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TREATMENT OF WASTEWATER IN PLATING INDUSTRY BY CHELATE EXTRACTION METHOD

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ABSTRACT

One of the most efficient methods to preserve environment is the procedure and method to achieve effective materials and the dual and reapplication of natural sources. In this research, After carrying out a vast study on the growing spot of Iranian oaks , of the 36 species and subspecies already identified, 10 species and subspecies were selected as they cover a vaster surface of Iranian woods in North Alborz Region, Arsabaran region and the western part of Zagros Mountains. 5 samples were collected of each type form diffrenet spots of the country. The effective and usable substances of oak fruit were extracted and isolated by the four methods of maceration, decoction, perculation and soxhlet tannin in one specie show no significant difference; However the amount of tannin in different species show significant differences in statistical term in such a way that the minimum and maximum tannin was found in Q.Brantii, Belangri with 9.7% tannin and Q.Macrantera, with 3.2 percent tannin. This amount was put in contact with wastewater various from plating industries in 20 to 50 mg/L amount. The mentioned wastewaters contained 50 to 750 mg/L heavy metals (chromium, nickel, zinc, copper and silver). The results were analyzed through a biofactor variance analysis model with repetition in each house (Toki) Results showed that the removal output for zinc, copper, nickel and chromium without considering the initial concentration of the metal were 91 to 95 percent, 71 to 83.5 percent, 59 to 90 percent, and 84 to 85 percent, respectively. Also, with repect to the studies carried out with different methods and comparing the percentage of metals removal it was concluded that the removals output through applying 50 mg/L. hydroxide calcium along with 50 mg/L tannin with 82.3 percent average had the most effects in entire metals.

Key word: Wood, oak tree, extraction, heavy metals, plating wastewater

INTRODUCTION

The west and northwest forests of Iran covers approximate 5.2 million acres of Iran, representing more than 49 percent of the entire forest area of Iran. Out of 12.3 million acres of forests in Iran, 85 percent is in form of thin and scattered forests and there is only 15 percent dense woods (Fatahi, 1994). The area of scattered and open forests in Iran is twice the average number of open woods. This points out the sensitivity of forest and vegetation cover in different regions of Iran. According to the reports of Forests and Pastures Organization, the size of restoration of woods in Iran until 1979 was around 100,000, from 1979 to 1989 it was only 20,163 acres, to 1995 it was 356,507 and until 1998 the restoration covered 392787 acres (State woods and pastures, 1999).On the other hand, the statistics of annual destruction process of Forests in Iran is 130,000 acres per year (Fatahi, 1994). These figures reveal that despite considerable efforts of concerned organizations, for many reasons, the process of this deforestation is still expanding (State woods and pastures, 1999). Generally, the woods in west and northwest of Iran expand to 20 provinces of the state and have made their special forest communities in valleys, hillsides and mountain parts, according to the nature of their species (State woods and pastures, 1999).Based on the statistics of the General Department of Natural Resources, oak-trees form around 80 percent of the 750,000 acre woods of Kohgiloyeh and Bouyerahmad Province (Fatahi, *et al.*, 1969). The amount of oak nuts during acorn period from this region was about 260,000 Tons. Value of each kg of oak nuts is

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about 200 Rials; therefore, the yield will reach around 50 billion Rials (pub. Min. Agr. Rural, Dev., 1982). The number of yielding oak-trees in this province is 13 million grown trees and the amount of fruit products during yielding year is around 20 kg each tree. It has to be noted that as we proceed towards northern regions like Kermanshah ad Kurdistan, because of sensitivity of the specifications of the region (Q. infectoria), several drought years in the past few years, harvested for fuel and provisions for animals and shortness of precipitation during the year, have all influenced on the number of yielding trees and most trees have transformed to plane branches. Although the woods of these regions are not used as plantation for industrial purpose, the function of these woods in some instances is much more valuable than the woods of northern region of the Country in term of national economic, as far as environment , soil and water preservation may concern. Forty percent of the surface waters of Iran flows in Zagros region. There are 7 first grade rivers that pass through Zagros woods to the cultivating valleys. The area of the woods and the twigs of trees are the source of cattle provision for the tribal groups of region. Also, in Arsabaran, oak fruit is used to keep turkey and goat. The oak-tree trunk is usually used as fuel in rural and tribe areas (Fatahi, *et al*., 1994). Arsabaran growing region is in north part of East Azarbaijan province, in the area of Aras River catchment .The area of the woods in this region is assessed to be 164000 acres. The existence of special climate and a semi-humid and foggy air have led to formation of semi-dense and bulk forests in this region. With respect to the genetic variety of the species, this area is registered as the genetic reserves of world. More than 100 wooden species are in this area, the most important of which are oak, beech, white maple, mountain elm, yew, ash tree, quince leaved medlar, almond, maple, juniper, blueberry and etc. (Javanshir, 1976, Mosadeq, 1996). It has to be noted that the growing region of Northern Zagros has semi-humid and cool climate with cold and Alpine winters. This region recieves relatively good sum of snow in winter with annual percipitation of 800 mm(Alijani,K.,1989). The soil in this is multilayer and mixed with lime materials and pH is 6-8 (Fatahi, 1994). However, South Zagros has dry and hot climate and by proceeding to east, the rate of rain drop decreases (excluding parts of Lorestan to Zardkouh Bakhtiari). These regions recieves scattered area of precipitation with less than 550 mm per year (Alijani, 1989). In more southern regions, close to desert parts, the rate of rain drop reaches to 150 mm per year(Fatahi, *et al*., 1994). The type of soil is brown, lithocell and colluvium and the thickness is less than the northern region.The total area of growing land in Zagros region is assessed to be around 5.2 million acres and they are in Kurdistan, Kermanshah, Lorestan, Chahrmahal Bakhtiari, Khuzestan, Ilam, Kohgilouyeh and Bouyerahmad along with some part of Fars (Sabeti, 1976).

MATERIALS AND METHODS

In order to updated the information's of oak-tree woods in west and northwest Zagros, along with environmental applications, oak fruit is considered as an assest to preserve oak species by Javanshir, *et al*., 1976. In this project, was studied and over reviewed (Fatahi, 1994). Asar Region in northeast Lorestan, Javanroud region in north part of Kermanshah Province, Sovin region in Baneh, surrounding areas of Sardasht and Marivan, Dareh Boiylapoush valley around Khoy in West Azarbaijan, regions around Caliber in north of Tabriz, Khoda Afarin, Kalaleh Regions in borderlines of Iran and Republic of Armenia, Makini region in north Kalibar in East Azarbaijan, Talesh region in North Gilan and Naharkhoran region in Gorgan were stadied. Field studies were carried out by photography and direct sampling of oak trees, including the twigs with fruit and calyx. All oak tree fruit samples were pressed and extracted liqued. The samples were dried up with a suitable way. (picture 1-4) (Javanshir, 1970). Then, the species of oak tree fruit were identified via identification keys (Javanshir, 1970). In conclusion, following identification of origin species, the amount of yield of each tree in each region is determined and compared with the yields of previous years (Pub. Min. Agr. Rural, Dev., 1982). Among the current methods of liquid extraction from oak tree fruits, the maceration method was chosen and performed as follows:

Fig. 1: Quercus

Fig. 2: Q. Infectoria (sub) latifolia

Fig. 3: Quercus magnosquamata

Fig. 4: Quercus libani

- 10 g semi-smashed fruit is weighed and put inside a 250 mL Beaker containing 100 mL distilled water and is left for 24 hours.
- The specimen is then warmed up in the hot water bath for two hours and the filtration was performed.
- The left over of filteration was extracted one more time (Fatahi, 1969) (Makkar, H. P., *et al.*, 1991).
- For measurement of tannin available in the fruit of different species of Iranian oak trees, a number of methods were studied, namely, blood

hemoglobin (Hagerman, 1978), chorimertry, vanillin acid digestion and Leuin Shawl's methods (Robbers, 1996) and with respect to the available devices and the exactness of the test, the Leuin Shawl's method was chosen. Now, using the following formula, the total restorable amount an also, the total restorable amount besides tannin is calculated and by following formula according to a procedure by robbers J. E.(1996).

$$(V_1 - V_0) = \frac{P}{1000} * \frac{250}{10} * \frac{100}{M_0} * \frac{1000}{D}$$

(Total restorable amount besides tannin)- (total restorable amount by permanganate) = (Percent of pure tannin)

$(V_1 - V_0)$: The difference between the amount of permanganate used for the main subject and the witness group

P : Weight quantity of permanganate per mg. in mL of distilled water

M: Weight quantity of the species used in extraction per gram

D: Amount of dry material of the species.

With respect to the results obtained in table (Alijani, 1989), line 1 and 5 for Quercus Branti sub Belangri species, collected from Kouh Green region in Nahavand and Uraman region of Paveh city, show the highest amount of tannin (9.7 percent; therefore in all stages of subsequent tests, the oak extractions of those regions were used (Table 1). In this stage, with respect to the subsequent studies made on plating industry, the most important metals that can be measured in the waste water of those industries were known to be chromium, nickel, copper and zinc (Patterson, 1985). Based on this finding, 8 electro-plating unit that particularly used the above-mentioned plating procedures were chosen and samples were taken from their discharged wastewater in a standard form (Tsalev, *et al*., 1983). The samples were prepared in accordance with the standard procedures of preparation method and the amount of initial concentration of metals were measured by using atomic absorption spectrometry. The amount of metals elliminated in treated wastewater was assessed by Jar test device and under mix, clotting and residual procedures (Walter, *et al*., 1972). Flocc formation of heavy metals with tanin was optimized through exprimental process in accordance with the standard time is available with 50 mg/L hydroxide calcium, 50 mg/L. of hydroxide sodium and 50 mg/L extraction of oak essence (Thomas, *et al*., 1985).

Table 1: The percentage of pure tannin in various species of Iranian oak

Quercus	(V_1-V_2)	P/1000	250/10	100/M	100/D	percentage of tannin
Q.Brantii sub Belangri	28.78 ± 0.44	1.33 / 1000	25	10	100 / 96.3	10.1 – 9.8
Q.Brantii sub Brantii	15.04 ± 0.59	0.00133	25	10	100 / 97.9	5.3 – 4.9
Q.Infectoria sub Boisseri	15.12 ± 1.13	0.00133	25	10	100 / 96.4	5.6 – 4.8
Q.Infectoria sub Latifolia	21.98 ± 1.17	0.00133	25	10	100 / 97.1	7.93 – 7.12
Q.Brantii sub Belangri	27.8 ± 1.34	0.00133	25	10	100 / 96.3	10.1 – 9.1
Q.Infectoria sub Boisseri	12.18 ± 0.86	0.00133	25	10	100 / 96.4	4.5 – 3.9
Q.Magnosquamata	15.68 ± 0.51	0.00133	25	10	100 / 96	5.6 – 5.2
Q.Libanii	16.68 ± 1.34	0.00133	25	10	100 / 98.1	6.1 – 5.2
Q.Libanii	14.04 ± 1.25	0.00133	25	10	100 / 98.1	5.2 – 4.3
Q.Longipes	5.56 ± 0.34	0.00133	25	10	100 / 99.1	2 – 1.75
Q.Castanifolia	7.06 ± 0.68	0.00133	25	10	100 / 97.3	2.6 – 2.2
Q.Macrantera	8.68 ± 0.15	0.00133	25	10	100 / 98.6	3 – 2.8
Q.Macrantera	9.68 ± 0.59	0.00133	25	10	100 / 98.6	3.5 – 3
Q.Castanifolia	5.82 ± 0.68	0.00133	25	10	100 / 97.3	2.2 – 1.75
Q.Castanifolia	10.98 ± 0.41	0.00133	25	10	100 / 97.3	3.9 – 3.6
Q.Macrantera	10.42 ± 1.3	0.00133	25	10	100 / 98.6	3.9 – 3.1
Q.Castanifolia	10.82 ± 0.04	0.00133	25	10	100 / 97.3	3.7 – 3.6
Q.Kamarovii	19.32 ± 0.33	0.00133	25	10	100 / 96.5	6.8 – 6.5

The turbidimeter type HACH, model 2001A was used and the measured amounts which were in Nephlometric turbidity unit (NTU) term. Also, the pH meter device type Beckman, model 310 was used for pH measurement and the atomic absorption spectrometry type Oriental Instruments, model ANA 180 was used for this test. Four testing procedures as follows were used for all metals:

In the first stage 50 mg/L tannin-contained extract plus 50 mg/L hydroxide calcium, in the second stage 50 mg tannin-contained extract plus 50 mg/L hydroxide sodium, in the thrid stage 50 mg tannin extract, alone and in the fourth stage, 50 mg hydroxide calcium along were used. The findings of the above-mentioned tests are summarized

Table 2: The specifications of zinc reduction from raw wastewater of plating industries unit No. 1

Metal	Concentration of primary metal PPM	Primary tubidity NTU	secondary tubidity NTU	Primary pH	Secondary pH	$Ca(OH)_2$ mg/L	NaOH mg/L	Tannin extraction PPM	Concentration of secondary metal PPM
Zn	330	155	17	12.7	12.85	50	-	50	27.5
Zn	330	155	21	12.7	12.89	-	50	50	28.3
Zn	330	155	60	12.7	12.90	50	-	-	100
Zn	330	155	30	12.7	12.57	-	-	50	60

Table 3: The specifications of zinc reduction from raw wastewater of plating industries unit No. 2

Metal	Concentration of primary metal PPM	Primary tubidity NTU	secondary tubidity NTU	Primary pH	Secondary pH	$Ca(OH)_2$ mg/L	NaOH mg/L	Tannin extraction PPM	Concentration of secondary metal PPM
Zn	532	250	27	10.26	10.43	50	-	50	27
Zn	532	250	33.5	10.26	10.28	-	50	50	29
Zn	532	250	96	10.26	10.40	50	-	-	95
Zn	532	250	48.3	10.26	10.16	-	-	50	48

Table 4: The specifications of copper reduction from raw wastewater of plating industries unit No. 3

Metal	Concentration of primary metal PPM	Primary tubidity NTU	secondary tubidity NTU	Primary pH	Secondary pH	$Ca(OH)_2$ mg/L	NaOH mg/L	Tannin extraction PPM	Concentration of secondary metal PPM
Cu	143	96	20	6.12	7.24	50	-	50	42
Cu	143	96	35	6.12	7.2	-	50	50	53
Cu	143	96	56	6.12	7.5	50	-	-	100
Cu	143	96	42	6.12	6.05	-	-	50	73.5

Table 5: The specifications of copper reduction from raw wastewater of plating industries unit No. 4

Metal	Concentration of primary metal PPM	Primary tubidity NTU	secondary tubidity NTU	Primary pH	Secondary pH	$Ca(OH)_2$ mg/L	NaOH mg/L	Tannin extraction PPM	Concentration of secondary metal PPM
Cu	386	260	54	10.2	10.43	50	-	50	64
Cu	386	260	94	10.2	10.28	-	50	50	93
Cu	386	260	151	10.2	10.5	50	-	-	214.3
Cu	386	260	112	10.2	10.16	-	-	50	82.4

Table 6: The specifications of Nickel reduction from raw wastewater of plating industries unit No. 5

Metal	Concentration of primary metal PPM	Primary tubidity NTU	secondary tubidity NTU	Primary pH	Secondary pH	$Ca(OH)_2$ mg/L	NaOH mg/L	Tannin extraction PPM	Concentration of secondary metal PPM
Ni	250	144	75	7.82	8.25	100	-	50	103
Ni	250	144	86.4	7.82	8.74	-	200	50	108
Ni	250	144	101	7.82	8.66	100	-	-	163
Ni	250	144	93.6	7.82	7.53	-	-	50	132.7

Table 7: The specifications of Nickel reduction from raw wastewater of plating industries unit No. 6

Metal	Concentration of primary metal PPM	Primary tubidity NTU	secondary tubidity NTU	Primary pH	Secondary pH	$Ca(OH)_2$ mg/L	NaOH mg/L	Tannin extraction PPM	Concentration of secondary metal PPM
Ni	514	290	87	10.26	10.34	50	-	50	50
Ni	514	290	114	10.26	10.3	-	50	50	97
Ni	514	290	145	10.26	10.5	50	-	-	115
Ni	514	290	101.5	10.26	10.19	-	-	50	92

Table 8: The specifications of Chromiun reduction from raw wastewater of plating industries unit No. 7

Metal	Concentration of primary metal PPM	Primary tubidity NTU	secondary tubidity NTU	Primary pH	Secondary pH	$Ca(OH)_2$ mg/L	NaOH mg/L	Tannin extraction PPM	Concentration of secondary metal PPM
Cr	160	278	26	10.32	10.62	50	-	50	24
Cr	160	278	32	10.32	10.53	-	50	50	31
Cr	160	278	27	10.32	10.75	50	-	-	58
Cr	160	278	24	10.32	10.15	-	-	50	29

Table 9: The specifications of Chromiun reduction from raw wastewater of plating industries unit No. 8

Metal	Concentration of primary metal PPM	Primary tubidity NTU	secondary tubidity NTU	Primary pH	Secondary pH	$Ca(OH)_2$ mg/L	NaOH mg/L	Tannin extraction PPM	Concentration of secondary metal PPM
44	50	-	50	9.32	8.52	43	461	265	Cr
56	50	50	-	9.2	8.52	53	461	265	Cr
96	-	-	50	9.43	8.52	45	461	265	Cr
48	50	-	-	8.14	8.52	40	461	265	Cr

Table 10: The specifications of reduction different metals percentng of raw wastewater plating industries unit 1 to 8

Total mean percentage	Cr	Cr	Ni	Ni	Cu	Cu	Zn	Zn	Total omitting different metals
82.3	84	85	90	59	83.5	71	95	91*	50 mg/L $Ca(OH)_2$+50 mg/L Tanin extraction
77.8	79	80	81	57	76	63	94.5	92.5	50 mg/L NaOH+50 mg/L Tanin extraction
53.2	64	60	78	35	45	31	82	31	50 mg/L $Ca(OH)_2$
74	82	82	82	47	79	49	91	82	50 mg/L Tanin extraction

RESULTS

West and northwest of Zagros is the main source of reserving water in the six-catchments of Small Zab, Sefidroud, Persian Gulf (Karkheh), Dez, Simareh and Hendijan. Any destruction and moving back of forests in these regions will affect the area of surface water in the catchment reserves of a vast part of Iran (Badrifar, 1989; Golegolab, 1957). The Q. infectoria, Q. Persica species grow in lower altitudes (less than 1600 meter) in South Zagros which is drier and has a lower rate of rain drop (Tabatabaei, *et al.*, 1966). Because of more accessibility these species are in risk of destruction; while Q.marcanteera, which grow in higher regions (above 1600 meters) in northern Azarbaijan with higher rate of rain drop are not easily accessible and are in lower risk of destruction.Although one should not disregard the fact that because of development of borderlines during imposed war in Baneh, Marivan and Sardasht, the main region of Q.libani, the process of destruction is exceeding (Fig.s 5-7). Percentage of pure tannin extracted from various species of oak trees wer in range of (2.2 to 10.1). Highest amount of tannin was extracted from Q.Brantii sub Belangri specie.

Fig. 5: Baneh region, Marivan, Sourian District

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Fig. 5: Baneh region, Marivan, Sourian District

DISCUSSION

Forests restoration in the region during the past 20 years and the annual destruction rate of 130,000 acres and also, because of the area of the region one may conclude that preserving and saving these national resources require new policy making and relying on local inhabitants and people. Furthermore, developing additional applications for other yields of oak trees will justify the preservation of these trees by giving economic motivation to people. For example, with regard to the extractable Tannin substances from the various galls on Q.Infectoria following the insect stings , consists of 60 to 70 percent tannin, while this substance is imported in large amounts and is used in leather making and other industries. Such economic aspects not only provides necessary motivation for local people to preserve these species, but also, it can be a source of supplying the domestic needs of the Country along with planning for exportation. With respect to the findings obtained from the bi-factor variance analysis with one repetition in each box (Toki test), it was revealed that the pH controller leave same effect on the secondary concentration average of zinc, copper and nickel metals and in another word, both hydroxide calcium and hydroxide sodium leave same effects in the mean amount of residue of the above-mentioned metals ($p= 0.856$). Also, the above-mentioned compounds do not leave same effect on the secondary average of chrome concentration and in another word, hydroxide calcium and hydroxide sodium do not have same effects in the average of residue. ($p= 0.006$). Furthermore, the average of secondary concentration of zinc, copper, nickel and chrome metals- considering the separate effects of hydroxide calcium and tannin- show that both substances do not have same effect in the average of residuals of above-mentioned metals ($p= 0.035$).

Studying the findings show that if 50 mg/L hydroxide calcium and 50 mg/L tannin-contained extraction comes to contact with the discharged water, the best flocc formation of zinc, copper, nickel and chromium, without consideration to their amount of initial concentration in the waste water, is achieved and if the pH of the environment has a relatively neutral or a little alkaline reaction, a 50 gr/L tannin-content extract, too, can give the same results like the first test (Sarmad, *et al.*, 1991; Mosadeq, 1996) reported the procedure of flocc formation for different metals from the raw waste water of plating industries, as measured for units 1 to 8 during March 2003- March 2004. As the Table reveals, the output of zinc flocc formation from raw wastewater, without considering the initial concentration of zinc, is 91 to 95 percent for this metal, the output of copper flocc formation from raw wastewater, without considering the initial concentration of copper, is 71 to 83.5 percent, the output of nickel flocc formation from raw wastewater, without considering the initial concentration of nickel , is 59 to 90 percent and the output of chromium flocc formation from raw wastewater, without considering the initial concentration of chrome, is 84 to 85 percent for this metal. Also, with respect to the studied conducted with various methods and by comparing the percent of metals flocc formation in them, it can be concluded that the flocc formation output through applying 50 mg/L hydroxide calcium along with 50 mg/L. tannin-content extraction with 82.3 percent output showed the highest effects on all metals, using 50 mg/L hydroxide sodium along with 50 mg/L tannin-content extract with 77.8 percent had the second rank of importance, 50 mg tannin-content extract without pH controlling substances had the third rank with 74 percent average and finally, using 50 mg/L hydroxide calcium without tannin-content extract with 53.2 percent got the fourth rank in flocc formating heavy metals from different plating industries.

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