# Applicability of Charged Membranes in Purification of Polluted Water

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## Abstract

Water pollution occurs when harmful substances often chemicals or microorganism contaminate a stream, rivers, lakes, ocean and other water bodies. Around the world about 80 percent of wastewater is released in the environment without proper treatment. This impacts our ecosystems and jeopardize public health. With water demand growing in every sector from agriculture to industry, it is judicious to use wastewater after proper treatment to tide over water crisis.

To understand the mechanism of ionic diffusion through membranes the membrane resistance, capacitance and impedance of parchment supported charged membranes have been analyzed. The electrical resistance($R_x$), and capacitance($C_x$) developed across these membranes have been measured at different electrolyte concentrations and oscillator frequencies, were found to be dependent on the concentration of bathing electrolyte and applied oscillator frequencies. The results are interpreted in terms of changes produced in the electrical double layer at the membrane/electrolyte interface. The values of membrane resistance, capacitance and impedance have been computed by considering different equivalent electrical circuit models. The electrical impedance spectra have been found to deviate from the theoretical predictions at low frequencies due to non homogeneity and roughness of the membrane surface. The parchment supported charged membranes may be useful at higher oscillator frequencies.

**Keywords:** Ionic Diffusion, Charged Membranes, Oscillator Frequency, Membrane Resistance, Membrane Capacitance.

## Introduction

The world is facing a global water crisis. As water demand is increasing across the globe, the quantity of wastewater produced as a result of various human activities is also growing, the World Water Development Report 2017 points out. However, it is in our interest, to make use of this water rather than lose it. There are many causes driving this crisis, but it is clear that freshwater and coastal ecosystem across the globe, upon which humanity has depended for millennia, are increasingly threatened. The waste water management and recycling may be a solution of this crisis. Inorganic precipitate membranes have attracted a lot of attention in the past decade because of their wide applications in separation processes. Membrane separation is a "rate governed process" based on preferential permeation of the desired species through semi permeable membrane. Membrane technologies are the most common technology of desalination and water recycling [1]. In order to understand the mechanism of transport through inorganic precipitate membranes a number studies have been made by different workers [2-5] and charged membranes are one of the most advanced separation membranes, which have been widely used in various industries [6,7].

A flat electrical double layer is created by a charged plane in contact with an electrolyte solution composed of solvent molecules, counter-ions and co-ions. Impedance measurements provide a powerful diagnostic tool for the analysis of many electro-chemical system [8,9]. A common interpretation has been that the resistance term is connected with the ion permeable structures and the capacitance term with the ion impermeable parts of the membranes. This paper describes the analysis of impedance of parchment supported inorganic precipitate cupric carbonate, nickel carbonate and complex cupric nickel carbonate model membranes under various conditions of bathing electrolyte concentrations and applied oscillator frequency in order to understand the mechanism of ionic transport through these membranes.

**Experimental**

Parchment supported membranes have been prepared by the method of interaction as in previous studies [10]. The membranes thus obtained were cut into a circular disc form of unit cross sectional area and sealed between the two half cells of an electrochemical cell and the half cells were filled with electrolyte solutions to equilibrate the membrane. The solutions were then replaced by purified mercury without removing the adhering surface liquids. Air bubbles, if any, on the membrane surface were removed by tilting the cell assembly. Mercury is likely to be oxidized to form mercuric oxide which would form films on membrane faces and causes irreversibility. Use of purified mercury has eliminated this problem and has given reproducible results.

A universal LCR bridge-921 has been used to measure the electrical resistance and capacitance of the membranes. The membranes were equilibrated with a lower concentration of potassium chloride initially, Then the higher concentration while measuring the resistance and capacitance of the membranes. The effect of oscillator frequency on the resistance and capacitance values have been observed for all the membranes. A platinum wire coated with platinum black dipped in Hg was used as the electrode. A thermostat was used to maintain the temperature at $25 \pm 0.1^{\circ}C$.

**Results and Discussion**

The electrical resistance (Rx) and capacitance (Cx) across parchment-supported Cupric carbonate, Nickel carbonate and complex Cupric-Nickel carbonate membranes equilibrated with

different concentrations of potassium chloride electrolyte solution at 1 KHz frequency have been measured. The Rx and Cx were also determined for all three membranes equilibrated with 0.1 M solution of same electrolyte by applying different ac frequencies. These values are given in Tables-1and2.

Structural simulation is a newer approach to modeling for more complex electrochemical systems [11]. Armstrong [12,13] has attempted to adopt some of the theoretical models for aqueous electrolyte system in order to obtain simple model for metal/super-ion conductor. By using equivalent electrical circuits for the membrane electrolyte system [14], the values of membrane resistance (Rm), capacitance (Cm), and impedance (Z) are calculated with the help of the following equation and given in Tables-3 and 4.

$$R_m = R_x \left[ (1 + X_x/R_x)^2 \right] \qquad (1)$$

$$X_x = \frac{1}{\omega C_x} \qquad (2)$$

and

$$C_m = \left( \frac{X_x}{R_x} \right) \left( \frac{1}{\omega R_m} \right) \qquad (3)$$

$$Z = \sqrt{R_x^2 + X_x^2} \qquad (4)$$

where $\omega = 2\pi f$ and f is the frequency used to measure $R_x$ and $C_x$.

**Table - 1**

**Electrical Resistance ($R_x$) And Capacitance ($C_x$) Observed Across Parchment Supported Cupric Carbonate, Nickel Carbonate And Complex Cupric-Nickel Carbonate Membranes Equilibrated With Different Concentrations Of Potassium Chloride At 1 KHz (Temp $25 \pm 0.1^{\circ}C$)**

| Electrolyte Conc. (M/L) | Cupric carbonate Membrane | | Nickel carbonate Membrane | | Complex Cupric-Nickel carbonate Membrane | |
|---|---|---|---|---|---|---|
| | $R_X$ (Ω) | $C_X$ (μF) | $R_X$ (Ω) | $C_X$ (μF) | $R_X$ (Ω) | $C_X$ (μF) |
| $1 \times 10^{-4}$ | $14.80 \times 10^2$ | $6.65 \times 10^{-2}$ | $13.50 \times 10^2$ | $6.80 \times 10^{-2}$ | $14.30 \times 10^2$ | $6.80 \times 10^{-2}$ |
| $1 \times 10^{-3}$ | $9.60 \times 10^2$ | $8.20 \times 10^{-2}$ | $8.80 \times 10^2$ | $8.40 \times 10^{-2}$ | $8.90 \times 10^2$ | $8.70 \times 10^{-2}$ |
| $1 \times 10^{-2}$ | $5.45 \times 10^2$ | $11.15 \times 10^{-2}$ | $5.00 \times 10^2$ | $11.50 \times 10^{-2}$ | $6.00 \times 10^2$ | $10.70 \times 10^{-2}$ |
| $1 \times 10^{-1}$ | $2.55 \times 10^2$ | $16.70 \times 10^{-2}$ | $3.10 \times 10^2$ | $14.60 \times 10^{-2}$ | $2.90 \times 10^2$ | $15.30 \times 10^{-2}$ |
| $1 \times 10^0$ | $1.40 \times 10^2$ | $89.80 \times 10^{-2}$ | $1.90 \times 10^2$ | $80.11 \times 10^{-2}$ | $2.10 \times 10^2$ | $161.0 \times 10^{-2}$ |
| $2 \times 10^0$ | $0.78 \times 10^2$ | $221.0 \times 10^{-2}$ | $1.21 \times 10^2$ | $213.0 \times 10^{-2}$ | $1.30 \times 10^2$ | $201.0 \times 10^{-2}$ |

**Table - 2**

**Electrical Resistance ($R_x$) And Capacitance ($C_x$) Observed Across Parchment Supported Cupric Carbonate, Nickel Carbonate And Complex Cupric-Nickel Carbonate Membranes Equilibrated With 0.1 M Pottassium Chloride At Different Oscillator Frequencies (Temp $25 \pm 0.1^{\circ}C$)**

| Oscillator Frequency (Hz) | Cupric carbonate Membrane | | Nickel carbonate Membrane | | Complex Cupric-Nickel carbonate Membrane | |
|---|---|---|---|---|---|---|
| | $R_X$ (Ω) | $C_X$ (μF) | $R_X$ (Ω) | $C_X$ (μF) | $R_X$ (Ω) | $C_X$ (μF) |
| $1 \times 10^3$ | $2.60 \times 10^2$ | $16.70 \times 10^{-2}$ | $3.10 \times 10^2$ | $14.60 \times 10^{-2}$ | $2.90 \times 10^2$ | $15.30 \times 10^{-2}$ |
| $2 \times 10^3$ | $1.60 \times 10^2$ | $12.10 \times 10^{-2}$ | $2.10 \times 10^2$ | $12.50 \times 10^{-2}$ | $1.70 \times 10^2$ | $11.10 \times 10^{-2}$ |
| $3 \times 10^3$ | $1.20 \times 10^2$ | $9.40 \times 10^{-2}$ | $1.60 \times 10^2$ | $9.70 \times 10^{-2}$ | $1.30 \times 10^2$ | $9.20 \times 10^{-2}$ |
| $4 \times 10^3$ | $0.97 \times 10^2$ | $8.10 \times 10^{-2}$ | $1.30 \times 10^2$ | $8.40 \times 10^{-2}$ | $1.20 \times 10^2$ | $7.70 \times 10^{-2}$ |
| $5 \times 10^3$ | $0.82 \times 10^2$ | $7.30 \times 10^{-2}$ | $1.00 \times 10^2$ | $7.60 \times 10^{-2}$ | $0.95 \times 10^2$ | $6.80 \times 10^{-2}$ |
| $6 \times 10^3$ | $0.75 \times 10^2$ | $6.30 \times 10^{-2}$ | $0.90 \times 10^2$ | $7.10 \times 10^{-2}$ | $0.82 \times 10^2$ | $6.50 \times 10^{-2}$ |

Although the values of Rm and Cm can be easily computed for simple membranes from bridge readings of Rx and Cx, such calculations cannot be

done for complex membranes because it cannot be described by a definite equivalent electrical circuit. However, in an ideal case, if simple cupric carbonate,

nickel carbonate and complex cupric-nickel carbonate membranes by retaining their identity, it may conform

to the circuit shown below –

**Fig.1  The equivalent electrical circuit for an ideal complex membrane built from simple membranes $R_A$, $R_B$ and $C_A$, $C_B$ are simple membrane resistances and capacitances, respectively.**
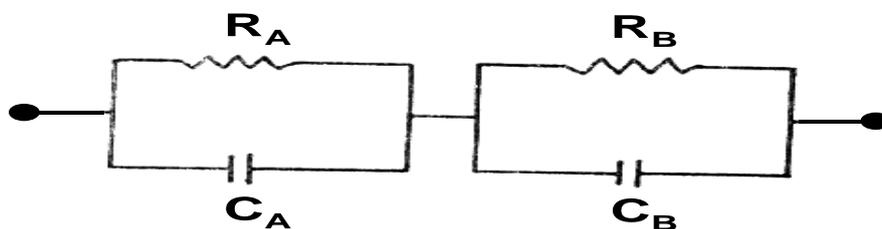


The impedance of the circuit given in Fig-1 for two units is given by equation

$$\left( \frac{R_A}{1 + J\omega \, C_A.R_A} \right) + \left( \frac{R_B}{1 + J\omega \, C_B.R_B} \right) \quad (5)$$

Where J= √-1, $R_A$, $R_B$ and $C_A$, $C_B$ are the membrane resistances and capacitances of the two units (Cupric carbonate and Nickel carbonate). Separating the real and imaginary parts of Eq. (5) yields.

$$R_e = \left( \frac{R_A}{1 + \omega^2.C^2_A.R^2_A} \right) + \left( \frac{R_B}{1 + \omega^2.C^2_B.R^2_B} \right) \quad (6)$$

$$X_e = \left( \frac{\omega C_A.R^2_A}{1 + \omega^2.C^2_A.R^2_A} \right) + \left( \frac{\omega C_B.R^2_B}{1 + \omega^2.C^2_B.R^2_B} \right) \quad (7)$$

However, another equivalent electrical circuit has been used to represent the complex membranes in view of uncontrolled simultaneous deposition of cupric carbonate and nickel carbonate in the interstices of parchment paper [15] as shown in Fig.2

**Fig.2: The equivalent electrical circuit for the complex membrane**



In this case the identity of simple cupric carbonate and nickel carbonate are lost, and the impedance of the circuit is given by

$$\left[ \frac{R_A + R_B}{1 + J\omega \left( \dfrac{C_A.C_B}{C_A+C_B} \right) (R_A + R_B)} \right] \quad (8)$$

Now separating the real and imaginary parts of Eq. (8), we have,

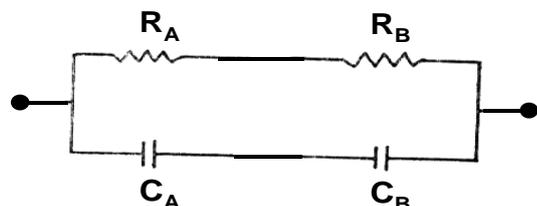$$R_e = \frac{(R_A + R_B)}{\left[ 1 + \omega^2 \left( \dfrac{C_A \, C_B}{C_A+C_B} \right)^2 (R_A + R_B)^2 \right]} \quad (9)$$

and

$$X_e = \frac{\omega \left( \dfrac{C_A.C_B}{C_A+C_B} \right) (R_A + R_B)^2}{1 + \omega^2 \left( \dfrac{C_A.C_B}{C_A+C_B} \right)^2 (R_A + R_B)^2} \quad (10)$$

The values of Re and Xe determined from Eqs-6 and 7 or Eqs-9 and 10 equilibrating the membranes with different concentrations of various electrolytes at 1KHz oscillator frequency are given in Table-5. These values are comparable to the observed values of $R_e$ and $X_e$ of the complex membrane particularly at higher concentrations of bathing electrolyte. The calculated values of $R_e$ and $X_e$ determined from Eqs-6and7 or Eqs-9and10 at different frequencies equilibrated with 0.1 M solutions of various electrolytes are given in Table-6. These values are also comparable to the observed values of $R_e$ and $X_e$ of the complex membrane particularly at high oscillator frequencies.

*Remarking An Analisation*

Table - 3
Calculated Values of Membrane Resistance ($R_m$), Capacitance ($C_m$), and Impedance (Z) For Parchment Supported Cupric Carbonate, Nickel Carbonate and Complex Cupric-Nickel Carbonate Membrane Equilibrated With Different Concentrations of Potassium Chloride at 1 KHz (Temp 25 ± 0.1ºc)

| Electrolyte Conc. | Cupric Carbonate Membrane | | | Nickel Carbonate Membrane | | | Complex Cupric-Nickel Carbonate Membrane | | |
|---|---|---|---|---|---|---|---|---|---|
| (M/L) | $R_m$ (Ω) | $C_m$ (µF) | Z(Ω) | $R_m$ (Ω) | $C_m$ (µF) | Z(Ω) | $R_m$ (Ω) | $C_m$ (µF) | Z(Ω) |
| $1 \times 10^{-4}$ | $58.22 \times 10^2$ | $4.60 \times 10^{-2}$ | $29.35 \times 10^2$ | $54.09 \times 10^2$ | $5.10 \times 10^{-2}$ | $27.02 \times 10^2$ | $52.62 \times 10^2$ | $5.00 \times 10^{-2}$ | $27.02 \times 10^2$ |
| $1 \times 10^{-3}$ | $48.84 \times 10^2$ | $6.50 \times 10^{-2}$ | $21.65 \times 10^2$ | $50.12 \times 10^2$ | $6.80 \times 10^{-2}$ | $21.00 \times 10^2$ | $46.36 \times 10^2$ | $7.00 \times 10^{-2}$ | $21.00 \times 10^2$ |
| $1 \times 10^{-2}$ | $42.28 \times 10^2$ | $9.70 \times 10^{-2}$ | $15.28 \times 10^2$ | $43.30 \times 10^2$ | $10.20 \times 10^{-2}$ | $14.71 \times 10^2$ | $43.25 \times 10^2$ | $9.10 \times 10^{-2}$ | $14.71 \times 10^2$ |
| $1 \times 10^{-1}$ | $38.16 \times 10^2$ | $15.00 \times 10^{-2}$ | $9.86 \times 10^2$ | $41.42 \times 10^2$ | $13.50 \times 10^{-2}$ | $11.31 \times 10^2$ | $43.19 \times 10^2$ | $14.20 \times 10^{-2}$ | $11.33 \times 10^2$ |
| $1 \times 10^0$ | $3.63 \times 10^2$ | $55.40 \times 10^{-2}$ | $2.25 \times 10^2$ | $3.96 \times 10^2$ | $41.20 \times 10^{-2}$ | $2.76 \times 10^2$ | $2.55 \times 10^2$ | $29.10 \times 10^{-2}$ | $2.76 \times 10^2$ |
| $2 \times 10^0$ | $1.44 \times 10^2$ | $102.00 \times 10^{-2}$ | $1.06 \times 10^2$ | $1.66 \times 10^2$ | $58.60 \times 10^{-2}$ | $1.41 \times 10^2$ | $1.78 \times 10^2$ | $54.30 \times 10^{-2}$ | $1.41 \times 10^2$ |

Table - 4
Calculated Values of Membrane Resistance ($R_m$), Capacitance ($C_m$), and Impedance (Z) For Parchment Supported Lead Carbonate, Cobalt Carbonate Membrane and A Complex Lead-Cobalt Carbonate Membrane Equilibrated With 0.1 M Potassium Chloride Solution At Different Oscillator Frequencies (Temp 25 ± 0.1ºC)

| Oscillator Frequency | Lead Carbonate Membrane | | | Cobalt Carbonate Membrane | | | Complex lead-cobalt Carbonate Membrane | | |
|---|---|---|---|---|---|---|---|---|---|
| (Hz) | $R_m$ (Ω) | $C_m$ (µF) | Z(Ω) | $R_m$ (Ω) | $C_m$ (µF) | Z(Ω) | $R_m$ (Ω) | $C_m$ (µF) | Z(Ω) |
| $1 \times 10^3$ | $38.16 \times 10^2$ | $15.50 \times 10^{-2}$ | $9.86 \times 10^2$ | $41.42 \times 10^2$ | $13.50 \times 10^{-2}$ | $11.30 \times 10^2$ | $40.20 \times 10^2$ | $14.20 \times 10^{-2}$ | $10.80 \times 10^2$ |
| $2 \times 10^3$ | $28.14 \times 10^2$ | $23.40 \times 10^{-2}$ | $6.60 \times 10^2$ | $21.96 \times 10^2$ | $22.60 \times 10^{-2}$ | $6.71 \times 10^2$ | $31.94 \times 10^2$ | $21.00 \times 10^{-2}$ | $7.36 \times 10^2$ |
| $3 \times 10^3$ | $27.49 \times 10^2$ | $26.90 \times 10^{-2}$ | $5.76 \times 10^2$ | $20.39 \times 10^2$ | $26.60 \times 10^{-2}$ | $5.73 \times 10^2$ | $26.36 \times 10^2$ | $26.20 \times 10^{-2}$ | $5.92 \times 10^2$ |
| $4 \times 10^3$ | $25.82 \times 10^2$ | $31.20 \times 10^{-2}$ | $5.00 \times 10^2$ | $18.73 \times 10^2$ | $31.10 \times 10^{-2}$ | $4.93 \times 10^2$ | $24.66 \times 10^2$ | $29.10 \times 10^{-2}$ | $5.32 \times 10^2$ |
| $5 \times 10^3$ | $24.00 \times 10^2$ | $35.20 \times 10^{-2}$ | $4.43 \times 10^2$ | $18.23 \times 10^2$ | $35.70 \times 10^{-2}$ | $4.33 \times 10^2$ | $24.00 \times 10^2$ | $32.70 \times 10^{-2}$ | $4.78 \times 10^2$ |
| $6 \times 10^3$ | $20.71 \times 10^2$ | $39.60 \times 10^{-2}$ | $3.94 \times 10^2$ | $16.35 \times 10^2$ | $40.20 \times 10^{-2}$ | $3.83 \times 10^2$ | $21.42 \times 10^2$ | $38.10 \times 10^{-2}$ | $4.19 \times 10^2$ |

**Table – 5**

**Theoretical and Observed Values of Resistive and Reactive Components of Impedance of Complex Cupric-Nickel Carbonate Membrane Equilibrated With Different Concentration Of Potassium Chloride At 1 Khz (Temp $25 \pm 0.1^{\circ}$C)**

| Electrolyte Conc. (M/L) | Theoretical Values for Fig. 2 | | Theoretical Values for Fig. 3 | | Experimental Values | |
|---|---|---|---|---|---|---|
| | $R_e$ ($\Omega$) | $X_e$ ($\Omega$) | $R_e$ ($\Omega$) | $X_e$ ($\Omega$) | $R_e$ ($\Omega$) | $X_e$ ($\Omega$) |
| $1 \times 10^{-4}$ | $28.76 \times 10^2$ | $49.06 \times 10^2$ | $28.79 \times 10^2$ | $49.12 \times 10^2$ | $14.30 \times 10^2$ | $23.41 \times 10^2$ |
| $1 \times 10^{-3}$ | $18.80 \times 10^2$ | $38.81 \times 10^2$ | $18.77 \times 10^2$ | $38.78 \times 10^2$ | $8.95 \times 10^2$ | $18.30 \times 10^2$ |
| $1 \times 10^{-2}$ | $10.52 \times 10^2$ | $28.09 \times 10^2$ | $10.71 \times 10^2$ | $28.62 \times 10^2$ | $6.00 \times 10^2$ | $14.95 \times 10^2$ |
| $1 \times 10^{-1}$ | $5.83 \times 10^2$ | $20.71 \times 10^2$ | $5.84 \times 10^2$ | $20.76 \times 10^2$ | $2.90 \times 10^2$ | $10.40 \times 10^2$ |
| $1 \times 10^{0}$ | $3.34 \times 10^2$ | $3.75 \times 10^2$ | $3.35 \times 10^2$ | $3.78 \times 10^2$ | $2.10 \times 10^2$ | $0.98 \times 10^2$ |
| $2 \times 10^{0}$ | $2.04 \times 10^2$ | $1.55 \times 10^2$ | $2.03 \times 10^2$ | $1.47 \times 10^2$ | $1.30 \times 10^2$ | $0.79 \times 10^2$ |

**Table - 6**

**Theoretical And Observed Values Of Resistive And Reactive Components Of Impedance Of Complex Cupric-Nickel Carbonate Membrane Equilibrated With 0.1 M Potassium Chloride At Different Oscillator Frequencies (Temp $25 \pm 0.1^{\circ}$C)**

| Oscillator Frequency (Hz) | Theoretical Values for Fig. 2 | | Theoretical Values for Fig. 3 | | Experimental Values | |
|---|---|---|---|---|---|---|
| | $R_e$ ($\Omega$) | $X_e$ ($\Omega$) | $R_e$ ($\Omega$) | $X_e$ ($\Omega$) | $R_e$ ($\Omega$) | $X_e$ ($\Omega$) |
| $1 \times 10^{3}$ | $44.00 \times 10^2$ | $18.02 \times 10^2$ | $44.30 \times 10^2$ | $18.60 \times 10^2$ | $44.00 \times 10^2$ | $18.00 \times 10^2$ |
| $2 \times 10^{3}$ | $0.96 \times 10^2$ | $6.79 \times 10^2$ | $1.00 \times 10^2$ | $7.00 \times 10^2$ | $1.00 \times 10^2$ | $7.00 \times 10^2$ |
| $3 \times 10^{3}$ | $0.33 \times 10^2$ | $1.77 \times 10^2$ | $0.39 \times 10^2$ | $1.88 \times 10^2$ | $0.36 \times 10^2$ | $1.80 \times 10^2$ |
| $4 \times 10^{3}$ | $0.14 \times 10^2$ | $1.68 \times 10^2$ | $0.16 \times 10^2$ | $1.70 \times 10^2$ | $0.15 \times 10^2$ | $1.70 \times 10^2$ |
| $5 \times 10^{3}$ | $0.07 \times 10^2$ | $1.54 \times 10^2$ | $0.10 \times 10^2$ | $1.58 \times 10^2$ | $0.20 \times 10^2$ | $1.60 \times 10^2$ |
| $6 \times 10^{3}$ | $0.04 \times 10^2$ | $0.32 \times 10^2$ | $0.08 \times 10^2$ | $1.30 \times 10^2$ | $0.04 \times 10^2$ | $1.30 \times 10^2$ |

It may be concluded that the two equivalent circuits may be utilized or represent the behavior of complex membrane at least in higher concentration ranges. The deviation in dilute concentrations may be assigned to the interfacial polarization and structural changes in the interfacial double layers. Thus electrically charged membranes maybe useful in filtration and desalination process.

**Endnotes**

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