Preparation of Selective Hazardous Metal Ion Adsorbents from Acrylic Monomer Grafted PET Films

Nazia Rahman[1]*, N. C. Dafader[1], Salma Sultana[1], M. F. Alam[1], Kazuhiro Hara[2]

[1]Nuclear and Radiation Chemistry Division, Institute of Nuclear Science and Technology, Atomic Energy Research Establishment, Savar, Dhaka-1349, Bangladesh

[2]Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University
744 Moto-oka, Nishi-ku, Fukuoka 8189-0395, Japan

*E-mail: naziabaec@gmail.com

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Introduction

Heavy Metal Pollution → Minamata disease (organic mercury poisoning); Itai-itai disease (cadmium poisoning)

Conventional methods for removal of hazardous metal

- Precipitation — Produces toxic sludge
- Ion exchange — High operational cost
- Electro-deposition method — Requires electricity
- Activated carbon adsorption — Difficult to regenerate

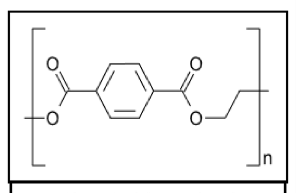
It is necessary to develop alternative low cost effective adsorbents

One of the new developments in recent years

Functional monomer grafted synthetic polymers as adsorbent
Relatively high adsorption capacity, selectivity and easy to regenerate

Poly(ethylene terephthalate), PET

PET is a thermoplastic polymer resin of the polyester family which is exclusively used in synthetic fibers; beverage, food and other liquid containers; and engineering applications



Chemical formula of Polyethylene Terephthalate

Beverage, food and other liquid containers of PET

Reasons behind choosing PET to prepare adsorbent

- Excellent chemical and thermal stability and mechanical properties of PET
- Desirable properties can be imparted to PET by grafting with functional monomers
- Future prospect of recycling of waste PET materials in heavy metal removal

Bales of crushed PET bottles

PET bottle flakes

Grafting of functional monomers on PET to bring metal adsorption property

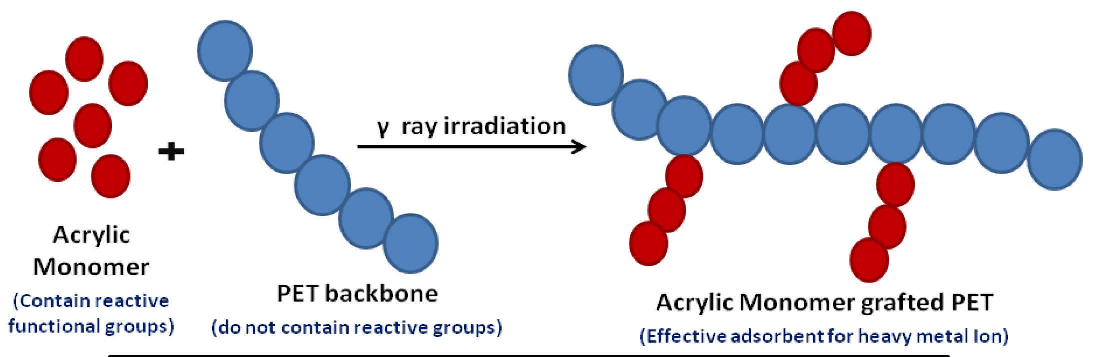


Figure: Graft co-polymerization of acrylic monomers on PET

Commonly used methods for initiating graft co-polymerization

- Ionizing radiation
- Ultraviolet light
- Plasma treatment
- Chemical initiators

Advantage of radiation grafting
- extensive penetration
- rapid formation of radicals
- less atmospheric pollution

Selective metal adsorption: Recovery of a specific metal ion from mixture of metal ions

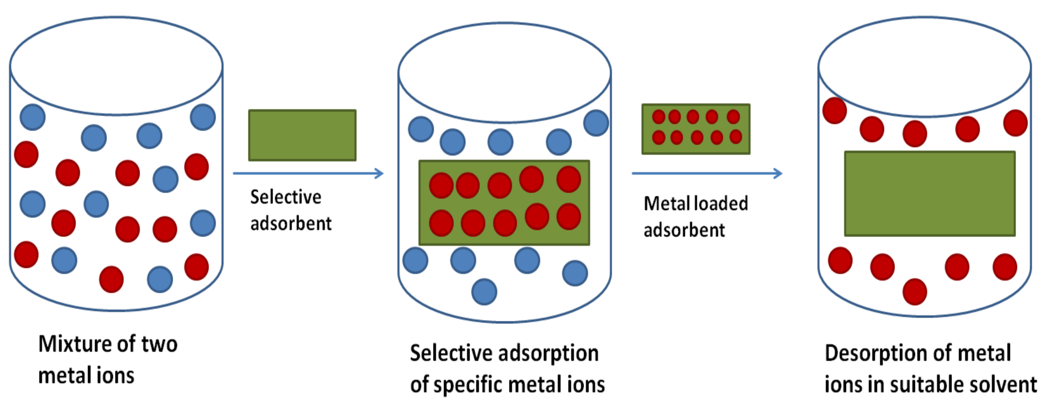


Figure: Schematic presentation of selective metal adsorption process

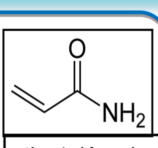
Advantages of selective metal ion adsorption

- Permits recovery of a specific metal ion from mixture of metal ions.
- Allows the re-use of the recovered metal ion.
- Reduces risk of secondary pollution by the recovered metal ions.

Overview of present study

Grafting of different acrylic monomers on PET film by γ - ray irradiation for selective adsorption of metal ions

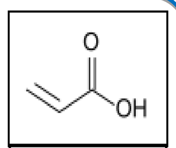
Part-1: Development of a method for γ - ray induced grafting of acrylamide (AAm) on PET film → Analysis of effects of grafting on the surface structures and on the Hg(II) adsorption properties



Chemical formula of acrylamide

Part-2: Application of AAm grafted PET film for selective adsorption of Hg(II) from mixture of Hg(II) and Pb(II) → Investigate the selective Hg(II) adsorption process: isotherm and kinetics, desorption and reusability

Part-3: Grafting of acrylic acid on PET film for selective adsorption of Cu(II) from mixture of Cu(II), Co(II) and Ni(II) → Investigate the selective Cu(II) adsorption process: isotherm and kinetics, desorption and reusability



Chemical formula of acrylic acid

Part-1: The effect of hot DMSO treatment on γ -ray induced grafting of acrylamide onto PET film

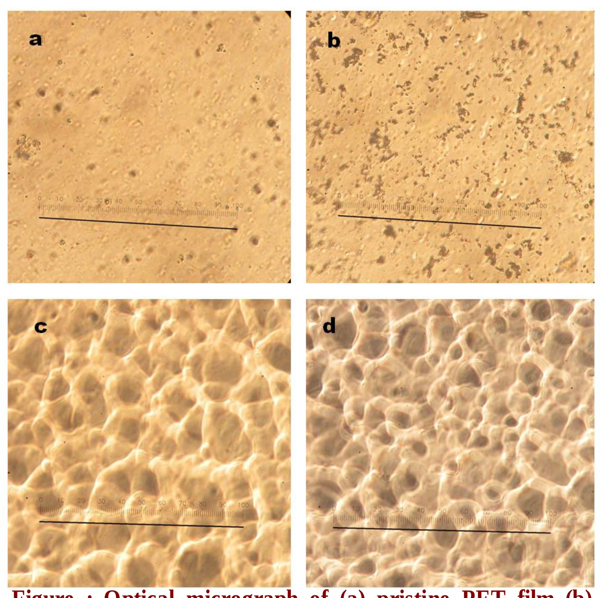


Figure : Optical micrograph of (a) pristine PET film (b) DMSO treated (at 140 °C) wet film (c) AAm-soaked wet film (d) AAm-grafted-PET film [The black line inside each figure represent 100 micrometer].

- A new method for the γ -ray induced grafting of acrylamide (AAm) onto PET films through DMSO pretreatment was developed.

- Much higher graft yield was achieved than previous studies

Table: A comparison of graft yield and/or graft density derived in the present study (γ-ray) with the results obtained using other methods.

Grafting method	Graft yield (%)	Graft density (μg/cm²)
UV radiation [Coşkun et al., 2006]	0.3 [a]	-
UV radiation [Bağ et al., 2000]	-	10 [a]
CO_2 laser [Bozkaya et al., 2012]	-	359 [a]
SI-ATRP [Coşkun et al., 2006]	1.37 [a]	-
SI-ATRP [Zhang et al., 2010]	2.52 [a]	-
γ-ray irradiation [present study]	15.5 (± 0.5)	1125 (± 25)

[a] the values are described as they appear in the references

Part-2 : Selective Hg(II) adsorption from aqueous solutions of Hg(II) and Pb(II) by hydrolyzed acrylamide-grafted PET films

- AAm grafted PET films were hydrolyzed by KOH treatment and the hydrolyzed film was used to study adsorption of Hg(II) and Pb(II) ions from aqueous solutions.

Table: Hg(II) and Pb(II) adsorption capacity of AAm-grafted PET and hydrolyzed AAm-grafted PET compared with those of other adsorbents (from single metal solutions)

Adsorbent	Hg(II) adsorption capacity (mg/g)	Pb(II) adsorption capacity (mg/g)
Pristine PET film [present study]	0 [a]	0 [a]
AAm-grafted PET film [present study]	15.0 [a]	1.3 [a]
Hydrolyzed AAm-grafted PET film [present study]	70.0 [a]	8.0 [a]
4-Vinyl pyridine/2-hydroxyethylmethacrylate-grafted PET fiber [Temoçin et al., Water Air Soil Pollut. 2010]	15.72 [b]	1.21 [b]
Activated carbon obtained from palm oil by products [Wahi et al., World Appl. Sci. J. 2009,]	52.67 [b]	48.96 [b]
Modified starch-based adsorbent [Huang et al., J. Hazard. Mater. 2011]	131.2 [b]	123.2 [b]
Poly(HEMA/chitosan) membranes [Bayramoglu et al., J. Appl. Polym. Sci. 2007]	39.5 [a]	37.0 [a]

a: experimental value, *b*: calculated from the Langmuir model

Part-2 : Summary

- The newly developed hydrolyzed AAm-grafted PET films were found to show high adsorption-selectivity for Hg(II) (~ 24 mg/g) over Pb(II) (~ 5 mg/g) from their mixture solution (initial metal concentrations of 100 mg/L and pH 4.5.
- The adsorbent can be regenerated and re-used for selective Hg(II) adsorption.

Part-3: Selective Cu(II) adsorption from aqueous solutions including Cu(II), Co(II) and Ni(II) by modified acrylic acid grafted PET film

- Grafting of AAc acid onto PET films was carried out by gamma irradiation and grafted film were modified by KOH treatment.
- The modified film was used to study adsorption of Cu(II), Co(II) and Ni(II) ions from aqueous solutions.

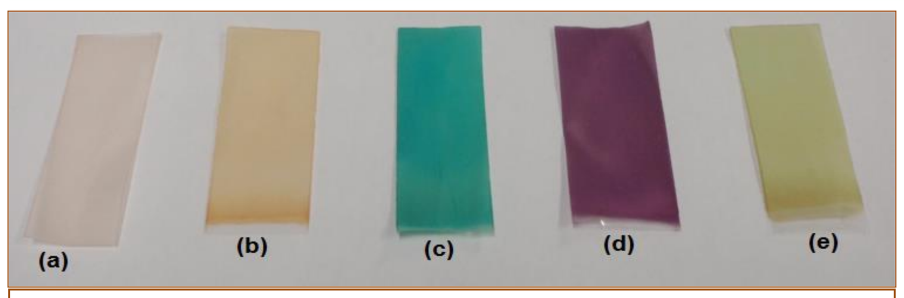


Figure: (a) AAc grafted PET (b) AAc grafted PET after modification by KOH treatment (c) Modified AAc grafted PET film after Cu(II) loading (d) Modified AAc grafted PET film after Co(II) loading (c) Modified AAc grafted PET film after Ni(II) loading.

Table : Metal ion adsorption capacity of AAc grafted PET, modified AAc grafted PET compared with some other adsorbents (From single metal solution).

Adsorbent	Cu^{2+} adsorption capacity (mg/g)	Co^{2+} adsorption capacity (mg/g)	Ni^{2+} adsorption capacity (mg/g)
Pristine PET film [present study]	0 [a]	0 [a]	0 [a]
AAc graft PET film [present study]	10.0 [a]	7.0 [a]	8.0 [a]
Modified AAc graft PET film [present study]	100.0 [a]	67.0 [a]	85.0 [a]
Itaconic acid/acrylamide graft PET fiber [Çoşkun et al., 2006- React. Funct. Polym]	7.73 [b]	14.81 [b]	13.79 [b]
Methacrylic acid/acrylamide graft PET fiber [Çoşkun et al., 2006- Sep. Purif. Technol]	31.25 [b]	27.17 [b]	43.48 [b]
Cross-linked and non cross-linked chitosan [Schmuhl et al., 2001]	>80 [b]	-	-

a: experimental value, *b*: calculated from the Langmuir model

Part-3: Summary

- The prepared AAc grafted film showed high selectivity towards Cu(II) (~55 mg/g) over other heavy metals (~ 7.8 mg/g for Co(II) and ~ 9.0 mg/g for Ni(II)) from their solution (initial metal concentrations 2000 mg/L and pH 4.
- The film can be used repeatedly for selective Cu(II) sorption.

Conclusive remarks of present study

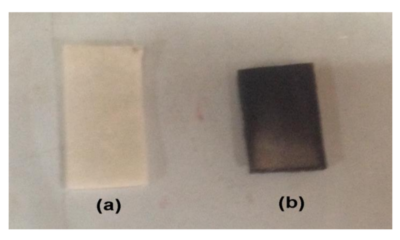
Thus present study shows

- The preparation of new selective adsorbents through γ - ray induced grafting of functional monomers on PET film.
- Adsorption of specific metal ions selectively and effectively from binary and ternary metal solutions.

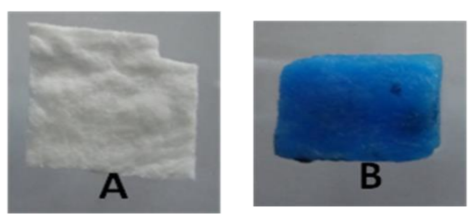
Some other ongoing research work

(1) Amidoxime adsorbent has prepared by radiation induced grafting of acrylonitrile on polyethylene film. The prepared adsorbent is very effective for Cr(VI) removal, The highest adsorption capacity obtained was 200 mg/g of adsorbent.

(2) Iminodiacetate group containing adsorbent (IDA) has prepared from GMA-g-PE non-woven fabric. The chromium adsorption of IDA adsorbent reached to 83.75 mg/g of adsorbent.



Amidoxime adsorbent before (a) after (b) Cr(VI) adsorption



Physical appearance of IDA adsorbent before (A) and after (B) copper adsorption