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Amphetamine, Methamphetamine, Morphine @ AuNPs Kit Based on PARAFAC

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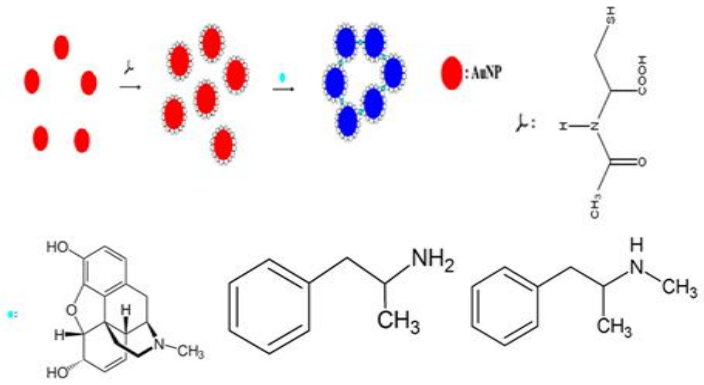
ABSTRACT

The SPR of AuNPs as a new analytical method is used to detect drugs and species through the aggregation of Au-nanoparticles. In this study, we reduced gold ion using citrate and then the surface of nano particles was modified by L-cysteine. The multivariable calibration method of SPR provide a highly precise, somehow can make an error less than 6%. As compared a highly selective and sensitive ternary determination of Amphetamine (AMP), Methamphetamine (M-AMP) and Morphine (MOR) based on the proposed method is not needed to a higher amounts of the reagents. As rose the concentration of the addictive, the color of AuNPs altered from a wine red to blue which was easily noticeable by naked eyes. A multivariable calibration model along with three dimensional methods like PARAFAC, within the analysis of SPR adsorption could provide a high functionality as they fitted the experimental data as well. A chemical based kite and chemo metrics provide an excellent kite, which in turn is unique as experimentally confirmed. PARAFAC is a multi-way method rooted in psychometrics. Notably, chemometrics and the related areas due to various reasons such as high computational power, consciousness of the method and its potentials, the improved complication of data, have received much more considerations.

GRAPHICAL ABSTRACT



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Introduction

Au nanoparticles (Au NPs), thanks to high stability in various aspects like biosensors, biomedicine, catalysis, and other vital applications, are widely utilized [1-6]. Since we are interested in TRLFS data obtained as a function of chemical conditions, the entire data set can be expressed by a three-way array, X, where the under bar is used to signify the multi-way (>2-way) nature of the array. In three-way parallel factor analysis (PARAFAC), the assumption is that X follows the trilinear. During this analysis, the primary mode of X matches the chemical settings of the adsorption samples, such as pH, the concentration of salt used, or Eu^{3+} concentration, and the second and third modes to the spectral and temporal profiles, respectively. Column-wise orthogonal random matrices were used for the initialization, and local minima were checked using the PARAFAC model to ensure the achieved model's stability and interpretability [7]. Internal production, due to internal management of overhead and manpower costs, in many respects, can compete with foreign samples while being used in the domestic environment; they also compete with their foreign counterparts in terms of quality [8]. Various analytical methods have been developed for determining AMP, M-AMP, and MOR.. Methamphetamine and 3,4-methylenedioxy-N-methamphetamine are amphetamine-type stimulants [9, 10]. Determining ketamine, methamphetamine, and 3,4-methylenedioxy methamphetamine in human hair by flash evaporation-gas chromatography/mass spectrometry [11]. Qualitative screening tests using immunochromatographic cartridges to monitor M-AMP, AMP, and MOR [12]. Simultaneous determination of illicit drugs in human urine by liquid chromatography-mass spectrometry was evaluated according to reference [13]. The method for simultaneous determination and confirmation of illicit drugs (e.g., methamphetamine, amphetamine, ephedrine, methyl ephedrine, morphine, morphine-3-glucuronide, morphine-6-glucuronide, 6-acetylmorphine, cocaine, and benzoylecgonine) in human urine by thermospray liquid chromatography-mass spectrometry (LC-MS) was studied. And some kinetic spectrophotometry method for determining morphine, nalbuphine, and naltrexone drugs in bulk and pharmaceutical formulations was performed in near years [14]. A few methods have been described to detect nalbuphine in pharmaceutical formulations; they include GC coupled to electron-capture detection [15] or mass spectrometry [20], HPLC with electrochemical detection, and ion-selective electrode. Although the GC methods [15, 16] are sensitive, they involve expensive equipment and time-consuming sample preparation and are not readily available for regular drug monitoring. On the other hand, the reported HPLC methods do not provide adequate sensitivity. In this work, urine specimens were first tested and then controlled in-person situations using tape tests within hydrolysis and screening tests; then, the positive cases were tested using the AuNPs @ spectroscopy @ chemometrics software method and based on the available instructions for this kit.

Experimental

Materials and methods

All chemicals used in this research were of analytical grade without purification. Tri sodium citrate dehydrates, hydrochloride acid, and hydroxide sodium were purchased from the Merck Company.

Synthesis of gold nanoparticles

The Au seeds were synthesized according to Ferns method. Briefly, 100 mL of 1 mmol L^{-1} aqueous solution of $HAuCl_4$ was heated to boil

with stirring; then 10 mL 1% (w/v) aqueous sodium citrate was added suddenly. The color of the mixed solution changed from yellow to red wine in several minutes, indicating the formation of AuNPs. The boiling and stirring were continued for 15 min [2]. The seed solution was cooled to room temperature and was stored in a dark bottle at 4 °C. Figure 1 exhibits the typical UV visible spectra of the prepared nanoparticle (NPs). The solution of prepared citrate-capped AuNPs (Figure. 1) is wine red and has a characteristic LSPR absorption band of AuNPs at 520 nm (.max) with a narrow peak. In solution, mono disperse AuNPs appear red and exhibit a relatively narrow surface Plasmon absorption band centered at around 520 nm in the UV-Vis spectrum.

In contrast, a solution containing aggregated AuNPs appears blue, corresponding to a characteristic red shift in the surface Plasmon resonance to a high wavelength of 660 nm [1–6]. It is interesting changing the color and size when AuNPs aggregated. (Figures 2A-C).

Characterization

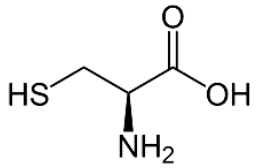
The adsorption analysis was implemented using an Agilent 8453 UV-Visible spectrophotometer with 1cm quartz cells. To observe and evaluate the surface morphology of synthesized nanoparticles, the transition electron microscopy used. A pH meter (model 713) to measure the pH of samples was used. We also employed an ultrasonic (RoHS, Korea) for dispersing the nanoparticles into solution. The corresponding calculations using MATLAB (Hyper-cube Inc. Version10) program were done

Results and Discussion

Optimization part

Optimization of L-cysteine concentration

The concentration of L-cysteine plays a significant role in the aggregation process and the selectivity agent. Thus, as a modifier could harness the distance between nanoparticles, the functional groups could somehow provide a H-bonding interaction with materials [8]. Thus, a simultaneous measurement is provided. We investigated the alterations of L-Cysteine concentration and the concentration of 10^{-5} mol. L^{-1} as an optimum concentration was selected and considered for further analysis. The corresponding experiments considering a ratio of 10/20 for nanoparticles and drugs were implemented, and the ratio used led to an enhanced selectivity. Highlighting the importance of the amount of nanoparticles determined and used caused the interaction between nanoparticles and drugs to be significantly increased. Therefore, enhanced selectivity can be attributed mainly to nanoparticles' interaction and accumulations [17]. As heavy metal ionophores, ligands with various donor atoms have been studied. The hard, functional groups of L-Cysteine can help to an effective drug loading, as it is expected [18].



Scheme 1. Structure of L-Cysteine

Optimization of NaCl concentration

As proven, ion strength could contribute to the nanoparticles aggregation and plays a leading role in this end. Harnessing the electric multilayer is potentially affected by the strong electrolytes formed in the aqueous solution. Give that experimentally confirmed an increment in ion strength as a highly effective parameter led to nanoparticle aggregation, highlighting the importance of forming ion strength for nanoparticle accumulations. The ion strength of made solution affects the nanoparticle accumulation [1-6]. As demonstrated from figure

3, 1 mmol. L^{-1} as an optimum concentration of NaCl has been considered and utilized.

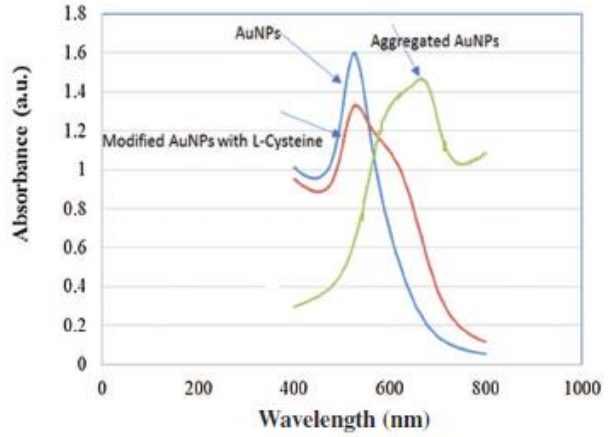


Figure 1. Typical UV visible spectra

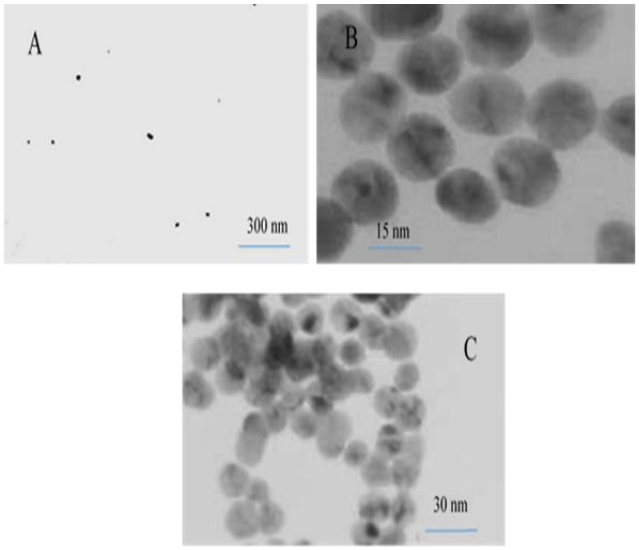


Figure 2. TEM images of Au nanoparticles (A) Au nanoparticles modified with L-cysteine (B). The aggregated Au nanoparticles (C)

Optimization of pH

pH is a crucial parameter affecting the amount of drug adsorption and plays an essential and pivotal role. Therefore its effect must be substantially considered. It should be noted that the functional groups corresponding to drugs as an anorectic group could also fundamentally alter depending on the alteration of pH. The selected drugs via the weak Van der Waals forces and the electrostatic interaction can be effectively and physically loaded on the surface of nanoparticles, resulting in an effective drug loading [1-6]. To boost the electrostatic interactions, the drug molecule must be accumulated in the nanoparticles surrounding as a strong interaction is considered. As demonstrated in Figure 4, the synthesized AuNPs have high stability within the pH range 6, and a remarkable interaction is observed in pH 6. Given that experimentally confirmed within the high pH range, the drugs and metallic ions on the surface of nanoparticles could form hydroxide sediment, which in turn could significantly affect drug loading, highlighting the importance of pH as a desired loading considered [1-6]. Upon further investigation, we selected pH 6 as an optimum condition for further study (Figure 4).

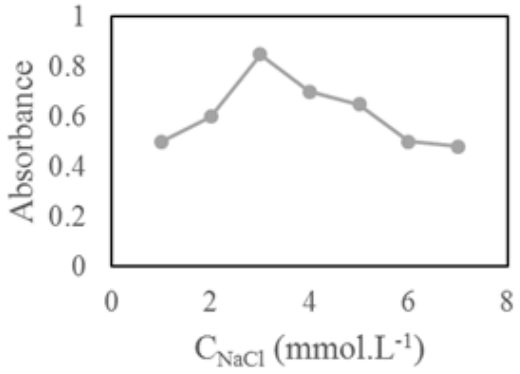


Figure 3. Evaluation and optimization of the ionic strength considering the experimental conditions used (pH of 6, ionic strength 1- 7 m mol. L^{-1}, time 10 min, L-cysteine 10^{-5} mol. L^{-1}, injection of 200 µL of turnery drugs (10^{-4} mol. L^{-1}): AuNPs, 10 nmol. L^{-1})

Optimization of incubation time

The incubation time as an effectiveness parameter was considered to find the optimum value. While M-AMP, MOR, and AMP, we mixed under an optimum condition, in which the

process corresponding to drug loading and nanoparticles accumulated are formed. A considerable alteration of spectra was observed within the optimum time 10 min. We considered 10 min as the optimum incubation time, and a considerable spectral alteration is seen at this time. As illustrated in Figure 5, for kinetic study, the time of 10 min as an optimum time was selected and considered.

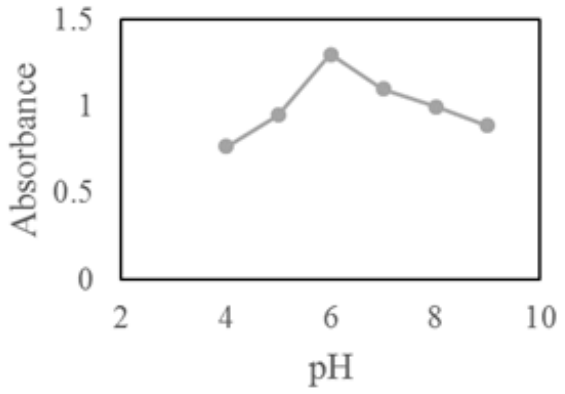


Figure 4. The effect of pH on drugs adsorption amount (pH from 4-9, time 10 min, ionic strength 3 m mol. L^{-1} L-cysteine 10^{-5} mol. L^{-1}, injection of 200 µL of M-AMP, MOR, and AMP (10^{-4} mol. L^{-1}): AuNPs, 10 n mol. L^{-1})

PARAFAC model development

According to Kiers [19], The PARAFAC model is widely used as a restricted version of the Tukers 3 model. Also, the Tukers3 model would, in turn, retunes to a restricted version of two dimensional PCA model. The Tucker3 and PARAFAC models could also predicate the experimental data. As the PARAFAC model provides a lower freedom degree toward other models defined, it could provide high performance as it is considered and utilized. It was found that the PCA model better fitted the experimental data than the Tukers3 model, and the PCA model demonstrated a better functionality against the PRAFAC model. Among three models considered PARAFAC model, Tucker3, and two-way PCA, we selected the model that could provide a higher degree of freedom. Therefore the selected model would, in turn, could contribute to enhanced performance as it is expected. Therefore, using the simplest possible model turns out to be the best option. However, it is not a trend anymore, as this principle dates back to the fourteenth century (Occam's razor) [20]. The PCA model is the most flexible and complex model and uses the utmost degrees of freedom, while PARAFAC is a constrained and simple model.

This model, inanition to a seamless fit, would equal the data and use all degrees of freedom outstandingly. Therefore, the higher structure will result in a poorer fit and t simpler model. Using multi-way methods is not to attain a better fit but rather to create more interpretable, acceptable, and vigorous models. There is the same difference between partial least squares regression (PLS) and multiple linear regressions (MLR) for multivariate calibration.

Depending on the alterations of the calibration data, the MLR model could also provide remarkable and desired results.

PLS as a constrained version of PLS and .L.R.MLR could focus on the systematic part of data. In other words, multivariable models provide less sensitivity toward the noise. They can also provide the loadings directly affected by other models. The PCA model is also utilized for predicting the complex process.

In the case of a F-component P.C.A. solution, I X J X K array unfolded to a Z X JK matrix, the PCA model consists of F (I + JK> parameters (scores and loading elements).

A corresponding Tucker model with an equal number of components in each mode would consist of F (Z +Z + K) + F 3, and PARAFAC F (Z + J + K) parameters. To have a hypothetical example, consider a 10 X 100 X 20 array modeled by a 5-component solution. A two-way PCA model of the 10 X 2000 unfolded arrays consists of 10050 parameters, a Tucker model of 775, and a PARAFAC model of 650 parameters. The

interpretation of PCA model is much more complicated than the multi-way models.

Compared to other models, the PARAFAC model needs a higher time for calculating and analyzing of data determined. For the PARAFAC model, in addition to the mentioned cases, its algorithm has been formed based on alternating.

Least squares (ALS), with interring a random amount of the defined program stars to run. The number of steps and interaction of calculation is significantly affected by the ALS algorithm PARAFAC, and somehow, this model could decrease the number of calculations up to 20 times [21-23].

In the next step, we evaluated and investigated the three-way models. These models could provide a good result for any order as they are considered utilized. Considering all conditions, the PARAFAC model and the models with higher orders could provide a good and considerable functionality as they are used. Therefore, their applicability in fitting experimental data could be substantially considered despite current limitations.

The present report helps us to find out the addicted patients. They depend on psychotropic and addictive drugs such as amphetamine, methamphetamine, and morphine in the medical diagnostic labs**.**

It is important to note that using this device in diagnostic laboratories as a substitute for ELISA tests or western blot testing is strictly prohibited. Therefore, the provision of these kits in pharmacies and their usage as a means of self-examination in individuals due to the importance of non-separation testing of advice is not allowed [24].

Kinetic modeling

In this study, based on the aggregation of citrate-capped L-cysteine modified gold nanoparticles (Au NPs), a new method was introduced for Tramadol, Methadone, and buprenorphine, AuNPs-kit using software and chemometrics tools. The absorbance increases and decreases versus time have been used as a technique for simultaneous determination of these molecules at 520 and 650 nm. Depending on the kinetic parameter, the developed and fabricated kit could affect drug loading as experimentally confirmed for Tramadol, Methadone, and buprenorphine. As observed, the alteration of the concentration of drugs (Tramadol, Methadone, and buprenorphine) affects adsorption. Somehow, the peaks corresponding to adsorption in 520 nm and 650 nm have been decreased and increased, respectively. According to the adsorption analysis and as the time altered, it was observed that the peak intensity was also altered over time. We recorded the adsorption peaks' alteration over time using the PARAFAC model. The drug simulation measurements were done. Figure 6 reveals the kinetic profile of M-AMP, MOR, and AMP @AuNPs. We analyzed the increase of absorbance versus time at 650 nm.

All three addicted drugs used have demonstrated different results (Figure 7-8). According to Figures 6 and 7, we could take some information on the kinetic difference of M-AMP, MOR, and AMP. Therefore, we can conclude that these three drugs could determine simultaneously in a software-based kit.

In PARAFAC mfile; (factor) is unknown Concentrations, (it== iteration) is the repeat of model and (err) give us the error of model (Table 1).

In right section input is classified to X== 3-way data of absorbance, Fac is number of factors==3 and for unimodality and non-negativity must to use options=0 and const= [2 3 2].

Table 1. PARAFAC analysis

Conc of AMP (ng. mL^{-1})	Conc of M-AMP (ng. mL^{-1})		Conc of MOR (ng. mL^{-1})		
Real amount	Predicted amount	Real amount	Predicted amount	Real amount	Predicted amount
400	500	300	312	34	31
650	651	610	609	0	0.01
		([factor, it, err] =parafac (X, 3, 0, [2 3 2])			
490	487	730	731.5	5	5.04
err= 4.01 it=70					

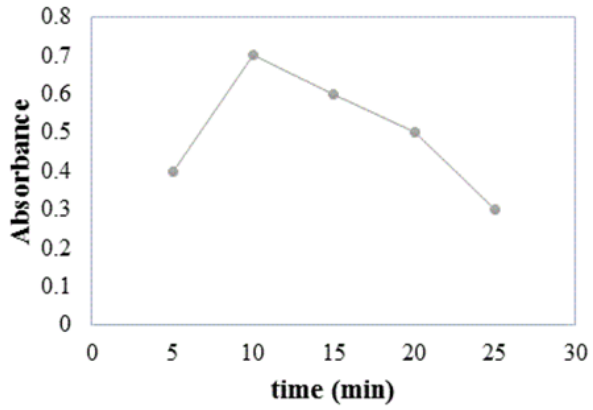


Figure 5. The effect of the incubation time on drugs adsorption (pH of 6, time 5-25 min, ionic strength 3 m mol.L^{-1} L-cysteine 10^{-5} mol. L^{-1}, injection of 200 µL of M-AMP, MOR, and AMP (10^{-4} mol.L^{-1}): AuNPs, 10 n mol. L^{-1})

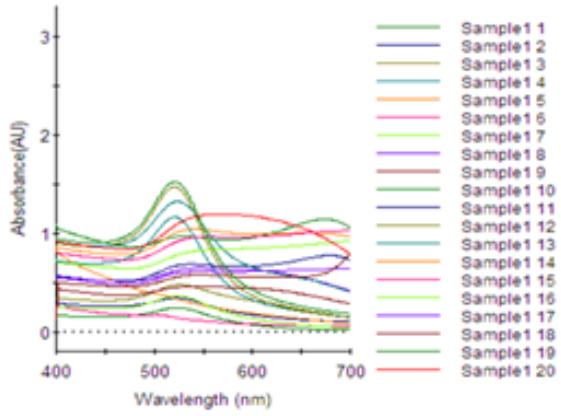


Figure 6. Kinetic profile of M-AMP, AMP, and MOR @AuNPs

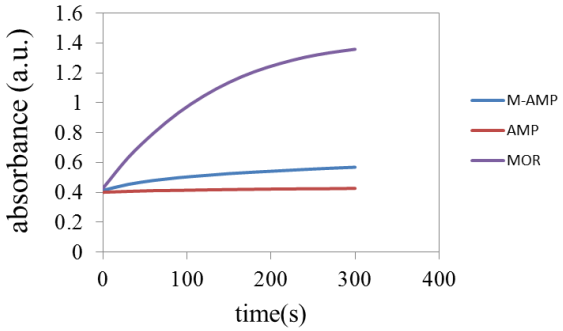


Figure 7. Kinetic difference of AuNPs absorbance spectra at 520 and 650 in the presence of M-AMP, MOR, and AMP

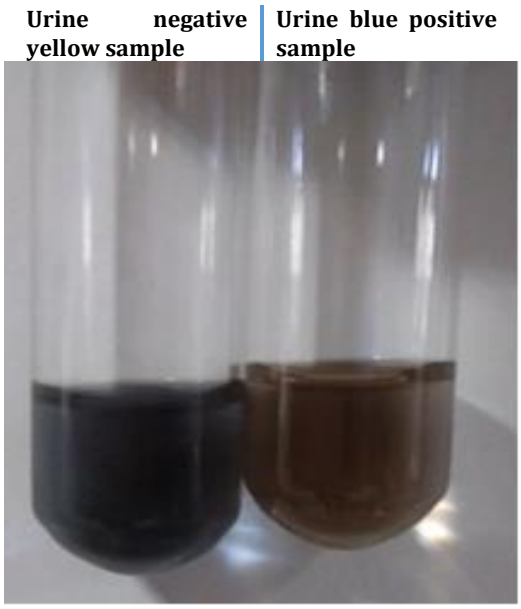


Figure 8. Color alteration of the synthesized sample color changed in positive and negative actual samples

Interference study

In the current study, the effect of the components with a similar structure on drug measurements was substantially investigated. Table 2 summarizes some components with similar structures affecting AMP, M-AMP, and MOR. The results showed no significant interference of K^+, Na^+, NO_3^-, I^-, Cl^-, Mg^{2+}, Fe^{3+}, Ca^{2+}, and codeine. Some of the components used such as NH_2OH, Mn^{2+}, SO_4^{2-}, Ca^{2+}, Zr^{2+}, Co^{2+}, Zn^{2+}, Ni^{2+}, Al^{3+}, Fe^{2+}, Cu^{2+}, Tramadol, Buprenorphine, and Methadone within the high concentration range were evaluated and investigated.

As shown in Table 2, the presented method provided a higher selectivity against other components investigated.

Real sample analysis

The applicability of the suggested model was evaluated as AMF, M-MAF, and MOR was injected into the urine sample, and results indicated an excellent recovery percent (98.4-105.1%). As seen in Table 3, the presented method suggesting highly potential in detecting AMF, M-MAF, and MOR was for actual samples.

Table 2. The effect of interference materials on simentanus measurements of drugs under optimum conditions determined (ionic strength 3 mmol. L^{-1}, time 10 min, L-Cysteine concentration 10^{-5} mol. L^{-1}, pH of 6: AuNPs, 10 nmol. L^{-1})

Substance	Tolerable concentration (ng. mL^{-1}) (analyte (MOR: AMP: M-AMP): interfering ion)
K^+, Na^+, NO^{3-}, I^-, Cl^-, Mg^{2+}, Fe^{3+}, Ca^{2+}, codeine	**1:1:1:400**
, NH_2OH, Mn^{2+}, SO_4^{2-}, Ca^{2+}, Zr^{2+}, Co^{2+}, Zn^{2+}, Ni^{2+}, Al^{3+}, Fe^{2+}, Cu^{2+}, Morphine, Amphetamine, Methamphetamine	**1:1:1:200**

Table 3. Investigation and analysis of the amount of drugs t into the urine sample under the mentioned condition (ionic strength 1 mmol/L, time 10 min, pH of 6: AuNPs, 10 nmol. L^{-1})

Sample	Add			Found			Recovery(%)		
	M-AMP ng. mL^{-1}	MOR ng. mL^{-1}	AMP ng. mL^{-1}	M-AMP ng. mL^{-1}	MOR ng. mL^{-1}	AMP ng. mL^{-1}	M-AMP	MOR	AMP
Urine	0	0	0	302	101.2	10.02	-	-	-
	320	120	20	319.5	122.0	21.02	99.8	101.7	105.1
	330	130	30	332.1	128.2	30.5	100.6	98.6	101.6
	340	140	40	341.03	141.0	40.5	100.3	100.7	101.2
	350	150	50	348.2	151.02	49.2	99.5	100.7	98.4

Conclusion

To sum up, in this study for detecting and measuring drugs, we successfully synthesized the gold-based nanoparticles, which then functionalized with L-Cysteine. To this end, the SPR absorption of nanoparticles accumulated in the presence of drugs was evaluated and investigated. According to the results of adsorption analysis, it was found that the PARAFAC model has been fitted to the experimental data of SPR adsorption as well, and a prediction error lower than 6% was computationally revealed, highlighting the high performance of the model used. Experiments also demonstrated that by adding drugs to nanoparticles, a color alteration of a solution is formed. Somehow the color solution altered from red win to blue, suggesting the strong interaction between the drug molecules and the surface of nanoparticles which could contribute to detecting the drug concentrations used. The PARAFAC model used as a multivariable

calibration model accurately fitted the experimental data and could measure the drug concentration separately. It can be concluded that the developed kite having a unique feature in detecting drugs has significant potential, and its applicability can be fundamentally considered. PARAFAC as a multi-way technique has its origin in psychometrics. Recently researchers have paid much attention to it as it can increase the computational power, method - awareness, and possibilities. The suggested method has the following pluses:

It is a highly selective and sensitive method. There is no need for any preliminary sample preparation. In spectrophotometry combined with chemometrics and this method, minor sample preparation is needed, and samples are at the same value. Increased drug concentration led to a considerable color alteration of AuNps from wine der to blue.

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Disclosure Statement

The authors reported no potential conflict of interest.

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