Data Article

Dataset on aqueous solid-liquid extraction of gossypol from defatted cottonseed in acidic medium using green solvent, its kinetics and thermodynamics study and mass transfer effects

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A B S T R A C T

Extraction of gossypol from cottonseed is essentially required to produce cottonseed free from gossypol for animal feed and or human applications. The focus of the present research was to determine the percentage gossypol extraction after extracting the defatted cottonseed using environment friendly green solvent ethanol-water (95:5 v/v) acidified with 0.5 M oxalic acid. The cottonseed samples were taken according to the fixed solvent to seed ratio and were extracted in batch process using round bottom flasks maintained at required temperatures for different extraction times ranging from 5 to 180 mins. After extraction the samples were filtered and dried and subjected to total gossypol analysis using BIS method. One factor at a time (OFAT) experimental design was employed to optimize the different process parameters like acid type and concentration, solvent to seed ratio, temperature and contact time. The obtained data was studied for analysis of kinetics of extraction using three different kinetic models, calculation of activation energy, evaluating values of

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kinetic parameters and thermodynamic parameters. The data was also analyzed for evaluation of mass transfer effects viz. liquid film diffusion and internal solid diffusion and calculation of diffusion rate constants for the extraction of gossypol from cottonseed. The present dataset demonstrated the analysis of experimental data for determining the type of kinetics, thermodynamic parameters and mass transfer effects of the solvent extraction for future researchers.

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### Specifications Table

| Subject | Filtration and Separation |
|---------|---------------------------|
| Specific subject area | Chemical Engineering-Separation Technology |
| Type of data | Table and Figure |
| How data were acquired | UV-Vis Double beam Spectrophotometer (Systromic, model 2202) |
| Data format | Raw, analysed |
| Parameters for experimental data collection | The data was experimentally obtained at fixed pressure of 0.974 bar (731 mm Hg) and at respective temperatures as mentioned in the text i.e. figures and tables etc. |
| Description of Experimental Data Collection | The experimental data for solvent extraction was obtained using batch extraction and total gossypol was analysed using UV-Vis spectrophotometer using standard BIS method. |
| Data source location | Chandigarh-160014, India. |
| Data accessibility | Raw data is given in this article. |

### Value of the data

- Kinetics and mass transfer analysis is essential for any solid-liquid extraction process. This data provides methodology to apply kinetic models and their applicability in extraction, gives evaluation and effect of diffusion rate constants and effect of process parameters on kinetics.
- This data can be used by researchers/scientists/investigators who work in the field of solid-liquid extraction and separation science and/or technology.
- The mass transfer effect on extraction and evaluation of thermodynamic parameters has been elucidated in simple and descriptive manner which will be useful for all the fellow researchers.
- This dataset can be used as a tool to identify the kinetics of extraction and mechanisms affecting the solid-liquid extraction process.

### 1. Data Description

This dataset contains 12 Figures and 13 tables that represent the solid-liquid extraction data, kinetics of extraction, thermodynamics and diffusion rate constants for the extraction of gossypol from defatted cottonseed using ethanol-water (95:5v/v) solvent. One factor at a time (OFAT) experimental design was used to optimized the process parameters. Fig. 1 shows the effect of acid concentration on gossypol extraction using three different acids at 348 K and solvent to seed ratio (SR) of 15 in 180 min. Fig. 2 shows the effect of solvent to seed ratio on gossypol extraction using three different acids at 0.5 M conc., 348 K temperature and in extraction time of 180 mins. Fig. 3 shows the effect of temperature on gossypol extraction at different solvent to seed ratios. Fig. 4 shows effect of time at different temperatures on extraction at SR15 and 180
Fig. 1. Effect of acid concentration on gossypol extraction at 348 K, SR15 and 180 min.

Fig. 2. Effect of solvent to seed ratio using different acids (180 min, 348 K)

The average % error was found out to be 1.4%. The optimum conditions obtained were 0.5 M oxalic acid, SR 15, 348 K temperature and 180 min contact time.

Tables 1-4 correspond to raw data of Fig. 1 to Fig. 4. Fig. 5 shows the fitting of experimental data into pseudo first order kinetic model for gossypol extraction at SR 15 and 348 K. Table 5 gives the raw data for Fig. 5.

Table 6 gives the values of evaluated first order rate constant and extraction capacity at saturation. Fig. 6 shows the graph of extraction of gossypol (Ct) vs time at 348 K at SR 15. Fig. 7 shows the pseudo second order kinetics by plotting a graph between t/Ct and time. Tables 7 and 8 correspond to raw data of Fig. 6 and Fig. 7. Table 9 gives the values of kinetic parameters for gossypol extraction for pseudo second order model at SR15 and different temperatures. Fig. 8 describes the Elovich kinetics model and Table 10 corresponds to raw data of Fig. 8. Table 11 gives the values of Elovich kinetic model constants. Fig. 9 shows the plot of ln (k) vs
Fig. 3. Effect of temperature on gossypol extraction at different solvent to seed ratios using ethanol-water solvent acidified with 0.5 M oxalic acid in 180 min.

Fig. 4. Effect of time at different temperatures on extraction using Ethanol-water solvent acidified with 0.5 M oxalic acid at SR15 and 180 min

Table 1
Effect of acid concentration on gossypol extraction at 348 K, SR15 and 180 min.

| S. No. | Acid Concentration(M) | % Gossypol Extraction |
|--------|------------------------|------------------------|
|        |                        | Citric Acid | Phosphoric acid | Oxalic acid |
| 1      | 0.3                    | 78.94       | 84.2         | 87.71       |
| 2      | 0.4                    | 84.55       | 88.41       | 90.52       |
| 3      | 0.5                    | 89.11       | 92.98       | 93.33       |
| 4      | 0.6                    | 89.47       | 93.33       | 93.68       |
Table 2
Effect of solvent to seed ratio on gossypol extraction at 348 K, SR15 and 180 min.

| S. No. | Solvent to Seed Ratio (SR) | % Gossypol Extraction |
|--------|---------------------------|-----------------------|
|        |                           | Citric Acid | Phosphoric acid | Oxalic acid |
| 1      | 5                         | 77.88       | 80.69          | 84.55       |
| 2      | 10                        | 85.96       | 87.36          | 88.41       |
| 3      | 15                        | 90.52       | 92.98          | 93.33       |
| 4      | 20                        | 90.87       | 93             | 93.68       |

Table 3
Effect of temperature on gossypol extraction at different solvent to seed ratios with Ethanol-water acidified with 0.5 M oxalic acid in 180 min

| S. No. | Temperature (K) | % Gossypol Extraction (mean values) |
|--------|----------------|-------------------------------------|
|        |                | SR-5 | SR-10 | SR-15 | SR-20 |
| 1      | 318            | 67.71| 73.67 | 83.5  | 83.85 |
| 2      | 328            | 73.32| 81.39 | 85.61 | 86.31 |
| 3      | 338            | 80.69| 85.00 | 90.87 | 91.92 |
| 4      | 348            | 84.55| 88.41 | 93.33 | 93.7  |

Table 4
Effect of time at different temperatures on extraction at SR15 and 180 min

| S. No. | Time (min) | % Gossypol Extraction with Temperature (K), mean values |
|--------|------------|--------------------------------------------------------|
|        |            | 318 K | 328 K | 338 K | 348 K | 358 K |
| 1      | 30         | 65.95 | 72.62 | 75.78 | 77.53 | 78.6  |
| 2      | 60         | 70.86 | 77.53 | 81.04 | 83.15 | 84.9  |
| 3      | 120        | 77.88 | 81.74 | 86.31 | 89.11 | 89.8  |
| 4      | 180        | 83.5  | 85.61 | 90.87 | 93.33 | 94.03 |

Fig. 5. Pseudo first order kinetics of gossypol extraction at SR 15 and 348 K

1/T to evaluate activation energy. Fig. 10 describes the Vantt Hoff's plot i.e. ln (K_e) vs 1/T for evaluating thermodynamic parameters for gossypol extraction. The raw data for Fig. 9 (value of k) and Fig. 10 (value of K_e) had been taken from Tables 9 and 12 and used after taking natural logarithm of k and K_e values respectively. Table 12 gives values of thermodynamic parameters for gossypol extraction. Fig. 11 describes mass transfer effect i.e. intraparticle diffusion model depicting plot of C_t vs t^{1/2}. Fig. 12 explains the mass transfer mechanism of solid-liquid extrac-
Table 5
Pseudo first order kinetics data for gossypol extraction at 348 K and SR15

| S. No. | Time (min) | Log (Ce-Ct) |
|--------|------------|-------------|
| 1      | 5          | −0.58486    |
| 2      | 15         | −0.72285    |
| 3      | 30         | −0.92702    |
| 4      | 60         | −1.11748    |
| 5      | 120        | −1.49894    |
| 6      | 180        | −2.52288    |

Table 6
Rate constant (k) and extraction capacity (Ce) using pseudo first order model

| SR, mL/g | Temp, K | Slope (x 10^2) | k (min⁻¹) | Intercept | Ce (mg mL⁻¹) | R²    |
|----------|---------|----------------|-----------|-----------|--------------|-------|
| 15       | 348     | −1.01          | 0.0233    | −0.5358   | 0.2912       | 0.9603|

Fig. 6. Plot of extraction of gossypol (Ct) vs time at 348 K at SR 15 for Ethanol-water (95:5)-0.5 M oxalic acid

tion. The raw data for Fig. 11 had been used from Table 7. Table 13 gives the values of diffusion rate constants i.e. liquid film diffusion constant and internal solid diffusion constant (Table 8).

2. Experimental Design, Materials, and Methods

2.1. Experimental design

One factor at a time (OFAT) experimental design was used in this work to optimize the process parameters i.e. acid type and concentration, solvent to seed ratio, temperature and contact time. The obtained experimental data was analysed to determine the kinetics of extraction, thermodynamic parameters and mass transfer effect.
Table 7
Plot of extraction of gossypol (Ct) vs time at 348 K at SR 15

| S. No. | Time (min) | Ct  |
|-------|------------|-----|
| 1     | 0          | 0   |
| 2     | 5          | 0.4369 |
| 3     | 15         | 0.5077 |
| 4     | 30         | 0.5787 |
| 5     | 60         | 0.6207 |
| 6     | 120        | 0.6653 |
| 7     | 180        | 0.6967 |

Table 8
Pseudo second order kinetics of gossypol extraction at different temperatures (t/Ct vs time)

| S. No. | Time (min) | t/Ct (min mL/mg) |
|--------|------------|------------------|
|        |            | 318 K 328 K 338 K 348 K |
| 1      | 30         | 60.97 55.36 53.04 51.84 |
| 2      | 60         | 113.47 103.68 99.19 96.67 |
| 3      | 120        | 206.44 196.67 186.25 180.37 |
| 4      | 180        | 288.79 281.67 265.32 258.33 |

Table 9
Kinetic parameters for gossypol extraction for pseudo second order model at SR15

| Temp, K | Ei (mg mL⁻¹ min⁻¹) | k(mLmg⁻¹ min⁻¹) | Ce (mg/mL⁻¹) | R²  |
|---------|-------------------|-----------------|--------------|-----|
| 318     | 0.0630            | 0.1513          | 0.6329       | 0.9996 |
| 328     | 0.0752            | 0.1609          | 0.6711       | 0.9993 |
| 338     | 0.0824            | 0.1694          | 0.6976       | 0.9997 |
| 348     | 0.0893            | 0.1784          | 0.7155       | 0.9998 |

Table 10
Elovich kinetics plot (ln(t) vs Ct) for kinetics of gossypol extraction at 348 K

| S. No. | Ct  | Ln (t) |
|--------|-----|--------|
| 1      | 0.4369       | 1.609438 |
| 2      | 0.5077       | 2.70805  |
| 3      | 0.5787       | 3.401197 |
| 4      | 0.6207       | 4.094345 |
| 5      | 0.6653       | 4.787492 |
| 6      | 0.6967       | 5.192957 |

Table 11
Elovich kinetic model constants

| SR, mL/g | Temp, K | Slope | β (mLmg⁻¹) | Intercept | α (mg mL⁻¹ min⁻¹) | R²  |
|----------|---------|-------|------------|-----------|-------------------|-----|
| 15       | 348     | 13.62 | 0.0734     | −4.3304   | 9.917             | 0.9952 |

Table 12
Thermodynamic parameters for extraction of gossypol employing Ethanol-water (95:5 v/v) solvent acidified with 0.5 M oxalic acid at SR 15.

| Temperature K | Equilibrium constant (Ke) | Gibbs free energy (ΔG°) J/mol | ΔH° J/mol | ΔS° J/mol K |
|---------------|--------------------------|-----------------------------|-----------|-------------|
| 318           | 1.1160                   | −290.031                    | 8333.122  | 27.14       |
| 328           | 1.2358                   | −577.304                    |           |             |
| 338           | 1.3505                   | −844.445                    |           |             |
| 348           | 1.4647                   | −1104.07                    |           |             |
Fig. 7. Pseudo second order kinetics of gossypol extraction at different temperatures \((t/C_t \text{ vs time})\) for Ethanol-water (95:5)-0.5 M oxalic acid at SR15

\[ y = 13.627x - 4.3304 \]
\[ R^2 = 0.9952 \]

Fig. 8. Elovich kinetics model for kinetics of gossypol extraction at 348 K \((\ln(t) \text{ vs } C_t)\) for Ethanol-water (95:5)-0.5 M oxalic acid

| Table 13 | Mass transfer model rate constants for gossypol extraction |
|----------|----------------------------------------------------------|
| Gossypol extraction; **SR** (at 348 K) | Diffusion rate constants, \((\text{mg/ml} \cdot \text{min}^{0.5})\) |  |
| Liquid film diffusion constant | Internal solid diffusion rate constant |  |
| 15 mL/g |  |  |
| \(K_1\) | 0.0437 | 0.0134 |
| Intercepts on x-axis | 0.3388 | 0.5172 |
2.2. Materials

Cotton variety used was RCH-776- BT cotton (G. Hirsutum) hybrid variety procured from local market. Ethanol, 3-amino-1-propanol, glacial acetic acid, oxalic acid, citric acid, phosphoric acid and N, N dimethyl formamide were purchased from Merck Specialities Private Ltd, India. All stock solutions were prepared using double distilled water made in laboratory. Gossypol standard was purchased from Sigma Aldrich, India. All chemicals used were of analytical grade.

2.3. Gossypol extraction procedure

Known amount of defatted cottonseed sample with ethanol-water (95:5 v/v) solvent at desired solvent to seed ratio was taken in a flat bottom flask. The mixture was extracted at desired
temperature (318, 328, 338 and 348 K) using temperature controlled hot plate kept in glass (closed) enclosure, a stir bar (250 rpm) was utilized for proper contact. After known periods of extraction time the sample was filtered using buchner funnel. The filtered sample was then dried at a temperature of 50 °C using a convection oven for 12 hours. The dried sample was then analysed for total gossypol using UV-Vis Double beam Spectrophotometer.

2.4. Analysis of Total Gossypol

The analysis of total gossypol content was determined using UV-Vis Double beam Spectrophotometer as per BIS standard method IS: 4876-1986 [1,2]. The percentage gossypol extraction was calculated from total gossypol content of the sample extracted and initial seed.

2.5. Kinetics and thermodynamics of gossypol extraction

The kinetics of gossypol extraction was analysed using three different models namely pseudo first order model, Elovich model and pseudo second order model as thoroughly explained by
H.A. Harouna-Oumarou et al., 2007 [3] and other researchers [2–8]. The thermodynamic parameters were evaluated as discussed by Singh et al., 2019 [2] and others [6,9–10].

2.6. Green solvent

Ethanol–water was chosen as green solvent as it is environment friendly and also ethanol qualifies as a green solvent as per CHEM21 solvent selection guide [11].

The values of activation energy, E and specific rate constant, $k_0$ were calculated from the slope and intercept of graphical plot between $\ln(k)$ and $1/T$ as shown in Fig. 9 [2]. The obtained values of $k_0$ and E were 0.012 mLg$^{-1}$min$^{-1}$ and 5.021 kJ mol$^{-1}$ respectively.

2.7. Mass transfer effects

The effect of mass transfer on solid-liquid extraction of gossypol was analysed using intra-particle diffusion model [12] and mechanism of solid-liquid extraction as per discussed by Harouna-Oumarou et al., 2007 [3].

2.7.1. Mass transfer model

To study the effect of mass transfer on solid liquid extraction kinetics of Ethanol-water system a graph of $C_t$ vs $t^{1/2}$ (intra-particle diffusion model) at optimum conditions was plotted as shown in Fig. 11.

The first part of the curve is attributed to liquid film effect i.e. liquid film diffusion (slope $K_1$) taking place at solid-liquid interface, while the second linear part indicates internal solid diffusion (slope $K_2$). The diffusion rate parameters $K_1$ and $K_2$ as obtained are shown in Table 5. The diffusion rate parameters indicate that the internal solid diffusion controls the extraction rate; which is the slowest step in extraction.

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Conflict of Interest

The authors declare no conflict of interest.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.dib.2020.105620.

References

[1] Bureau of Indian Standards, Specification for edible cottonseed flour (solvent extracted), BIS, 1986, pp. 1–16. August 1986 https://ia800409.us.archive.org/8/items/gov.in.is.4876.1986/is.4876.1986.pdf (Accessed on 24 January 2020).
[2] Surinder Singh, S.K. Sharma, S.K. Kansal, Batch extraction of gossypol from cottonseed meal using mixed solvent system and its kinetic modeling, Chemical Engineering Communications 206 (12) (2019) 1608–1617 doi:org/10.1080/00986445.2018.1558214.
[3] H.A. Harouna-Oumarou, H. Fauduet, C. Porte, Y.S. Ho, Comparison of kinetic models for the aqueous solid-liquid extraction of Tilia sapwood a continuous stirred tank reactor, Chem. Eng. Commun 194 (4) (2007) 537–552, doi:10.1080/00986440600992511.

[4] Y.S. Ho, C. McKay, Pseudo-second order model for sorption processes, Process Biochem 34 (5) (1999) 451–465, doi:10.1016/S0032-9592(98)00112-5.

[5] E. Simeonov, I. Tsibranska, A. Minchev, Solid-liquid extraction from plants- experimental kinetics and modelling, Chem. Eng. J 73 (3) (1999) 255–259 doi:org/10.1016/S1385-8947(99)0030-3.

[6] Y.-S. Ho, Removal of copper ions from aqueous solution by tree fern, Water Research 37 (2003) 2323–2330.

[7] L. Rakotondramasy-Rabesiaka, J.L. Havet, C. Porte, H. Fauduet, Solid-liquid extraction of protopine from Fumaria officinalis L.-Analysis determination, kinetic reaction and model building, Sep. Purif. Technol 54 (2) (2007) 253–261, doi:10.1016/j.seppur.2006.09.015.

[8] L. Prat R.Wongkittipong, S. Damronglerd, C. Gourdon, Solid-liquid extraction of andrographolide from plants-experimental study, kinetic reaction and model, Sep. Purif. Technol 40 (2) (2004) 147–154 doi:10.1016/j.seppur.2004.02.002.

[9] D.K. Saxena, S.K. Sharma, S.S. Sambi, Kinetics and thermodynamics of gossypol extraction from defatted cottonseed meal by ethanol, Polish J. Chem. Technol 14 (2) (2012) 29–34, doi:10.2478/v10026-012-0067-4.

[10] D.K. Saxena, S.K. Sharma, S.S. Sambi, Kinetics and thermodynamics of gossypol extraction from defatted cottonseed meal by ethanol Acidified by Oxalic acid, Int. J. Sci. Res 4 (8) (2015) 1967–1971.

[11] D. Prat, A. Wells, J. Hayler, H. Sneddon, C.R. Mcelroy, S. Abou-shehada, P.J. Dunn, CHEM21 selection guide of classical- and less classical-solvents, Green Chem 18 (2016) 288–296, doi:10.1039/c5gc01008j.

[12] Kelvin O. Yoro, Mutiu K. Amosa, Patrick T. Sekoai, Jean Mulopo, O. Michael, Daramola Diffusion mechanism and effect of mass transfer limitation during the adsorption of CO2 by polyaspartamide in a packed-bed unit, International Journal of Sustainable Engineering (2019), doi:10.1080/19397038.2019.1592261.