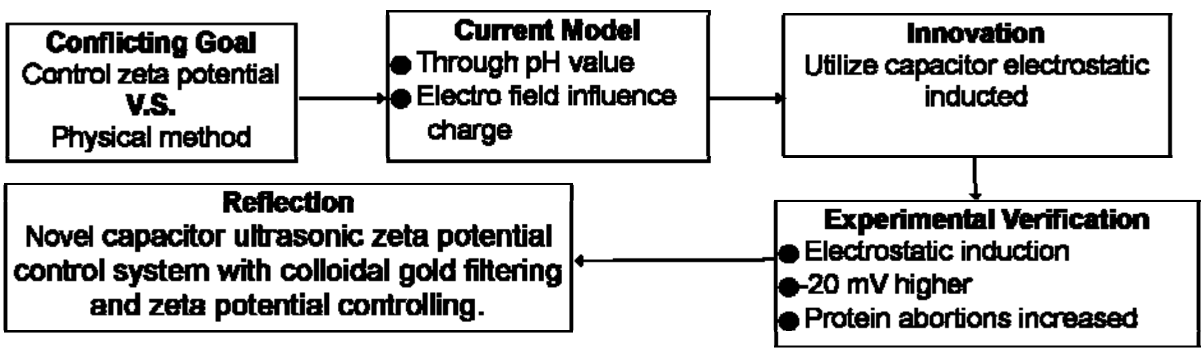




The relations between average zeta potential and charging time, the peak value was shifted form -29.5 mV to -59 mV, which means the zeta potential of colloidal gold was raised successfully through Capacitor Ultra-sonic Zeta Potential Controller. The longer the changing time, the higher peak volume will be. The saturation is about -59 mV.



The relation between charging time and conjugated protein quantity on colloidal gold, the absorption was analyzed with SDS PAGE, the typical protein molecular weight assay. The quantities were counted trough software Biotech by comparing the band's area with standard. The curve trend is similar to Fig 1. The result shows that through controlling zeta Potential, The conjugated protein quantity will be controlled as well.



Conclusions

According to the protein binding experimental result, zeta potential plays an important role in the property of colloid. Our conclusion is:

The capacitor ultrasonic zeta potential control Controller were successfully designed and produced. The zeta potential peak volume of gold colloid can be raised from -30 mV to -59 mV. The saturation of average zeta potential is -50 mV.

Tube Well Mass spectrum (TW-MS), Continual-filtering Centrifuge (CoCe.) and settling system.

The results shows that colloidal gold can be filtered to 100 nm through settling. By using CoCe. and TW-MS, the mean size can be filtered to 30 nm. The peak volume zeta potential of gold colloid can be raised from -30 mV to -59 mV. Which is -20 mV higher than the saturation of the conventional pH-changing way (-40 mV).

The protein conjugated experimental result shows that colloidal with higher zeta potential conjugate more protein.

The filtering techniques of colloidal gold have been developed with the physical method. Three main results show that by using Settling System, colloidal gold can be filtered to 100 nm. Then with Continual-filtering Centrifuge and Tube Well Mass Spectrometer, colloidal gold could be filtered to 30 nm. The results demonstrate these techniques could separate the different diameters of the colloidal gold successfully.

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評語

1. 從當紅的金粒子應用發想，進行系統化的研究是相當值得鼓勵。
2. 利用電容與超音波的原理研究其對界達電位的影響，應更清楚地在題目上表達。目前題目恐有混淆疑義。
3. 簡報同學的報告及問答表現十分優良。
4. 對應用方面應顯示更多的資訊。