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ABSTRACT

Title of Thesis : The Separation of Proteins via Charged Electromembrane Process

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Thesis Directed by : Dr. Cheng-Chong Lin

A new method for separation and concentration of protein and other biocolloids such as hemoglobin and albumin was developed by a process called the charged electromembrane (CEM) process. The process is characterized by using a metallic membrane as a filter which can carry either positive or negative electric current.

The effect of various factors on the separation coefficient and filtration efficiency were investigated experimentally. These factors include strength of electric field, pH of colloidal solution, electric current through membrane filter and flow rate.

It was found out that the thickness of the gel layer formed on the membrane could be largely eliminated by electrophoresis and the expulsive force caused by the negatively charged electromembrane.

THE SEPARATION OF PROTEINS VIA CHARGED ELECTROMEMBRANE PROCESS

BY

CHI-WEN LIN

Thesis Submitted to the Faculty of the Graduate School
of the
New Jersey Institute of Technology
In Partial Fulfillment of the Requirements for the Degree
of
Master of Science in Chemical Engineering

APPROVAL SHEET

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I. INTRODUCTION

A new method for separation or concentration of protein and other biocolloids such as hemoglobin and albumin is developed by a process named charged electromembrane (CEM) process which was invented by Professor C. C. Lin. In this research, both batch and continuous processes with charged electromembrane (CEM) were experimentally investigated at various operating conditions.

This review focuses on a relatively new class of membrane separation processes in which an electric field applied across a charged membrane can control the permeability of that membrane to charged and neutral solutes. The process is characterized by metallic membranes with optional pore size which can carry either positive or negative electric current. This differs from the use of polymer membranes in other traditional processes. The liquid phase is a solution of proteins which is under the influence of electric field.

Filtration is a unit operation designed to separate suspended particles from a fluid stream by passing the solution through a porous medium. As fluid is forced through the voids or pores of the medium in the traditional process solid particles are retained on the medium's surface or, in some cases, on the walls of the pores, while the fluid, called filtrate, passes through.[1]

In order to separate and to thicken colloidal solution, ultrafiltration and electro-ultrafiltration have been used in the chemical industry, the food industry, the fermentation industry, the pharmaceutical industry, waste water treatment and many other chemical engineering processes. But one of the major bottlenecks in these filtrations of colloidal solutions such as those of protein is gel formation, which exhibits very large filtration resistance.[2]

The charged electromembrane process is very effective in increasing the filtration flux, because the thickness of the gel layer formed on the membrane might be largely eliminated by electrophoresis and the driving force caused by negatively charged membrane. The sources of the driving force for processing by ultrafiltration and CEM are obviously distinctive. In the case of the CEM filtration process, the reason for suspended particles being retained in the concentrated solution is not the pore size of the membrane but the expulsive function of the electromembrane which carries the same charge as suspended colloids do.

Protein is a polypeptide which is made by condensing various kinds of amino acids and it exhibits amphoteric properties. The electrobinetic potential (so called ζ-potential) of protein particles dispersed in water and the polarity of the potential are natually changed by

the value of the hydrogen exponent, pH, of the dispersion medium. Therefore, in the process of electromembrane separation of a colloidal solution of amphoteric material such as protein, PH of the dispersion medium affects filtration flux.

The charged electromembrane (CEM) process is used to improve the efficiency in separating colloidal particles such as protein from colloidal solution. In this process, it is important to find suitable operating conditions in order to increase filtration flux by decreasing the thickness of the gel layer formed on the membrane and to enhance the reflection coefficient of colloidal particles defined as $R = 1 - (C/C_0)$ and filtration efficiency defined as $\eta = C/C_0 - 1$.

Electrodialysis is the transport of ions through membranes as a result of an electrical driving force. Several variants of the basic electrodialysis process, coupled with ion exchanger and/or neutral membranes have been developed to overcome some of the problems that have been encountered with the basic process. The classical cation-anion membrane used in separation or concentration of proteins via electrodialysis poses two potential problems with respect to fouling of the membrane. These problems are precipitation of the less soluble calcium minerals, and deposition of protein fractions on anion - exchange membrane

surfaces.[3],[4]

Filtration by the charged electromembrane process is a complex operation involving the intricate interaction of many factors, some of which may as yet be unrecognized. In numerous applications, however, negatively charged electromembrane filters have proved reliable. It may well be that our understanding of the total processes involved in filtration does not yet account for that high degree of dependability. Particle concentration by negatively charged electromembranes is, however, a well contributed practice if it is properly performed. This success augurs a growing future for the filtrative technique and its expanding applications.

II. THEORETICAL CONSIDERATION

A. Fundamentals

In the first experiment, a fluid mixture of hemoglobin and Bovine albumin was examined using water as solvent in the CEM process. It is well known that the above proteins have isoelectric points $I_H(6.9)$ and $I_A(4.8)$ respectively. Proteins carry both negatively and positively charged groups and can normally move toward anode and cathode respectively due to electrophoresis. At low pH the net charge of protein is positive while at high pH it is negative. At the point of zero net charge, the isoelectric point, substances are not moving toward any directions as migration ceases.(5)

Hemoglobin is a relatively small (65,000 M.wt) and highly charged molecule. Bovine albumin, with a molecular weight of about 69,000, is the smallest and most aboundant of serum proteins. In the case of milk proteins, they are known to form complexs with small ions and molecules, to bind water, and to form complexes with other proteins or macromolecules. The protein in normal milk or fresh whey is a high molecular weight albumin (approximately 40,000). Amino acids and proteins acquire their charge mainly through the ionization of carboxyl and amino groups to give COO^- and NH_3^+ ions. The ionization of these groups and the net molecular

charge, depends on the pH of the solution. At low pH an amino acid or protein molecule will be positively charged and at high pH it will be negatively charged. The presence of charges on the particles or macromolecules of the dispersed phase of a colloidal solution will result in an electrical double layer at the interface, as suggested by Helmholtz of opposite sign to that of macromolecules.(6)

Like many other solutions of food substances, protein is extremely pH sensitive. The normal pH of protein is about 4.7. Shifts of pH away from the isoelectric point tend to denature protein. On the acidic side of the isoelectric point, this denatured protein in agglomerated as an insoluble point, protein is also denatured, but under these alkaline conditions protein fractions are quite soluble. Shifts of pH that accompany polarization and water splitting at membrane interfaces, when limiting currents are exceeded, frequently result in deposition of thin layers of denatured protein on membrane faces.(6)

In the presence of an external electrical field, the particles of the dispersed phase move relatively to the dispersing medium in a direction determined by the sign of the charge and the direction of the applied field.

When electric current passes through the solution

compartments and membranes, negatively charged colloids tend to migrate toward the cathode and positively charged colloids tend to migrate toward the anode.(7)

In addition to separating colloids from solution on the basis of their charge sign, membrane systems can be used to separate ions of like charge on the basis of their different rates of transport through membranes.

When an electric field is applied to a normal membrane system

anode | solution(C) | membrane | solution(c) | cathode

the current is carried through the membrane by the ions whose movement will be facilitated if the membrane is highly conducting (high fixed charge density). Both cations and anions are transfered across it.(8)

When an ion moves from the solution bath into the generally hydrophobic membrane it may lose waters of hydration. To do so, it requires energy. Thus, one expects an energy barrier at the membrane surface that would tend to keep ions out of the membrane.

In the area of membrane transport, electrical forces can couple to charged membrane and interstitial fluid (e.q., electrokinetic interactions), or directly to charged solute species (e.q., solute migration). It is therefore very

important to distinguish between the effect of an electric field on the membrane matrix versus that on the solutes to be transported. To make this point, several contrasting systems including one in which the spacing between molecules of a membrane matrix, and hence the effective pore radius of the membrane, can be controlled by an applied electric field. More specifically, the electric field in the latter configuration alters the concentration or pH inside the membrane by means of an electrodiffusion mechanism. This modulates electrostatic repulsion forces between charged membrane molecules and fibrils, thereby changing the interstitial separation distances which determine the effective permeability of the membrane to other charged solute probes.[9]

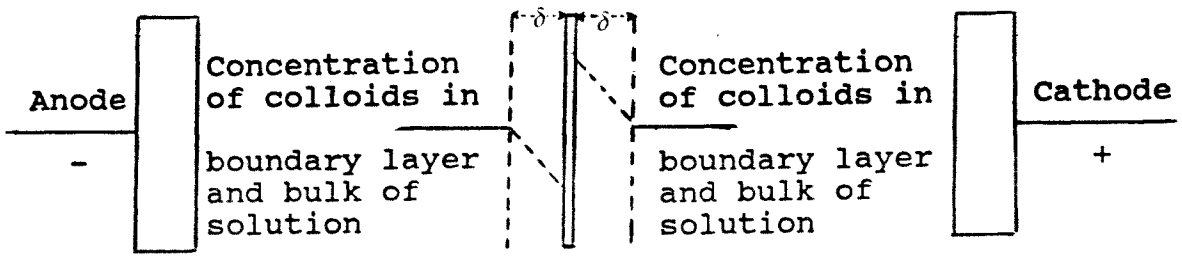
In the experiments of Pefferborn et al.[10] the membrane pores were rigid but could be blocked by concentration changed in absorbed, flexible macromolecules. In contrast, others have studied membranes made entirely of flexible molecules in which ionization of fixed charge groups can result in bulk swelling of the membrane. Hence they concluded that their membrane's structure and physical properties were closely linked to the membrane's transport properties.[11]

Gliozzi, Vittoria and Cifferri found that swelling, electrical resistance, streaming potential and water

transport changed significantly with bath pH. Therefore, the increases in swelling and dimensional changes that occured when the pH was lowered. Titration curves of intact bovine corium collagen show that collagen possesses a positive net fixed charge density for pH < 5, negative fixed charge for pH > 9, and is essentially neutral (isoelectric) between pH 5 and 9. The resulting electrostatic repulsion and osmotic swelling forces generated by collagen fixed charge groups are capable of inducing the observed increase in the volume and changes in the dimensions of the membranes.[12]

B. Limitations

The major limitation of the production rates achievable in the conventional electromembrane process is concentration polarization at the surface of the electromembrane. Concentration polarization occurs because of differences in the transport numbers of colloids in the solutions and in the electromembranes. To illustrate the source of concentration polarization that occurs at an electromembrane, consider the fact that the transport number of negatively charged colloids in a solution is lower than it is in an electromembrane. Because of the lower transport number, the number of negative colloids transporting through the solution to the surface of the conventional electromembrane by the electrical current is not sufficient to make up for the negative colloid removed from the surface and transfering through the membrane. This deficiency of negative colloids results in a reduction of the concentration of colloids in the solution at the surface of the membrane.(13)



E : electromembrane

δ : thickness of idealized boundary layer

As the electric current density is increased these interfacial concentrations become lower (on the entering side) and higher (on the other side). As the current density is increased still more, the concentration of colloids at the entering interface approaches zero. At this current density (termed the limiting current density) H^+ and OH^- ions formed by ionization of water begin to be conducted through the solution and through the membrane. The OH^- ions transfered through the membrane can cause deleterious changes of pH in the membrane and in the solutions in the boundary layers. Also, the layer of the almost deionized water in the boundary increases in resistance. Thus concentration polarization imposes a real limitation on the production rate (i.e., transport of desired colloids) in a conventional electromembrane unit.

The flux and selectivity of the conventional electromembrane filtration process have been improved by combining it with an electrophoteric force which acts on the retained solutes to control concentration polarization. During CEM filtration, the steady-state solution flux occurs when the convectic transport of retained solute toward the membrane is balanced by the backtransport of retained solute from the membrane, according to the simple film theory of concentration polarization.

C. Design Equations

A particle on one side may eventually travel to the other side of the diffusion volume as a result of thermal motion, although interactions with the matter in the diffusion volume may interfere. We assume that the probability for this transfer is the same for movement in both directions across the element. Therefore, if more particles are on one side than on the other, there will be a net flux toward the dificient side. Since all particles have equal probability to travel across the elememt, the flux in a given direction will be proportional to the concentration at the side where the particles leave. That is,[13]

$$\frac{1}{A}\frac{dN}{dt} = Pn - P(n + dn) = -Pdn$$

is the mole flux moving in the positive direction, where P is a quantity that defines the probability that a particle makes it across the diffusion volume element. The larger the thickness, dx, of the diffusion volume the smaller P should be, because there would be more interfering material.

$$P = \frac{D}{dx}$$

Where the proportionality constant, D, is called the diffusion coefficient. Then

$$\frac{1}{A}\frac{dN}{dt} = -D\frac{dn}{dx}$$

If the particles are charged the flux corresponds to a diffusion current density

$$j_{diff} = FZ \frac{1}{A} \frac{dN}{dt}$$

so,

$$j_{diff} = -FZD \frac{dN}{dx}$$

Since we are now dealing with ions, any electric field present in the volume element will also influence the movement of the charged particles. To obtain the mole flux due to the electric field consider a volume element of material across which there is no concentration gradient. The ion will reach some terminal or drift velocity, V, in the element because of their interactions with the material. The current density due to the field will then be

$$j_{field} = Fznv$$

Now define mobility per unit valence, U:

$ZU = \frac{V}{E}$, where E is the electric field.

Thus, the field current density is

$$j_{field} = FZ^2unE$$

(We always assume that U is constant across the membrane). A given concentration gradient moves particles in a given direction, independent of their charge: a different sign for charge then gives a different sign for current density.

In general a concentration gradient and an electric field are both present. Then the total current density is

$$j = FZ^2unE - FZD\frac{dN}{dx}$$

According to the Helmholtz-Smoluchowski equation, electrophoretic velocity is directly proportional to the electrical field strength (Volts per meters)[2],[14]

$$U_E = \frac{\zeta DE}{6\pi\mu}\left(\frac{1}{300}\right)^2 = kE$$

The one-dimensional, steady-state, solute balance in the fluid boundary layer above a membrane which completely retains the solute is

$$JC = -D\frac{dc}{dx} + kEC$$

Where D is the solute diffusivity. Integrating across the boundary layer gives

$$J = K \ln \frac{C_i}{C_o} + kE$$

Where C_i and C_0 are the solute concentrations in the so called inside solution and outside solution respectively, and K is an average mass transfer coefficient.[14]

III. EXPERIMENTAL AND APPARATUS

PART 1. Separation of Hemoglobin and Albumin

A separation cell unit is set up by arranging the two electrodes connected with a DC-power supply. The cell unit consists of four compartments separated by one electric membrane and two ion-exchange membranes as shown schematically in figure 1. The cell unit is set in a cooling bath kept at 10°C.

In these experiments, the fluid mixture is made up of two proteins of hemoglobin and albumin. This first part of the separation process offers us some hints to further the possibility of concentration of proteins.

Samples were analyzed by a spectrophotometer (The Bausch & Lomb, Spectronic®710) for hemoglobin and albumin content. This was obtained by means of two magnetic stirrers (stirring speed of 50 rpm). The peak adsorption for hemoglobin in water and for albumin in water occurs at 403 nm with visible light source and 280 nm with the hydrogen light source. The peak adsorption at 280 nm for the ternary system was due to hemoglobin and albumin and the albumin adsorption was obtained by substracting the hemoglobin adsorption from the total adsorption. On the other hand, the hemoglobin concentration from the transmittance value at 403 nm was

obtained from the calibration curve.

Albumin may be measured directly, however, by selective and reversible complexation to a dye such as methyl orange, bromcresol green, or HABA (2 - (4' - hydrozyazobenzene) - benzoic acid). The Bio-rad protein assay is a dye-binding assay on the differential color change of a dye in response to various concentrations of protein.

Buffer used for this experiment is Tris, obtained as Trizma base from Sigma Chemical Co. Tris buffers were prepared of known concentration and adjusted to pH 5.5 with concentrated HCl solution. Deionized water was used in all solutions. The pH and conductivity of buffers were measured periodically and adjusted when pH changed by more than 0.5 pH units.

The buffer and protein solutions were cooled by a cooling bath as necessary to remove any heat generated during the process. The applied voltage during electromembrane separation was measured between the electrodes and across the retentate compartment.

The following variables are measured or controlled:

. D-C voltage and current supplied to each electrode
. pH value
. resistor connected to electromembrane

PART 2. Concentration of Protein

A concentration cell which is a little bit similar to that of the previous part is set up. A schematic diagram of the experimental appratus is shown in figure 2. It consisted mainly of a rectangular electrofilter module, feed tank, suck pump and a D.C. electric power supply unit. This module was made of proper stainless steel membrane which carried negative current. The colloidal solution was fed into the concentration compartment at a constant flow rate, regulated with a control valve. The filtrate was sucked out by a pump so that the liquid levels in every compartment were kept constant. All experiments were carried out at 10^{o}C. The conditions of colloidal particle concentration, feed rate, resistor to the electromembrane and electric field strength were varied.

The concentration of protein in the colloidal solution was measured by a spectrophotometer (The Bausch & Lomb, Spectronic®710)at a wavelength of 280 nm. The calibration for the relation between absorbancy, A_s, and gelatin concentration, C_b, was experimentally obtained as follows: $C_b = 0.001202A_s$ in the range of $0.02\% \leq C_b \leq 0.15\%$ (g/100c.c).

In this part, the effect of pH of colloidal solution,

the strength of electric field and the flow rate on the filtration efficiency of colloidal particles are investigated experimentally. The hydrogen exponent, pH, of the colloidal solution was controlled with a buffer solution of sodium phosphate. The isoelectric point of gelatin dispersed in water is about 4.7, and therefore the ζ-potential of the gelatin particle is negative if the pH of solution is larger than 4.7.

The following variables are measured or controlled:

. D-C voltage and current supplied to each electrode
. pH value
. resistor connected to electromembrane
. initial concentration
. flow rate

IV. RESULTS AND DISCUSSION

PART I

A. Effect of Electric Field Strength on Separation

The effect of electric field strength, E, on the reflection coefficient of colloidal particles, defined as $R = 1 - (C/C_0)$, was experimentally investigated. Figure 4 shows the relation between R and E with resistor as a parameter under the condition of constant initial concentration and pH values. From the result shown in figure 4, it is confirmed that R increases with increasing E. This will be reasonably explained by the Helmholtz-Smoluchowski equation:

$$U_E = \frac{\zeta DE}{6\pi\mu}\left(\frac{1}{300}\right)^2 = kE$$

Electrophoretic velocity is directly proportional to the electrical field strength (volts per meters).

The model showed that film resistance could be controlled in part by the applied field. One can observe that an applied electric field is able to control the build up of retained macrosolutes at a membrane surface, and thereby increase flux.

At a given applied electric resistance, the increase in

separation is more rapid for an electric field strength of 680 V/m versus 440 V/m, 280 V/m as seen in figure 5-7 which show the relations between R and time(T).

B. Effect of Initial Concentration of Colloidal Solution on Separation

Figure 8 shows how the increasing rate in separation is affected by the initial concentration of hemoglobin. As a constant electric field strength and resistance applied to the electromembrane, an initial concentration of the more dilute 0.02% hemoglobin increases more rapidly than those particle solutions of 0.04% and 0.10%.

C. Separation of Hemoglobin and Albumin Particles

As the pH value (5.5) is above the isoelectric point of bovine albumin (I_A = 4.8), the colloids in albumin solution with positive charge do not migrate toward the anode. In other words, they will be not only be attracted by the cathode but also will be stopped migrating toward contact with the cathode by the neutral ion-exchange membrane. Figure 9 shows the results. This result enhances the possibility for separation of hemoglobin and albubin.

Figure 10 shows the contribution of the electromembrane which successfully takes the place of the selectivity of traditional ion-exchange membranes. Curve 4 ascends sluggishly while curve 1,2 ascend more sharply.

The electromembrane which carries negative current with the smaller electric resistor (500 ohm) contributes more than those with large electric resistors (1000 ohm, 2700 ohm) in the process of separation. The reason for connecting an appropriate electric resistor to electromembrane is to keep the electric current through the electromembrane much smaller than which through anode.

The electric current through the electromembrane is great enough to prevent the gel formation while small enough not to stop the negatively charged hemoglobin colloids migrating toward the anode. This result is shown in figure 11.

PART II

A. Effect of Electric Field Strength on Filtration

The changes in concentration with time for an initial 2500 c.c, 0.2 wt% solution of protein are shown in figure 12 for negative electromembrane filtration and non-charged electromembrane (membrane carries no current) filtration. As the applied electric field strength E is increased up to about 700 V/m, the time required to increased the concentration by a given factor decreases drastically. This is due to a higher flux for the negatively charged electromembrane during the concentration operation as seen in figure 12.

The reason for a larger filtration flux with the negative electromembrane than with the non-charged electromembrane in the concentration of proteins is considered as to be follows:
Dispersed negative particles of protein are influenced by two driving forces. One caused by the electrical field will make particles move in the reverse direction to the membrane. Another one caused by continuously input/output flow (drag force) tends to carry dispersed particles in the filtrated solution moving toward the electromembrane. The electromembrane carrying a negative current is to prevent the possibility of deposition of protein particles on the membrane caused by convectional flow.

B. Effect of pH of Colloidal Solution on Filtration Flux

The effect of pH of colloidal solution on filtration flux was examined under conditions where U and C_o are constant. Figure 13 shows the plot of total filtration flux vs pH of the colloidal solution in both the non-charged electromembrane (filtrating membrane carries no current) filtration and the negatively charged electromembrane filtration operated as a batch process. In the range of pH smaller than the isoelectric point, the filtration flux with non-charged electromembrane is larger than that with the negatively charged electromembrane. At the isoelectric point, the filtration flux is not enhanced electrically because the electrophoretic velocity of dispersed particles in the reverse direction to the membrane is smallest. Consequently, the filtration flux with negatively charged the electromembrane becomes smaller than that with the non-charged electromembrane. At the range of pH smaller than the isoelectric point, the positively charged protein-particles migrate to the membrane owing to electrophoresis and so the thickness of the gel layer formed on the membrane in the negatively charged electromembrane is larger than that in non-charged electromembrane.

C. Effect of Flow Rate on Filtration

Figure 14 shows two phenomena under the same conditions but different flow rates and employed electric field strength, high potency of filtration favors low flow rate but high voltage. This comes to be opposite to the tranditional ultrafiltration process which needs larger shear force caused near the membrane so that the gel layer formed on the membrane and the concentration polarization layer near the membrane become thin. Higher flow rate causes a stronger drag force which tends to carry dispersed particles in filtrated solution passing through charged electromembrane.

D. Effect of Electric Resistance on Filtration

Figure 15 shows that the charged electromembrane contributes more in filtration efficiency connected with a 500 ohm resistor than connected with 1000 ohm or 2700 ohm resistor. It seems that the charged electromembrane contributes more in eliminating the formation of the gel layer with a larger electric current density than with a smaller one.

V. CONCLUSION

The negatively charged electromembrane can hopefully take the place of the selectivity of traditional ion-exchange membranes. It becomes experimentally very effective in increasing the separation potency and filtration flux because the fouling of the membrane is decreased by the expulsive ability caused by the chargeable electromembrane which generally carries a negative current.

The experiment gave the following conclusions:

1) The separation coefficient and filtration efficiency of colloidal particles are affected by electric field strength, concentration of dispersed particles, pH of dispersion medium as well as charge on the electromembrane in the CEM process.
2) The effect of input/output flow rates in the electromembrane filtration process is remarkable. The operating conditions should include economic factors.
3) A layer electric current density over the electromembrane contributes more in increasing the filtration efficiency than a smaller electric current density does.
4) Negative charge enhances filtration flux in the CEM process, because gel layer formation on the membrane is prevented by electrophoresis.

A list for comparison of the charged electromembrane with some other existing membranes is shown in Table 9.

The conditions of colloidal particle concentration, flow rate, electric field strength and pH value were varied to identify the optimal condition for performance. While the general processes involved in the electromembrane permeation are now understood, it is not yet possible to write and solve the appropriate equations which will enable us to predict permeation rate without significant experimentation. The vast amount of data developed on a number of specific systems must be expanded to the almost infinite combinations of suspended particles, penetrant, and conditions in order to develop general theories and equations. As this is accomplished, the great abilities of electromembranes to make biological and industrial separations may be more widely utilized.

VI. GLOSSARY

ζ	:	electrokinetic potential of protein particle [V]
I_H	:	isoelectric point of haemoglobin [-]
I_A	:	isoelectric point of albumin [-]
R	:	reflection coefficient of colloidal particle [-]
η	:	filtration efficiency of colloidal particle [-]
δ	:	thickness of idealized boundary layer [m]
P	:	probability of a particle acrossing the diffusion volume element [-]
j_{diff}	:	flux corresponding to a diffusion current density
j_{field}	:	flux corresponding to a electric field [current/area]
E	:	ELECTRIC FIELD STRENGTH [V/m]
r	:	resistor series to electro membrane [Ω]
As	:	absorbancy measured by spectrophotometer [-]
C_o	:	initial concentration of bulk solution [wt%]
C	:	concentration of colloidal solution [wt%]
D	:	solute diffusivity [$(\text{Length})^2(\text{time})^{-1}$]
K	:	average mass transfer coefficient [m/s]
U	:	mobility per unit valence [$(\text{charge})(\text{time})(\text{mass})^{-1}$]

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TABLE 1

Concentration of Hemoglobin in CEM Process

Using 1000 ohm Resistor

CONDITION : Initial concentration 0.02%

pH 5.5

Resistor 1000 ohm

RUN 1 : 280 V/m

RUN 2 : 440 V/m

RUN 3 : 680 V/m

TIME	CONCENTRATION (wt%)		
(MIN.)	RUN 1	RUN 2	RUN 3
10	0.0189	0.0180	0.0180
20	0.0178	0.0163	0.0161
30	0.0169	0.0147	0.0140
40	0.0160	0.0136	0.0118
50	O.0151	O.0118	0.0102
60	0.0145	0.0109	0.0091
70	0.0129	0.0101	0.0081
80	0.0116	0.0099	0.0084
90	0.0116	0.0091	0.0072
100	0.0118	0.0088	0.0068
110	0.0108	0.0074	0.0068
120	0.0104	0.0071	0.0063

TABLE 2

Concentration of Hemoglobin in CEM Process

Using 500 ohm Resistor

CONDITION : Initial Concentration 0.02

pH 5.5

Resistor 500 ohm

RUN 4 : 280 V/m

RUN 5 : 440 V/m

RUN 6 : 680 V/m

TIME	CONCENTRATION (wt%)		
(MIN.)	RUN 4	RUN 5	RUN 6
10	0.0167	0.0151	0.0150
20	0.0136	0.0114	0.0110
30	0.0126	0.0109	0.0092
40	0.0121	0.0104	0.0085
50	0.0109	0.0088	0.0071
60	0.0095	0.0070	0.0059
70	0.0086	0.0074	0.0050
80	0.0074	0.0079	0.0046
90	0.0074	0.0063	0.0047
100	0.0072	0.0056	0.0049

TABLE 3

Concentration of Hemoglobin in CEM Process

Using 2700 ohm Resistor

CONDITION : Initial concentration 0.02%

pH 5.5

Resistor 2700 ohm

RUN 7 : 280 V/m

RUN 8 : 440 V/m

RUN 9 : 680 V/m

TIME	CONCENTRATION (wt%)		
(MIN.)	RUN 7	RUN 8	RUN 9
10	0.0192	0.0189	0.0189
20	0.0185	0.0183	0.0172
30	0.0172	0.0177	0.0164
40	0.0166	0.0164	0.0155
50	0.0157	0.0149	0.0138
60	0.0150	0.0144	0.0126
70	0.0144	0.0131	0.0119
80	0.0136	0.0116	0.0113
90	0.0135	0.0117	0.0121
100	0.0134	0.0115	0.0111

TABLE 4

Concentration of Hemoglobin in CEM Process with Different Initial Concentration

CONDITION : E 680 V/m

pH 5.5

Resistor 500 ohm

RUN 10 : initial concentration 0.04%

RUN 11 : initial concentration 0.01%

TIME	CONCENTRATION (wt%)	
(MIN.)	RUN 10	RUN 11
20	0.0244	0.0800
40	0.0228	0.0689
60	0.0160	0.0543
80	0.0134	0.0490
100	0.0098	0.0421

TABLE 5

Absorbance Value for Albumin-Buffer System in CEM Process at pH 5.5

CONDITION : Initial concentration 0.02%
pH 5.5
E 680 V/m
RUN 12 : Resistor 500 ohm
RUN 13 : Resistor 1000 ohm

TIME	ABSORBANCE	
(MIN.)	RUN 12	RUN 13
10	0.632	0.659
20	0.631	0.657
30	0.629	0.583
40	0.631	0.678
50	0.628	0.676
60	0.651	0.678
70	0.651	0.671
80	0.655	0.669
90	0.657	0.670
100	0.648	0.670

TABLE 6

Concentration of Hemoglobin-Albumin-Buffer System in CEM Process with Different Values of Resistor

CONDITION : Initial concentration
0.02% hemoglobin, 0.02% albumin
pH 5.5
E 440 V/m

RUN 14 : Resistor 500 ohm

RUN 15 : Resistor 1000 ohm

RUN 16 : Resistor 2700 ohm

RUN 17 : neutral electromembrane

TIME	CONC. OF HAEMOGLOBIN (WT%)			
(MIN.)	RUN 14	RUN 15	RUN 16	RUN 17
20	0.01950	0.01908	0.01965	0.01964
40	0.01896	0.02082	0.01952	0.01908
60	0.01872	0.01748	0.02284	0.01900
80	0.01636	0.01720	0.01897	0.01896
100	0.01500	0.01692	0.01892	0.01808
120	0.01324	0.01400	0.01752	0.01768
140	0.01352	0.01268	0.01704	0.01644
160	0.01000	0.01168	0.01524	0.01684
180	0.01308	0.00924	0.01568	0.01500
200	0.00700	0.00920	0.01508	0.01480

TABLE 7

Concentration of Milk Protein in CEM Process with Different Electric Field Strength

CONDITION : Initial concentration 0.2%
pH 7.4
Flow Rate 10 c.c/min
Resistor 500 ohm

RUN 18 : E 770 V/m

RUN 19 : E 440 V/m

RUN 20 : neutral electromembrane

TIME	CONCENTRATION (wt%)		
(MIN.)	RUN 18	RUN 19	RUN 20
20	0.2309	0.2181	0.2980
40	0.2931	0.2183	0.5162
80	0.4112	0.3082	0.5416
150	0.5627	0.4627	0.6342
200	0.7860	0.4621	0.6349
250	1.0727	0.6482	0.6489

TABLE 8

Comparison of Final Concentration of Protein in CEM Process & in Non-charged Electromembrane Process at Various of pH Values

CONDITION : Initial concentration 0.5%
E 770 V/m
Flow Rate 10 c.c/min
Run Time 60 min.
RUN 21 : Resistor 500 ohm
RUN 22 : neutral electromembrane

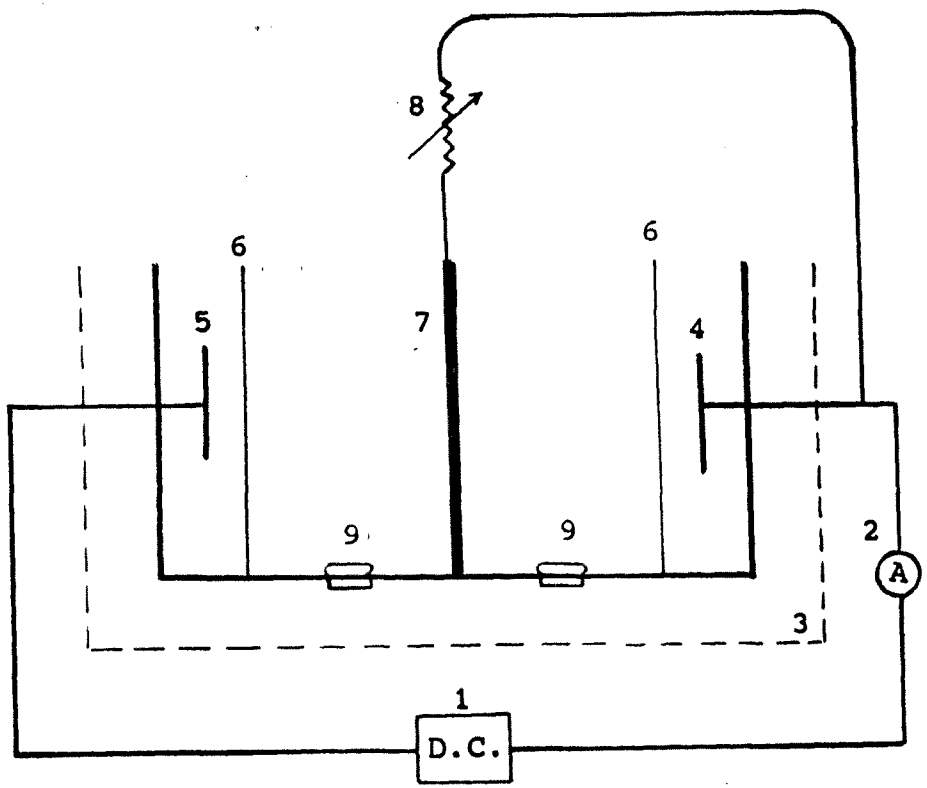
	FINAL CONC.(wt%)	
PH	RUN 21	RUN 22
3	0.615	0.625
4	0.600	0.625
5	0.625	0.661
6	0.712	0.677
7	0.856	0.712
8	0.992	0.795

TABLE 9

Comparison of Charged Electromembrane Process

with some other Existing Membrane Processes

Process / Item	Ultrafiltration (UF)	Reverse Osmosis (RO)	Electrodialysis (ED)	Charged Electro-membrane (CEM)
Molecular Size	0.001 - 0.02 μ	0.0001 - 0.001 μ	0.0001 - 0.001 μ	0.0001 - 0.02 μ
Driving Force	Machanical	Osmosis	Electroosmosis	Electro-phoresis
Pressure Applied	0 - 10 atm	60 - 100 atm	0 - 10 atm	0 atm
Separation Principle	Pore Size	Molecular Permeability & Selectivity	Ion Exchange	Electric Charges
Characteri-stics	Pure water Transfer	Pure water Transfer	Iones Transfer	Charged Transfer
Comment	1. Cake Formation 2. Coarse Particle Only 3. Relatively High Membrane Cost	1. High Pressure 2. Salt and Low Molecules 3. High Membrane Cost	1. Fouling 2. Salt Only 3. High Membrane Cost	1. No Cake & Fouling 2. High to Low Charged Colloids 3. Low Membrane Cost



1. Regulated D.C. power supply
2. Ampere meter
3. Cooling bath
4. Cathode
5. Anode
6. Ion-exchange membrane
7. Electromembrane
8. Adjustable resistance
9. Magnetic stirrer

Figure 1 Experimental set-up for separation of hemoglobin and albumin

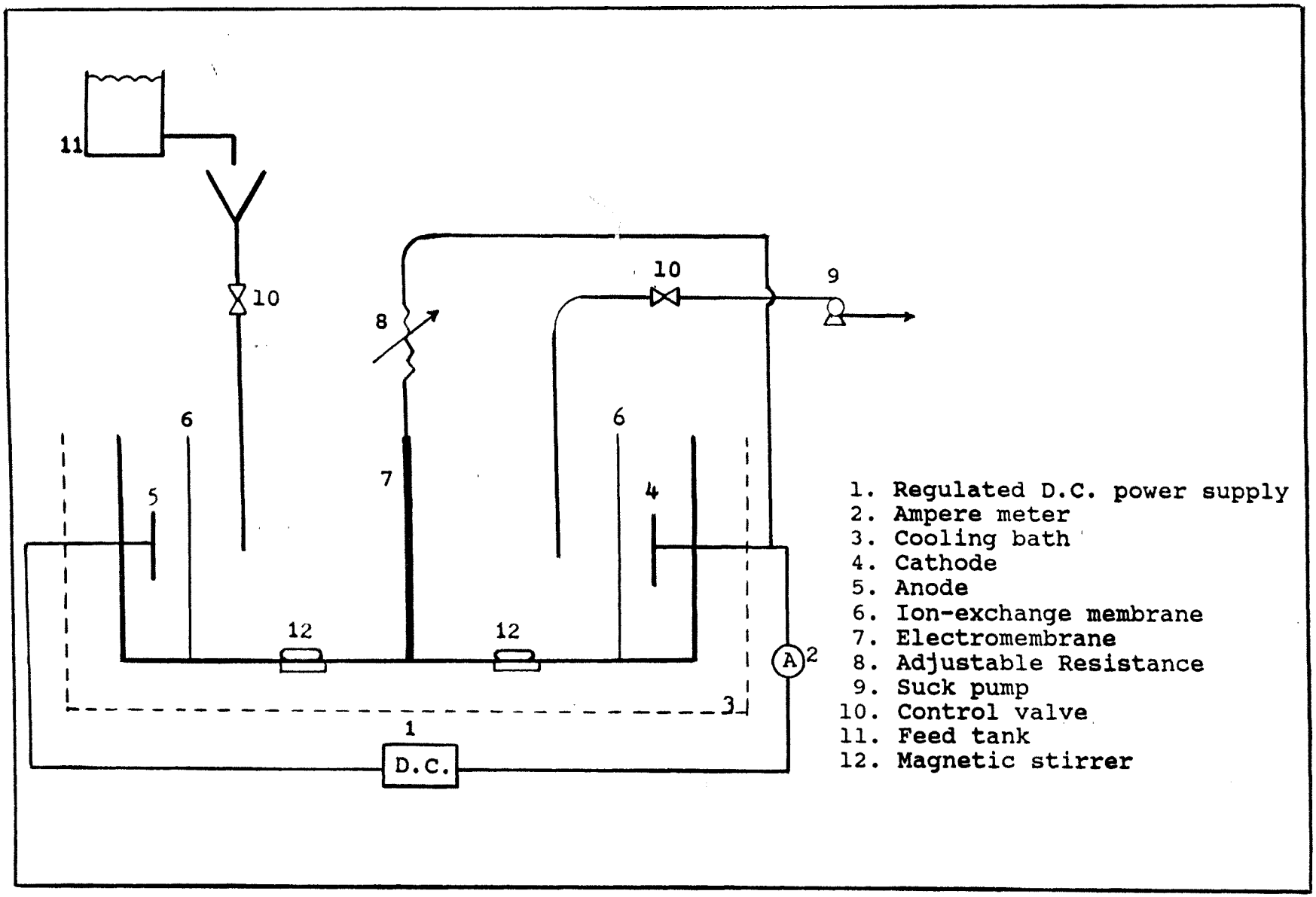


Figure 2 Experimental set-up for concentration of protein

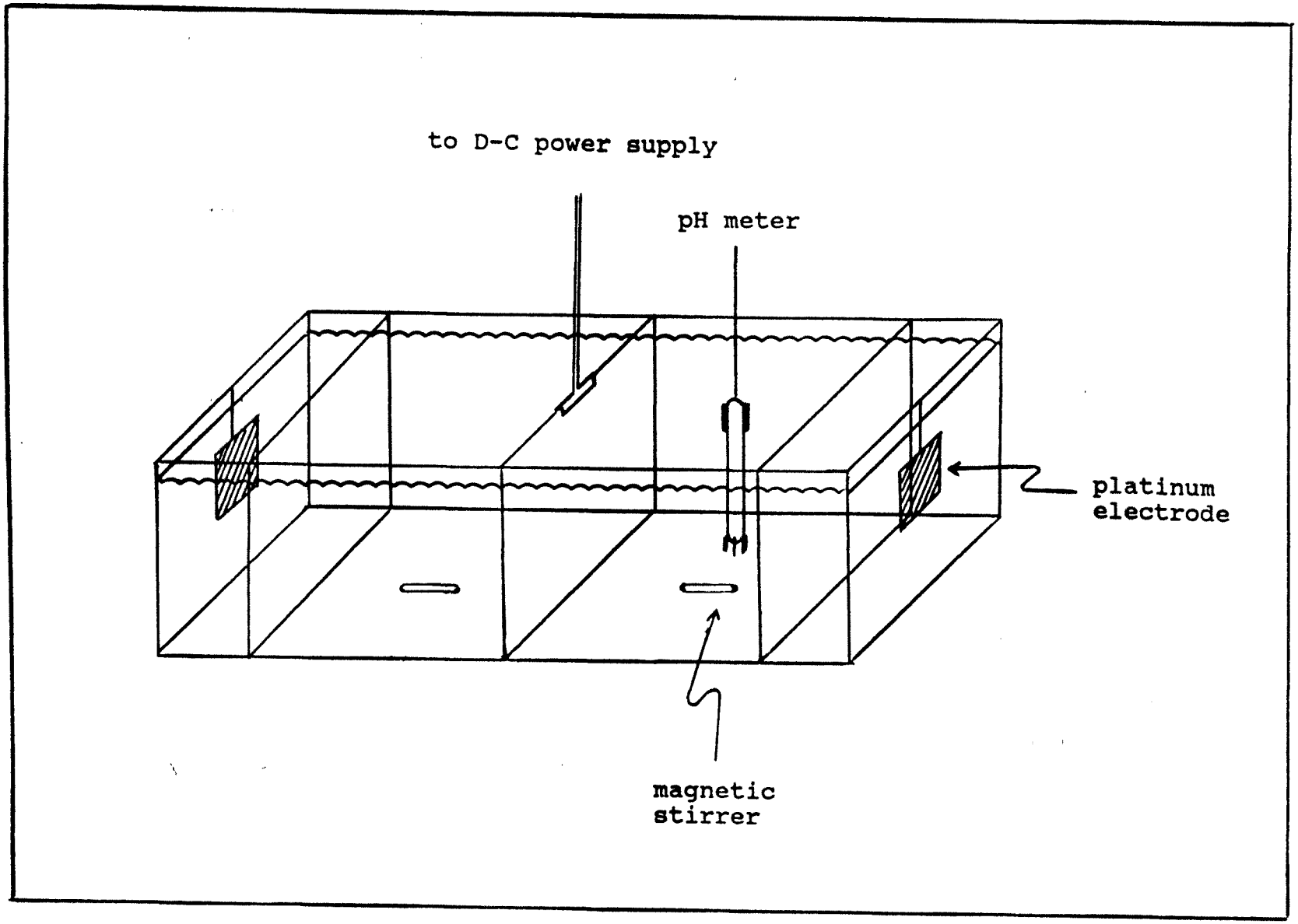


Figure 3 Schematic of transport cell for concentration of protein

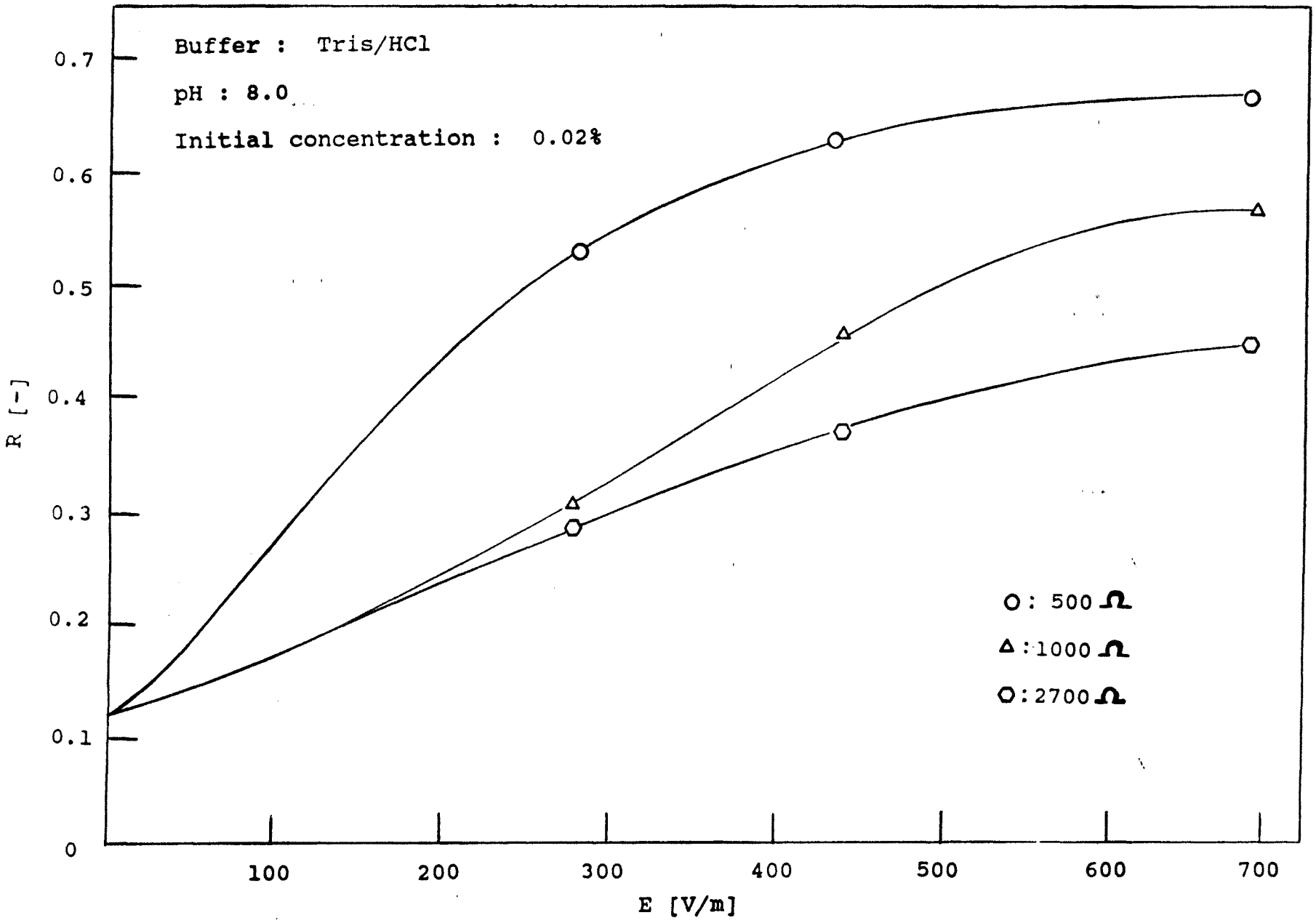


Figure 4 Effect of electric field strength on reflection coefficient for hemoglobin-buffer system with resistor as a parameter

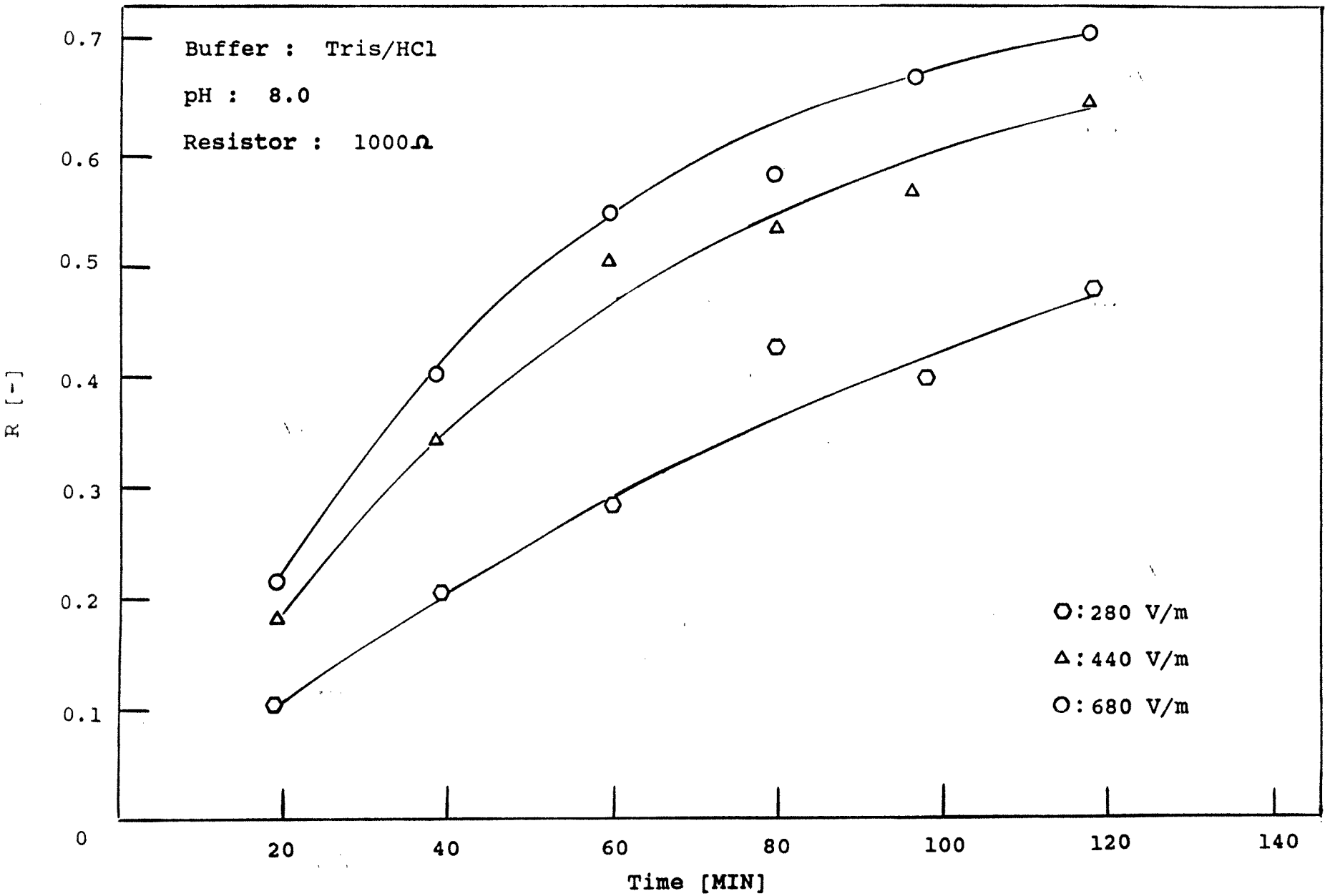


Figure 5 Variation of reflection coefficient for hemoglobin-buffer system having a 1000 ohm resistor with electric field strength as a parameter

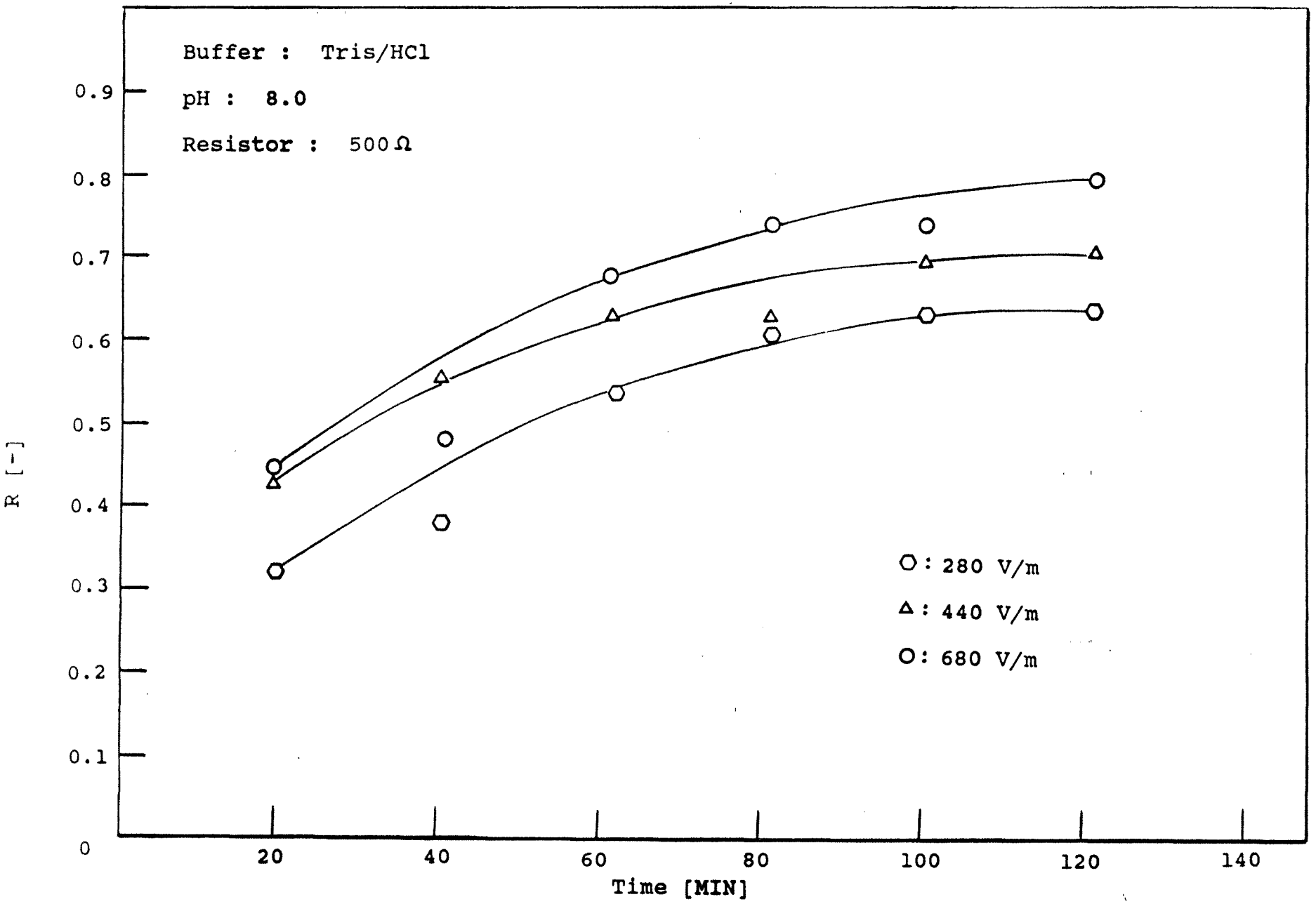


Figure 6 Variation of reflection coefficient with time for hemoglobin-buffer system having a 500 ohm resistor with electric field strength as a parameter

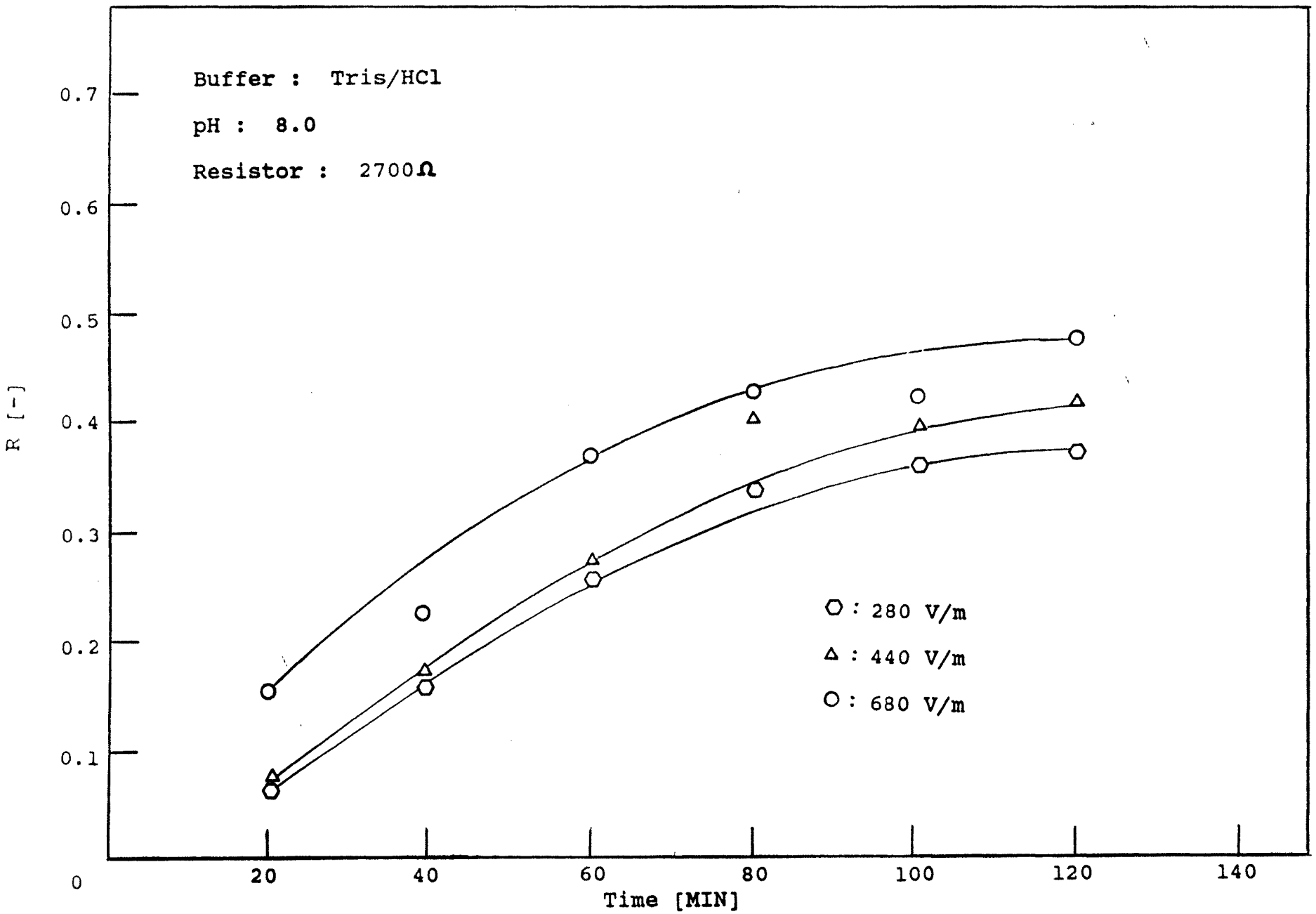


Figure 7 Variation of reflection coefficient with time for hemoglobin-buffer system having a 2700 ohm resistor with electric field strength as a parameter

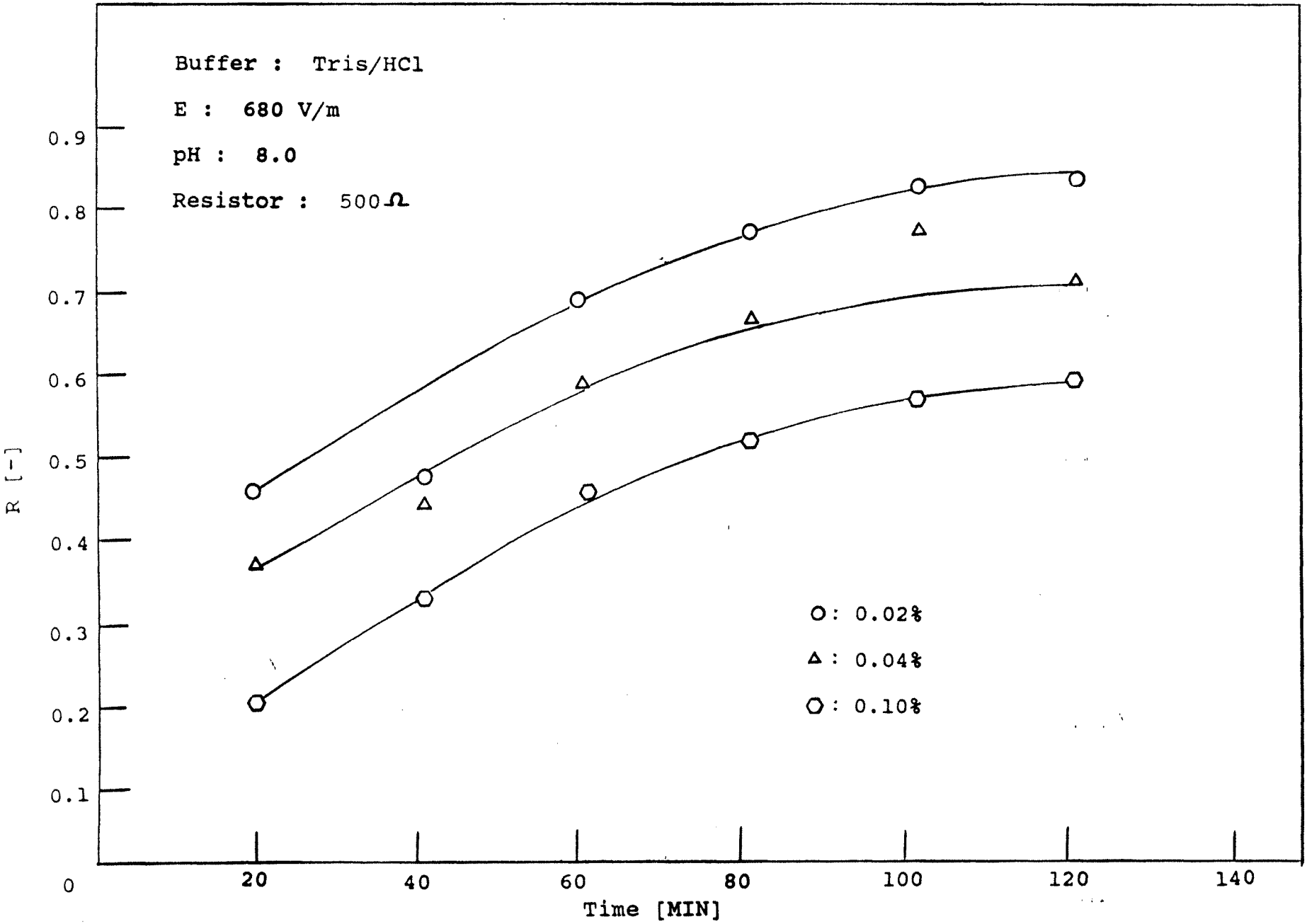


Figure 8 Variation of reflection coefficient with time for hemoglobin-buffer system with initial concentration as a parameter

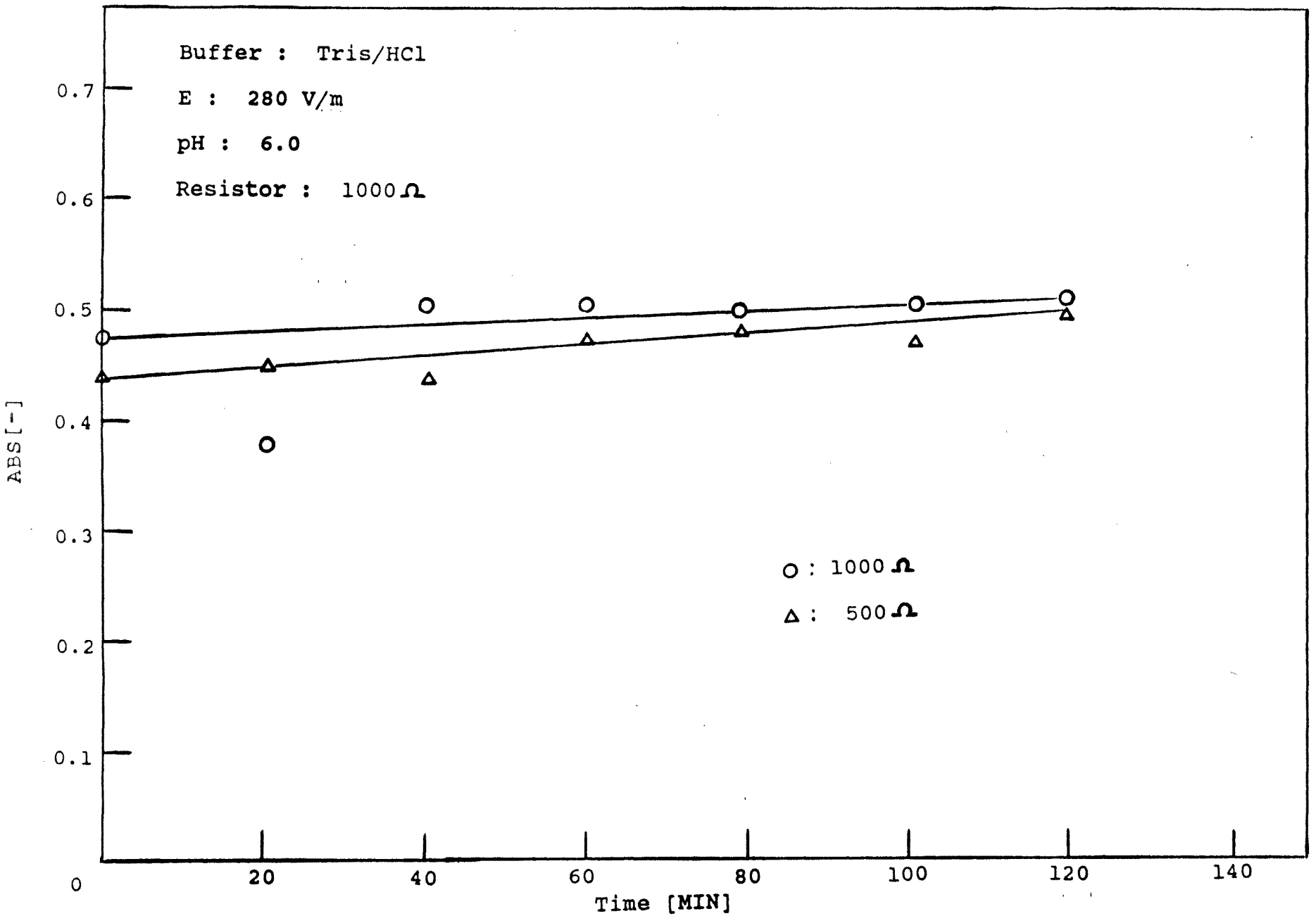


Figure 9 Variation of reflection coefficient with time for albumin-buffer system with resistor as a parameter

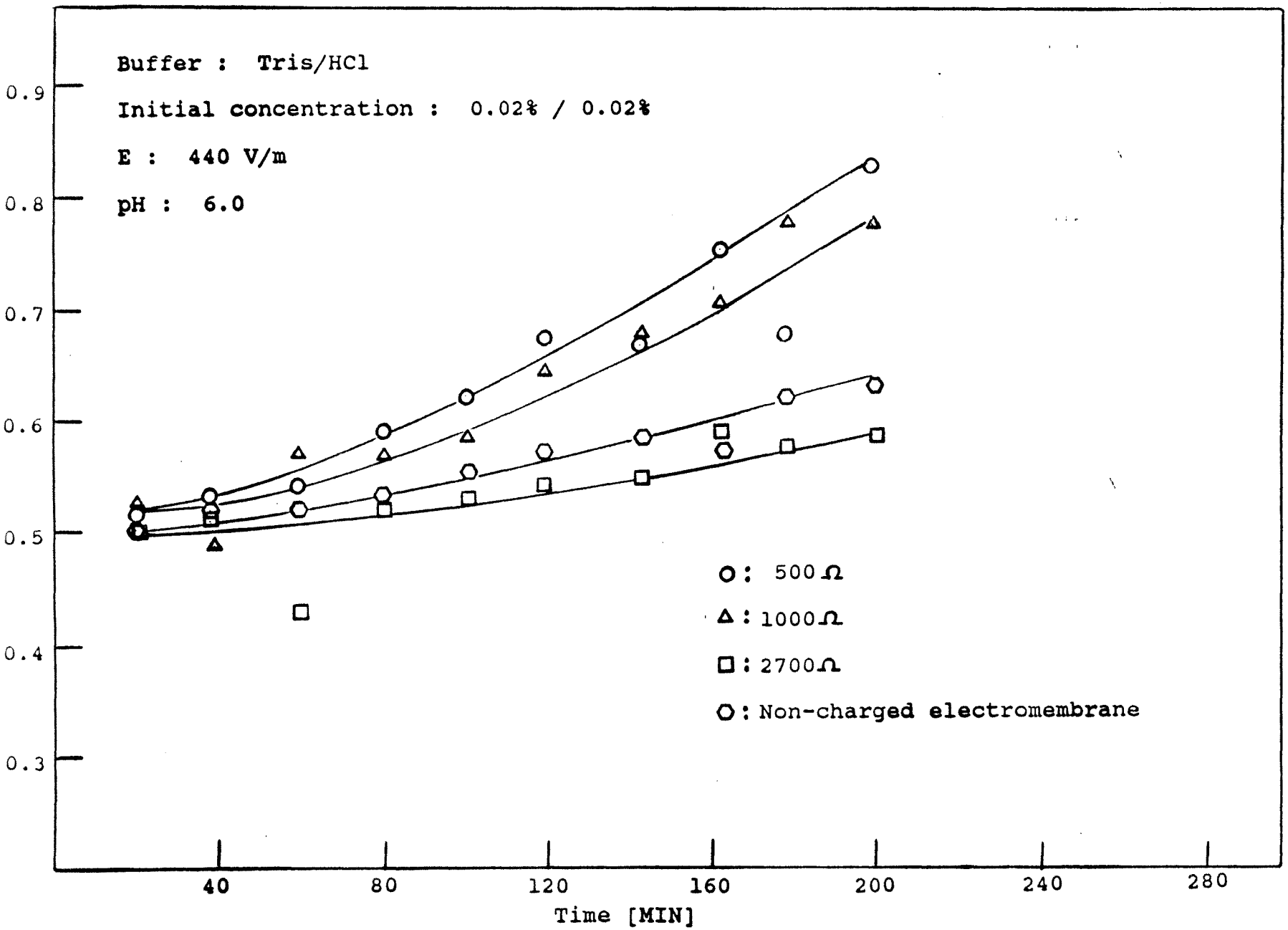


Figure 10 Variation of reflection coefficient with time for hemoglobin-albumin-buffer system with resistor as a parameter

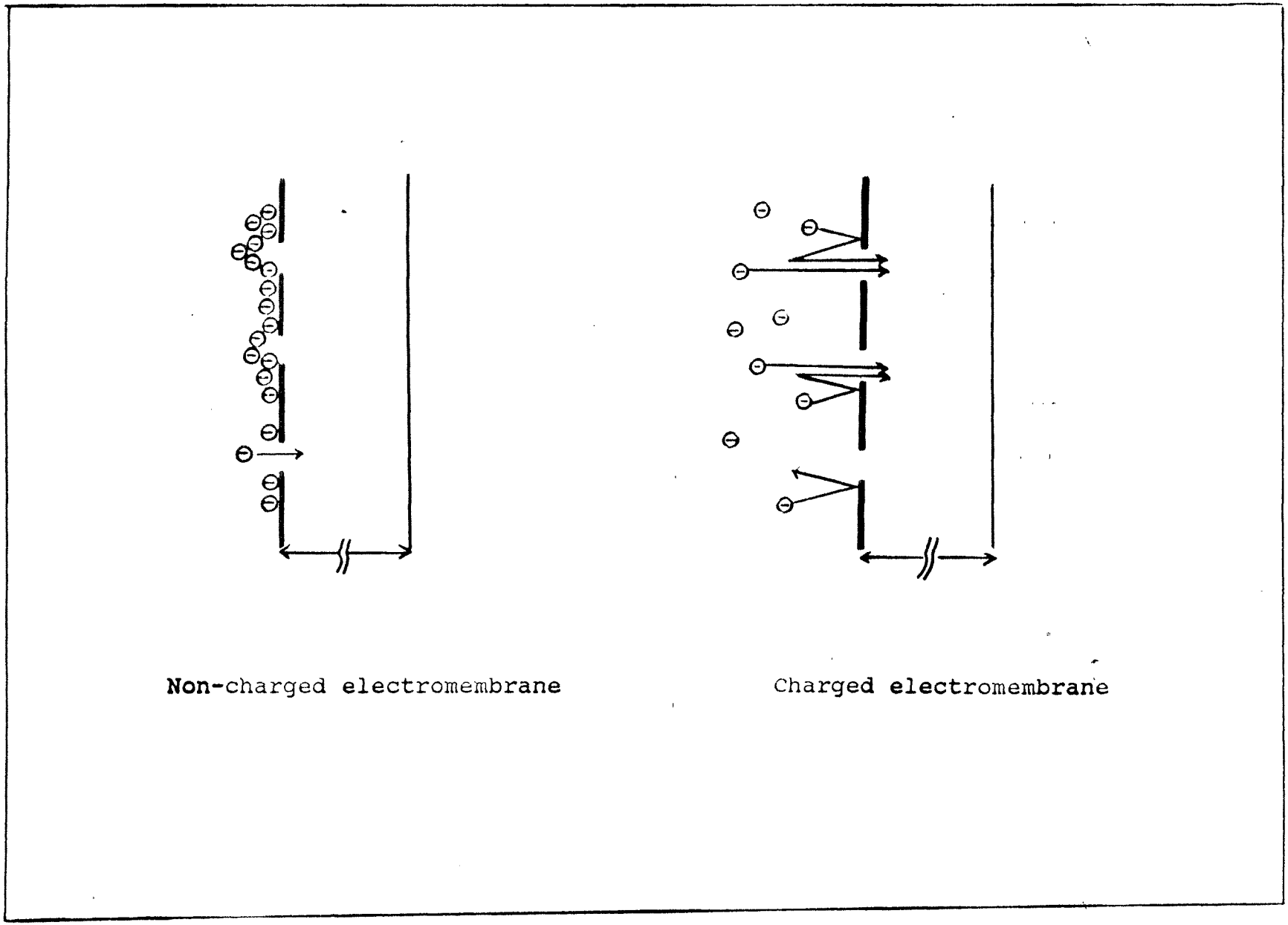


Figure 11 Comparison of characters by scheme for separation by non-charged electromembrane and negatively charged electro-membrane

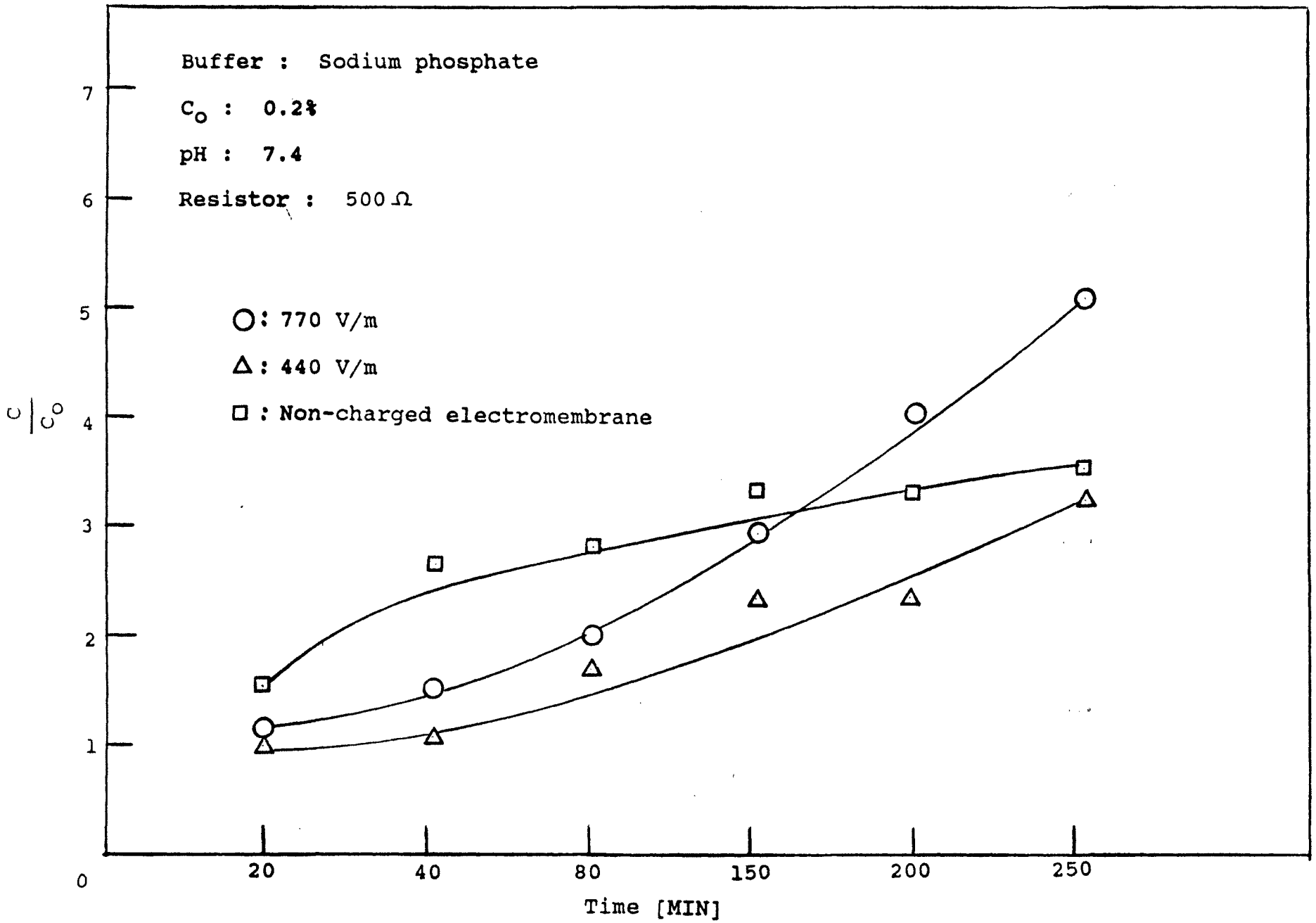


Figure 12 Variation of concentration ratio of milk protein with time in CEM process having different electric field strength

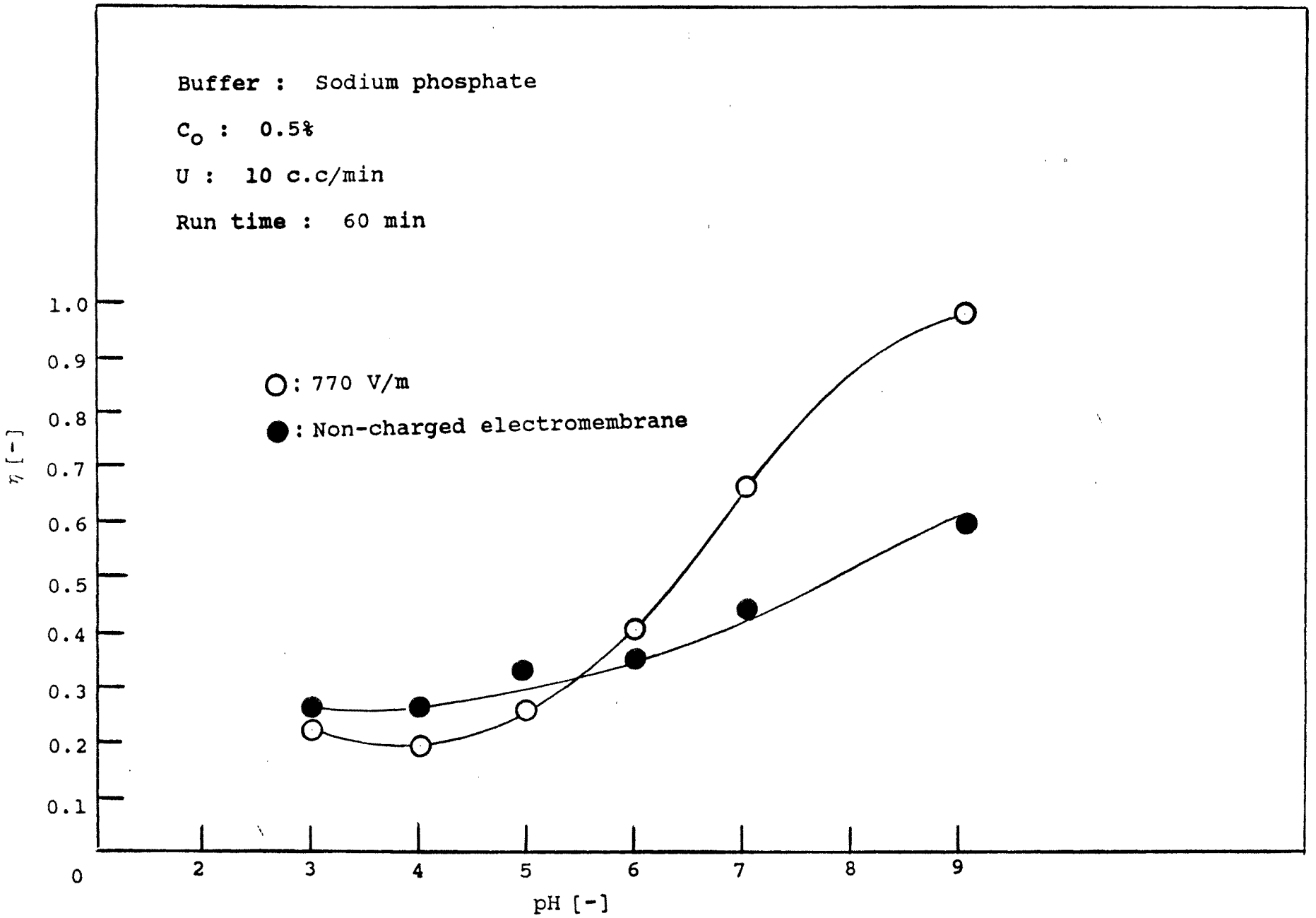


Figure 13 Effect of pH value on filtration efficiency for protein-buffer system

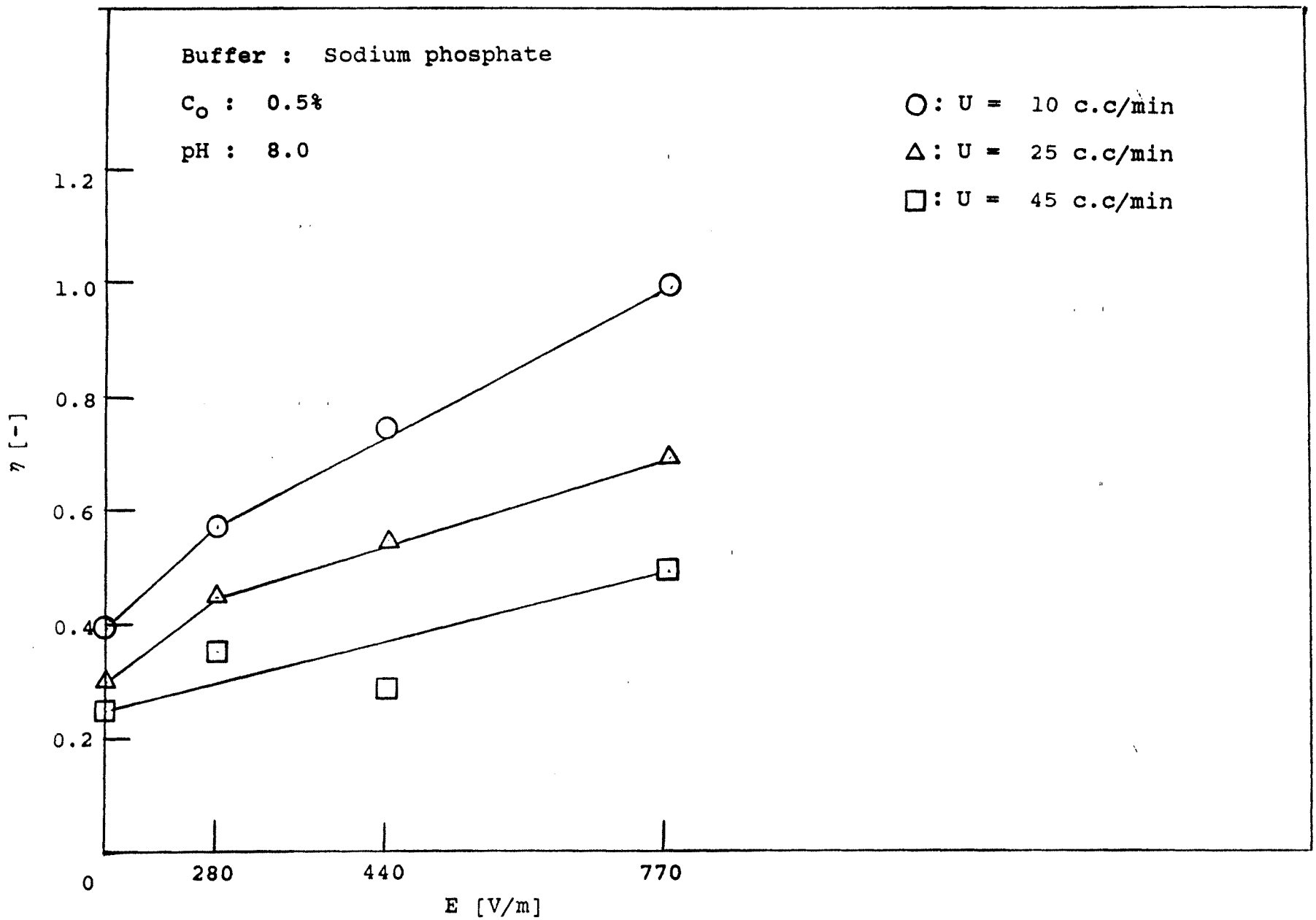


Figure 14 Effect of electric strength field on filtration efficiency for protein-buffer system with flow rate as a parameter

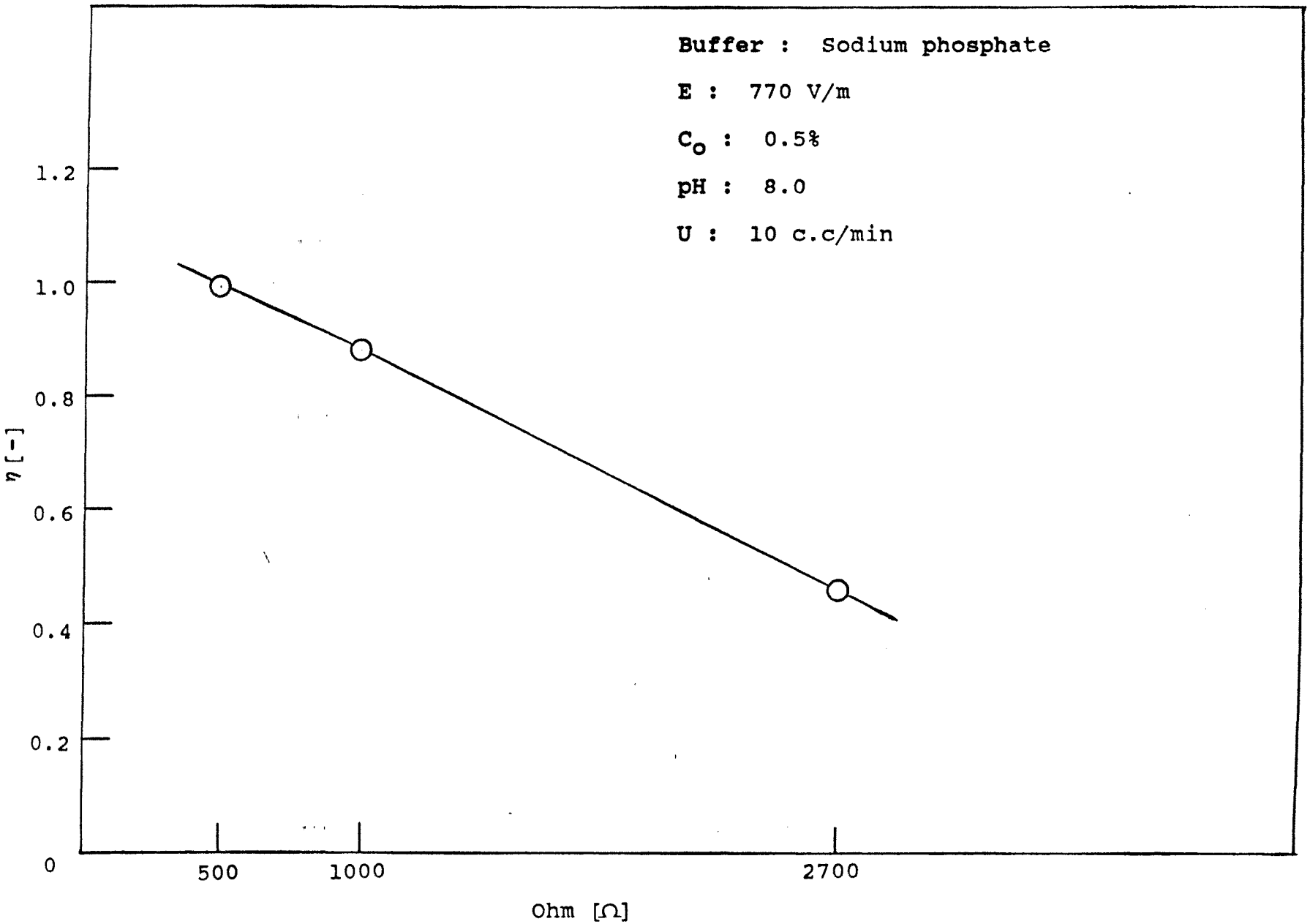


Figure 15 Effect of resistor connected to electromembrane on filtration efficiency for protein-buffer system