Food Chemistry 170 (2015) 62–73

Contents lists available at ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem

Analytical Methods

Tackling correlated responses during process optimisation of rapeseed meal protein extraction

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ARTICLE INFO

Article history:
Received 14 August 2013
Received in revised form 21 July 2014
Accepted 11 August 2014
Available online 21 August 2014

Keywords:
Response Surface Methodology
Principal Component Analysis
Desirability function
Multiple Linear Regression
Rapeseed press-cake

ABSTRACT

Setting of process variables to meet the required specifications of quality characteristics is a crucial task in the extraction technology or process quality control. Simultaneous optimisation of several conflicting characteristics poses a problem, especially when correlation exists. To remedy this shortfall, we present multi-response optimisation based on Response Surface Methodology (RSM)-Principal Component Analysis (PCA)-desirability function approach, combined with Multiple Linear Regression (MLR). Experimental manifestation of the proposed methodology was executed using a multi-responses-based protein extraction process from an industrial waste, rapeseed press-cake. The proposed optimal factor combination reflects a compromise between the partially conflicting natures of the original responses. Prediction accuracy of this new hybrid method was found to be better than RSM alone, verifying the adequacy and superiority of the said approach. Furthermore, this study suggests the feasibility of the exploitation of the waste rapeseed oil-cake for extraction of valuable protein, with improved colour properties using simple, viable process.

1. Introduction

Currently, interest in finding the optimal experimental conditions for more than one response by using Response Surface Methodology (RSM) is frequent in studies employing product quality improving techniques. Often in industries, it is difficult to analyse the results obtained from RSM when several dependent variables (responses) of interest, especially of opposing nature, are involved. This fact makes optimisation procedure a challenging issue in industrial processes. The analysis of data from a multi-response experiment requires careful consideration of the multivariate nature of the data. Interrelationships that may exist among the responses can render univariate investigation meaningless. Several publications have presented approaches addressing multiple quality characteristics but very few published papers have focused primarily on the existence of correlation. If we desire to optimise several correlated responses simultaneously, we would end up in obtaining futile separate individual optima; optimal condition for one response may be far from real optimum or even physically impractical for the others or may produce unsatisfactory results for the remaining responses. Thus, if correlations among quality characteristics are ignored, the designers may miss finding design variable settings that simultaneously improve the quality of all the responses, which in-turn could lead to an unrealistic solution causing model instability, over-fitting and errors of prediction.

Principal Component Analysis (PCA) has been shown to mitigate the problem of correlation among multiple quality characteristics; however, principal component regression (PCR) cannot adequately model the non-linear relationships, in spite of its success in many applications. Meanwhile, RSM, despite its great features and successful performance in optimising non-linear data, is not able to deal with correlated responses, which is one of its drawbacks. So, it is expected that either PCA or RSM alone is inadequate for predicting such responses. In this work, a two-fold technique based on PCA and RSM was applied as strategy for tackling this problem, the basic mechanism of which is described below (Section 1.1). Recently, Salmasnia, Kazemzadeh, and Niaki (2011) proposed a new methodology called desirability function-based PCA (DF-PCA), which claims to combine the advantageous features of both PCA and desirability function in a single method.

Taking into account all the information mentioned above, it seemed prudent to investigate the performance of the combined approach modeling (RSM-PCA) along with desirability function. The application of this new hybrid method is likely to improve the predictive accuracy of the model over the individual methods

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http://dx.doi.org/10.1016/j.foodchem.2014.08.053
0308-8146/

because it amalgamates each method's unique features to capture different patterns or features in the data set.

1.1. Theoretical explanation of multi-response optimisation using PCA and RSM

The idea is to implement PCA for obtaining uncorrelated interpretable components, which are the substitution of the original responses (Ribeiro, Teófilo, Augusto, & Ferreira, 2010). By doing so, significant information contained in the original variables is replaced by a small set of new variables (called the principal components, PCs) and redundant information is thus eliminated.

Fig. 1 displays the underlying principles involved in this two-fold technique (Ribeiro et al., 2010). In first step, by exploring the correlation structure among the responses, all p responses are transformed into k PCs ($k < p$) and then using central composite design (CCD), PCs are fitted by choosing a polynomial equation with appropriate degree. Desirability function is used for judging the predicted PCs under optimum condition. The optimum values of the causal factors (independent variables) are determined from the value of the individual desirability functions that maximise overall composite desirability (D). In second step, under suggested optimal experimental conditions, Multiple Linear Regression analysis plays a key role for finding predicted responses from predicted PC scores.

1.2. Experimental manifestation of the proposed optimisation technique

As an experimental demonstration for the proposed methodology, optimising the extraction of light-coloured protein from rapeseed press-cake, a major bulk waste from oil industries, have been chosen. Deriving protein concentrates and isolates from *Brassica* oilseeds for food, drug or cosmetic applications, have been attempted over the past few years for its superior-quality, high energy value and also for curving the exorbitant cost of protein from animal sources. Unfortunately, the rapeseed/canola protein isolates produced from the reported methodologies/techniques mainly suffers from poor commercial appearance due to its unappealing dark brown–black colour, caused by association of oxidised or polymerised polyphenolic compounds with protein matrices, especially during conventional alkaline extraction. None of the proposed processes (ultrafiltration, diafiltration, ion-exchange and protein micellar mass methods) have been commercially exploited or upscaled to practice so far, most probably due to membrane fouling problems, incomplete polyphenols removal, loss of soluble protein, excessive cost, high water consumption or tedious processing, involving cumbersome and error-prone multi-steps (Sadeghi, Appu Rao, & Bhagya, 2006). An effective search for efficient and cheap means for obtaining light-coloured rapeseed protein still remains a challenge.

Although several experimental design investigations for vegetable protein extraction can be found in the literature, examples of optimisation to improve the extraction of light-coloured protein from rapeseed press-cake are scarce. Many studies from protein extraction literature illustrate the use of correlated responses. However, most authors choose to ignore the influence of the correlation structure that exists in the original data set. In view of the current discussions, the aim of this work is to apply a strategy based on CCD and PCA to find the suitable extraction parameters (extraction time, solvent:meal ratio, NaCl and sodium sulphite concentrations) that simultaneously optimises the protein yield and colour characteristics of the rapeseed protein, extracted under alkaline condition. The influence of these extraction parameters on the protein yield and colour parameters has had few arguments and studies. The choice of the extraction parameters and the influencing responses has been guided by a number of factors as indicated in the literature. "RSM-PCA-desirability function" trio for multiple responses has garnered appreciation in different fields of civil engineering and manufacturing technology; however, it is yet to be explored by the biological and food scientists. In the domain of food processing technology, very few articles have discussed optimisation of multiple correlated responses, probably due to complexity in the methodology involved. To the best of our knowledge, the idea of implementing Multiple Linear Regression (MLR) along with Response Surface Methodology (RSM)-Principal Component Analysis (PCA) is attempted for the first time in this paper. Moreover, the study mathematically proves the superior predictive ability of RSM-PCA combination method over RSM alone using experimental data in Section 3.3. The present investigation attempts a methodical approach for multiple responses, particularly when the responses are correlated.

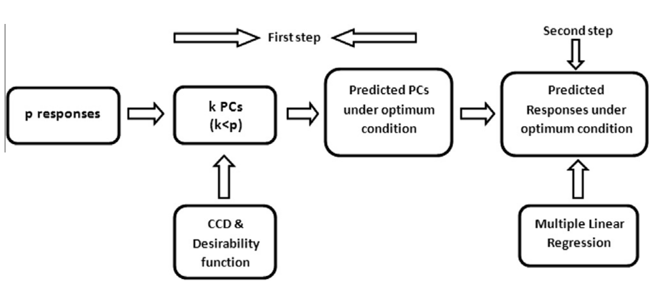

Fig. 1. Architecture of a multi-objective optimisation approach based on PCA, RSM and desirability function.

2. Materials and methods

2.1. Materials

Rapeseed press-cake was obtained from Assam Khadi and Village Industries Board, Guwahati, Assam, India. The press-cake was ground to pass through 60 mesh size sieve, and then stored at −18 °C for further analysis. All solvents and reagents were obtained from Merck® (India), of analytical grade. Bovine serum albumin (BSA) was procured from Sigma Chemicals Co. (St. Louis, MO, US).

2.2. Preparation of defatted, partially dephenolised meal (treated meal)

Ground meal was defatted using hexane:diethyl ether (1:1 v/v) solvent mixture for reducing the lipid content to <0.1% (by Soxhlet method). Since the dark colour of oilseed protein is mainly caused by its phenolic compounds, as a result of the removal of them the colour of the product is expected to become lighter. So, defatted meal was extracted with acidified methanol:acetone (1:1 v/v) mixture at a meal-to-solvent ratio of 1:20 (w/v) by mixing the suspension at 200 rpm for 2 h (at 25 °C) in an orbital shaker, according to our earlier report (Das Purkayastha et al., 2013). Suspension was then centrifuged (SIGMA 3–18 K Centrifuge) at 10,000 rpm for 20 min (at 4 °C), and the residue (treated meal) was dried in a vacuum oven under reduced pressure (150 mm Hg) at 35 ± 2 °C for 24 h and was ground again to pass through a 60 mesh sieve to obtain fine powder and then stored at −18 °C until use.

2.3. Optimisation of extraction parameters

Rapeseed protein was extracted with selected 31 combinations of independent variables such as extraction time (1–5 h), solvent:meal ratio (10:1–30:1 v/w), NaCl concentration (0–0.2 M)