Wat. Sci. Tech. Vol. 26, No. 5-6, pp. 1265–1274, 1992.
Printed in Great Britain.

0273–1223/92 $15.00

TREATMENT OF WASTEWATER WITH HIGH ENERGY ELECTRON BEAM IRRADIATION

S. Farooq*,†, C. N. Kurucz**, T. D. Waite*,
W. J. Cooper***, S. R. Mane*** and J. H. Greenfield***

* *Department of Civil Engineering, University of Miami, FL 33124, USA*
** *Department of Management Sciences, University of Miami, FL 33124, USA*
*** *Drinking Water Res. Cen., Florida International University, Miami, FL 33199, USA*

ABSTRACT

The objective of this study was to determine the effectiveness of high energy electron beam irradiation in the disinfection of wastewater, and removal of organic matter through measurement of BOD and COD values. The raw wastewater and secondary effluents were transported in a 22.7m^3 tank truck to the research facility built at the Virginia Key Wastewater Treatment Plant, Miami, Florida. The facility is equipped with a 1.5 MeV, 50 ma electron accelerator capable of treating the wastewater at a volumetric flow rate of 7.57 l/sec. Some preliminary disinfection was also carried out using a ^{60}Co gamma radiation source. Removal of coliphage, total coliforms, and standard plate counts was found on the order of two to three orders of magnitude in raw wastewater. Gamma irradiation gave slightly better inactivation. However, in all cases it was found that coliphage is more resistant than the other two organisms investigated. Twenty to 30 percent removal of BOD was observed in the case of raw wastewater, however, no significant change was observed in removal of COD.

KEYWORDS

High energy electrons; electron beam; gamma irradiation; disinfection; BOD; COD; coliform; plate counts; coliphage.

INTRODUCTION

Treatment of wastewater with high energy electrons is a promising new technology. The technology has been used successfully for years in disinfection of surgical instruments, food preservation, generation of X-rays and in treatment of human diseases (Cleland, et al., 1984). While several sources of radiation are available for use in treating water and wastewater, and many have been tested, the electron beam has the advantage of not requiring a radioactive source. This fact alone has made this form of radiation much more acceptable to the general public.

The electron beam treatment process consists of the interaction between high-energy ionizing radiation and wastewater that is passed through the beam. Electron accelerators or ^{60}Cobalt are used in order to generate fast electrons. Once electrons are introduced into the stream of wastewater, they

† Permanent address: King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia

produce electrons of decreasing amounts of energy through their interaction (in less than 10^{-12} sec) with the wastewater. After high energy (fast) electrons impact an aqueous solution, with or without particulate matter present, reactive transient species are formed (Bielski and Gebicke, 1970; Draganic and Draganic, 1971; Buxton, et al., 1988; Kurucz, et al., 1991; Singh, et al., 1985.) The three transient species of most interest are the aqueous electron e^-_{aq}, the hydrogen radical, $H^\cdot$, and the hydroxyl radical, $OH^\cdot$. The relative concentrations of these radicals in an irradiated solution of pure water are 44, 10 and 46%, respectively. These highly reactive species in turn attack both living and non-living structures to promote their oxidation, reduction, dissociation, and degradation.

The amount of energy that is absorbed by an irradiated material, per unit mass, is known as a dose and is expressed in rads. Depending on the properties of the material, and the strength and time of the beam treatment, the dose absorbed by the material will be affected. The temperature difference between influent and effluent streams is converted to absorbed dose by the relationship that a change of 1°C in water is equivalent to a dose of approximately 418.6 krads.

Interest in using penetrating ionizing energy for the disinfection of wastewater and sludges has been growing for several decades. Touhill, et al. (1969) studied the disinfection capabilities of irradiation on municipal wastewater at Chicago Metropolitan Sanitary District. The population of coliform bacteria was reduced by two orders of magnitude with complete inactivation at a dose of 74 krads.

Levaillant and Gallien (1979) reported 90% reduction of total coliforms in water at a dose of as low as 11 krads, and complete sterilization at a dose of 80 krads. However, they also indicate full scale studies have demonstrated high dose requirements for 90% inactivation of coliform bacteria. Shah (1976) observed a dose of 300 krads was sufficient to produce a 99.99% kill in the total population present in sewage sludge. The coliforms and other gram negative bacteria including salmonella and shigella in sludge proved to be the most susceptible to radiation. A complete inactivation of these bacteria was obtained with total dosage of less than 250 krads.

A biodegradability study conducted by Alexander (1969) using gamma radiation at 300 krads on a primary effluent revealed that the irradiation did not significantly enhance the rate of oxygen demand or the total biodegradability, however, about 35% BOD removal was observed. He also observed that no significant change in COD was produced by irradiation.

Condren (1969) performed experiments by using gamma radiation on selected organic compounds such as phenol, alanine, sucrose, and tannic acid. A linear rate of removal in both COD and TOC was observed with respect to the total absorbed dose and the removals were proportional to the concentration of the solution. The maximum organic carbon removals rated for phenol, alanine, sucrose and tannic acid were found to be 17, 17, 23, and 28 mg/l per mrad, respectively, and the COD removals for phenol were about 22 mg/l of COD per mrad.

From the literature review it is certain that radiation technology is effective in disinfection of wastewater and sludges and also in the destruction of some of the toxic compounds. However, no full scale study has been conducted using an electron beam process to establish its effectiveness in the treatment of wastewater. The purpose of this study was to investigate the effectiveness of a high energy electron beam in full scale disinfection, and to observe any changes in the characteristics of the waste with respect to the values of BOD, COD and BOD rate constants.

MATERIAL AND METHODS

The full-scale research facility built at the Virginia Key Wastewater Treatment Plant in Miami, Florida, is equipped with a 1.5 MeV, 50 mA electron accelerator. Varying the beam current changes the absorbed dose in a linear fashion, allowing for experimentation at doses from 0 to 800 krads. The electron beam is scanned at 100 Hz to give a coverage 122cm wide and 5cm high. The volumetric capacity for irradiating the wastewater is 7.57 l/sec. Settled raw sewage and secondary effluent were transported to the facility in a $22.7m^3$ tank truck, and hooked directly to the influent pump. The details of the process and diagrams are given elsewhere (Kurucz, et al., 1991).

Preliminary research work on the effects of gamma radiation for disinfection was also conducted using a ^{60}Co gamma source located at the University of Miami Radiation Control Center. Here screw capped vials holding wastewater sample are placed at an appropriate distance from the source for a predetermined amount of time to give the desired applied radiation dose. A linear regression of a ln/ln plot of distance versus dose rate was generated to determine the dose rate at any distance from the ^{60}Co source and thus the required time to obtain a given dose.

The BOD kinetic experiments were performed using an Otel electrolytic respirometer, Model ER 100.Six cells available for experimentation were utilized for samples before and after irradiation at 0, 30 and 50 mA of beam current, where samples at 0 mA served as a control. A total sample volume of one liter without any dilution was used for analysis. The seed, nutrients and nitrification inhibitor were provided and the entire experimental setup was placed in an incubator and maintained at 20° C. The elapsed time and the BOD in mg/l were recorded by an electric strip chart recorder at an interval of one hour. The COD measurements were carried out using Hach COD vials and reactor.

Standard plate counts were determined using Pour Plate Method (#9215 B), coliform bacteria by Total Coliform Multiple-Tube (MPN) Fermentation Technique (#9221B), and coliphage according to the proposed Coliphage Detection Method (#9221D) recommended in APHA Standard Methods (1989).

RESULTS AND DISCUSSION

Fig. 1 shows the inactivation of the natural populations of total coliforms, standard plate counts, and coliphage at various doses of gamma irradiation in a raw sewage. The original populations were of the order of 7, 8, and 7 logs per 100 ml, respectively. Two logs inactivation of coliphage, and four to five logs for total coliforms and standard plate counts were observed, respectively, at an absorbed dose of 462 krads. Inactivation of coliphage increases gradually with an increase of radiation dose. However, the inactivation rates of coliforms and standard plate counts were higher initially and then stabilized at the reduced level. The survival curve for coliphage indicates that it is more resistant to gamma irradiation as compared to the coliform and standard plate counts at lower levels of irradiation.

Disinfection results of secondary effluent with gamma irradiation are given in Fig. 2. The original populations of coliform, standard plate counts, and coliphage present were of the order of 5, 6, and 4 logs per 100ml, respectively. Inactivation of all three organisms investigated reaches the reduction of three orders of magnitude at a dose of 500 krads, which is approximately one order of magnitude more for coliphage than the raw sewage.For coliform and standard plate counts the inactivation is one to two orders of magnitude less effective, respectively, than in the raw sewage. This behavior of poor inactivation in the cases of coliforms and plate counts is contrary to the expected results as secondary effluent contains less

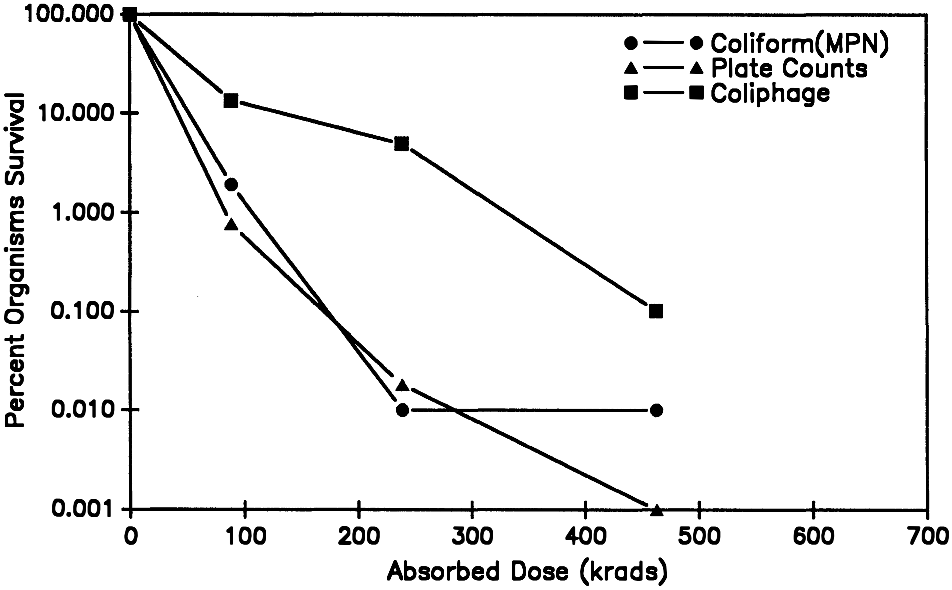

Fig.1. Disinfection of Raw Wastewater with Gamma Irradiation

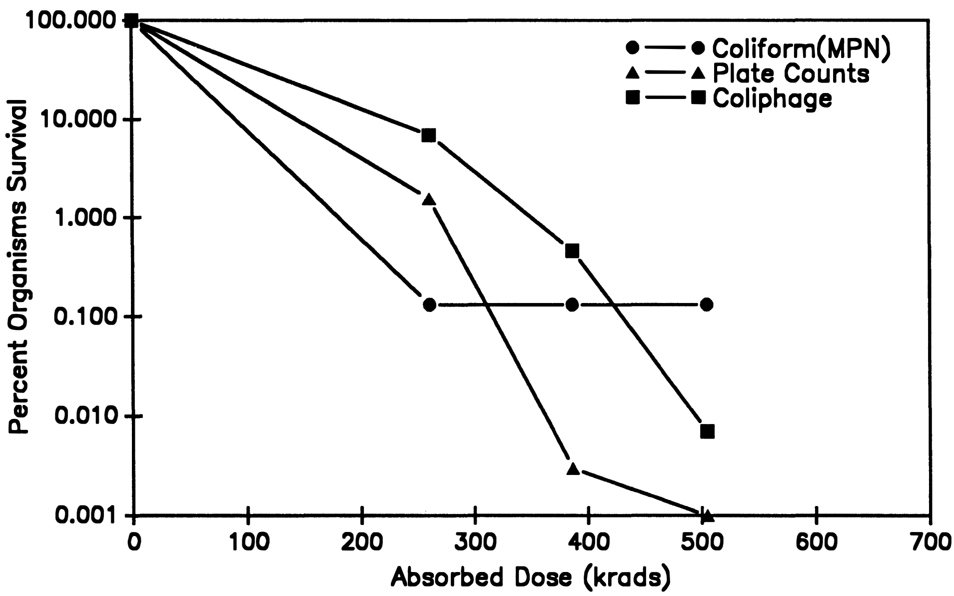

Fig.2. Disinfection of Secondary Effluent with Gamma Irradiation

oxidizable matter and other interfering substances in the form of particulate matter, which can shelter the organisms from direct irradiation and, therefore, resulting in better inactivation. The only possible explanation is the formation of some radicals in the raw sewage which are more efficient in inactivation of the microorganisms. Incidentally, similar results of improved removal of organics are observed in the case of raw sewage as compared to the secondary effluent.

The results of full-scale electron beam experiments with raw sewage are given in Fig. 3. Between two to three orders of magnitude of reduction was observed for all three organisms at absorbed irradiation doses of 462 to 650 krads. Inactivation of coliphage is more gradual than coliform and standard plate counts, which are inactivated rapidly at lower dosage. Similar to the results of gamma irradiation, coliphage is more resistant in survival than the other two microorganisms. Inactivation data for all three organisms in the case of electron beams appear to be less efficient as compared to the gamma irradiation. The primary reason is that the sample is exposed to the gamma irradiation over the longer time period (several hours in case of higher dosage) to get the similar dose as in the case of electron beam, where the exposure is only the fraction of a second.

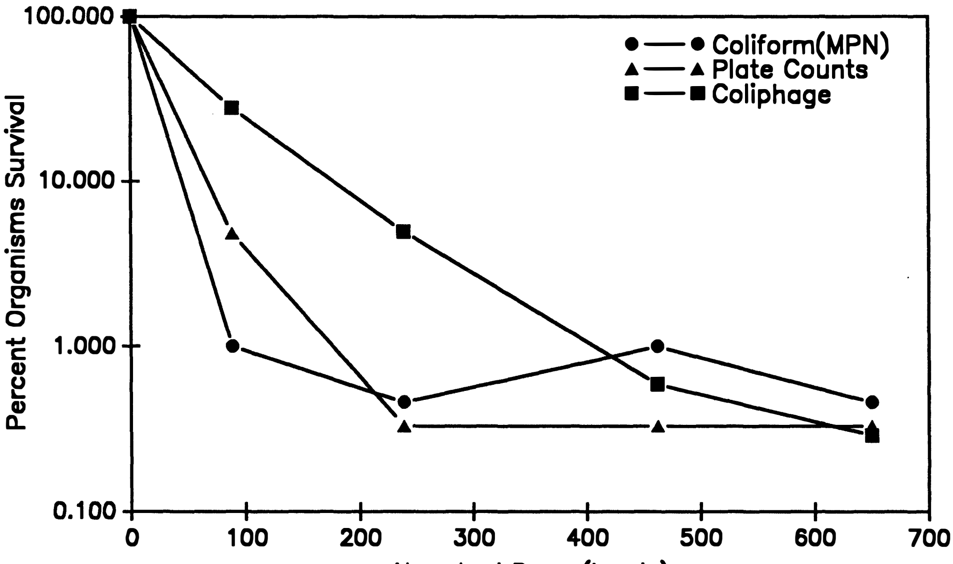

Fig.3. Disinfection of Raw Wastewater with E–Beam

The actual mechanism of inactivation of microorganisms by electron beam irradiation is not clear. It is known that many free radicals are formed but their role in inactivation is not clear. The very short half-life of these oxidants indicates that perhaps the electron itself may be the major inactivating mechanism. There is a stable hydrogen peroxide residual formed upon radiation, and this is monitored routinely during experimentation. Comparison of these data with other disinfection studies using electron beam radiation is difficult because of the scale of the experimental system and different ways of reporting absorbed dose.

BOD kinetic studies have been conducted in raw wastewater using an electrolytic respirometer. The rate of biodegradation and ultimate BOD after three and ten days were determined by the two distinct phases of biological oxidation. The first is referred to as the synthesis phase and the second is known as the endogenous metabolic phase. Fig. 4 shows the effect of irradiation on samples incubated for ten days at total absorbed dose of 500 and 813 krads, respectively. A marked reduction in BOD was observed at both doses with no significant change in the control (i.e., at zero dose). The reduction in BOD, after ten days, from 142 to 115 mg/l at 500 krads and from 151 to 107 mg/l at 813 krads, is apparently due to the oxidation of organic compounds in the wastewater. The same experiment was repeated four different times to obtain reliable kinetic data. Fig. 5 summarizes the data from all four different experiments with respect to the percent removal against absorbed dose of electron beam irradiation. A regression analysis was performed to obtain the best fit for these percent removal data. The Y intercept is 329 and the slope of the line is equal to 0.028. The coefficient of determination (R^2) for the best fitted line was 0.867.

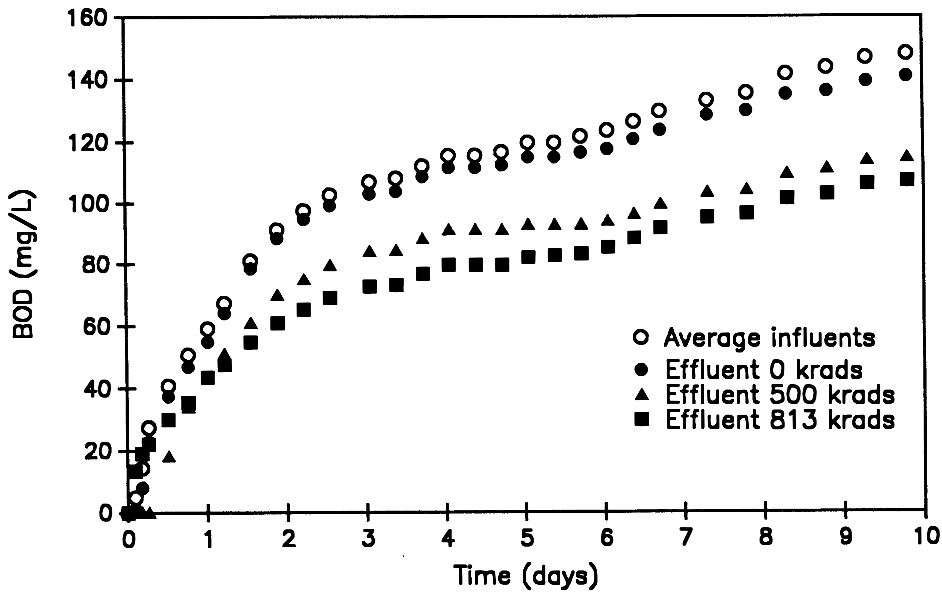

Fig.4. BOD as a Function of Time at 0, 500, and 813 krads

The BOD rate constants were calculated using non-linear regression analysis of the following first-order BOD model.

$$Y = L_o \,(1-e^{-kt}),$$

where k is the first-order rate constant (day^{-1}), Y is the BOD exerted at any time t (mg/l) and L_o is the ultimate BOD (mg/l). The average influent BOD rate constants for all four experiments are given in Table 1. Also given are the average of the differences in the rate constants for corresponding influent and effluent samples (Inf-Eff), along with 95% confidence interval estimates of the true mean differences.

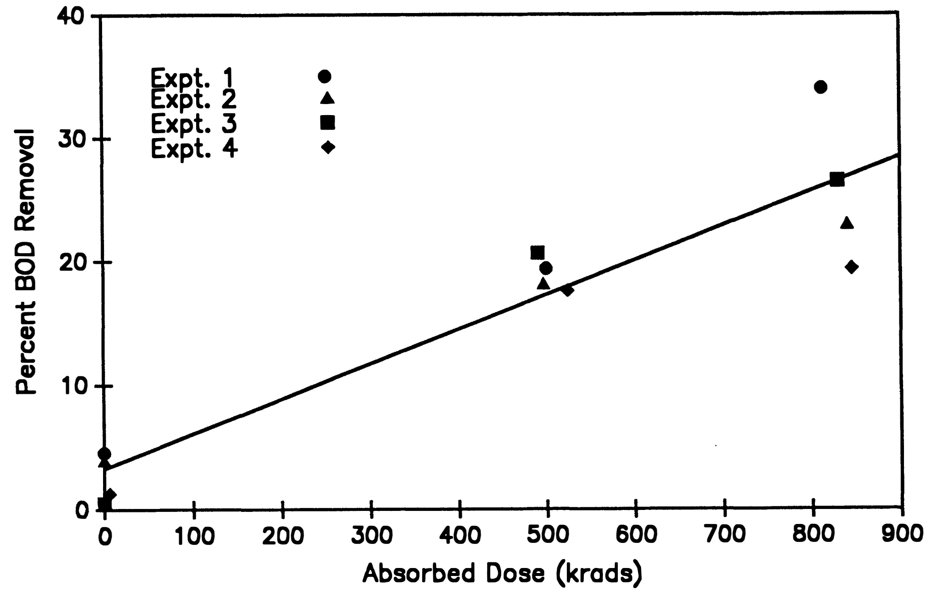

Fig.5. BOD removal as a Function of Dose

The rate constants at 500 and 813 krads tended to decrease slightly as indicated by the positive influent minus effluent averages. Since most of the confidence intervals contain zero, however, the decreases are, in general, not statistically significant. Only the ten day rate constant at 500 krads is marginally significant at the α =.05 level.

The rate constants for the influents in the first three days were observed to be approximately 0.8 day^{-1} and then dropped to about 0.5 day^{-1} when calculated over ten days. This is because the rate of biodegradation in the endogenous metabolism is often less than 0.1 day^{-1} (Eckenfelder, 1980). The values of k vary from 0.34 to 0.64 day^{-1} for raw wastewater as compared to the values of 0.14 to 0.23 day^{-1} for high degree biotreatment effluent.

The ultimate BOD values were also estimated from the non-linear regression analysis at all doses tested. Decreases in ultimate BOD values at 500 and 813 krads were in the range of 13 to 25 percent. The decreases were all statistically significant at the 5% level of significance (α=.05) as indicated by the fact that the confidence intervals on the difference in influent and effluent values do not include zero. The percent removal of ultimate BOD was in the same range as observed at BOD_5 and followed a similar linear function.

In summary the data of this study suggest that irradiation did not cause any significant change in the rate of the biodegradation process, suggesting that irradiation does not break up the non-biodegradable compounds into easily biodegradable substances. However, the removal of BOD observed may be due to oxidation caused by OH$^{\cdot}$ radical. Cooper et al. (1990) also discussed the role of the OH$^{\cdot}$ radical in oxidizing organic compounds.

TABLE1 Estimates of Three Day and Ten Day BOD Rate Constants and Ultimate BOD

		Three Day		Ten Day	
Sample Calculation	Dose (krads)	Rate Constant k (day^{-1})	Ultimate BOD L_o (mg/l)	Rate Constant k (day^{-1})	Ultimate BOD L_o (mg/l)
Influent	0	0.80	105.3	0.53	125.1
Inf-Eff	0	0.04	4.5	-0.02	4.4
		(-0.17,0.09)	(-3.1,12.1)	(-0.05,0.02)	(-3.2,11.9)
Influent	500	0.86	103.5	0.56	122.4
Inf-Eff	500	0.20	13.5	0.09	19.2
		(-0.03,0.43)	(1.5,25.4)	(0.01,0.17)	(12.5,25.9)
Influent	813	0.84	109.0	0.52	127.7
Inf-Eff	813	0.03	31.2	0.03	30.9
		(-0.25,0.31)	(6.7,55.6)	(-0.02,0.08)	(16.9,44.8)

95% Confidence intervals on the mean differences between corresponding influents and effluents are given below the average differences (Inf-Eff).

Two experiments were conducted to study the effects of the electron beam on BOD kinetics of the secondary effluent at three different doses of 166, 500, and 830 krads. No significant difference was observed in the values of influent and effluent BOD at all three doses investigated. The BOD_5 of the treated effluents studies were as low as 14 mg/l and since most of the biodegradable organic matter is removed by the biological treatment process, no effect of the electron beam irradiation was observed. This indicates that the irradiation may be more effective in removing the oxidizable organic matter present in the raw sewage as compared to inert and oxidized organic matter in the secondary effluent.

The wastewater samples subjected to electron beam irradiation and used for BOD experiments were also tested to determine any effect on COD. Thirteen experiments conducted on raw wastewater and secondary effluents demonstrated that no significant changes occurred in COD due to irradiation. Other studies regarding the irradiation of specific organic compounds and their breakdown mechanisms are reported elsewhere (Cooper 1991). Overall the results discussed agree with the results obtained by Alexander (1969) using gamma irradiation on primary effluents. He observed about 35 percent BOD removal at the end of ten days, incubation period at an absorbed dose of 300 krads. He also did not find any change in biodegradability, rate of oxygen demand, and COD.

CONCLUSIONS

The following specific conclusions may be drawn from this study.

1. Two logs inactivation of coliphage, and four to five logs for total coliform and standard plate counts were observed at a gamma irradiation dose of 563 krads in raw sewage. In the case of secondary effluent, the

inactivation of all organisms was approximately three orders of magnitude for a similar dose of gamma irradiation.

2. Electron beam irradiation appears to be less effective than gamma irradiation as the overall inactivation for all three organisms investigated was approximately two to three orders of magnitude in the raw wastewater.

3. In all cases investigated, coliphage appears to be more resistant than total coliforms and standard plate counts to gamma and high energy electron beam irradiation.

4. Percent BOD removal by electron beam irradiation increased linearly with an increase in total absorbed dose, giving removals of 20 to 30 percent at doses of 500 and 830 krads, respectively, in raw sewage.

5. Non-linear regression analysis performed on BOD kinetics data showed that there was no effect of irradiation on the rate constants of raw wastewater. The average rate constants for influent raw wastewater ranged from 0.52 to 0.55 day^{-1} and the difference between these values and those for irradiated raw wastewater were not statistically significant. This indicates no specific change in the nature of the raw wastewater after irradiation.

6. Irradiation of secondary effluent at doses of 166.5 and 840 krads did not produce any changes in BOD values, indicating the resistant nature of the organics in the effluent.

7. Similarly no significant change was observed in the values of COD of raw wastewater and secondary effluent after the electron beam irradiation.

ACKNOWLEDGEMENT

The authors are grateful to the National Science Foundation for supporting this research work. The cooperation of the Miami-Dade Water & Sewer Authority is appreciated in completion of this work. Dr. Farooq would like to thank King Fahd University of Petroleum and Minerals, Dhahran 312161, Saudi Arabia which supported him through sabbatical leave to work on the project.

REFERENCES

Alexander, M. (1969). Biodegradability of Sewage Following Gamma Irradiation. M.S. Thesis, Purdue University, Lafayette, Indiana.

APHA, AWWA, and WPCF (1989) Standard Methods for the Examination of Water and Wastewater. American Public Health Association, Washington, D.C.

Bielski, B.H.J. and Gebicke, J.M. (1970). Species in Irradiated Oxygenated Water. Adv. Radiation Chemistry, 2:177-279.

Buxton, G.V., Greenstock, C.L. Helman, W.P., and Ross, A.B. (1988). Critical review of Rate Constants for Reactions of Hydrated Electrons, Hydrogen Atoms and Hydroxyl Radicals (·OH/·⁻o) in aqueous solution. Journal of Physical and Chemical Reference Date, 17, 513-886.

Cleland, M.R., Fernald, R.A. and Maloof, S.R. (1984). Electron beam process design for the treatment of wastes and economic feasibility of the process. Special Issue of Radiation Physics and Chemistry, 24, N1, 179-190.

Condren, A.J. (1969). Radiation induced oxidation of selected organics in wastewater. Thesis, Purdue University, Lafayette, Indiana.

Cooper, W.J., Nickelsen, M.G., Meachem, D.E., Cadavid, E.M., Waite, T.D., and Kurucz, C.N. (1991). High Energy Electron Beam Irradiation:An innovative process for the Treatment of Aqueous Based Organic Hazardous Wastes. J. of Environ. Science & Health (Toxic & Hazardous Substance Control). In Press.

Draganic, I.G. and Draganic, Z.D. (1971). The Radiation Chemistry of Water. Academic Press, N.Y., N.Y., 242.

Eckenfelder, W.W. Jr. (1980). Principles of Water Quality Management. CBI Publishing Co., Boston, MA, 15-17.

Kurucz, C.N., Waite,T.D., Cooper, W.J., and Nickelsen, M.G. (1991). High Energy Electron Beam Irradiation of Water, Wastewater, and Sludge.In: Advances in Nuclear Science and Technology, Lewins, J., Jr., and Becker, M. (Eds). Plenum Press, N.Y., N.Y. In Press.

Levaillant, C. and Gallien, C.L. (1979). Sanitation methods using high energy electron beams. Radiation Phys. Chem., 14, 309-316.

Shah, D.N. (1976). High energy electron irradiation of wastewater liquid residuals. M.S. Thesis, M.I.T., Cambridge, MA.

Singh,A., Sagert, N.H., Borsa, J., Singh, H., and Bennet, G.S. (1985). The use of high-energy radiation for the treatment of wastewater: A review. Proceedings of the 8th Symposium on Wastewater Treatment, Montreal, Canada, 191-209.

Touhill, C.J., Martin, E.C., Fujihara, M.P., Olesen, D.E., Stein, J.E. and McDonnel, G. (1969). The effects of radiation on Chicago Metropolitan Sanitary District municipal and industrial wastewater. J. Wat. Poll. Control Fed. 41. R44.