# *Recovery of Iodine with Activated Carbon from Dilute Aqueous Solutions*

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*Recuperación de yodo con carbón activado a partir de disoluciones acuosas diluidas*

*Recuperació de iode amb carbó activat a partir de dissolucions aquoses diluïdes*

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## RESUMEN

Se presenta un método para la extracción de yodo de disoluciones acuosas diluidas, tales como salmueras naturales y de salinas, mediante carbón activado granulado, seguido de desorción de yodo del carbón activado usando una disolución de hidróxido sódico. Se observa que la adsorción de yodo de disoluciones diluidas se puede correlacionar bien con la ecuación de Freundlich, y que la cinética de adsorción sigue aproximadamente una reacción irreversible de primer orden. Los resultados muestran que el pH de la disolución en la etapa de adsorción y la concentración de la disolución de hidróxido sódico en la etapa de desorción tienen la máxima influencia en las cantidades de yodo que se adsorbe y que se desorbe, respectivamente. Además, se observa que las propiedades de adsorción y desorción del carbón regenerado son comparables a las que tiene en su estado inicial, si bien se aprecian algunas pérdidas mecánicas.

**Palabras clave:** Carbón activado, Adsorción, Desorción, Salmuera, Yodo.

## SUMMARY

A method for iodine extraction from dilute aqueous solutions, such as natural brines and bitterns, by a granulated activated carbon, and then desorption of iodine from activated carbon using sodium hydroxide solution is presented. It was found that adsorption of iodine from dilute solutions can be well correlated with Freundlich equation, and kinetics of adsorption follows approximately a first-order irreversible reaction. The results showed that pH of solution at the adsorption stage and concentration of sodium hydroxide solution in the desorption stage have the most influence on the amounts of iodine adsorption and desorption, respectively. Furthermore, it was found that adsorption and desorption properties of regenerated carbon are comparable with its initial state, although some mechanical losses were also observed.

**Keywords:** Activated carbon, Adsorption, Desorption, Brine, Iodine.

## RESUM

Es presenta un mètode per a l'extracció de iode de dissolucions aquoses diluïdes, tals com salmorres naturals i de salines, mitjançant carbó activat granulat, seguit de desorció de iode del carbó activat emprant una dissolució d'hidròxid sòdic. Es troba que l'adsorció de iode de dissolucions diluïdes es pot correlacionar bé amb l'equació de Freundlich, i que la cinètica d'adsorció segueix aproximadament una reacció irreversible de primer ordre. Els resultats mostren que el pH de la dissolució en l'etapa d'adsorció i la concentració de la dissolució d'hidròxid sòdic en l'etapa de desorció tenen la màxima influència en les quantitats de iode que s'adsorbeix i que es desorbeix, respectivament. A més, es troba que les propietats d'adsorció i desorció del carbò regenerat són comparables a les que té al seu estat inicial, tot i que s'observen algunes pèrdues mecàniques.

**Mots clau:** Carbó activat, Adsorció, Desorció, Salmorra, Iode.

## 1. INTRODUCTION

The natural brines and bitterns have historically been viewed as a waste product and proper disposal of them is a significant portion of the cost attendant to well operation. The process of disposal is expensive and from an environmental management viewpoint, recovery of this resource is a more desirable alternative. So the various oil field brines with various iodine contents, usually in the range of 10-150 mg/l, can be considered as sources for the commercial production of iodine. A well-known method of recovering iodine from natural brines consists of acidifying the brine, oxidizing to liberate iodine in the elemental state, adsorption of the liberated iodine by activated carbon, separating the activated carbon from the brine and finally desorption of iodine from activated carbon [1-10]. In recent years, many studies have examined the preparation of different kinds of activated carbons; also the characterization and different uses of the produced adsorbents have been extensively investigated [12-20]. In this paper, extraction of iodine from dilute aqueous solutions by a granulated activated carbon is investigated, and the iodine is then desorbed from activated carbon using sodium hydroxide solution. Using Taguchi method, design of adsorption and desorption experiments is performed and the optimum operating conditions to reach the highest percentages of adsorption and desorption is found.

## 2. EXPERIMENTAL

### 2.1. Materials and Methods

Iodine adsorption experiments were performed using a commercial activated carbon; its specifications are summarized in table 1. The selection of this granulated activated carbon was due to its acceptable surface area, high density, low attrition loss, high hardness, simplicity of filtration and separation processes and ease of regeneration process.

*Table 1. Specifications of the activated carbon*

| Parameter | AC |
|---|---|
| Iodine Number ($mgI_2/gC$) | 850 |
| BET surface area ($m^2/g$) | 900 |
| Particle diameter (mm) | 3-4 |
| Apparent density ($g/cm^3$) | 0.5 |
| Hardness (%) | 99 |
| Ash content (%) | 10 |
| Humidity (%) | 5 |

The iodine stock solution of 0.1N was prepared as follows: 12.700 g of iodine and 19.100 g of potassium iodide (KI) were weighed and mixed in a beaker. 2 to 5ml of water was added to the beaker and stirred well. By adding small increments of water continually (approximately 5ml each) and stirring, the total volume was reached 50 to 60ml. The solution was allowed to stand a minimum of 4h to ensure that all crystals are thoroughly dissolved. The mixture was transferred to a 1l volumetric flask and filled to the mark with distilled water [11]. Analysis of the iodine solution was performed using the UV spectrophotometer.

## 3. RESULTS AND DISCUSSION

### 3.1. Adsorption Experiments

#### 3.1.1. Optimization

In the present work, optimization of adsorption tests was carried out using Taguchi method by Qualitek-4 software. Taguchi method allows for the collection of the necessary data to determine which factors most affect process quality with a minimum amount of experimentation, thus saving time and resources. “Optimization” that implies “determination of best levels of control factors” is one of the important goals of this approach. This experimental design involves using orthogonal arrays to organize the parameters affecting the process and the levels at which they should be varied. The arrays are selected by the number of parameters (variables) and the number of levels (states) [21]. $L_9$ is one of the most common orthogonal arrays which was used for designing experiments with 4 control factors each of them varied at 3 levels (Table 2).

*Table 2. Arrangement of 4 factors varied in 3 levels in $L_9$ orthogonal array*

| Experiment | Factor 1 | Factor 2 | Factor 3 | Factor 4 |
|---|---|---|---|---|
| 1 | Level 1 | Level 1 | Level 1 | Level 1 |
| 2 | Level 1 | Level 2 | Level 2 | Level 2 |
| 3 | Level 1 | Level 3 | Level 3 | Level 3 |
| 4 | Level 2 | Level 1 | Level 2 | Level 3 |
| 5 | Level 2 | Level 2 | Level 3 | Level 1 |
| 6 | Level 2 | Level 3 | Level 1 | Level 2 |
| 7 | Level 3 | Level 1 | Level 3 | Level 2 |
| 8 | Level 3 | Level 2 | Level 1 | Level 3 |
| 9 | Level 3 | Level 3 | Level 2 | Level 1 |

Many parameters affect the amount and rate of iodine adsorption, such as type of activated carbon, pH of solution, temperature, agitation speed, contact time, initial concentration of iodine solution, the presence of complexing ions such as chlorides in solution etc [1]. In the present work, using a $L_9$ array, the influences of four factors including pH of the solution, agitation speed, particle size of carbon and carbon dosage are studied. In order to investigate the influences of acidic, basic and natural conditions on adsorption, three levels of pH were selected (2, 5, and 9). Because of difficult filtration of activated carbons with small particle size, this factor was studied at three levels (0.3-0.5, 0.5-1.2 and 1.2-1.7 mm). Agitation speed was also investigated at three levels (100, 200 and 300 rpm). It is obvious that increasing the carbon dosage, on one hand, increases the rate and amount of adsorption, but on the other hand increases filtration and desorption costs; so the optimum carbon dosage should be calculated. The selected levels for this factor were 1, 1.5 and 2g/l. According to the arrangement of factors in table 3, the adsorption experiments were carried out, and adsorption percentage obtained in each run was determined.

The optimal operating conditions for adsorption experiments determined by the software are as follows; pH of solution: 2, agitation speed: 200rpm, particle size: 0.5-1.2mm and carbon dosage: 2g/l. As it was expected, it is observed that at acidic conditions iodine adsorption is more than other pHs. It is established that, among the various species such as $I^-$, $I_3^-$ and $I_2$ that are present in the iodine solution, only $I_2$ can be adsorbed on the activated carbon. According to equation (1), the acidification of iodine solution can cause the progress of reaction in the forward direction, which leads to the formation of more iodine molecules, and so increase of iodine adsorption [1].

*Table 3. Arrangement of factors and their levels in $L_9$ array and percentages of adsorption obtained for each run*

| Run | pH of the solution | Agitation speed (rpm) | Carbon Particle size (mm) | Carbon dosage (g/l) | PA%* |
|---|---|---|---|---|---|
| 1 | 2 | 100 | 0.3-0.5 | 1 | 85.1 |
| 2 | 2 | 200 | 0.5-1.2 | 1.5 | 94.8 |
| 3 | 2 | 300 | 1.2-1.7 | 2 | 95.2 |
| 4 | 5 | 100 | 0.5-1.2 | 2 | 89.4 |
| 5 | 5 | 200 | 1.2-1.7 | 1 | 77.1 |
| 6 | 5 | 300 | 0.3-0.5 | 1.5 | 90.6 |
| 7 | 9 | 100 | 1.2-1.7 | 1.5 | 36.7 |
| 8 | 9 | 200 | 0.3-0.5 | 2 | 47.5 |
| 9 | 9 | 300 | 0.5-1.2 | 1 | 27.4 |

* The percentage of iodine adsorption

$$5I^- + IO_3^- + 6H^+ \Leftrightarrow 3I_2 + 3H_2O \quad \textbf{(1)}$$

The contribution percent of each factor affecting on adsorption also predicted by Taguchi software are as follows; pH of solution: 93.4%, agitation rate: 0.2%, carbon size: 0.7% and carbon dose: 5.7%. It can be concluded that pH of solution has the greatest effect on iodine adsorption percent and the effects of particle size and agitation speed are negligible. The agitation speed does not have any considerable affect on the equilibrium adsorption; it only reduces the equilibration time and increases the adsorption kinetic rates. Performing the experiment under the optimal conditions showed that 97.5% of iodine can be adsorbed on activated carbon that is almost equal to the amount predicted by the software.

**3.1.2. Equilibrium**

In this section, a series of 100ml of 600 mg/l iodine solutions, were treated with various carbon doses of 0.1, 0.125, 0.15, 0.2, 0.25, 0.5, 1 and 2.5g. The samples were agitated at 200rpm agitation speed at 30ºC. After 24h, the samples were filtered and the filtrates analyzed to determine the equilibrium concentration. The amount of iodine adsorbed by the activated carbon is determined as:

$$q = \frac{V(C_0 - C)}{M} \quad \textbf{(2)}$$

And the percentage of Iodine adsorption (PA %) is calculated as:

$$PA\% = \frac{C_0 - C}{C_0} \times 100 \quad \textbf{(3)}$$

Where q (mg adsorbate/g carbon) is the amount of adsorbate on the adsorbent, $C_0$ and C (mg/l) are the initial and equilibrium concentrations of adsorbate in the aqueous solution respectively, V (ml) is the volume of the aqueous solution, and M (g) is the mass of the adsorbent used in the adsorption [22]. From Fig. 1, it can be seen that iodine adsorption isotherm is regular, positive and concave to the concentration axis. Considering the slope of the initial portion of the curve, it can be concluded that sorption of iodine corresponds to L-type sorption isotherms in the Giles classification system. Initially the adsorption is quite rapid, which is followed by a slow approach to equilibrium at high concentrations [23].

The most widely used models for solid-liquid adsorption are Freundlich and Langmuir isotherms. Freundlich isotherm describes the heterogeneous surface energies by multilayer adsorption, and can be expressed as:

$$q = k_F C^{1/n} \quad \textbf{(4)}$$

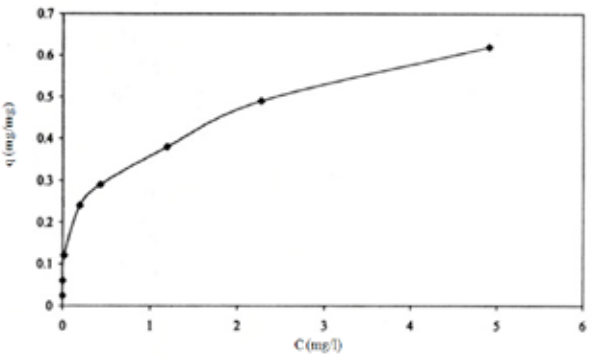


*Fig. 1 Adsorption isotherm of iodine*

Where q (mg adsorbate/g carbon) is the amount of adsorbate adsorbed at equilibrium and C(mg/l) is the equilibrium concentration of adsorbate in solution. In this model, the mechanism and the rate of adsorption are functions of the constants 1/n and $k_F$ (l/g) [22]. The Freundlich plot for the adsorption of iodine is shown in Fig. 2 that gives a good fit ($R^2$=0.9986).

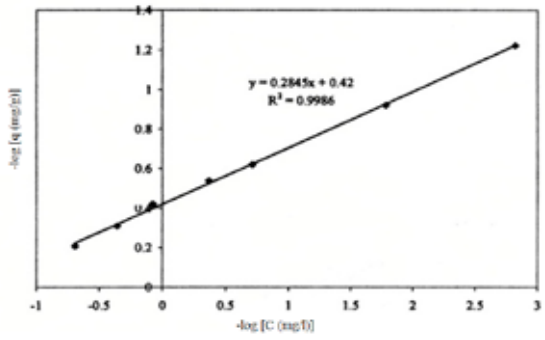


*Fig. 2 Freundlich plot*

The basic assumption of Langmuir equation is a homogeneous surface of the solid adsorbent. For dilute solutions, this model may be represented as:

$$q = \frac{q_m . k_L . C}{1 + k_L . C} \quad (5)$$

In the model, $q_m$ (mg/mg) is the amount of adsorption corresponding to complete monolayer coverage, i.e., the maximum adsorption capacity and $k_L$ (l/mg) is the Langmuir constant that is related to the intensity of adsorption

or adsorption energy [22]. A linear plot of (C/q) against C would give $q_m$ and $k_L$ from the slope and intercept (Fig. 3). As it can be seen, the experimental data gives almost a good fit in this case ($R^2$=0.9699).

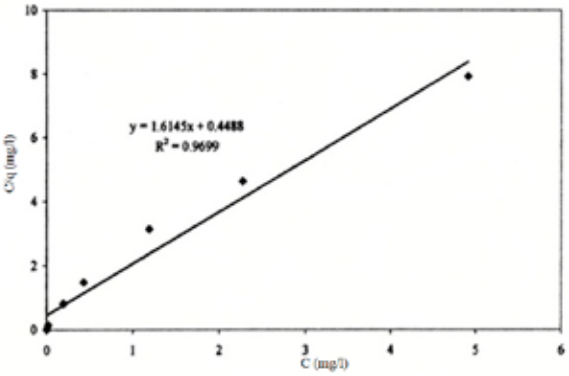


*Fig. 3 Langmuir plot*

### 3.1.3. Kinetics

In these experiments, a series of 100 ml of 200 mg/l iodine solutions, were treated with 0.2 g of carbon (mesh: 0.3-0.5) for 5, 10, 15, 30, 60, 120, 180 and 360min. The solutions with pH of 2 were agitated at 200rpm agitation speed at room temperature (30°C).

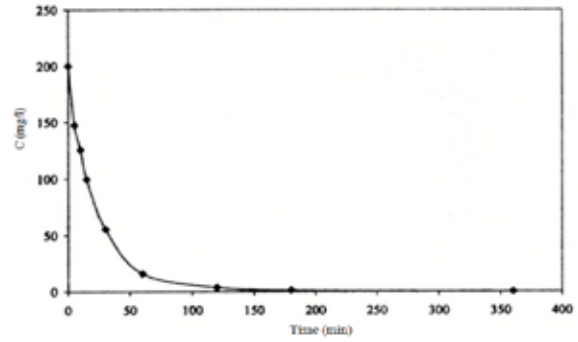


*Fig. 4 Iodine concentration as a function of contact time*

According to Fig. 4, the time to reach equilibrium in the adsorption stage is 200min, and after this time the concentration change is negligible. Investigation of three kinetic models (zero-order, first-order and second-order) for iodine adsorption, showed that the first-order equation is able to better describe this process than the zero- and second-order models. The first-order rate constant $k_1$(min$^{-1}$) can be obtained from the slope of the plot of -ln($C/C_0$) vs time, as shown in Fig. 5 ($R^2$=0.9914, $k_1$=0.027 min$^{-1}$).

$$-\ln\frac{C}{C_0} = k_1 t \quad \textbf{(7)}$$

Where $C_0$ and C (mg/l) are the concentration of iodine solution and at time t=0 and t=t.

### 3.2. Desorption Experiments

### 3.2.1. Optimization

Optimization of desorption tests was also carried out using Taguchi method; desorption factors and the selected levels are as follows: temperature of NaOH solution (30, 55 and 80°C), concentration of NaOH solution (0.1, 0.5 and

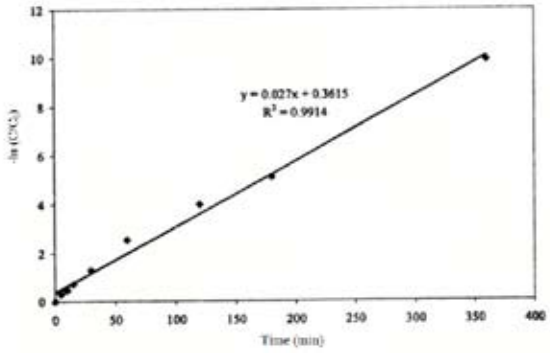


*Fig. 5 First-order kinetic plots*

1%) and agitation speed (100, 200 and 300 rpm). The maximum level of temperature was 80°C, because higher values of temperature degrade the granular form of activated carbon and breaks up the iodide. The suitable orthogonal array for these conditions is also $L_9$ array that is shown in table 4. Table 5 shows the arrangement of investigated factors in this work, and the percentage of desorption obtained for each run.

*Table 4. Arrangement of 3 factors varied in 3 levels in $L_9$ orthogonal array*

| Experiment | Factor 1 | Factor 2 | Factor 3 |
|---|---|---|---|
| 1 | Level 1 | Level 1 | Level 1 |
| 2 | Level 1 | Level 2 | Level 2 |
| 3 | Level 1 | Level 3 | Level 3 |
| 4 | Level 2 | Level 1 | Level 2 |
| 5 | Level 2 | Level 2 | Level 3 |
| 6 | Level 2 | Level 3 | Level 1 |
| 7 | Level 3 | Level 1 | Level 3 |
| 8 | Level 3 | Level 2 | Level 1 |
| 9 | Level 3 | Level 3 | Level 2 |

The optimal desorption conditions obtained by the software are as follows: temperature of NaOH solution: 50°C, concentration of NaOH solution: 0.5 % and agitation speed: 300rpm. Desorption experiments under these optimal conditions showed 98.9% desorption. The Taguchi design software results also showed that in the desorption section, the contributions of each factor on desorption percentage are as follows: concentration of NaOH solution (55%), temperature of NaOH solution (39.4%) and agitation speed (4.6%). It can be concluded that for both adsorption and desorption stages, agitation speed has a negligible influence on the response.

### *3.2.2. Effect of contact time*

The experimental conditions for iodine desorption from spent carbons were as follows: 2 g of activated carbon loaded with the maximum amount of iodine were placed into desorption medium containing 100 ml NaOH (0.5 %). The amounts of iodine desorbed in 15, 45, 90, 180, 360 min and 24 h were measured. Agitation of the samples was followed at 200 rpm and constant temperature of 30°C. The percentage of desorption (PD %) is determined as:

$$PD\% = \frac{V'.C'}{V.(C_o - C)} \times 100 \quad \textbf{(9)}$$

Where $V$ (ml) and $C$ (mg/l) are the volume and concentration of iodine solution after desorption, respectively; V(ml), $C_o$(mg/l) and C(mg/l) are the volume, initial concentration and final concentration of the iodine solution which has

*Table 5. Arrangement of factors and their levels in L9 array and the percentages of desorption for each run*

| Run | Temperature of NaOH solution (°C) | Concentration of NaOH solution (%) | Agitation speed (rpm) | PD%* |
|---|---|---|---|---|
| 1 | 30 | 0.1 | 100 | 79.1 |
| 2 | 30 | 0.5 | 200 | 90.6 |
| 3 | 30 | 1 | 300 | 85.9 |
| 4 | 55 | 0.1 | 200 | 88.4 |
| 5 | 55 | 0.5 | 300 | 98.4 |
| 6 | 55 | 1 | 100 | 90.9 |
| 7 | 80 | 0.1 | 300 | 88.6 |
| 8 | 80 | 0.5 | 100 | 94.8 |
| 9 | 80 | 1 | 200 | 90.5 |

* The percentage of iodine desorption

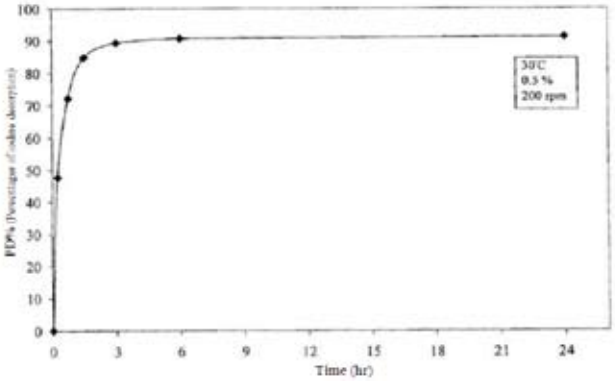


*Fig. 6 Iodine desorption as a function of contact time*

been used for adsorption [1]. According to Fig. 6, the necessary time for equilibrium desorption is obtained as 6h.

### 3.3. Recovery Experiments

Finally the possibilities of adsorbent recovery instead of using a new activated carbon (reusing of activated carbon after iodine desorption in a new adsorption-desorption cycle) was investigated. The experiment was carried out on a used activated carbon sample which was already applied in the iodine adsorption and desorption processes, both under the optimal operating conditions (Table 6).

This carbon was dried for 24h at 110°C, and then the adsorption, desorption and regeneration cycles were replicated on it, similar to the first cycle. It is obviously observed that the amount of regenerated carbon is always less than the amount of initial carbon. Most of these losses appear in the agitation and filtration stages, and the mechanical loss in each cycle sometimes reaches to 10%. It is found that, the amount of iodine desorption from regenerated carbon decreases a little, with increasing the number of cycles; in contrast the amount of iodine adsorption increases somewhat. This negligible increase might be due to the modification of activated carbon surface by halogenation with the residual iodine on the activated carbon surface. It is established that one of the ways for modification of activated carbons structure and increasing their adsorption capacity is treatment with halogens [24]. According to Lee et al., (2004) halogenation of activated carbons especially surface treatment with iodine can be carried out at low temperatures even at room temperature [25].

4. Conclusion

The present paper showed that the granular activated carbon can be used for adsorption and recovering iodine from dilute aqueous solutions. Since activated carbons do not have affinity to adsorb sodium and chloride, it can be concluded the mentioned method can be used for iodine extraction from natural brines and the like, having iodine concentration more than 50 mg/l.

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*Table 6. The optimal operating conditions for adsorption and desorption stages*

| Adsorption stage | | Desorption stage | |
|---|---|---|---|
| **Factor** | **Optimum level** | **Factor** | **Optimum level** |
| pH of iodine solution | 2 | NaOH solution temperature (°C) | 55 |
| Agitation speed ( rpm) | 200 | NaOH solution concentration (%) | 0.5 |
| Particle size of carbon (mm) | 0.5-1.2 | Agitation speed ( rpm) | 300 |
| Carbon dosage (g/l) | 2 | Time (h) | 6 |
| Time (h) | 3 | (NaOH solution volume/ iodine solution volume) | 0.1 |
| Initial Concentration of iodine solution (mg/l) | 30 | (Washing water volume/ iodine solution volume) | 0.1 |
| PA% | 97.5 | Number of washing stages | 2 |
| | | PD% | 98.9 |

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