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Mean centering of ratio spectra and concentration augmented classical least squares in a comparative approach for quantitation of spectrally overlapped bands of antihypertensives in formulations

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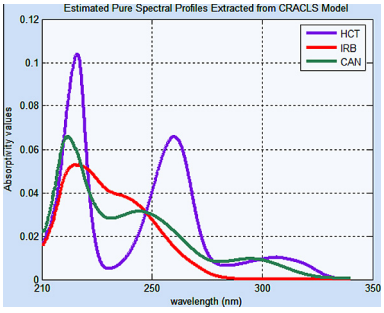
HIGHLIGHTS

- HCT is commonly co-formulated with IRB or CAN.
- MCR and CRACLS are applied in a comparative approach for resolution of the three drugs.
- One calibration curve and one model were used to predict HCT in both formulations.
- Separation of three drugs was achieved by simple and smart methods.
- Both MCR and CRACLS were successfully determines the three drugs in tablets.

GRAPHICAL ABSTRACT



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ABSTRACT

Two different methods manipulating spectrophotometric data have been developed, validated and compared. One is capable of removing the signal of any interfering components at the selected wavelength of the component of interest (univariate). The other includes more variables and extracts maximum information to determine the component of interest in the presence of other components (multivariate). The applied methods are smart, simple, accurate, sensitive, precise and capable of determination of spectrally overlapped antihypertensives; hydrochlorothiazide (HCT), irbesartan (IRB) and candesartan (CAN). Mean centering of ratio spectra (MCR) and concentration residual augmented classical least-squares method (CRACLS) were developed and their efficiency was compared. CRACLS is a simple method that is capable of extracting the pure spectral profiles of each component in a mixture. Correlation was calculated between the estimated and pure spectra and was found to be 0.9998, 0.9987 and 0.9992 for HCT, IRB and CAN, respectively. The methods were successfully determined the three components in bulk powder, laboratory-prepared mixtures, and combined dosage forms. The results obtained were compared statistically with each other and to those of the official methods.

Introduction

Hydrochlorothiazide (HCT) (Fig. 1a) or 6-chloro-1,1-dioxo-3,4-dihydro-2*H*-1,2,4-benzothiadiazine-7-sulfonamide is a diuretic drug of the thiazide class that acts by inhibiting the kidneys' ability to retain water and is frequently used for the treatment of

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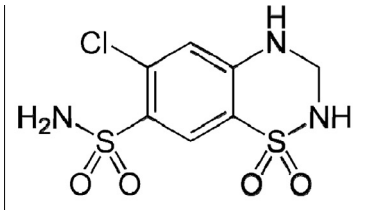
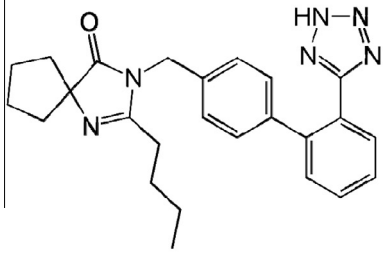
hypertension. Irbesartan (IRB) (Fig. 1b) or 2-butyl-3-({4-[2-(2*H*-1,2,3,4-tetrazol-5-yl)phenyl]phenyl}methyl)-1,3-diazaspiro[4.4] non-1-en-4-one is an angiotensin II receptor antagonist and used mainly for the treatment of hypertension. Candesartan (CAN) (Fig. 1c) or 2-ethoxy-1-({4-[2-(2*H*-1,2,3,4-tetrazol-5-yl)phenyl]-phenyl}methyl)-1*H*-1,3-benzodiazole-7-carboxylic acid is an angiotensin II receptor antagonist that is used mainly for the treatment of hypertension. Both IRB and CAN is also available in a combination formulation with a low dose thiazide diuretic, invariably hydrochlorothiazide, to achieve an additive antihypertensive effect [1].

Few methods have been reported for the simultaneous determination of HCT and IRB in their pharmaceutical formulations as HPLC [2–9] and spectrophotometry [2–13]. Different analytical methods have been reported for the determination of HCT and CAN which include HPLC [14–18], HPTLC [19] and spectrophotometry [20–24]. While for IRB and CAN, only two HPLC methods were reported [25,26].

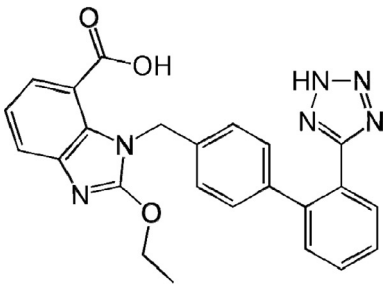
As both IRB and CAN are co-formulated with HCT to achieve maximum therapeutic effect and no method was reported for their determination in ternary mixture. So, our aim was to simultaneously determine the three drugs without prior separation by a simple and smart method that could be applied for determination of any drugs combination. The developed methods can be successfully applied in routine analysis and QC laboratories. Two different techniques were applied; the first is a univariate method, namely mean centering of ratio spectra (MCR) which is based on cancelling the contribution of other components than the analyte. While, the second technique is a multivariate calibration methods namely concentration residual augmented classical least squares (CRACLS) which is based on determination of all components in the presence of each other through spectral resolution.



a. Hydrchlorthiazide (HCT)



b. Irbesartan (IRB)



c. Candesartan (CAN)

Fig. 1. Structural formulae of the studied compounds.

Mean centering of ratio spectra method (MCR)

It is a well-established method that has the advantage of being applicable to binary and ternary mixture, as well it enhances the signal to noise ratio. The theory of the method is well known and based on mean centering as a processing step to eliminate the interference of components present in a mixture rather than the component of interest [27].

Concentration augmented classical least squares method (CRACLS)

CRACLS is a recently developed method for the resolution of complex mixtures. Unlike CLS, CRACLS is an alternative method that estimates absorptivity ($\hat{S}$) by a process of repetitive approximation as shown in the following steps [28,29]:

Step 1: $\hat{S}$ is calculated: $\hat{S} = (C\_C) - 1C'A$.
Step 2: $\hat{S}$ is used to predict C_: $C\_ = A\hat{S}'(\hat{S}\hat{S}') - 1$.
Step 3: Error in C′: $E = C' - C$.
Step 4: One vector of E is augmented to the original C (E is considered as a new component).
Step 5: Step (1) is repeated using the augmented C until no further improvement in prediction is achieved. CRACLS model was built for HCT, IRB and CAN.

Experimental

Samples

Pure samples

Pure samples were kindly donated by October Pharm, the percentage purity was found to be 99.83, 100.28 and 99.67 for HCT, IRN and CAN according to their official methods [1].

Pharmaceutical formulations

Kansartan Plus® tablets, Batch No. 120536A (150/12.5), 120537A (300/12.5) of IRB and HCT, respectively, manufactured by CHEMIPHARM. Atacand Plus® tablets Batch No. 130352 (16/12.5) of CAN and HCT, respectively, Manufactured by SANOFI AVENTIS. Both formulations were obtained from local market.

Solvents

Methanol (E. Merck, Darmstadt, FRG).

Apparatus

Shimadzu (Columbia, MD) 1605 UVPC spectrophotometer using 1.00 cm quartz cells. Scans were carried out in the range of 220–300 nm at 0.5 nm intervals.

Software

All computations were performed in Matlab (Natick, MA) for Windows™ Version 6.5 [30]. The PLS procedure was taken from PLS_Toolbox [31] for use with Matlab 6.5.