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Evaluation of Different Pretreatment Methods for Enhancement of Biohydrogen Production from Fruit and Vegetable Waste (FVW) Using Mixed Anaerobic Cultures

AJAY DWIVEDI^{a*}, A. N. VAIDYA^a AND M. SURESH KUMAR^b

Efficacy of six individual pretreatment methods, microwave, ozone, alkali, acid, steam explosion and thermal, for a mixture of fruit and vegetable waste (FVW) before dark fermentation has been assessed for augmentation of hydrogen production. The effectiveness of pretreatment was determined by calculating the increase in soluble chemical oxygen demand (SCOD) and reducing sugars (RS) of FVW after pretreatment and then performing batch biohydrogen potential experiments. Microwave pretreatment achieved the highest increase in SCOD and reducing sugars of 68% and 50%, respectively compared to control. Nethermost results were observed for thermal pretreatment; increase in SCOD and reducing sugars 45% and 35% respectively. Among the six individual pretreatment methods used for determination of biohydrogen potential, microwave pretreatment attained highest hydrogen yield (cumulative hydrogen) 350 mL, and heat pretreatment exhibited lowest hydrogen yield of 80 mL, whereas FVW without pretreatment obtained a hydrogen yield of 20 mL.

Key words: *Biohydrogen production, fruit and vegetable wastes, pretreatment, solubilisation*

Introduction

Estimated production of fruits and vegetables in India is 150 million tonnes with a total waste generation of 50 million tonnes per annum (Ravidra, 2015)¹⁹. Of this total produce, a large quantity of waste is generated in numerous processes associated with its production lifecycles such as harvesting, transportation, storage, marketing, processing and post consumption (Shen et al., 2013)²². These wastes are not properly managed in the existing infrastructure of solid waste management giving rise to problems such as soil, surface and groundwater pollution from leachate as well as uncontrolled methane emissions as a potent greenhouse gas (Surendra et al., 2014)²⁴. FVW due to its highly biodegradable nature and rich organic composition has the potential for energy generation by the environment-friendly and cost-effective process of anaerobic digestion by dark fermentation (Kapdan and Kargi, 2006; Venkata Mohan et al., 2009)^{14,25}. Traditionally, carbon-rich organic wastes such as FVW have been utilized for methane production via dark anaerobic fermentation (Demirel and Scherer, 2008; Garcia-Peña et al., 2011; Gunaseelan, 2004; Singh et al., 2012)^{4,8,9,23} but in recent years, the focus has been shifted towards the generation of more energy-rich products such as hydrogen from dark anaerobic fermentation. Hydrogen is regarded as the fuel of future due to its high calorific value (122kJ/g) and clean combustion nature producing only water as a by-product (Alibardi and Cossu,

2015; Kapdan and Kargi, 2006; Romero Aguilar et al., 2013; Venkata Mohan et al., 2009)^{1,14,20,25}. Production of hydrogen via dark anaerobic fermentation can act as an efficient tool in FVW management, and also can provide hydrogen as a valuable energy resource. However anaerobic digestion is marked by a number of limitations, of which the chief is biotransformation of complex FVW, therefore the first step must be physical, chemical or biological pretreatment for breaking complex molecules into simple monomers to increase the solubility of organic material and improve the efficiency of anaerobic treatment (Ariunbaatar et al., 2014; Jia et al., 2014; López Torres and Espinosa Llorens, 2008)^{2,12,16}.

Some pretreatment methods have been used and their application individual or combined have yielded varied results under varied conditions for varied substrates, thus creating discrepancies in their effectiveness (Elbeshbishy et al., 2011)⁶. Thermal, steam explosion, acid, alkali, ozone, microwave are the pretreatment methods studied for a wide variety of substrates namely food waste, algal biomass, fruit and vegetable waste, wastewater (Ariunbaatar et al., 2014; Elbeshbishy et al., 2011; Gadhe et al., 2014; Liu et al., 2013; Roy et al., 2014)^{2,6,7,15,21}. However, a very few of these pretreatment methods have been studied on FVW. The present study aims to determine and optimize the effect of thermal, steam explosion, acid, alkali, ozone, and microwave pretreatment on

^a Solid and Hazardous Waste Management Division, CSIR- National Environmental Engineering Research Institute (CSIR-NEERI), Nagpur, India

^b Environmental Impact and Sustainability Division, CSIR- National Environmental Engineering Research Institute (CSIR-NEERI), Nagpur, India

* Corresponding author: ajayhdwivedi9857@gmail.com

Adsorption of Textile Dye Effluents through *Chara* Species

SUNIL KUMAR^{1*}, A. S. AHLUWALIA¹ AND PRADIP KUMAR²

Dyes and pigments represent chronic ecological problems as they are toxic and carcinogenic. Removal of dyes from the effluents is a major problem. Biosorption through algal biomass is an effective and cheap process for the removal of dyes from industrial effluents. In the present study, biosorption of red dye textile effluent has been carried out using different doses of the dried biomass of *Chara* sp. High dose of algal biomass (100 mg) showed the higher percent adsorption as compared to lower dose (50 mg) of algal biomass. Maximum specific uptake value was observed at 40% red dye effluent through 100 mg biomass and at 80% red dye effluent through 50mg biomass. The results of Freundlich isotherm model showed better biosorption than Langmuir isotherm model. FTIR studies demonstrated that aromatic azo, C=S, sulfur and carbonyl groups could be associated with the adsorption mechanism of red dye.

Key words: Textile dye effluent, biosorption, *Chara* sp., adsorption isotherms

Introduction

Water pollution is caused by the release of various pollutants as a consequence of industrial progress (Ayangbenro and Babalola, 2017)¹ which has now become a serious threat to lives dependent on it. A lot of chemicals including dyes, pigments and aromatic molecular structural compounds are extensively used for several industrial applications such as textiles, printing, pharmaceuticals, food, toys, paper, plastic and cosmetics (Mohana *et al.*, 2008., Boudechiche *et al.*, 2016)^{22,4}.

Worldwide over 10,000 different dyes and pigments are used in dyeing and printing industries (Pandya *et al.*, 2017)²⁷. The total world colorant production is estimated to be 8, 00,000 tons per year and at least 10% of the used dyestuff enters the environment through waste (Palmieri *et al.*, 2005; Kalaiarasi *et al.*, 2012)^{26,14}. The dyes of synthetic origin are of complex aromatic structure and specifically designed to be recalcitrant with poor biodegradability, they are very stable and difficult to degrade by conventional aerobic biological treatments, such as the activated sludge process (Nadaoglu *et al.*, 2013., Kalkan *et al.*, 2015)^{23,15}.

Various physical, chemical and biological methods, namely adsorption, biosorption, coagulation, precipitation, membrane filtration, solvent extraction, chemical oxidation, and photochemical degradation have been used for the treatment of dye containing wastewater. Among these methods, the adsorption process using low-cost adsorbent materials is proved to be an effective process for color removal from wastewater (Ravikumar *et al.*, 2006, Elizalde-gonzalez *et al.*, 2009)^{28,9}. Worldwide more than 50,000 algal species have

been identified and the list is increasing in number with the development of new technologies in the field of algal taxonomy (Kumar and Ahluwalia, 2017)¹⁷. Identification and selection of an ideal algal species are fundamentally important for effective and economic bioremediation process of pollutants e.g. dyes, heavy metals etc. An ideal species should have the high growth rate, easy to harvest, wide range of tolerance of environmental stress. Algae have been found potential and suitable biosorbent because of their fast and easy growth as well as their wide availability. Biosorption using biomass of photosynthetic aquatic organisms, such as algae and aquatic plants or mosses, represents an alternative of cheap and readily available sorbents for the removal of contaminants from wastewaters or polluted water systems (Renuka *et al.*, 2015)²⁹. Algae are ubiquitous naturally and serve as one of the biomaterials with high capacity for removing dye from contaminated waters (Daneshwar *et al.*, 2007)⁷.

Microalgae are known to remove dyes by bioadsorption, biodegradation and bioconversion. Microalgae degrade dyes for nitrogen source by removing nitrogen, phosphorus and carbon from water, it can help in reducing eutrophication in the aquatic environment (Olguin, 2003; Ruiz *et al.*, 2011)^{25,30} and are unique in sequestering carbon dioxide (Mata *et al.*, 2011)²¹. Both living and non-viable algae have been used in color removal from wastewaters. Microalgae have been identified as potent metal and dyes biosorbent due to presence of binding sites such as carboxyl, sulfonate, amine and hydroxyl groups (Davis *et al.*, 2003; Celekli *et al.*, 2011 and 2013)^{8,5,6}. Microalgae *Chara* sp. was investigated as a viable biomaterial for biological treatment of wastewater dye effluent. *Chara* sp. has also been reported for degradation of

¹ Department of Botany, Panjab University, Chandigarh (India)

² Department of Biotechnology, C.C.S. University, Meerut (U. P., India)

* Corresponding author: sunilkumarbotany@gmail.com

Biosorptive Removal of Colour from Direct Dyes by Dead Macrofungi (*Pleurotus sajor-caju*)

K.THASILU **, M.SRIMURALI** AND J. KARTHIKEYAN **

Potential of non-viable pulverized fungi as a biosorbent for removal of colour from textile dyes is the subject matter of this research communication. Three direct dyes, C.I. Direct Red 28, C.I. Direct Brown 1:1 and C.I. Direct Yellow 8 were investigated by batch adsorption tests employing non-viable macro fungi as biosorbent for colour removal. Various sorption process parameters like contact time, pH, particle size were investigated and desorption and interruption studies were also conducted. The study indicates that all the three direct dyes are amenable for their colour removal by sorption onto fungi thus indicating the potential of fungi as a sorbent. Kinetic studies advocated an equilibrium time of 2 hours. Sorbent of size 504.97 μm yielded better results than other sizes of sorbent investigated. Lower pH conditions resulted in higher colour removal. At pH 2.5, removal of colour of C.I. Direct Red 28, C.I. Direct Brown 1:1 and C.I. Direct Yellow 8 was 98%, 80% and 75% respectively. Adsorption isotherms followed Langmuir adsorption pattern indicating formation of a monolayer of dye molecules onto fungi sorbent. Desorption studies suggest that the sorption to be of the type chemisorption. Interruption studies denote the rate limiting step to be film diffusion. The study indicates that all the three direct dyes are amenable for their colour removal by sorption onto fungi.

Key words : Biosorption, colour removal, desorption, interruption, isotherms

1. Introduction

Rapid industrialization and urbanization are resulting in generation of large quantities of aqueous effluents. Many of these effluents contain high levels of toxic pollutants like dyes^{1,2}. Synthetic dyes are used extensively for colouration of natural and synthetic materials and are widely used in several industries like textile, pulp and paper, etc for colouration. Discharge of coloured effluents into receiving waters imparts colour and impairs its intended beneficial use. A number of physicochemical, biological and chemical processes have been investigated for removal of colour from synthetic coloured effluents.

Biosorption is a selective concentration process wherein the pollutant under consideration gets accumulated onto/in the sorbent, i.e. viable or non-viable biota either by a single process or a combination of the processes of sorption, assimilation and accumulation³. Feasibility studies for large scale applications have demonstrated that, biosorptive processes using non-living biomass are more applicable than the bioaccumulate processes that use living microorganisms as living organisms require a nutrient supply and complicated bioreactor systems². Biosorption employing *Trametes versicolour* and immobilized fungus *Versicolour* was investigated for removal of colour from pulp and paper industry

effluents and 70% of the colour was removed after 4 days⁴. Macro fungi (*fomitopsis carnea*) possess excellent biosorption capacity and the dye uptake was better at higher pH⁵. Decolourisation of a chemical industry effluent by *white-rot-fungus* was investigated and decolourisation of 80% was obtained in 3 days⁶. Decolourisation of textile effluents and dye amended soils by Fungi *Aspergillus niger* and *Mucor mucedo* was studied and 92% colour removal was observed⁷. Decolourization of textile dyes Astrazone Blue, Red, Black by *fungus pellets* was observed to be more than 75% colour removal⁸. Removal of indigo carmine from an aqueous solution by the fungus *pleurotus ostreatus* was 93% efficient⁹. Biosorption of Methylene Blue dye from aqueous solution using *papaya peel* investigated¹⁰ and reported that 93% colour removal at a contact time of 35 min with dye concentration of 1mg/100m and adsorbent dosage of 0.3g/100mL at a pH 8. *Soybean hull* was investigated for adsorption of safranin dye and reported relatively large adsorption capacity and effective removal of dye from the wastewater¹¹. Biosorption of Direct Red 89 and Reactive Green 12 by nonliving *Macrophyte L. Gibba L* were found optimal at acidic pH¹².

Biosorption using non-viable (dead) cells of naturally occurring biological species overcome the economic constraints of activated carbon adsorption⁵. In the present study, application of non-viable pulverized macro fungi

* Lecturer, Govt Polytechnic, Satyavedu, Department of Technical Education, A.P., India

** Professor, Department of Civil Engineering, SVU College of Engineering, Sri Venkateswara University, Tirupati - 517 502, A.P., India.

+ Corresponding author : kthasil@gmail.com

Preparation and Characterization of Cost Effective Adsorbent: Treatment of Wastewater from Dyeing Industry in Sanganer, Jaipur (India)

ALPANA KATEJA¹ AND BHANUPRIYA MORDHIYA^{2*}

Water pollution due to effluents from the textile dyeing industry is a serious concern. The study assesses the impact of dyeing and printing industry situated near Sanganer region of Jaipur (India). The effluent of these industries has very high concentration of pollutants. The wastewater of these units gets discharged on land without any treatment and comes into the Amanishah Nallah through small water courses. The quality of the Nallah wastewater, which generally contains domestic wastewater, is getting deteriorated due to the mixing of wastewater from these industrial units. It is characterized by very high TDS, BOD and COD. The impact of the wastewater of the dyeing and printing units is clearly noticeable in the study area. The work also includes the synthesis and characterization of the activated carbon (AC) from *Prosopis cineraria* and its use for wastewater treatment from dyeing industry in Sanganer region of Jaipur (India). The AC were synthesized by a physico-chemical activation method and analysed using different techniques such as Attenuated Total Reflection (ATR), Scanning Electron Microscopy (SEM), Energy Dispersive Spectroscopy (EDS) and X-ray Diffraction Method (XRD). Present research also focuses on understanding adsorption process and developing cost-effective technology for the treatment of dyes contaminated industrial wastewater. Comprehensive characterization of parameters indicates that *Prosopis cineraria* is a good adsorbent to treat wastewater.

Key words : BOD, COD, SEM, EDS, ATR, TDS, XRD, *Prosopis cineraria*, Sanganer, Amanishah drainage, cost effective adsorbent

Introduction

The textile industry is the largest consumer of dye material. During the coloration process a large percentage of the synthetic dye does not bind and is lost in the waste stream. Approximately 10-15 per cent dyes are released into the environment during dyeing process making the effluent highly coloured and aesthetically unpleasant.¹ Effluent from textile industries thus carries a large number of dyes and other additives which are mixed during the colouring process. In Sanganer (Jaipur) dyeing and printing of textile is a traditional industry.² Nilgar / Rangrej and Chhipa communities are mainly involved in this sector.³ Wastewater samples of Sanganer region were studied to find out the pollution load of wastewater generated from dyeing and printing units and its impact on the quality of domestic wastewater of the Amanishah Nallah. The wastewater of these units gets discharged on land without any treatment and enters into the Amanishah Nallah through small watercourses, which is difficult to remove through conventional methods. They may also undergo degradation to form products that are highly toxic and carcinogenic.⁴

Activated carbon (AC) is a type of carbonaceous material. It normally differs from elemental carbon by the oxidation of carbon atoms that are present on the inner and outer surfaces.⁵ Recently, these materials have become an important research area due to their excellent properties. These involve their large specific areas, nontoxicity, high porosity, and the ability of their big surface to contain different functional groups.^{6,7} By virtue of these properties, activated carbons have been widely used as adsorbent materials for a wide range of applications such as in wastewater treatment, purification of drinking water, gas phase adsorption in air pollution, and liquid phase adsorption.⁸ Commercial AC has been relatively expensive. So the synthesis of a relatively cheap AC for wastewater treatment is required⁹. This aim can be achieved by using inexpensive and widely available agricultural raw materials with high carbon contents and low inorganic contents.¹⁰ Currently, an important application of AC is the removal of polluted dyes from industrial wastewater. However, AC is widely used as a standard adsorbent for the removal of a wide range of polluted dyes from coloured wastewaters. Recently, activated carbon has been considered

¹ Professor, Department of Economics, University of Rajasthan, Jaipur (alpanakateja@gmail.com)

² Assistant Professor Department of Chemistry, University of Rajasthan, Jaipur (bmordhiya@gmail.com)

* Corresponding author : bmordhiya@gmail.com

Correlation Study amongst Water Quality Parameters of Paiswani River in Chitrakoot (M.P.), India: An Approach to Water Quality Management

ASHOK KUMAR TIWARI* AND SURYA KANT CHATURVEDI**

To find out an approach to water quality management through correlation studies between various water quality parameters, the statistical regression analysis was carried out for six sampling stations of Paiswani river in Chitrakoot (M.P.), India. The comparison of estimated values with WHO drinking water standards revealed that water of the study area is very much affected due to anthropogenic activity with reference to a number of physico-chemical parameters studied. Regression analysis suggests that the most informative correlations were with total hardness, which is a useful tracer for Paiswani river. A strong correlation between total hardness and other typical parameters such as calcium as calcium carbonates, magnesium hardness, total alkalinity, chloride and dissolved oxygen was observed. Correlation was also found among other parameters, like total hardness, which are physically, biologically or chemically inactive. The present study may be treated one step forward towards the water quality management.

Key words : *Correlation, parameters, regression equations, water quality management*

Introduction

Water is a quite essential natural vital element for all kinds of life. Rivers play a significant role because they serve the purpose of various human activities, such as bathing, disposal of sewage, irrigation, electricity generation, industrial production and disposal of industrial wastes, etc. During the process of cultural evolution, natural resources have been brutally exploited by man. The loss in agriculture, the gain in industrialization and population explosion have stressed the natural resources^{1, 2}.

It is well known fact that clean water is absolutely essential for healthy living. Adequate supply of fresh and clean water is a basic need for all human being on the earth, yet it has been observed that millions of people worldwide are deprived of this^{3, 4}.

The statistical regression analysis has been found to be a highly useful tool for correlating different parameters⁵⁻¹⁰. Correlation analysis measures the closeness of the relationship between chosen independent and dependent variables. If the correlation coefficient is nearer to +1 or -1, it shows the probability of linear relationship between the variable x and y. Analysis attempts to establish the nature of

the relationship between the variable and thereby provides a mechanism for predication or forecasting.

Study area

River Paiswani originates in the western uplands of Majhgawan block in district Satna, M.P., India and travels about 39 km before reaching at Sati Anusuiya. About 35 km of stream is seasonal that flows only in rainy season. From Sati Anusuiya it becomes perennial with minimum flow 1.2 m³/sec, where a number of springs feed it all the year¹¹. The river travels through various spots like Sati Anusuiya, Sphaticshila, Jankikund, Pramodvan, Goyankaghat, Bharatghat, Ramghat, Karwi, Bhairopaga, Surajkund and finally meets with river Yamuna near Rajapur, district Chitrakoot of U.P. after flowing about 74 km. The river has catchment area of 1956.3 sq km with major portion in Uttar Pradesh (U.P., India). Paiswani has four main tributaries i.e. Tiraha (50 sq km catchment), Jhuri (40 sq km catchment), Gedua (60 sq km catchment), Ohen (150 sq km catchment) and all are seasonal. This holy river is made-up of a series of pools with narrow rapids¹².

The water of Paiswani river is the main source of drinking water for the people of Chitrakoot and its surrounding areas from Sati Anusuiya to Rajapur. Previously the river water

* Environmental Scientist, Ayurveda Sadan, J.R.D.Tata Foundation for Research in Ayurveda & Yoga Science, Arogyadham, Deendayal Research Institute, Chitrakoot, Satna – 485 334 (M.P.)

** Associate Professor, Department of Biological Science, Faculty of Science and Environment, Mahatma Gandhi Chitrakoot Gramodaya Vishwavidyalaya Chitrakoot, Satna-485 334 (M.P.)

+ Corresponding author: gangagargi@gmail.com; ashokckt77@yahoo.com

Mathane and Nitrous Oxide Emission from Animal Dung / Faeces from Indian Livestock

MADHU MOHINI⁺, S. S. THAKUR, GOUTAM MONDAL, SUJATA PANDITA*

AND CHANDER DATT

The proportionate emission from manure storage, however, can vary widely depending on the storage type and climate. Total dung produced from Indian livestock was calculated by taking the census population at state level, body weight of different breeds available in various states and their DM consumed depending upon the feeding practices followed in different states for different categories of the livestock. Total DM of the dung was divided into three fractions as per its disposal system (making of dung cakes, spread in pastures or roads etc. and stored as heaps for some time) during different months of the year and in different states. IPCC default emission factors were taken for estimation of methane and nitrous oxide emission for different disposal systems. Total amount of dung produced was 313.69 million tons. Maximum contribution was by indigenous cattle (39%) and buffalo (37%). Among the states of India, Uttar Pradesh's contribution was maximum (15%) and then Rajasthan with 10%. Contribution of NEH region and UTs was negligible. Out of total dung produced, 32% was used for dung cakes, 49% as in pasture or otherwise and 19% is disposed off as solid. Total methane emission is 137.69 Gg and nitrous oxide emission is 87.45 Gg.

Key words: *Methane, nitrous oxide, Indian livestock, dung*

Introduction

Livestock is one of the major contributors of GHGs. It accounts for 18% of the global GHG emissions such as CH₄ from enteric fermentation, N₂O from manure and fertilizer¹. Live stock emissions of methane have two components, one from enteric fermentation and another from dung outside the animal system. About 37 % of anthropogenic methane is attributed to enteric fermentation by ruminants as part of their normal digestive processes. Livestock manure management is also a significant contributor of methane with global emissions accounting to 9.3 Tg/year². Livestock also contributes a small but significant emission of nitrous oxide (N₂O) from animal waste management systems.

Methane emission from storage of dairy cow manure has been estimated as 12-23% of total methane emission³. The proportionate emission from manure storage, however, can vary widely depending on the storage type and climate, and might only represent about 1/3 of the potential yield. Since enteric methane emission exceeds methane emission from manure storage, the primary efforts were mainly focused on reducing enteric methane emission, while manure storage has only received little attention.

Nitrous Oxide is also a potent greenhouse gas with a long lifetime of 150 years in the atmosphere and large

radioactive forcing potential than that of CO₂ is 310 times. Besides, nitrous oxide is also involved in the catalytic destruction of stratospheric ozone following its photolytic oxidation to nitric oxide (NO). The global atmospheric concentration of N₂O has increased from 270 ppb in the preindustrial period to 324 ppb in 2011⁴. Hence estimation of CH₄ and N₂O from fecal waste of livestock is equally important.

Material and methods

The estimates were made on the basis of the number of livestock reported in the Livestock Census 2007 (Department of Animal Husbandry and Dairying, Govt. of India) as per various categories and types of the animals (indigenous and crossbred cattle, buffalo, goat, indigenous and crossbred sheep, yak, mithun, horse, ponies, mules, donkeys, camels, indigenous and crossbred pigs). Body weight of different categories varied according to the age, breed and the availability of feeds. Nivsarkar⁵ has reported the birth weight and adult weight of male and female animals of each Indian breed of cattle and buffaloes.

Data about the digestibility of different Indian feeds in various species of animals have been published by animal nutritionists working in various laboratories throughout different Indian states^{6,7}. Therefore, the undigested feed i.e. dung voided by the Indian animals was worked out taking

Dairy Cattle Nutrition Division, National Dairy Research Institute-Karnal, India

* Dairy Cattle Physiology Division, National Dairy Research Institute-Karnal, India

⁺ Corresponding author: madhummd@yahoo.co.in