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Application of Taguchi method in activated carbon adsorption process of phenol removal from ceramic gasifier wastewater

Jagniyant Lunagariya[a], Karan Chabhadiya[b], Pankaj Pathak[c,*], Deepak Mashru[d]

[a] Department of Civil Engineering, Atmiya University, Rajkot, Gujarat 360005, India
[b] Research & Development, Vardhman Environmental Consultancy, Rajkot, Gujarat 360004, India
[c] Department of Environmental Science, SRM University-AP, Amaravati, Andhra Pradesh 522502, India
[d] Department of Communication Skills, Marwadi University, Rajkot, Gujarat 360003, India

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ABSTRACT

Ceramic gasifier wastewater (CGWW) contains organic toxic substances such as phenolic compounds and direct disposal of CGWW into the ground and surface water causes severe environmental impacts. Therefore, phenol removal from CGWW is utmost important to safeguard the natural water resources. Adsorption technique using activated carbon is employed to lessen the impacts of phenol. However, the pollution load on CGWW is very high and necessitates pretreatment using coagulation-flocculation process. Further, the sanctity of each parameter on phenol removal has to study by varying all-dominating parameters simultaneously and determined its significance. Therefore, Taguchi's L9 orthogonal array (OA) design is used in this study to optimize the carbon adsorption process on CGWW. The most governing parameters are time (30–60 min), temperature (30–60 °C) and liquid to solid, L/S (5–15) ratio and it is prioritized in the following order L/S>Temperature>Time. The optimized experimental conditions, i.e., time 30 min., temperature 45 °C at L/S 5 was predicted and that demonstrated 96% phenol removal from CGWW. The predicted results of Taguchi's OA is compared with an experimental analysis showed a good agreement ($R2 > 0.97$) with each other and vital significance. Accordingly, it is concluded that the Taguchi method is a promising, efficient, and cost-effective technique for the industry to minimize the experiments and maximize the phenol removing efficiency.

1. Introduction

Coal gasifiers are predominantly used in developing countries to produce conventional fuel gases (Kamble et al., 2019). However, a huge amount of hazardous and toxic organics such as phenols and polycyclic aromatic hydrocarbons (PAHs) are released during the gasification process, causing adverse environmental impacts (Ji et al., 2015, Sagbas et al., 2014). To mitigate this issue, generally, scrubbers are employed in the ceramic industry to remove organic pollutants from the coal gas and convert gaseous organics into liquid waste (Zhao et al., 2017, Ramakrishnan and Surampalli, 2012). The liquid waste has a high phenol concentration (4×10^3–8×10^3 mg/L) and is known as coal gasifier wastewater (CGWW). Discharging this wastewater into the ground causes severe geo-environmental hazards due to the presence of 60×10^3–90×10^3 mg/L of chemical oxygen demand (COD), 20×10^3–1×10^5 mg/L of total dissolved solids (TDS) (Adav and Lee, 2008, Wang et al., 2018, Jeong and Chung, 2006). However, the regulation made by the Ministry of Environment, Forests and Climate Change (MOEFCC) of India, stated that the maximum permissible limit of phenol in the industrial discharge should be at 1 mg/L, corresponding to the EPA standard. Therefore, the treatment of CGWW is of higher priority for industrial associates due to the strict government norms. Several physicochemical and biological treatment technologies such as acidification, coagulation, activated carbon adsorption, anaerobic hybrid reactor (AHR), anaerobic filters (AF), upflow anaerobic sludge blanket digestion (UASB), aerobic treatment such as cyclic activated sludge (CAS), membrane bioreactor (MBR), moving bed biofilm reactor (MBBR) and fluidized-bed bioreactor is employed to treat the CGWW (Ji et al., 2015, Adav and Lee, 2008, Fang et al., 2013, Gai et al., 2008, Hu et al., 2016, Maiti et al., 2019, Borghei and Hosseini, 2004). Amongst all, the physiochemical treatment is demonstrated to be rapid and easy compared to biological methods, but phenol removal efficiency is higher in biological methods (Hao et al., 2018, Hernandez and Edyvean, 2008). However, both treatment methods are susceptible to the characteristics of wastewater, and hence it is not adequate for industrial applications. However, industries require a quick, robust, and easy method for CGWW treatment; hence Taguchi analysis is used to design an experiment to achieve the targeted outcomes with minimum efforts (Pathak et al., 2014, Pathak et al., 2016, Phadke, 2008).

* Corresponding author.
E-mail address: pankajpathak18@gmail.com (P. Pathak).

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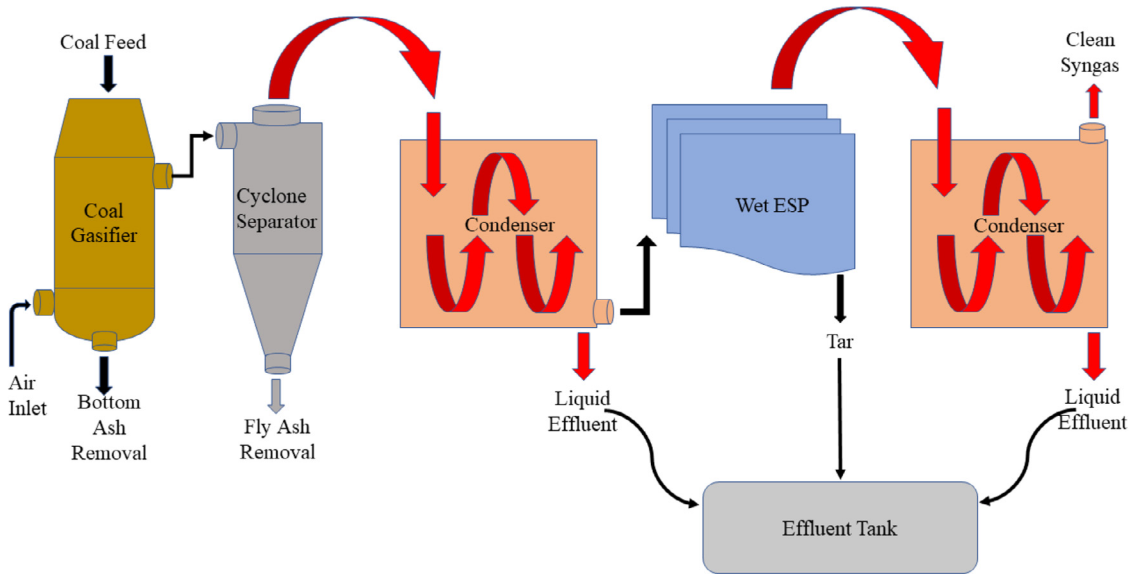

Fig. 1. Schematic diagram of coal gasification system.

Keeping in view, physiochemical treatment, i.e., carbon adsorption method, is designed to remove phenol from CGWW. Preceding this, coagulation-flocculation is performed to pre-treat the CGWW. Furthermore, it is reported that several parameters, viz., pH, interaction time, temperature, liquid to solid ratio (L/S), were identified, which has a significant influence on phenol removal (Zhao and Liu, 2016, Ramakrishnan and Gupta, 2006, Ramakrishnan and Surampalli, 2013). Identifying the influence of each parameter on phenol removal is a mandatory but very laborious task and necessitates performing sensitivity analysis (Phadke, 2008, Taguchi et al., 2005). Therefore, L9 orthogonal array (OA) of the Taguchi method was performed to understand the sanctity of each parameter on phenol removal. Further, experimental conditions were optimized to determine the robust and accessible approach for maximum phenol removal.

2. Description of study area

The Morbi city, Saurashtra region of Gujarat, India (22.8252° N, 70.8491° E) is selected as a study area where ~700 ceramic manufacturing industries of Morbi city, Gujarat, India contributes more than 90% of the total ceramic product of India. However, ceramic industries are causing severe air and water pollution in the environment. This study deliberates treating the wastewater obtained from ceramic coal gasifiers. Two condensers have been introduced in the outlet of the ceramic industry with an electrostatic precipitator scrubbing process to convert gaseous waste into liquid waste. During the scrubbing process, first, the gas passes through the primary condenser, where the temperature is set to condense moisture from the gas. The outlet of the primary condenser is wastewater.

Further, producer gas is sent to an electrostatic precipitator where tar is being captured, and then gas passes through the secondary condenser to remove the remaining moisture contents. After removing moisture and tar, producer gas is sent to the kiln for combustion. The wastewater generated from the primary condenser, electrostatic precipitator, and secondary condenser is collected in the effluent collection tank, as depicted in Fig. 1 (Cui et al., 2020, Lunagariya et al., 2019).

The physical and chemical characterizations of ceramic gasifier wastewater (CGWW) are carried out, and obtained results are presented in Table 1. Prior to chemical adsorption with activated carbon, preliminary treatment was done on CGWW using various coagulating agents such as Alum, $FeCl_3$, $FeSO_4$, $AlCl_3$, and non-ionic polyelectrolyte. The prime objective of the coagulation process is to reduce the total solids from the wastewater. All the chemicals used in the coagulation process are purchased from merck analytical grade, and commercial-grade quality activated carbon was used in the study, is from chemdyes corporation, India.

Table 1
Characteristics of waste water

Characteristics	Concentration (mg/L)
TDS	100000
TSS	1500
BOD	3000
COD	90400
Phenols	6122

3. Experimental

3.1. Characteristics of wastewater

Physical and chemical characteristics have been studied by following the standard method of USEPA 1684 (2001) (USEPA 1684, 2001), where the standard gravimetric method is employed to obtain total solids and total dissolved solids in CGWW (APHA, 5530), and results are presented in Table 1. On the other hand, phenol concentration is determined by using a UV-Visible spectrophotometer (Model TS2080Plus, Analytical Technologies Limited). The obtained results are presented in Table 1. Based on the primary characterization of wastewater, it is estimated that CGWW is very complex in nature.

3.2. Pre-treatment of CGWW

The jar test is used to pretreat the CGWW using different types of coagulating agents. In this test, four beakers of 1000 ml capacity were used, and each beaker is filled with 500 ml of CGWW. The desired pH values were achieved by adding NaOH and sulphuric acid as per the requirement after introducing coagulants. The coagulation process in the CGWW is done at 120 rpm for 1 minute, and then flocculation is started at 20 rpm for 20 minutes so that formed flocs should not break during mixing. Further, CGWW was kept for 2 hours to settle particles in it. Before and after the settlement of particles, turbidity and total dissolved solids (TDS) are determined using turbidity meter and TDS meter. It is found that $AlCl_3$ was found suitable to minimize the turbidity and total

Table 2
Value of Turbidity (in NTU) after pre-treatment at various pH and coagulating dose (in mg/L)

pHDose	5	5.5	6	6.5	7	7.5	8
1	46	37	34	38	81	51	64
1.5	48	37	72	36	82	49	68
2	**10**	**22**	**58**	**44**	**74**	**45**	**31**
2.5	12	69	61	56	61	62	59

Table 3
Pre-treatment analysis for removal of total solid (%) at various pH and coagulating dose (in gm/L)

pHDose	5	5.5	6	6.5	7	7.5	8
1	1	35	25	1	17	6	2
1.5	**2**	**61**	**1**	**6**	**13**	**7**	**7**
2	**4**	**46**	**61**	**7**	**19**	**8**	**6**
2.5	0	46	2	0	10	19	9

Table 4
Sensitivity analysis experiment design for L9 orthogonal array

Exp. No.	Input Parameters Time (min)	Temperature (°C)	L/S	Output Parameter Phenol Removal (%)
1	30	30	5	91.85
2	30	45	10	87.56
3	30	60	15	88.32
4	45	30	10	70.41
5	45	45	15	92.07
6	45	60	5	91.64
7	60	30	15	89.39
8	60	45	5	90.25
9	60	60	10	87.89

solids amongst all the coagulants. On the other hand, polyelectrolyte and alum also gave satisfactory results to settle the particles, but wastewater became viscous, and settling time was more as compared to $AlCl_3$. Thus, it cannot be used for the further treatment process.

Therefore, based on the observation, $AlCl_3$ is considered for further study to determine the optimum dose to remove total solids. The pH and $AlCl_3$ dose varied from 5–8 and 1–2.5 gm/L using Jar test and obtained results were presented in Tables 2 and 3; where initial turbidity of wastewater was 381 NTU (nephelometric turbidity unit).

3.3. Sensitivity analysis on carbon adsorption method for phenol removal

Sensitivity analysis using the Taguchi method was performed to design the experiment for phenol removal. Based on literature data, it was found that three parameters, i.e., time, temperature, and L/S, significantly influence phenol removal. Therefore, Taguchi's L9 orthogonal array (OA) is used to design experiments where nine experiments are conducted with three different parametric conditions, as shown in Table 4. Though, it should be noted that for examining the influence of these three parameters on the phenol removal, 3^3 (=27) experiments combination are needed, which is an arduous task. Hence, establishing the sensitivity of phenol removal is required on the characteristics of various parameters, as discussed above.

Taguchi's larger the better option is selected (refer Eq. 1) to analyze experimental conditions for phenol removal (Taguchi et al., 2005). The analysis of mean (ANOM) and analysis of variance (ANOVA) are determined by employing Minitab®18 software (Minitab 18.2.4 Inc., Pennsylvania) (Minitab 18.2.4, 2018), the input parameters of experiments are given in Table 4, where each input parameter has three levels, and each level comes three times in the OA. ANOM and ANOVA are used to prioritize the parameters and predict the best experimental conditions for maximum phenol removal (Pathak et al., 2014, Pathak et al., 2016).

$$\text{Larger the better characteristics}: \frac{S}{N} = -10 \cdot \log_{10} \cdot \frac{1}{n_R} \cdot \left[\sum \frac{1}{y^2}\right] \quad (1)$$

where, S and N refer to the signal (mean) and noise (standard deviation) of the input parameters, respectively, y is the data obtained from the experiments (i.e., the observed output) and n_R is the number of replicates (or observations). The S/N describes the extent of variability in the different parameters on phenol removal. Further, an additive model given by Taguchi et al. (2005) is used to forecast input parameters' influence on the target (phenol removal) referred as to y (Taguchi et al., 2005, Pathak et al., 2021). The additive model is shown in Eq. 2 and 3.

$$\eta\ (A_i, B_j, C_k, D_l, F_m) = \mu + a_i + b_j + c_k + d_l + e_m + \ldots + \varepsilon \quad (2)$$

$$\eta = -10 \cdot \log\left(\frac{1}{y^2}\right) \quad (3)$$

where, η is the objective function, μ is the overall mean of the signal to noise ratio (S/N) for the experimental region, a_i, b_j, c_k, d_l and e_m refer to the deviation from μ caused by the factors A, B, C, D and E respectively, subscripts i, j, k, l and m are the different levels and ε is experimental error.

Furthermore, activated carbon adsorption studies were conducted as per the orthogonal array-design of the experiment (OA-DOE) shown in Table 4. The 100 mL scale of a round-flat bottom 3-neck flask of 250 mL in capacity was used in the adsorption study. The pH electrode and sampling point were static with the flask and kept on a hotplate *cum* magnetic stirrer for heating (conditions designed) and stirring (250 rpm) (30). After the completion of the carbon adsorption experiment, the solution was filtered, and the filtrate was studied using UV-visible spectrophotometer for determining the phenol removal efficiency, as follows:

$$\%\text{Phenol Removal} = \left(\frac{C_L}{C_S}\right) \times 100 \quad (4)$$

where C_S and C_L are the phenol contents in the mass of carbon sample and output volume of filtrate, respectively.

4. Results and discussions

4.1. Effect of coagulant dose

Turbidity is effectively measured by adding coagulating agents into wastewater. It is observed from Table 2 that with increasing coagulating dose, turbidity was decreasing up to 2 gm/L of dose, and after then turbidity was increasing. It also indicates that turbidity removal is maximum in acidic conditions due to the dissolution of a colloidal particle at lower pH. The highest turbidity removal i.e., 10 NTU, was achieved at pH five by using a 2 gm/L dose of $AlCl_3$. On the other hand, total solid removal was also considered as a measuring parameter for the effectiveness of the coagulation and flocculation process. It was determined that the maximum solids removal was achieved in an acidic medium. The 61% removal of solids was achieved in two different cases: (1) pH 6 and 2 gm/L dose of coagulating agent, (2) pH 5.5 and 1.5 gm/L dose of coagulating agent (refer to Table 3). Considering both turbidity and total solid removal, 2 gm/L optimum coagulant dose was maintained at pH 6 for this study.

Further initial pre-treatment and different treatment techniques were given to wastewater. Subsequently, phenol concentration was determined on the pre-treated wastewater using UV-Visible Spectrophotometer. It is noticed that only 3.21% phenol was removed by using Eq. 4 through the pre-treatment process, which indicated that the removal of solids from wastewater shows an insignificant effect on phenol removal.

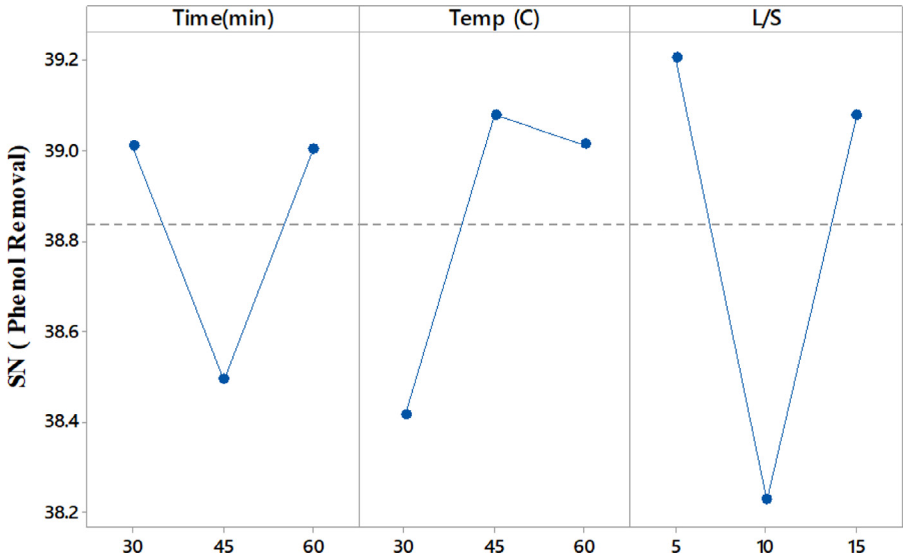

Fig. 2. Sensitivity analysis of phenol removal using L9 orthogonal array with larger the better options.

Table 5
Response Table for Signal to Noise Ratios

Parameter	Response (Average) value 1	2	3	Rank
Time (min)	39.01	38.49	39.20	3
Temperature (°C)	38.41	39.08	39.01	2
L/S	39.20	38.23	39.08	1

4.2. *Taguchi method*

Taguchi method is used to optimize the parameters viz., time, temperature, and L/S, which directly depend on phenol removal. The larger the better option of Taguchi method is selected as shown in Eq. 1 to compute S/N ratio and output parameter i.e., the measured phenol removal of each experiment of Taguchi's L9 OA is shown in Table 4. The prime objective of this study is to remove maximum phenol from CGWW; hence larger the better option was selected. Further, additive model (refer Eqs. 2 and 3) are employed to determine the influence of individual input parameters on removing phenol concentration. This analysis is termed as determination of the factor effect, and the results are presented in Table 5 and Fig. 2. The dotted line is shown in the figure represents the overall mean (μ) of the measured parameters. Further, S/N of phenol concentration is plotted on the Y-axis while the input parameters and their levels are plotted on the X-axis. It can be observed from the trends depicted in Table 5 that L/S ratio is the most influential parameter amongst all considered in this study, and the prioritization sequence is shown as L/S > T > Time. Further, the best experimental condition is predicted by Taguchi analysis to determine the maximum phenol recovery. Based on the prediction, the experiment is performed at L/S 5 in 30 minutes and 45 °C temperature yields 96% phenol removal. The experimental results and predicted results were compared and showed good agreement with each other i.e. R^2 0.97. The L/S ratio was found to be prominent amongst the other parameters due to the complexity of the phenolic compounds, and temperature is the second dominating parameter. It has been demonstrated that as temperature increases, the average kinetic energy of the molecules will effectively collide and settle. The optimized temperature was found 45 °C after the rate of reaction decreases with increasing temperature. Further, interaction time at 30 minutes plays a vital role in phenol removal. The reaction rate fastens at 30 minutes, and later phenol removal rate decreases.

Conclusions

The maximum amount of phenol is removed from CGWW using the carbon adsorption method. Preceding this coagulation-flocculation process is optimized with 2 g/L dose of $AlCl_3$ agent at pH 5 to remove total solids from CGWW. Accordingly, 61% of total solids and 3.21% of phenol were removed in this process. Further, the L9 OA of the Taguchi method is used to design the adsorption method by varying time, temperature and L/S with the minimum number of experimentation. Based on Taguchi's prediction, it has been found that 96% (R^2 0.97) of phenol can be removed at L/S 5 when interaction time is 30 minutes and temperature 45°C. This study provides optimized parameters for the efficient removal of phenol and can be effectively used in the ceramic industry.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

Adav, S.S., Lee, D.J., 2008. Physiological characterization and interactions of isolates in phenol-degrading aerobic granules. Appl. Microbiol. Biotecnol. 78, 899–905. doi:10.1007/s00253-008-1370-0.
APHA, 5530. Standard methods for the examination of water and wastewater.
Borghei, S.M., Hosseini, S.H., 2004. The treatment of phenolic wastewater using a moving bed bio-reactor. Process. Biochem. 39, 1177–1181. doi:10.1016/S0032-9592(02)00195-4.
Cui, H., Huang, X., Yu, Z., Chen, P., Cao, X., 2020. Application progress of enhanced coagulation in water treatment. RSC Adv. 10, 20231–20244. doi:10.1039/D0RA02979C.
Fang, F., Han, H., Zhao, Q., Xu, C., Zhang, L., 2013. Bioaugmentation of biological contact oxidation reactor (BCOR) with phenol-degrading bacteria for coal gasification wastewater (CGW) treatment. Bioresour. Technol. 150, 314–320. doi:10.1016/j.biortech.2013.09.119.
Gai, H., Jiang, Y., Qian, Y., Kraslawski, A., 2008. Conceptual design and retrofitting of the coal-gasification wastewater treatment process. Chem. Eng. J. 138, 84–94. doi:10.1016/j.cej.2007.05.032.
Hao, Z., Yuxing, H., Chunyan, X., Hongjun, H., Weiwei, M., 2018. Overview of the state of the art of processes and technical bottlenecks for coal gasification wastewater treatment. Sci. Total Environ. 637–638, 1108–1126. doi:10.1016/j.scitotenv.2018.05.054.

Hernandez, J.E., Edyvean, R.G.J., 2008. Inhibition of biogas production and biodegradability by substituted phenolic compounds in anaerobic sludge. J. Hazard. Mater. 160, 20–28. doi:10.1016/j.jhazmat.2008.02.075.

Hu, X., Chen, K., Lai, X., Yao, Q., Ji, S., Kaiser, K., 2016. Treatment of pretreated coal gasification wastewater (CGW) by magnetic polyacrylic anion exchange resin. J. Environm. Chem. Engg. 4, 2040–2044. doi:10.1016/j.jece.2016.02.018.

Jeong, Y.S., Chung, J.S., 2006. Simultaneous removal of COD, thiocyanate, cyanide and nitrogen from coal process wastewater using fluidized biofilm process. Process. Biochem. 41, 1141–1147. doi:10.1016/j.procbio.2005.12.010.

Ji, Q., Tabassum, S., Yu, G., Chu, C., Zhang, Z., 2015. Determination of biological removal of recalcitrant organic contaminants in coal gasification wastewater. Environ. Technol. 36, 2815–2824. doi:10.1080/09593330.2015.1049215.

Kamble, A.D., Saxena, V.K., Chavan, P.D., Mendhe, V.A., 2019. Co-gasification of coal and biomass an emerging clean energy technology: status and prospects of development in Indian context. Internation. J. Min. Sci. Technol. 29, 171–186. doi:10.1016/j.ijmst.2018.03.011.

Lunagariya, J., Gulabani, A., Mehta, N., Dhamani, N., Pathak, P., 2019. Biodegradation of phenolic compound from coal gasification wastewater, national environmental conference (NEC-2019). IIT, Bombay, pp. 1–4.

Maiti, D., Ansari, I., Rather, M.A., Deepa, A., 2019. Comprehensive review on wastewater discharged from the coal-related industries – characteristics and treatment strategies. Water Sci. Technol. 79, 2023–2035. doi:10.2166/wst.2019.195.

Minitab 18.2.4, 2018. Minitab, Inc., State College, PA, USA. https://www.minitab.com/en-us/products/minitab/ accessed 14 December 2020.

Pathak, P., Singh, D.N., Pandit, G.G., Apte, P.R., 2014. Establishing sensitivity of distribution coefficient on various attributes of soil-contaminant system. J. Hazard. Toxic Radioactive Waste 18, 64–75. doi:10.1061/(ASCE)HZ.2153-5515.0000199.

Pathak, P., Singh, D.N., Apte, P.R., Pandit, G.G., 2016. Statistical analysis for prediction of distribution coefficient (kd) of soil-contaminant system. J. Environ. Eng. 142, 1–11. doi:10.1061/(ASCE)EE.1943-7870.0001010.

Pathak, P., Singh, V.K., Chabhadiya, K., 2021. Sequential leaching of strategic metals from LNCM cathode material using organic and mineral acid lixiviants. J. Metals 73, 13861394. doi:10.1007/s11837-021-04631-z.

Phadke, M.S., 2008. Quality engineering using robust design. Prentice Hall Delhi, India.

Ramakrishnan, A., Gupta, S.K., 2006. Anaerobic biogranulation in a hybrid reactor treating phenolic waste. J. Hazard. Mater. 137, 1488–1495. doi:10.1016/j.jhazmat.2006.04.034.

Ramakrishnan, A., Surampalli, R.Y., 2012. Comparative performance of UASB and anaerobic hybrid reactors for the treatment of complex phenolic wastewater. Bioresour. Technol. 123, 352–359. doi:10.1016/j.biortech.2012.07.072.

Ramakrishnan, A., Surampalli, R.Y., 2013. Performance of anaerobic hybrid reactors for the treatment of complex phenolic wastewaters with biogas recirculation. Bioresour. Technol. 129, 26–32. doi:10.1016/j.biortech.2012.11.035.

Sagbas, S., Kantar, C., Sahiner, N., 2014. Preparation of poly(humic acid) particles and their use in toxic organo-phenolic compound removal from aqueous environments. Water, Air, Soil Pollut 225, 1809. doi:10.1007/s11270-013-1809-5.

Taguchi, G., Chowdhury, S., Wu, Y., 2005. Taguchi's Quality Engineering Handbook. John Wiley & Sons Inc., Hoboken, New Jersey.

USEPA 1684, 2001. Total, fixed, and volatile solids in water. Solids, Biosolids. https://www.epa.gov/sites/production/files/2015-10/documents/method_1684_draft_2001.pdf. accessed 16 April 2021.

Wang, C., Zhang, Q., Jiang, L., Hou, Z., 2018. The organic pollutant characteristics of lurgi coal gasification wastewater before and after ozonation. J. Chem. 1, 1–9. doi:10.1155/2018/1461673.

Zhao, Q., Liu, Y., 2016. State of the art of biological processes for coal gasification wastewater treatment. Biotechnol. Adv. 34, 1064–1072. doi:10.1016/j.biotechadv.2016.06.005.

Zhao, D., Lun, W., Wei, J., 2017. Discussion on wastewater treatment process of coal chemical industry. IOP Conf. Ser.: Earth Environ. Sci. 100, 012067.