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Biosorption of Copper and Zinc from Aqueous Solutions by Different Species of Macroalgae

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ABSTRACT

Pollution of watercourses by heavy metals represents a serious problem. As conventional removal methods have some drawbacks, there become urgent needs for sorbents of biological origin which are cheap, easily available and effective. Four dried marine macroalgae were evaluated as biosorbents of copper and zinc ions. *Dictyota dichotoma* (Phaeophyta), *Jania adhaerens*, *Liagora farinosa* and *Digenea simplex* which belong to Rhodophyta. Batch biosorption method was used. Different physical and chemical parameters were optimized for biosorption. The effects of initial metal concentration and time of contact were studied; *Liagora farinosa* biomass was the most effective in biosorption of Cu^{+2} and Zn^{+2} from the four prepared concentrations, as the adsorption capacity reached its maximum values; 55.62 and 115.26 mg/g after 120 min in 60 and 80 ppm, respectively. It was found that the percentage of adsorption of the two metals increased with increasing contact time from 30 to 120 min. Among the four algal species, *Liagora farinosa* had the highest potential for effective removal of Cu^{+2} and Zn^{+2}.The best adsorption capacity was at 40 ppm of the two metals tested.

Keywords: *Dictyota dichotoma*, *Digenea simplex*, *Jania adhaerens*, *Liagora farinosa*, biosorption

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INTRODUCTION

Water pollution is becoming a serious problem threatening human health and ecosystem[1].Aqueous heavy metal pollution represents an important environmental problem, because heavy metals are not biodegradable and tend to accumulate in living organisms causing various diseases and disorders as well as deleterious ecological effects [2].The main sources of heavy metals pollution are mining, sludge disposal, refining ores, fly ash from incinerators, the processing of radioactive materials, mineral processing operations, or electroplating, paints, alloys, batteries, pesticides, fertilizers or preservatives and surface finishing industries, energy and fuel production, these sources discharge a variety of toxic metals such as cadmium, chromium, copper, zinc, lead, mercury, selenium, arsenic, gold, silver and nickel into the environment. As a result, removal of these toxins from industrial effluents has become an important priority that is reflected in tightening and enforcement of environmental regulations [3].

There is a wide range of treatment methods which have classically been employed for stripping toxic metals from wastewaters such as chemical precipitation, ion-exchange, adsorption, membrane filtration, coagulation–flocculation, flotation and electrochemical methods [4-6]. However, these methods have disadvantages, like incomplete metal removal, high reagent or energy requirements, and generation of toxic sludge or other heavy metal-containing waste products that may sometimes be more toxic than their parent ones [7]. Hence, new technologies are required that can reduce heavy metal concentrations to environmentally acceptable levels at affordable cost. Biological approaches, especially application of biosorbents, have been suggested in the last decade.

Biosorption is a term that describes the removal of heavy metals by the passive binding to non-living biomass from an aqueous solution [8]. Biosorption as an innovative technology has gained importance due to its advantages over conventional separation techniques .These advantages are the reusability of biomaterial, low operating cost, improved selectivity for specific metals of interest, short operation time and no production of secondary compounds which might be toxic [9-11]. Regarding the environmental impact, biosorption can be considered to belong to clean technologies. It is known that clean technologies and products are tools for environmental sustainability, as well as environmental policy with regard to integrated pollution prevention and control [12, 13].

The biomass can be composed of algae, fungi, bacteria, and various plant species. Algal biomasses are advantageous because of its availability in almost unlimited amounts [14], besides its high metal uptake capacity due to its physicochemical characteristics [15].Marine algae have been found to be potential suitable sorbents because of their low cost, relatively high surface area and high binding affinity. The use of marine algae for heavy metal removal has been reported by several authors [16-20].

The aim of the present work was to evaluate the sorption capacity of four different algae; *Dictyota dichotoma*, *Jania adhaerens*, *Liagora farinosa* and *Digenea simplex* collected from the Egyptian Red Sea coast in respect of two different heavy metals: copper and zinc.

MATERIALS AND METHODS

1-Algal collection and processing:

Four algal species, namely *Dictyota dichotoma* (Phaeophyta), *Jania adhaerens*, *Liagora farinosa* and *Digenea simplex* which belong to Rhodophyta were collected from Red Sea on the coast of Hurghada, Egypt, transferred to laboratory in labelled polyethylene bags. The samples were washed several times with de-ionized water to remove dirt, and/or other impurities present in the raw materials. They were air dried for 10 days, then grinded and sieved at the pore size of 0.5 to 1 mm [21].

2-Heavy metal stock solution preparation:

Cu^{+2} and Zn^{+2} standard solutions were prepared from copper sulphate and zinc chloride, respectively. Primarily 1000 ppm (1000 mg/l) from each salt was prepared, and then it was serially diluted with distilled water to get 20, 40, 60 and 80 ppm concentrations.

3-Batch biosorption procedures:

Biosorption experiment was carried out at room temperature (24±2°C) in 500 ml Erlenmeyer flasks containing the prepared concentrations. 250 mg of each biomass was added to each flask and the mixtures were agitated on a rotary shaker at 180 rpm. Controls for each concentration without addition of biomass were also maintained. 100 ml was taken from each flask at different time intervals; 30, 60, 90 and 120 min. The solutions were separated from the biomass by filtration [22]. 10 ml concentrated nitric acid was added to each 100 ml volume of each sample for acidification. Digestion was carried out through heating the mixture on a hot plate until the entire volume reached 5 ml. De-ionized water was added to each sample to reach the volume of 20 ml. Samples were filtered again through Whatman No. 1 filter paper, then subjected to analysis using microwave plasma - atomic emission spectrometer (Agilent 4200 MP-AES) to determine heavy metal concentration.

4-Calculation of metal uptake:

To determine the adsorption capacity of tested biomasses, the following equation was applied:

$$qe = (C0 - Ce)V/m \qquad (1)$$

Where qe is the amount adsorbed at equilibrium (mg/g), C0 is the initial metal ions concentration (mg/L), Ce is the equilibrium metal ions concentration (mg/L), V is the volume of the aqueous phase (L), and m is the amount of algae used (g) [23].

The percentage of adsorption was calculated as follows:
Percentage of adsorption = C0-Ce / Ce * 100 (2)

RESULTS AND DISCUSSION

Amongst biological agents studied as biosorbents, marine macroalgae were shown to be extremely efficient in their ability to bind various metals from aqueous effluents. This was attributed to their relative high surface area and binding affinity. Many macroalgae were proven to be effective in biosorption of different heavy metals; *Colpomenia sinuosa* for Cu and Ni [24], *Corallina officinalis* for Cd [25], *Gracilaria* sp. for Pb, Cu, Cd, Zn and Ni [26], *Ulva reticulata* for Zn [27] and *Turbinaria conoides* for Pb [28].

In this study, we focused on two heavy metals; Cu^{+2} and Zn^{+2}. Copper is one of the most interesting water pollutants, because in small concentrations it is actually an essential nutrient for both animals and humans, but over a specific limit it can cause severe health damage [29- 31]. When it is found in high levels in our organs, copper can cause nausea, vomiting and damage to the stomach. High concentrations of copper can cause destruction of red blood cells, possibly resulting in anemia [32-34].

In the present study, it was observed that *Liagora farinosa* biomass was the most effective in biosorption of Cu^{+2} from the four prepared concentrations, as the adsorption capacity reached its maximum values; 38.2 and 55.62 mg/g after 120 min in 40 and 60 ppm, respectively (Table 1). *Digenea simplex* came in the second rank, followed by *Jania adhaerens* and finally *Dictyota dichotoma* in their adsorption capacities of Cu^{+2} (Table 1).Various literature studies recorded the success of different algal species in copper biosorption; *Laminaria japonica* [35], *Sargassum vulgare* [8], *S. fusiforme* [20], *S. filipendula* [36], *Palmaria palmate* [37], *Chondrus crispus* [38], *Fucus serratus* [39], *F. vesiculosus*[40] and *Durvillaea antarctica* [19].

Concerning Zn^{+2} , previous studies indicated zinc toxicosis in various mammalian species, including sheep, cattle, pigs, horses, and dogs, mostly taking the form of copper deficiency caused by excessive zinc intake [41, 42]. In human, ingestion of excessive amounts of zinc salts causes fever, nausea, vomiting, stomach cramps, and diarrhoea. In our study, *L. farinosa* biomass was the most effective reaching its maximum adsorption capacities; 60.16 and 115.26 mg/g after 90 and 120 min in 80 ppm, respectively. On the other hand, *J. adhaerens* was the second effective biosorbent in 20 ppm while *D. dichotoma* came after *L. farinosa* in its adsorption capacity in 80 ppm (Table 2). Many investigators recorded the efficiency of certain red algae (Rhodophyta) in heavy metal uptake; Hamdy [43] reported that *Jania rubens* was effective as a biosorbent of lead. Tamilselvan et al. [44] found that *Acanthophora spicifera* showed maximum biosorption of Cd (II)

(92.68%), Pb (II) (51.84%), and Cr (III) (50.29%). Metal uptake capacity of 60.09 mg/g was seen in *Gracilaria edulis* (Red algae) at 400 mg/L of initial chromium concentration, followed by 59.31mg/g for *Padina tetrastromatica* (brown algae) [45]. The adsorption of copper ions from aqueous solution using the biomass of the red alga *Gigartina acicularis* was studied by El Hassouni et al. [46]; they reported maximum adsorption capacity of 14.77 mg/g at room temperature. It was found that red algae contain cellulose, but their ability to bind heavy metals lies in the presence of sulphated polysaccharides made of galactans (agar and carraghenates) [38]. These compounds contain several functional groups (amino, carboxyl, sulphate and hydroxyl) which play an important role in the biosorption process as indicated by FTIR analysis [44, 46].

Other studies recorded the superior effectivity of brown algae over red algae, e.g. Holan and Volesky [47] who stated that the lead and nickel biosorbent uptake decreased in the following sequence: Phaeophyta> Rhodophyta. The brown algae were represented by *Ascophyllum nodosum, Fucus vesiculosus,* and *Sargassum natans* while red ones were represented by *Chondrus crispus, Galaxaura marginata,* and *Palmaria palmate*. *Fucus spiralis* as a brown alga resulted in the lowest Cu and Zn concentration levels in the solutions tested [38]. The presence of alginates in the cell wall of brown algae could be responsible for such behavior by anchoring the metal to the biomass [35, 48]. Cationic exchange mechanism through acidic functional groups at the algal surface is supposed to be responsible for such metal uptake [39].

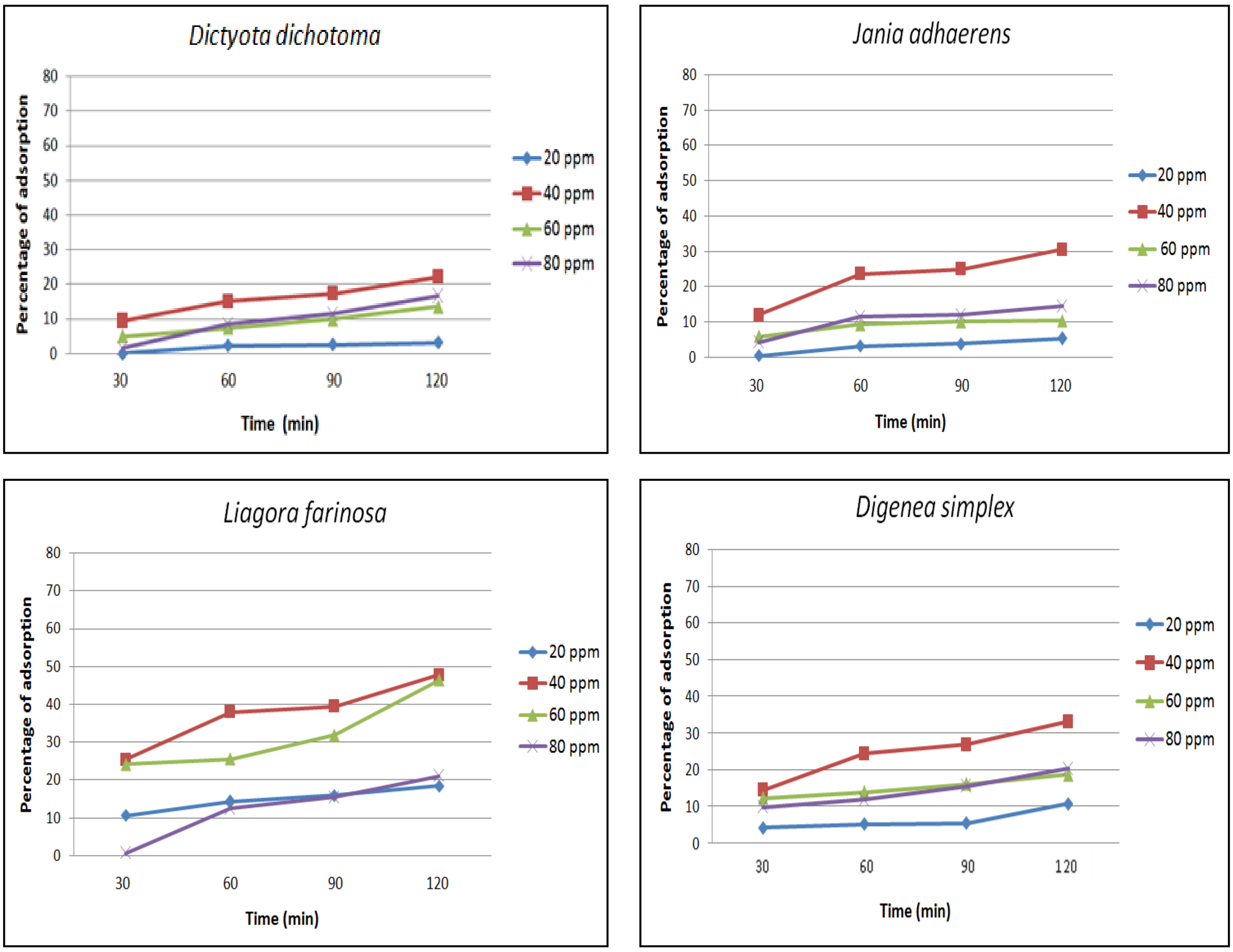

Fig. 1: Percentage of adsorption of Cu^{+2} by algal biomasses at different time intervals

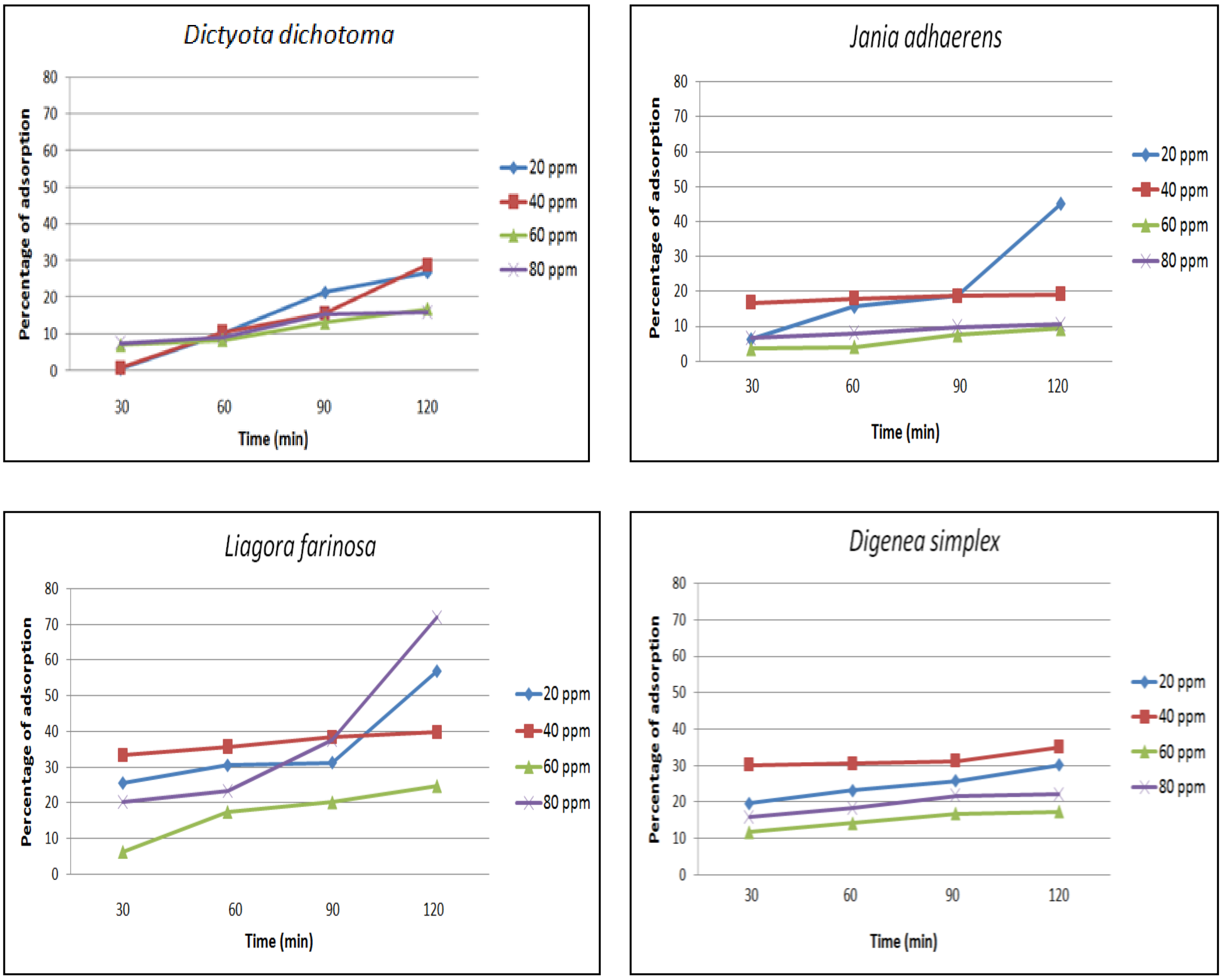

Fig. 2: Percentage of adsorption of Zn^{+2} by algal biomasses at different time intervals

Fig. 1 showed that the highest percentage of Cu^{+2}adsorption was at 40 ppm in the four investigated biomasses. Generally, the adsorption percentage of Zn^{+2} exhibited its highest values at 40 ppm, except that of *L. farinosa* after 120 min of exposure, as the percentages of 20 and 80 ppm adsorption exceeded that of 40 ppm (Fig. 2). Also the percentage of adsorption of 20 ppm by *J. adhaerens* was higher than that of 40 ppm after 120 min. In case of *D.* ***dichotoma***, the values of these percentages were so close (Fig. 2). Thus, the present results indicated that the increase in Cu^{+2} or Zn^{+2} concentrations didn't lead to subsequent increase in their adsorption. Similarly, Esmaeili et al. [49] found that the increase in initial concentration of Cr^{+6} from 10 to 60 mg L-1 led to decrease in the percentage of removal for both *Gracilaria* (red algae) and *Sargassum* sp. (brown algae). This may be due to saturation of the sorption sites on adsorbents. These results were in accordance with the observation made by Abdel-Aty et al. [50] on the biosorption of cadmium and lead from aqueous solution by fresh water alga, *Anabaena sphaerica* biomass. Such result can be attributed to less competition between the free bonding sites at low concentrations, which caused faster initial adsorption. When the unoccupied bonding sites decreased as the concentration increased, metal adsorption became more difficult and slower until equilibrium was achieved [51]. Contradictory, Li et al. [52] reported that the adsorption capacity of Cu (II) by the red alga ***Palmaria palmate*** increased with the increase of the initial copper concentration, Also, El-Hassouni et al. [46] showed that Cu (II) sorption capacity by *Gigartina acicularis* biomass increased with increasing initial metal concentration from 50 to 400 mg/L.

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Table (1): Adsorption capacity of Cu^{+2} concentrations by the four tested biomasses at different time intervals

Concentration (ppm)		20				40				60				80			
Time intervals (min)		30	60	90	120	30	60	90	120	30	60	90	120	30	60	90	120
Dictyota dichotoma	Concentration	20	19.55	19.51	19.37	36.26	33.98	33.12	31.18	57.03	55.61	54.06	51.95	78.86	73.35	70.84	66.74
	Adsorption capacity (mg/g)	0	0.9	0.98	1.26	7.48	12.04	13.76	17.64	5.94	8.78	11.88	16.1	2.28	13.3	18.32	26.52
Jania adhaerens	Concentration	19.93	19.38	19.24	18.96	35.2	30.61	30.05	27.8	56.49	54.73	53.95	53.85	76.67	73.19	70.5	68.52
	Adsorption capacity (mg/g)	0.14	1.24	1.52	2.08	9.6	18.78	19.9	24.4	7.02	10.54	12.1	12.3	6.66	13.62	19	22.96
Liagora farinosa	Concentration	17.9	17.16	16.83	16.32	29.86	24.85	24.24	20.9	45.52	44.74	40.94	32.19	79.54	70.12	67.69	63.18
	Adsorption capacity (mg/g)	4.2	5.68	6.34	7.36	20.28	30.3	31.52	38.2	28.96	30.52	38.12	55.62	0.92	19.76	24.62	33.64
Digenea simplex	Concentration	19.13	18.96	18.89	17.84	34.19	30.2	29.24	26.73	52.59	51.63	50.29	48.8	72.05	70.52	67.47	63.72
	Adsorption capacity (mg/g)	1.74	2.08	2.22	4.32	11.62	19.6	21.52	26.54	14.82	16.74	19.42	22.4	15.9	18.96	25.06	32.56

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Table (2): Adsorption capacity of Zn^{+2} concentrations by the four tested biomasses at different time intervals

Concentration (ppm)		20				40				60				80			
Time intervals (min)		30	60	90	120	30	60	90	120	30	60	90	120	30	60	90	120
Dictyota dichotoma	Concentration	19.91	17.95	15.72	14.66	39.71	35.78	33.77	28.45	55.77	55.05	52.14	49.89	74.02	72.66	67.63	67.3
	Adsorption capacity (mg/g)	0.18	4.1	8.56	10.68	0.58	8.44	12.46	23.1	8.46	9.9	15.72	20.22	11.96	14.68	24.74	25.4
Jania adhaerens	Concentration	18.75	16.87	16.24	11	33.32	32.84	32.58	32.35	57.88	57.61	55.53	54.45	74.69	73.69	72.2	71.68
	Adsorption capacity (mg/g)	2.5	6.26	7.52	18	13.36	14.32	14.84	15.3	4.24	4.78	8.94	11.1	10.62	12.62	15.6	16.64
Liagora farinosa	Concentration	14.91	13.92	13.76	8.62	26.6	25.68	24.62	24.09	56.25	49.53	47.93	45.18	63.89	61.45	49.92	22.37
	Adsorption capacity (mg/g)	10.18	12.16	12.48	22.76	26.8	28.64	30.76	31.82	7.5	20.94	24.14	29.64	32.22	37.1	60.16	115.26
Digenea simplex	Concentration	16.08	15.38	14.85	14	27.95	27.82	27.52	26.03	52.97	51.53	49.98	49.62	67.39	65.45	62.68	62.37
	Adsorption capacity (mg/g)	7.84	9.24	10.3	12	24.1	24.36	24.96	27.94	14.06	16.94	20.04	20.76	25.22	29.1	34.64	35.26

As the adsorbent dose, initial metal concentration, and temperature were kept constant, it was clear in the present study that both Cu^{+2} and Zn^{+2} uptake by the tested biomasses increased with increasing contact time from 30 to 120 min, and the optimum contact time was found to be 120 min. This is in agreement with Esmaeili et al. [49], they found that the removal of Cu^{+2} from wastewater by *Gracilaria corticata* was increased by time. Also, Esmaeili et al. [53] reported that Cr removal efficiency by *Gracilaria* and *Sargassum* sp. increased when contact time was increased from 15 to 120 min. The maximum biosorption by *Gracilaria corticata var cartecala* for Cr^{+3} was at 60 h, for Hg^{+2}, Pb^{+2}, and Cd^{+2} was after 24 h. [54]. Moreover, Christobel and Lipton [55] stated that the biosorption process was highly influenced by contact time, the adsorption of arsenic ions using 2g/100ml of *Ulva fasciata*, *Sargassum wightii* and *Gracilaria corticata* was rapid during the first 30 minutes, then it gradually increased, until the equilibrium was attained at 90 minutes. On the contrary, Xin et al. [56] found that the maximum adsorption (90%) of lead, copper, cadmium, zinc and nickel by the marine algae, *Sargassum* sp., *Ulva* sp. and *Gracilaria* occurred within 60 min. Similarly, fast adsorption of copper by *Palmaria palmata* occurred at the first 20 minutes and then slowed down until equilibrium was achieved [52]. The contact time could be evaluated as one of the important parameters affecting the biosorption efficiency. Different kinds of functional groups, with different affinities to the metal ions, are usually present on the biomass surface; the active binding groups with higher affinities are firstly occupied [57].

CONCLUSION

Liagora farinosa dried biomass was the most effective in biosorption of different concentrations of copper and zinc at different time intervals. Generally, the optimum concentration of Cu^{+2} to be adsorbed with the four tested macroalgae was 40 ppm. Few exceptions were observed in case of Zn^{+2} as the adsorption of 20, both 20 and 80 ppm exceeded that of 40 ppm after 120 min of exposure to *Jania adherens* and *Liagora farinosa*, respectively.

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