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Abstract

Many treatment processes have been applied for the removal of dyes from waste water such as physical, chemical and biological treatments [1]. Hydrogels are suitable for the removal of dyes due to their hydrophilic nature and three-dimensional polymeric network which can imbibe large amounts of water [2]. This study is divided into three points, the first one was preparation of (acrylamide/ starch) hydrogel by gamma radiation for removal of direct congo red (DCR) and direct blue (DB) dyes. The ionizing radiation technique seems an excellent method for the preparation of hydrogels, it is clean and more efficient than other techniques [3,4]. We report the influence of different parameters that affecting the adsorption. It was found that favorable adsorption was occurred at pH 3 for DCR and pH 10 for DB. The adsorption of dyes onto AAm/starch hydrogels is an endothermic process. Experimental data have been modeled by Freundlich isotherm. The second point was removal of the two dyes by degradation under the effect of gamma radiation. The degradation test of dyes was performed in aqueous solution under various radiation doses and pH and G-value was calculated. Destruction of 70% and 75% of the dye colour solutions was succeeded at radiation dose 40 and 30 kGy for DCR and DB, respectively. Comparing the removal percent of DCR and DB dyes by radiation degradation with that done by AAm/starch hydrogel adsorption was investigated. It was found that preferring removal of DCR and DB dyes by gamma radiation degradation. The novelty of the present study lays on the third point, where, adsorption of remaining dyes after their degradation to complete removal of direct red and direct blue dyes. It can be concluded that complete removal of DCR and DB was obtained by gamma radiation degradation followed by AAm/starch hydrogel adsorption.

Introduction

Adsorption is one of the most common methods used in wastewater treatment because it is economical, effective and simple in design. However, the adsorption method can only transfer the contaminants (dyes) from one phase to the other leaving the problem essentially unsolved. This problem can be solved by combining the conventional [with other methods]. Therefore, attention has to be focused on techniques that lead to the complete destruction of the dye molecules [5,6]. Gamma rays are one of the most promising advanced oxidation processes for environmental remediation where the produced hydroxyl radicals are the main oxidants for the degradation of dyes [7]. Hydrogels are polymeric materials that imbibe a considerable amount of water within a polymeric network without dissolution in water while keeping its three-dimensional stability. These hydrogels are suitable for dyes removal due to their hydrophilic nature and three-dimensional polymeric network which can imbibe large amounts of water [8]. In this study adsorption of DCR and DB dyes using acrylamide/ starch hydrogel (AAm/starch) that prepared by gamma radiation was investigated. Also degradation of the two dyes by gamma radiation was studied. After the dyes degradation the remaining was adsorbed by (AAm/starch) hydrogels to obtain a complete removal of both dyes.

References

- G. A. Mahmoud, S. E. Abdel-Aal, N. A. Badway, S. A. Abo Farha, E. A. Alshafei, *Starch/Stärke*, 65, 1–10 (2013).
- M. F. Abou Taleb, D.E. Hegazy, G. A. Mahmoud, *International Journal of Polymeric Materials and Polymeric Biomaterials*, 63, 840–845(2014).
- M.F. Abou Taleb, G.A. Mahmoud, S. M. Elsigeny, E.A. Hegazy, *Journal of Hazardous Materials*, 159, 372 (2008).
- G. A. Mahmoud, *Monatshefte für Chemie – Chemical Monthly* 145(2), 1-10 (2014).
- D. Georgiou, P. Melidis, A. Aivasidis, K. Gimouhopoulos, *Dyes and Pigments* 52, 69–78 (2002) *Chemical Reviews* 104, 1201–1217 (2004).
- L. A. Estroff and A. D. Hamilton, M. Pera-Titus, V. Garcia-Molina, M.A. Banos, J. Gimenez, S, Esplugas, *Applied Catalysis B* 47, 219–256 (2004)
- Y. Zheng, A. Wang, *Chemical Engineering Journal* 162, 186–193(2010).

Influence of initial pH

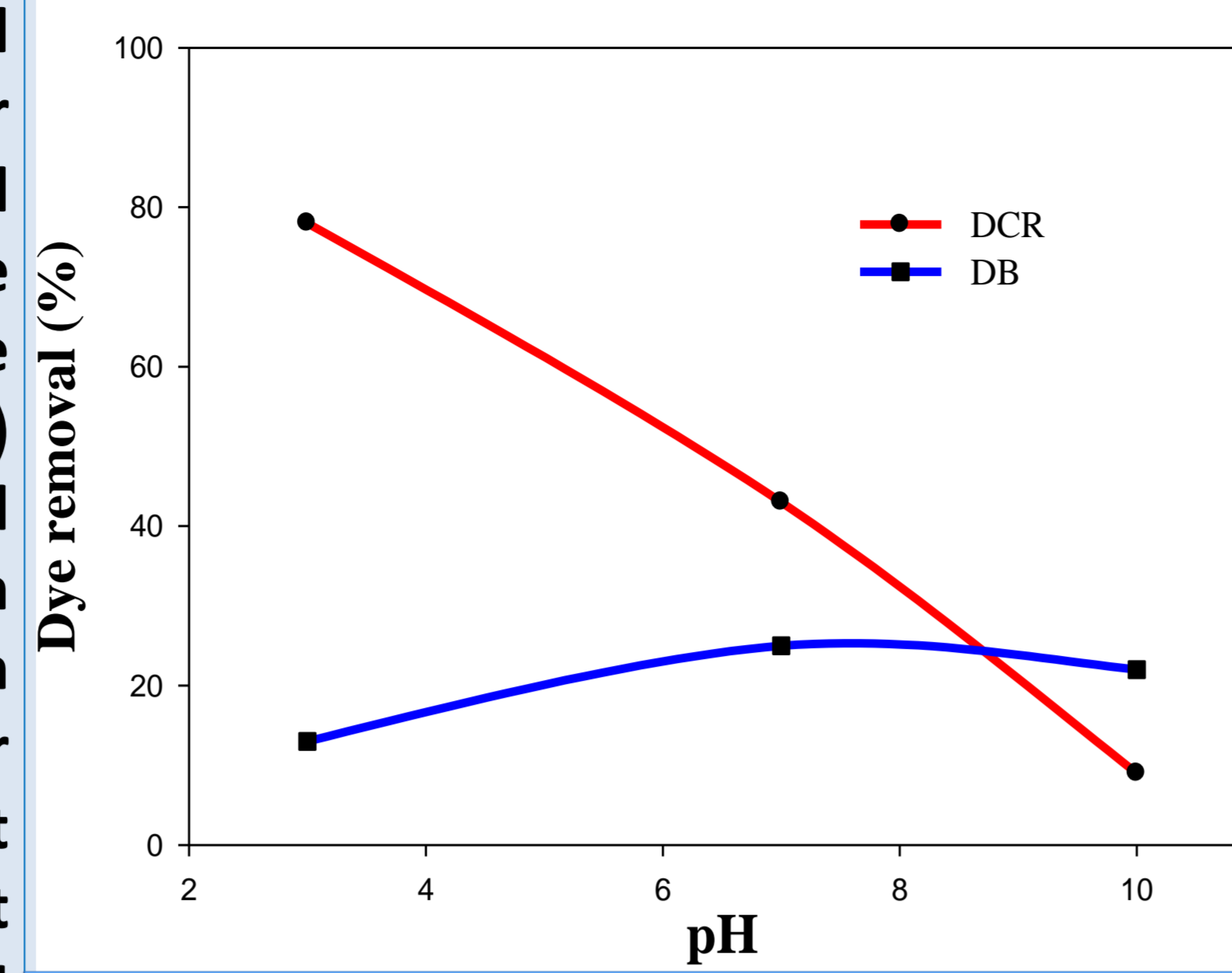


Figure (1): Effect of pH of medium on the removal percent of DCR and DB dyes onto AAm/starch hydrogels at room temperature and initial concentration 500 (mg/L).

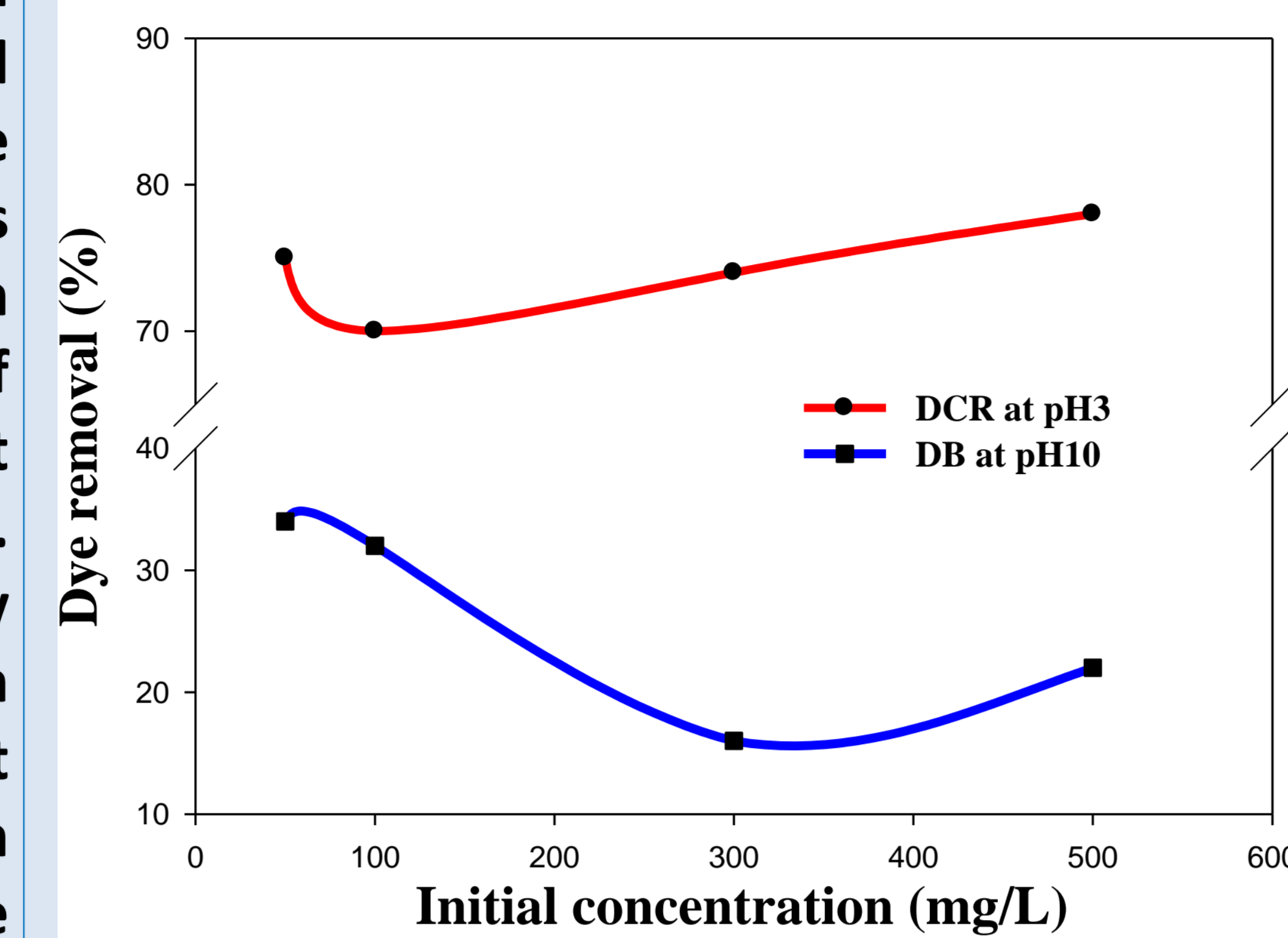


Figure (2): Effect of initial dye concentration on the removal percent of DCR and DB dyes onto AAm/starch hydrogels at room temperature.

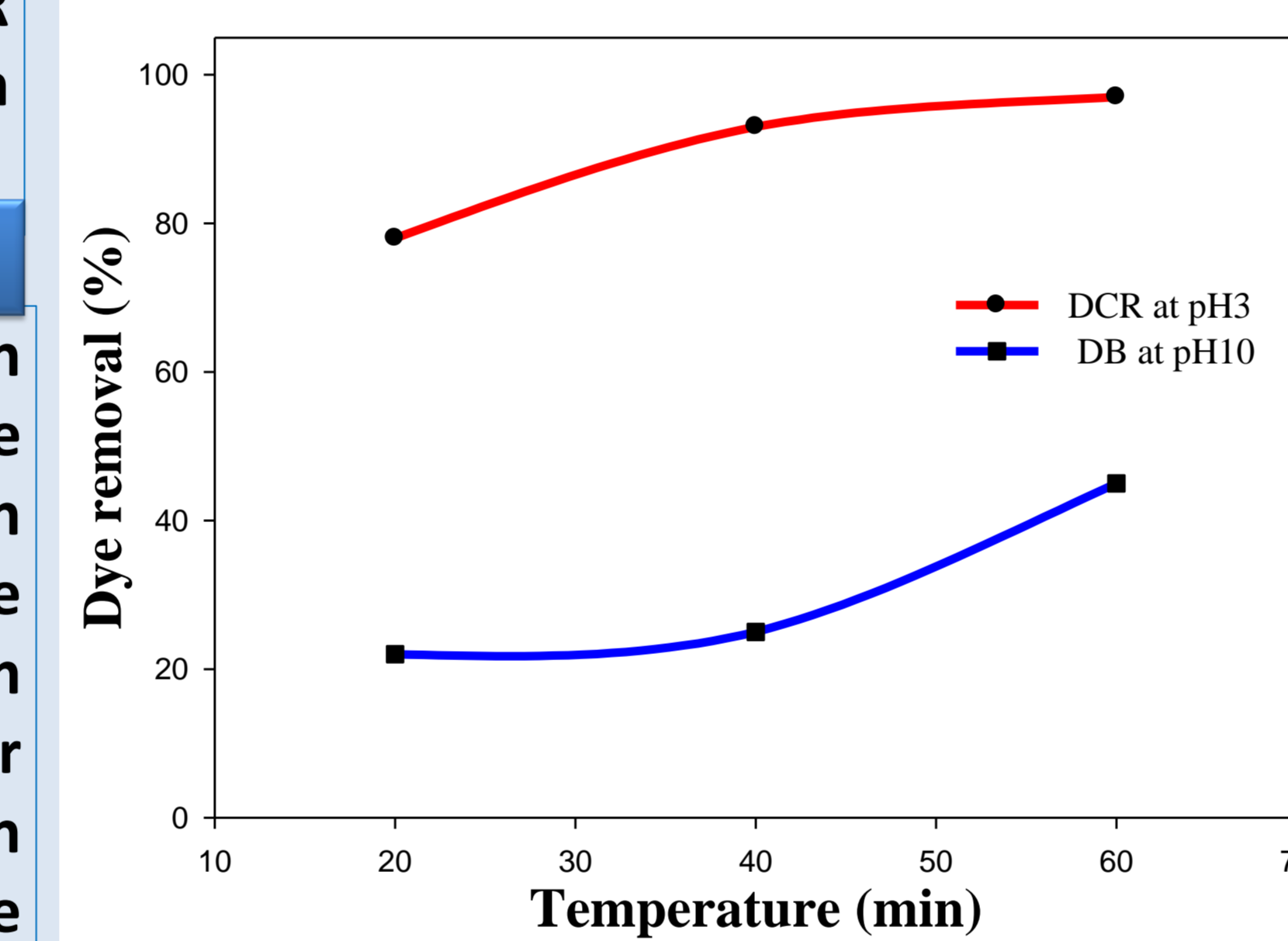


Figure (3): Effect of temperature on the removal percent of DCR and DB dyes onto AAm/starch hydrogels at initial dye concentration 500 mg/L.

Table 1: Isotherm constants of the Freundlich model for the adsorption of DCR and DB dyes onto AAm/starch hydrogel at various temperatures.

Isotherm parameters	DCR			DB		
	293k	313 k	333 k	293k	313 k	333 k
n	1.04	1.2	1.35	1.5	1.7	4.8
K_f ($mg^{1-1/n}L^{1/n}g^{-1}$)	1.2	3.6	10	0.2	2.82	1.7
R ²	0.92	0.98	0.97	0.96	0.98	0.98

Table 2: Thermodynamic parameters for adsorption of DCR and DB onto AAm/starch hydrogel

Initial concentration mg/L	ΔH kJ/mol	ΔS J/mol	ΔG_{293} kJ/mol	ΔG_{313} kJ/mol	ΔG_{333} kJ/mol
DCR dye					
50	15.6	64	-3.18	-4.46	-5.74
100	25.5	94	-2.04	-3.92	-5.8
500	39.7	163	-3.52	-6.78	-10.04
DB dye					
50	35.7	86	0.54	-1.86	-4.26
100	27.9	42	1.53	-0.27	-2.07
500	17.7	83.2	3.05	2.05	1.05

Degradation of dyes using gamma radiation

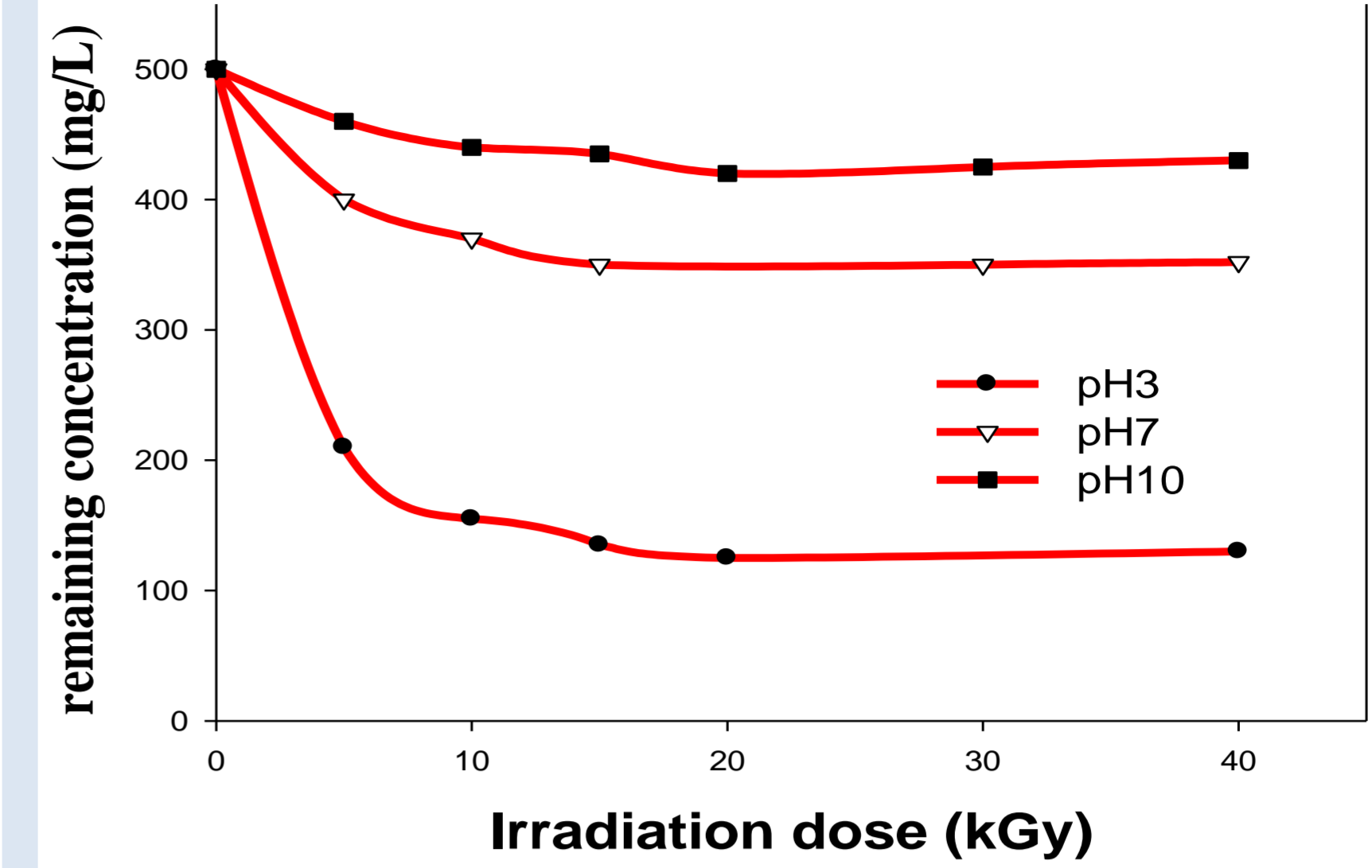


Figure (4): Effect of gamma radiation dose on degradation of DCR dye at initial concentration 500 mg/L and radiation dose rate 1.2 Gy/sec.

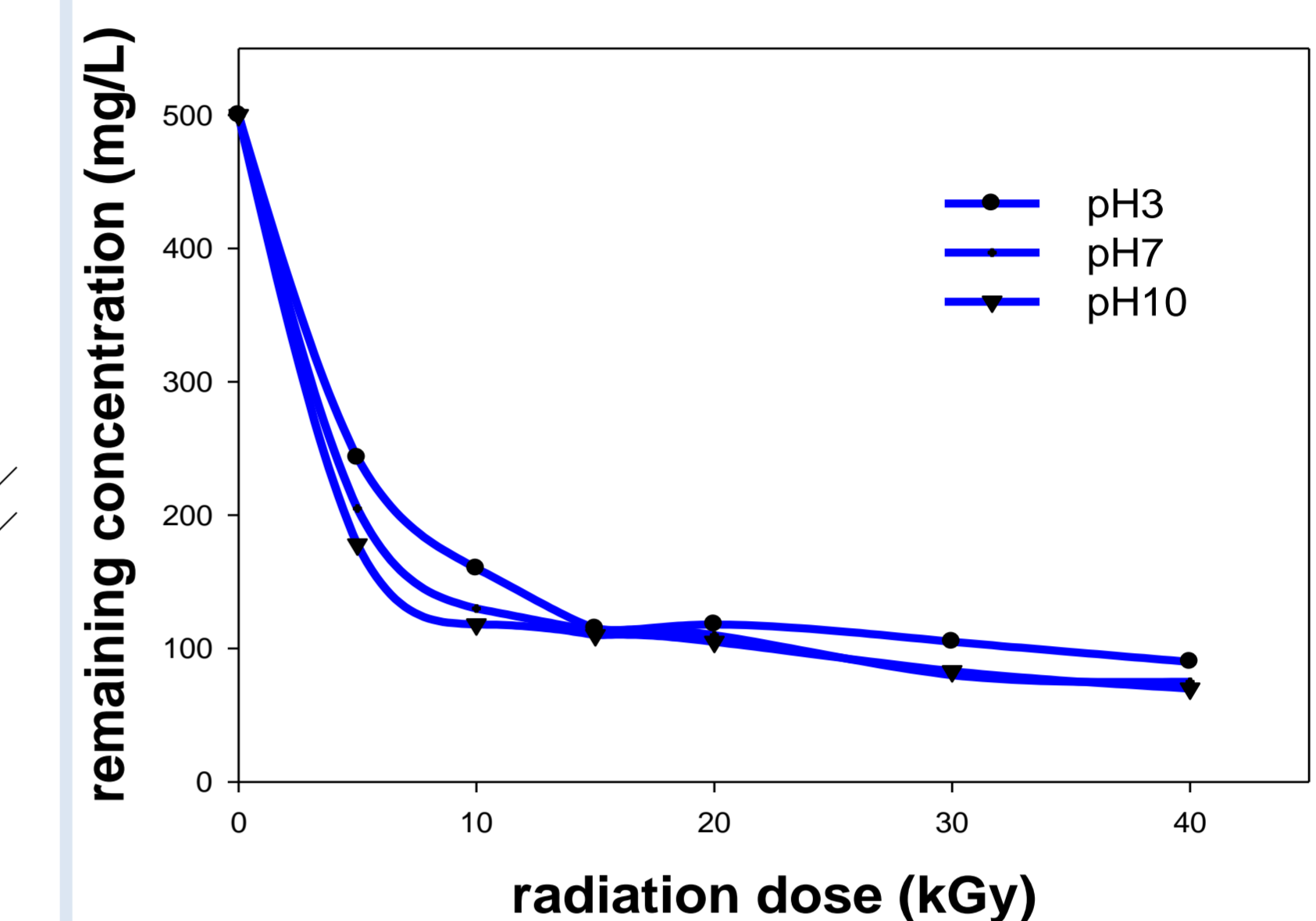


Figure (5): Effect of gamma radiation dose on degradation of DB dye at initial concentration 500 mg/L and radiation dose rate 1.2 Gy/sec.

Table 3: G values of DCR and DB dyes degradation by gamma radiation at various pH's and concentrations

pH	G values of dyes				
	Initial concentration (mg/L)				
	500	250	100	50	25
DCR dye					
3	0.26	0.14	0.014	0.012	0.011
7	0.14	0.056	0.034	0.02	0.014
10	0.035	0.042	0.031	0.020	0.0138
DB dye					
3	0.26	0.16	0.094	0.032	0.04
7	0.15	0.12	0.098	0.03	0.022
10	0.11	0.10	0.092	0.03	0.01

Conclusions

AAm/starch hydrogel prepared by gamma radiation was used for removal of DCR and DB dyes from aqueous solution. Favorable adsorption occurs at pH 3 for DCR and pH 10 for DB. The adsorption of dyes onto AAm/starch hydrogels is an endothermic process. Freundlich isotherm was made at different temperatures and their constants were calculated. Thermodynamic parameters such as (ΔH°), (ΔG°) and (ΔS°) were calculated. The negative values of ΔG° indicate the spontaneity of the process, whereas the positive values of ΔH° and ΔS° indicate the endothermic nature and increase in randomness of the process, respectively. The degradability of DCR and DB dyes at initial concentration of 500 mg/L at various pH's was examined and G- values were calculated. Comparing the removal percent of DCR and DB dyes by radiation degradation with that done by AAm/starch hydrogel adsorption is investigated. It can be observed that the removal percent of DCR and DB by radiation degradation was 70 and 75 % by adsorption was 78 and 28%, respectively. This means that preferring removal of DB dyes by gamma radiation degradation. Complete removal of DCR and DB was obtained by gamma radiation degradation followed by AAm/starch hydrogel adsorption.