

Recibido: 29 de febrero de 2024

Aceptado: 31 de julio del 2024

Publicado: 5 de agosto de 2025

### Cómo citar:

Cortés-Martínez, R., Mijares-Acosta, D. A., Flores, J. A., Martínez-Guerra, E., Viera-González, P. M., & Sánchez-Guerrero, G. E. "Plasmonic sensor as a method of measuring concentrations of binary solutions". *Artificial Intelligence on Electronics and Photonics* 1(1) pp.1-13. (2025).

## PLASMONIC SENSOR AS A METHOD OF MEASURING CONCENTRATIONS OF BINARY SOLUTIONS

Sensor plasmónico como método de medición de concentraciones en soluciones binarias

Rodolfo Cortés-Martínez<sup>1</sup> , Daniel Alberto Mijares-Acosta<sup>2\*</sup> , Jair Alejandro Flores Rodríguez<sup>2</sup> , Edgar Martínez-Guerra<sup>2</sup> , Perla Marlene Viera-González<sup>2</sup> , Guillermo Ezequiel Sánchez-Guerrero<sup>2</sup> 

<sup>1</sup> CICESE, Unidad Monterrey, Alianza Centro 504, PIIT, Apodaca, N.L. 66629, México

<sup>2</sup> Facultad de Ciencias Físico Matemáticas, Universidad Autónoma de Nuevo León, San Nicolás de los Garza, N.L. 664554, México

\* Autor de correspondencia: [daniel.mijaresa@uanl.edu.mx](mailto:daniel.mijaresa@uanl.edu.mx)

### KEYWORDS:

Surface plasmon resonance; transfer matrix method; binary solutions; Kretschmann configuration, plasmonic sensor.

### PALABRAS CLAVE:

Resonancia de plasmón de superficie; método de la matriz de transferencia; solución binaria; configuración de Kretschmann; sensor plasmónico.

## Abstract

This paper aims to showcase the behavior of Surface Plasmon Resonance (SPR). This optical technique can detect a small change in the refractive index of a sample and shows a shift in the reflective behavior, offering a method to measure precisely the concentration of a binary aqueous solution. The model used for this research is the Transfer Matrix Method (TMM) coded into a mathematical simulation. TMM has contemplated the Kretschmann configuration, composed of an SF10 prism and a gold layer of 50 nm beamed with a 633 nm wavelength laser on p-polarization, for the solution sample used a mix of CuSO<sub>4</sub> and water; by calculating the refractive index as a function of the binary solution concentration is possible to associate the reflective response to the concentration of the solution. The simulation presents a shift to the right into the angular response and, therefore, to the SPR angle to the increased concentration of the solution; this relationship is also shown in the SPR angle and the concentration of the solution in the calibration curve. These results may indicate the potential application of the technique as a sensor capable of measuring concentration in binary aqueous solutions, specifically, as a monitoring technology for water contaminants detection.

## Resumen

En este artículo se modela el comportamiento de la resonancia de plasmón superficie (SPR, por sus siglas en inglés), la cual puede detectar pequeños cambios en el índice de refracción de una muestra, mostrando un desplazamiento en el comportamiento reflectante, ofreciendo un método para medir con precisión la concentración de una solución acuosa binaria. El modelo utilizado es el Método de la Matriz de Transferencia (MMT) usado en una simulación matemática, se contempla la configuración de Kretschmann, compuesta por un prisma SF10 y una capa de oro de 50 nm excitada con un láser de 633 nm de longitud de onda en polarización p. Para la muestra de solución se utiliza una mezcla de CuSO<sub>4</sub> y agua, mediante el cálculo del índice de refracción como función de la concentración de la solución binaria, es posible asociar la respuesta reflectiva a la concentración de la solución. La simulación presenta un desplazamiento hacia la derecha en la respuesta angular y, por lo tanto, en el ángulo SPR ante los incrementos en la concentración de la solución, también se muestra la relación entre el ángulo SPR y la concentración de la solución en la curva de calibración. Estos resultados pueden indicar la aplicación potencial de la técnica como un sensor capaz de medir la concentración en soluciones acuosas binarias, específicamente como una tecnología de monitoreo para la detección de contaminantes en el agua.

## 1. Introduction

Surface Plasmon Resonance (SPR) is an optical phenomenon characterized by the collective oscillations of electrons at the interface between a metal and a dielectric material when illuminated by incident light [1]; these collective oscillations are known as Surface Plasmon Polaritons (SPPs) [1]. Plasmonic sensors are optical sensors based on SPR [2], which operate by detecting changes in the refractive index of the environment, altering the resonance conditions of SPPs [3]. The principle of operation of a plasmonic sensor involves detecting changes in the refractive index of the environment, thus affecting the resonance conditions of SPPs [3]. The excitation of surface plasmon polaritons can be achieved using coupling media such as prisms or gratings. The SPR coupling systems include the Kretschmann and Otto prism configurations, diffraction gratings, waveguides, and optical fibers [4,5]. The unique optical properties and high sensitivity of plasmonic sensors make them suitable for various applications, ranging from biology and medicine to environmental monitoring and energy conversion [6-8].

Binary solutions are homogeneous mixtures of two uniformly mixed or miscible components; they consist of a solute, the component in lesser quantity, and a solvent, the component in larger quantity. An example of a binary solution is table salt (sodium chloride) dissolved in water. Studying binary aqueous solutions helps us understand fundamental properties such as solubility, concentration, and the behavior of solutes in a solvent [9]. These solutions have applications in many scientific and engineering fields; for instance, the surface tension of aqueous solutions [10] is a critical physical property influencing the efficiency of distillation trays, the biological functioning of lung surfactants, the size distribution of aerosols produced by medical nebulizers, and the number of aerosol particles activated. The refractive index of a solution is an essential thermodynamic and optical property that can provide important information [11]. For example, it can lead to a better understanding of interactions between solute and solvent molecules [12]. A shift in the solution's refractive index, such as those caused by variations in solute concentration, modifies the resonance conditions for the SPPs [13]. Observing these alterations in resonance enables SPR to determine the solution's refractive index [14] and, consequently, track the solution's concentration.

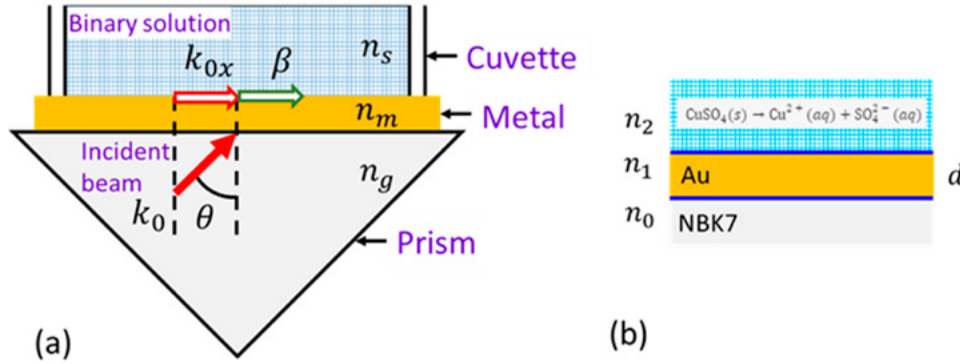
The SPR technique is employed to detect refractive indices or their changes. Using angular interrogation (reflectance vs. angle), the angle of minimum reflection, known as the *plasmon angle*, correlates with the refractive index [4]. Thus, the SPR system functions similarly to a refractometer. However, the primary advantage of the SPR technique lies in its ability to measure refractive index changes to monitor molecular reactions in real time (sensogram), enabling SPR biosensing [4, 15]. This process is done by performing an angular scan of a buffer solution, where analyte transport occurs, and positioning the

SPR system at the middle of the SPR curve slope; reflectance vs. time measurements are then conducted at a fixed angle. Therefore, the proposed system functions as a refractometer to measure the refractive index of various analyte concentrations. In this work, we use the Kretschmann configuration to numerically demonstrate the application of a plasmonic sensor for precise concentration measurements of binary solutions [16]. The Kretschmann configuration typically involves a layered structure, with each layer having a specific thickness and refractive index. The simulations in this paper were conducted using the Transfer Matrix Method (TMM), a powerful technique for modeling SPR in a layered structure [17]. The TMM works by using Fresnel coefficients to construct matrices representing each layer of the structure [18]. While many concepts used here are well known in SPR sensor research and similar work has been done previously, the innovation of this work lies in the chosen solution. In this paper, we use an aqueous solution of copper sulfate ( $\text{CuSO}_4$ ) for the numerical simulations due to its various industrial applications. For instance,  $\text{CuSO}_4$  can be used as a resistive element in liquid resistors and to add color to cement, ceramics, and some metals [19, 20].

Measuring concentrations is crucial in many disciplines, such as chemistry, pharmaceutical research, and environmental monitoring. Techniques such as chromatography [21, 22], mass spectrometry [23, 24], and others [25-35] are commonly used to detect contaminants in water samples [36]. While these methods offer high precision, they often require skilled staff and can be costly to implement. In contrast, the SPR technique may provide a promising alternative method to measure concentrations in binary solutions with excellent precision, versatility, low costs, and ease of operation. Our results demonstrate the potential of an SPR-based sensor for measuring concentrations in a binary aqueous solution of  $\text{CuSO}_4$ . The advantages of an SPR sensor include high sensitivity, versatility, and overall lower costs compared to other options. This technique may become another valuable tool for studying binary aqueous solutions, enhancing our understanding and application of optical phenomena in these solutions.

## 2. Methods

The Kretschmann configuration (Figure 1), also known as the Kretschmann-Reather configuration, is a standard setup to excite the surface plasmons; this setup allows the measurement of the SPP as a function of the incidence angle ( $\theta_{inc}$ ), and the prism used is known as the coupling prism.



**Figure 1.**

a) Standard setup of a SPR sensor in a Kretschmann configuration. b) Schematic structure consists of the semi-infinite prism (NBK7), a metallic thin layer (Au), and an output semi-infinite medium (solution sample).

With the setup, as shown in Figure 1a, under the total internal reflection condition, a component of the light beam travels parallel to the interface and, therefore, an evanescent wave is generated, which penetrates the metallic film, which has a wave vector parallel to the interface with a magnitude given by:

$$k_{\parallel} = \frac{2\pi}{\lambda} n_p \sin \theta_{\text{inc}} \quad (1)$$

where  $\lambda$  represents the wavelength of the incident light,  $n_p$ , the coupler prism refractive index, and  $\theta_{\text{inc}}$  the incident angle. This expression shows that  $k_{\parallel}$  is tunable with the angle  $\theta_{\text{inc}}$ , and the dependence on the incidence angle allows to match the evanescent wave with the surface plasmon wave vector  $\beta$  by making an angular scanning.

The SPPs are electromagnetic waves with a component perpendicular to the interface that decreases exponentially and has a wave vector  $\beta$ . Its magnitude is related to the dielectric constants of the outer medium, and the thin metallic film. The following expression gives the magnitude of  $\beta$ :

$$\beta = \frac{2\pi}{\lambda} \sqrt{\frac{n_1^2 n_m^2}{n_1^2 + n_m^2}} \quad (2)$$

where  $n_1$  is the refractive index at the vicinity of the interface (therefore, the dependence on the sample) and  $n_m$  is the refractive index of the metallic film. The evanescent wave excites the surface plasmon when  $k_{\parallel} = \text{Re}(\beta)$  where  $k_{\parallel}$  is the

component of the incident beam parallel to the interface and the plasmon wavevector which is a complex quantity.  $N_m^2 = (n + i\kappa)^2$  is the complex refractive index of the metal, where  $n$  is the real part and  $\kappa$  is the imaginary part (extinction coefficient) related to light absorption.  $n_1$  is the refractive index of the analyte. To satisfy the resonance condition, the illumination must be p-polarized (the electric field is parallel to the plane formed by the incident, reflected, and transmitted beams). This means each refractive index should be replaced by  $n/\cos\theta$ , which is related to the change in the propagation direction of light when it enters a medium at an angle. When this condition is fulfilled, the intensity of the reflected light decreases sharply the angle at which the minimum reflectivity occurs is the SPR angle,  $\theta_{SPR}$ . This phenomenon is the excitation of the surface plasmon resonance. The decay of the excited surface plasmon includes energy conversion to phonons or photons. As follows, the resonance angle relates to fixed values and  $n_m$ .

$$\theta_{SPR} = \sin^{-1} \left( \frac{1}{n_{glass}} \sqrt{\frac{n_1^2 n_m^2}{n_1^2 + n_m^2}} \right) \quad (3)$$

The Transfer-Matrix Method (TMM) is an analytical model utilized to analyze the propagation of waves through a stratified medium [37-39]. This project uses the TMM to analyze the stratified media in the Kretschmann configuration. We use this method to derive expressions describing the radiative properties of thin films, including reflectance and transmittance. Applying the TMM to the stratified structure provides a comprehensive understanding of how light interacts with and traverses the thin films in the Kretschmann configuration, enabling precise predictions of reflectance and transmittance characteristics. Figure 1b shows a multilayer structure (stratified medium) schematic where the  $n_j$  layer of thickness  $d_j$  has a complex refractive index,  $N_j = n_j + ik_j$ . Each layer of a multilayer coating has its transfer matrix, and the overall transfer matrix of the system is the product of individual transfer matrices, taken in the order in which the light propagates through the multilayer stack for each incident angle,  $\theta_i$ .

$$M_i = \prod_{j=1}^n M_{j-1,j} = \begin{pmatrix} m_{11,i} & m_{12,i} \\ m_{21,i} & m_{22,i} \end{pmatrix} \quad (4)$$

The interference matrix of the layer expresses the interference matrix of the layer is described by:

$$M_{j-1,j} = \begin{pmatrix} \frac{e^{i\varphi_j}}{t_{j-1,j}} & \frac{r_{j-1,j}e^{-i\varphi_j}}{t_{j-1,j}} \\ \frac{r_{j-1,j}e^{i\varphi_j}}{t_{j-1,j}} & \frac{e^{-i\varphi_j}}{t_{j-1,j}} \end{pmatrix} \quad (5)$$

Where:

$$r_{j-1,j} = \frac{n_{j-1}^2 k_x^j - n_j^2 k_x^{j-1}}{n_{j-1}^2 k_x^j + n_j^2 k_x^{j-1}} \quad (6)$$

$$t_{j-1,j} = \frac{n_{j-1}}{n_j} (1 + r_{j-1,j}) \quad (7)$$

$$\varphi_j = \frac{2\pi}{\lambda_0} n_j d_j \cos \theta_j \quad (8)$$

where  $r$  and  $t$  are the reflection and transmission coefficients,  $\varphi$  is the phase thickness, and  $d$  is the physical thickness. The total reflectance  $R(\theta_i)$  for each incident angle  $\theta_i$  is given by:

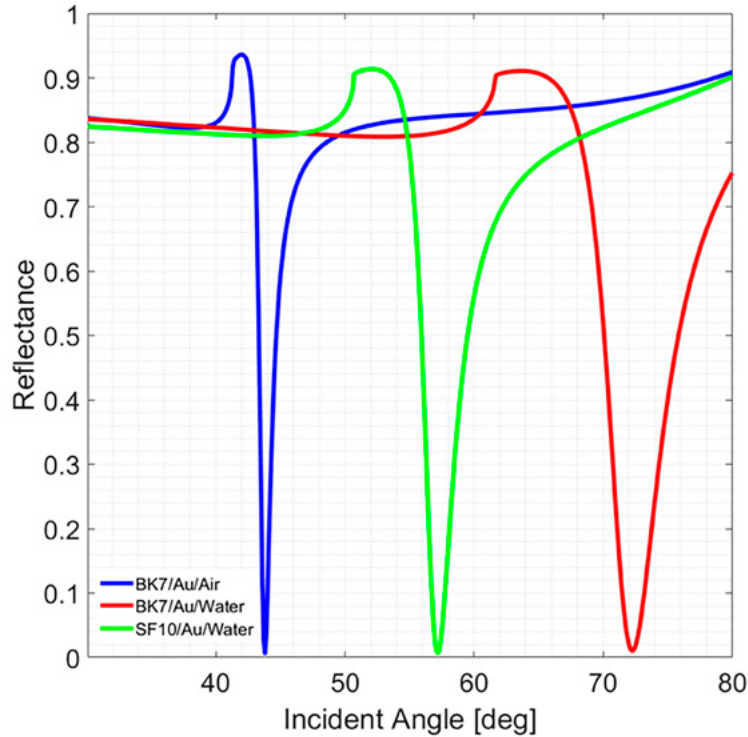
$$R(\theta_i) = \left| \frac{m_{(21,i)}}{m_{(11,i)}} \right|^2 \quad (9)$$

where  $m_{(11,i)}$  and  $m_{(21,i)}$  are the elements of transfer matrix  $M$  of the system.

For any given binary salt + water mixture, such as  $\text{CuSO}_4$  + water, the refractive index always decreases with increasing temperature and increases with the rising molality of salt. The following expression models these binary systems [40].

$$n_{BS} = n_{solvent} + \sum_{i=1}^N A_i m^i \quad (10)$$

where  $n_{BS}$  is the refractive index of the salt + water system,  $n_{solvent}$  is the refractive index of the solvent (water in this case  $n = 1.33317$ , sourced from [40]),  $m$  is the molality of the salt in the solution,  $A_i$  the fitting parameters, and  $N$  is the number of terms in the polynomial.



**Figure 2.**

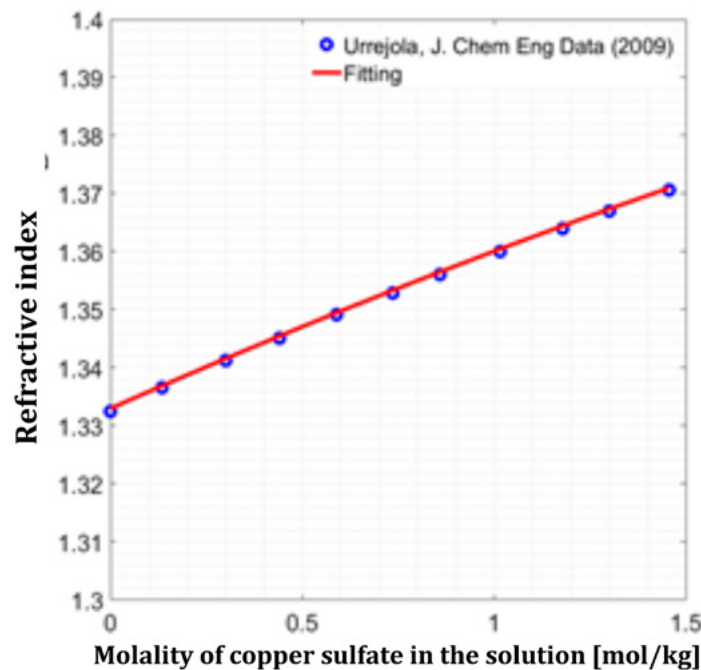
Simulation of the reflectance as a function of the incidence angle ( $\theta$ ), for an SPP excited with a 632.8 nm laser on a 50 nm thick Au film in a right angle prism (BK7,  $n = 1.51509$  [41]) against the air (blue line), the environment refractive index of water ( $n = 1.33317$ ) (red line) and a prism SF10 as coupling prism and water as sample (SF10,  $n = 1.72309$  [42], water,  $n = 1.33317$ ) (green line).

### 3. Results and discussion

As previously discussed, the SPR is sensitive to changes in the refractive index of the multilayer system. Additionally, a change in the concentration of the binary solution correlates to its refractive index; therefore, the plasmonic response changes with the concentration of the solution. As shown earlier in this article, the use of SPR as a measuring tool results in interesting [Figure 2](#) shows a reflective response against the angle of incidence for different concentrations. Each of the curves shows a dip in the reflectance caused by the SPP; the angle where this occurs is called the SPR angle. The reflectance curve in air (blue line) shows the SPR angle at 43.8 deg; this curve serves as a baseline reference for the SPR sensor without any specific environmental interaction. Transitioning the environment refractive index from air to water ( $n_{\text{water}} = 1.3317$ ) while using the BK7 prism ( $n_{\text{BK7}} = 1.5151$ ), results in a general rightward shift and



specifically to the SPR angle to 72.3 degrees (red line); additionally the SF10 prism ( $n_{SF10} = 1.7231$ ) curve still with water, shifts the SPR angle to 57.2 degrees (green line) showing that increasing the coupling prism refractive index, shifts the curve to the left. Researchers often optimize the experimental conditions, including the choice of materials and angles, to achieve maximum sensitivity and signal-to-noise ratio. In a Kretschmann configuration, the refractive index of the glass significantly impacts the angle at which SPR occurs; for this reason the prism is a critical parameter for optimizing sensor performance; in this case, researchers can adjust the glass refractive index to fine-tune the sensor response and optimize the sensor's sensitivity, stability, and reliability to measure the adjacent medium's refractive index changes. Figure 3 shows there is a quadratic dependence for the refractive index of the solution on concentration according Equation 9 [40],  $n_{BS} = -0.002177c^2 + 0.02924c + 1.3317$ . Physically, this curve describes how changes in the concentration of the binary solution impact its refractive index.



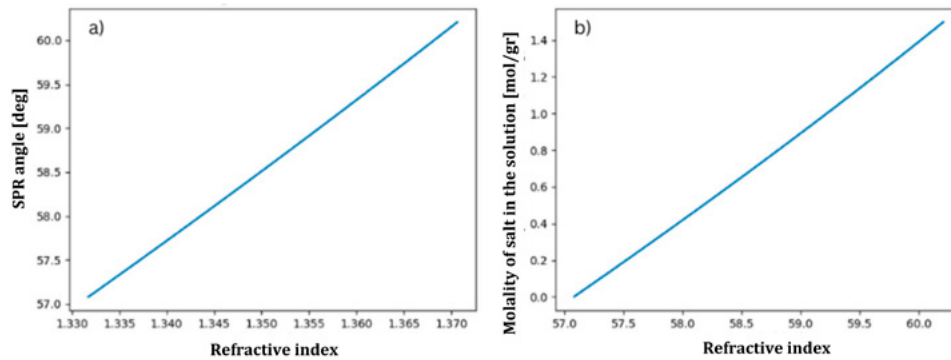
**Figure 3.**

Refractive index,  $n_{BS}$ , versus molality of copper sulfate in aqueous solution at room temperature.

As previously discussed, the SPR response depends on the environment's refractive index, and as Figure 3 shows, the refractive index varies with the concentration. Therefore, the SPR is sensitive to changes in the concentration.



In this case, the sensor's sensitivity can be optimized from . From a plot of refractive index versus SPR angle and the plot of molality versus refractive index is possible to relate the SPR angle with a specific associated molality, and therefore generate a calibration curve where only with the reflectance curve (and therefore the SPR angle) of a given binary solution, is possible to determinate the concentration of set solution.



**Figure 4.**

Steps to obtain the plasmonic sensor's calibration curve to determine the concentration of the binary solution ( $\text{CuSO}_4$  + water) from the resonance angle (b) by establishing a reference curve for the refractive index in terms of molality of the solution (Figure 3) and determining the resonance angle from a refractive index reference curve (a).

The calibration curve (Figure 4b) is the fundamental plot to generate an SPR sensor; the calibration curve proves that it is possible to associate a concentration for a determinate SPR angle, therefore showing the potential of a sensor based on SPR to measure concentrations on binary solutions, and proposing a new tool to understand optical characteristics and interactions within these solutions which can provide important information for material science, chemistry, biotechnology or environment monitoring.

## 4. Conclusions

In summary, our exploration of Surface Plasmon Resonance (SPR) as an optical sensor for binary solutions has revealed its robust responsiveness to concentration changes intricately tied to variations in the solution's refractive index. The reflectance curve, exemplified by the 50 nm thick Au film in Figure 1, serves as a foundational baseline, with the resonance angle at 43.8 degrees. Transitioning the environment's refractive index from air to water prompts a remarkable

shift to 72.3 degrees, underscoring the dynamic nature of SPR and dependence on the sample's refractive index. The Kretschmann configuration accentuates the crucial role of the glass refractive index, especially with SF10 glass at 57.2 degrees, offering a key parameter for optimizing sensor performance. Numerical simulations with a copper sulfate + water binary solution, depicted in Figure 3, showcase a quadratic dependence of the solution's refractive index on concentration, providing a quantitative tool for sensitivity optimization. The plot of refractive index versus resonance angle (Figure 4a) shows numerically the way the SPR angle depends on the environment's refractive index. Finally, the calibration curve, outlined in Figure 4b, encapsulates the three-step process, offering a robust methodology for concentration determination on the binary solution ( $\text{CuSO}_4$  + water). This comprehensive study advances our understanding of optical characteristics and interactions in binary solutions, with implications for material science, chemistry, biotechnology, and environmental monitoring applications.

## 5. Acknowledgments

R.C.M., my sincere thanks to Conahcyt for partial financial support through the Basic Scientific Research Grant No. A1-S-13587

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