

## Stability of Iodine in Iodized Salt

L.L.Diosady<sup>1</sup> and M.G. Venkatesh Mannar<sup>2</sup>,

<sup>1</sup> Professor of Food Engineering, Department of Chemical Engineering and Applied Chemistry, University of Toronto, Toronto, ON, Canada, M5S 3E5

<sup>2</sup> Executive Director, The Micronutrient Initiative, International Development Research Centre, 250 Albert St. P.O. Box 8500, Ottawa ON Canada, K1G 3H9

### ABSTRACT

Since iodine readily sublimates at ambient temperature, the effectiveness of salt iodization programs depends on the stability of the iodine carrier, typically potassium iodate. We examined the stability of iodine in typical salts available in 12 countries in a controlled laboratory setting, at high temperature (40°C) and controlled humidity (60 or 100%) for periods up to 12 months. Iodine was rapidly lost in most unrefined salt samples. The presence of moisture, due to hygroscopic impurities, and metal ion impurities, such as iron accelerated the loss of iodine. Packaging in a moisture-proof continuous film package significantly increased the iodine retention in all samples.

### 1. INTRODUCTION

The inadequate iodine content of the food supply is supplemented by the addition of iodine to salt in more than 100 countries world-wide as a result of the Universal Salt Iodization (USI) initiative. The virtual elimination of IDD in the world by the year 2000 has been set as a goal at several international fora including the World Health Assembly (1986, 1990), The World Summit for Children (1990) and the International Conference on Nutrition (1992).

Iodine is added to salt in the form of potassium iodide or iodate either as a dry solid or aqueous solution at the point of production. Iodate is typically used in tropical climates, due to its better resistance to oxidation. The actual availability of iodine from iodized salt at the consumer level can vary widely due to a number of factors, including the variability in the amount of iodine added during production, its uneven distribution within the batches or bags produced due to poor mixing, and due to losses in distribution, retail and in the household, during storage and meal preparation.

While there is a wide band of iodine intake levels that are safe to consume, it is important that adequate amounts are delivered to the consumer, while minimizing the costs of iodization. Therefore it is critical to have an accurate estimate of the losses in

iodine prior to consumption, so that the producers may compensate for the losses by the addition of excess iodine during production. We undertook a study to determine trends in iodine losses from typical salt samples from twelve countries under well controlled laboratory conditions, representing typical and extreme environmental conditions.

#### 1.2. Iodization Level

During the past few years the minimum goal of iodine fortification has been set at 150µg/day per person. Many countries aim at much higher levels. The actual iodate addition levels to salt is based on the average per capita salt consumption and anticipated losses of iodine during distribution. Specifically, Mannar and Dunn (1995), based on their earlier experience, recommended that the addition level be based on the assumption of 50% iodine loss between iodization and consumption. ICCIDD/WHO/ UNICEF (WHO 1990) have described desirable average levels of iodine in salt at various points in the salt distribution chain, taking into account level of salt intake, climatic conditions and packaging. In a 1996 booklet (WHO/NUT/96.13) the loss of iodine from production to household has been estimated to be

20%. The present study adds experimental data that should be useful for updating these recommendations.

### 1.3. Iodine Stability

Elemental iodine readily sublimates and is then rapidly lost to the atmosphere through diffusion. Potassium iodide is less stable than potassium iodate, as it can be oxidized to elemental iodine by oxygen or other oxidizing agents, especially in the presence of impurities, such as metal ions and moisture, which catalyze the reaction. Potassium iodate may be reduced to the elemental iodine by a variety of reducing agents in the salt, such as ferrous ions. Moisture is naturally present in the salt, or is abstracted from the air by hygroscopic impurities such as magnesium chloride. The pH of the condensed moisture on the salt is very much influenced by the type and quantity of impurities present, and this affects the stability of the iodine compounds. Elevated temperatures increase the rates of iodine loss.

The levels and types of impurities, moisture content and pH of salt produced for human consumption vary widely. Salt is produced from sea water, rock salt deposits and lake sediments, by solar evaporation of brines and dry or solution mining of rock salt deposits. Refining processes range from simple washing methods to large-scale mechanized vacuum evaporation systems which require trained operators and rigorous quality assurance.

Based on the chemical properties of salt aimed at human consumption, losses of iodine were not unexpected, and there have been numerous published and unpublished studies on iodine stability in salt during the past 75 years. A review of this literature showed that iodate is superior to iodide in terms of stability as a fortificant in salt. Published evidence of the stability of iodine, added in the form of iodate without stabilizers, is relatively meagre, but indicates iodine losses ranging from around 5% to 66% after 12 months.

The studies undertaken between 1923 and 1996, are difficult to compare. Variation between rates of iodine loss reflects impurities, moisture content, and processing methods. Conditions of packaging and storage, such as humidity and temperature also affect the final iodine content of the salt, yet these factors were not always clearly defined in earlier studies. Sample sizes

and reproducibility of results were not always reported, making it more difficult to assess the statistical significance of results.

A comprehensive review of the literature by Kelly (1953) concluded that the stability of iodine in salt is determined by (i) the moisture content of the salt and the humidity of the atmosphere (ii) light, (iii) heat (iv) impurities in the salt (v) alkalinity or acidity (vi) the form in which the iodine is present. He concluded that the iodine content will remain relatively constant if the salt is packed dry with an impervious lining, and kept dry, cool, and away from light. He recommended that iodate be used under adverse conditions such as found in developing countries where the salt being iodized is crude, unprocessed and usually not dried sufficiently.

Potassium iodate was stabilized by calcium carbonate in crude sea salt stored in hemp fibre sacks for up to eight months at ambient temperature and relative humidity between 70% and 84%. Only some 3.5% of the added iodine was lost. (Arroyave, et al. 1956)

Chauhan et al., (1992) compared iodine stability over 300 days in common salt iodized with iodate, packed in 5 kg solid high density polyethylene (HDPE) bags or left in open heaps. The relative humidity and temperature varied from 41 - 83% and 30-39 °C respectively. Both the salt packed in HDPE bags and in the open lost 9-10% of the added iodine within the first month, after which values remained practically constant.

While the actual magnitude of iodine losses were not well defined, a number of researchers investigated the effects of stabilizing compounds. Considerable variation in iodine stability were found in control samples without stabilizers.

Ranganathan, et al. (1986) found that coarse salt iodized with iodate at "normal" room temperature and humidity showed iodine losses of 20% after 12 months, while samples stabilized with calcium carbonate lost no iodine over an 18- month period.

In a later study analyses of five types of Indian salt (including powder and crystal) iodized with iodate showed losses of 28-51% after 3 months,

35-52% after 6 months, and up to 66% after 12 months. (Ranganathan, 1999).

Iodine losses from refined solar salt packed in open 1 kg plastic film bags, heated to 130°C for 2.5 hr. to simulate drying, and stored at ambient temperature were reported to be 5.7% after 12 months and 11% after 3 years (Zigong Institute, 1992).

#### **1.4. Packaging materials**

In most developing countries salt is sold packaged or in bulk. Packaging materials include paper, high and low density polyethylene, and woven bags made of jute, straw or high density polyethylene. The woven bags provide the most mechanical protection; however, they allow free flow of air and water to readily penetrate to the salt.

## **2. OBJECTIVES**

The purpose of the present study was to assess the effect of humidity and packaging materials on iodine stability in typical salt samples from countries with tropical and subtropical climates, under controlled conditions, in an effort to determine the range and timing of iodine losses that may be expected using iodine in the form of iodate under typical conditions, and to define cost-effective means of controlling or compensating for these losses.

## **3. EXPERIMENTAL METHODS**

### **3.1 Materials**

Potassium iodate, analytical reagent grade, was obtained from BDH, Toronto.

Samples of un-iodized salt were obtained through UNICEF country offices from routine production runs from local salt producers, from twelve countries, Bangladesh, Bolivia, China (2 levels of addition), Ghana, Guatemala, India (5 sources), Indonesia (4 sources), Pakistan, Philippines (3 sources), Senegal, Thailand and Tanzania . The samples were air shipped to Toronto.

A sample of un-iodized refined salt similar to typical Canadian household salt was obtained from Toronto Salt Chemical Co., Ltd. and used as a reference sample.

### **3.2 Sample treatment**

Salt samples with particle size less than 2 mm were used without pre-grinding. Larger crystals were ground by mortar and pestle, and sieved to pass through a 10 mesh sieve. (Diosady et al. 1998)

Two kg samples of salt from each source were fortified to contain ~50 mg/kg iodine using potassium iodate added as a 30g/L solution. The mixtures were blended to ensure uniformity using a 5L ribbon blender (LeRoy Somer - LSTronics).

### **3.3 Packaging materials**

Three packaging methods were tested. From each 2kg batch of iodated salt three 500 g samples were prepared, one each in woven high density polyethylene (HDPE) bags of 0.15 mm thickness, low-density polyethylene (LDPE) film bags of 0.07 mm thickness, and in open plastic containers. HDPE has a much higher tensile strength as it is made of longer molecular chains. HDPE bags were made by cutting sheets into thin strips and weaving these strips (1.5-2.5 mm wide) of HDPE film into a "cloth", which was then sewed to form bags. Eventhough the HDPE does not absorb water, the woven bags readily allow the passage of water through the weave.

### **3.4 Storage conditions**

The packages were stored under two conditions: elevated temperature and high humidity , (~ 40°C, 100% RH); and elevated temperature and medium humidity (~ 40°C, ~ 60 % RH).

The high temperature and high humidity was maintained by using a controlled temperature oven, in which the air was saturated with water vapour by exposure to a tray of water. The high temperature, medium humidity conditions were maintained in an environmental chamber manufactured by Associated Environmental Systems Division of Craig Systems Corporation.

### **3.5 Analytical Methods**

#### **3.5.1 Sampling**

Salt samples were analyzed at the start of the experimental series, and after 1, 2, 3, 6 and 12 months of storage. To obtain a representative samples for analysis, the bags of 500g salt were

repeatedly split by a two-stemmed powder funnel until approximately 15 grams of salt were collected. This sub-sample was used for the analyses. The technique was reported earlier, (Diosady et al. 1997). The remaining salt was replaced in its package, and returned to the controlled atmosphere. The bags were not heat sealed, as this would not be typical of local practice.

### 3.5.2. Moisture

The moisture content was determined gravimetrically. Samples of salt were weighed, then dried at 110°C for 16 hours and reweighed.

### 3.5.3 Iodine

Iodine content was measured by neutron activation analysis or titration, as described by Diosady et al. (1998)

#### Neutron Activation Analysis

Approximately 1.25 g of salt was accurately weighed and irradiated at 1 kW power using a neutron flux of  $5.0 \times 10^{11} \text{ cm}^{-2} \text{ sec}^{-1}$  for 3 minutes using the University of Toronto's SLOWPOKE nuclear reactor.

After 6 minutes delay the gamma emission at 443 keV was measured using a hyper pure germanium based gamma ray spectrometer. In the range of 5 to 250 mg iodine per kg salt the relative standard deviation of the analysis was 2%.

#### Titration

Ten grams of salt was dissolved in approximately 100 mL water. The pH was adjusted to 2.8 using 0.6% HCl. Next 30 mg KI powder was added, to convert all of the iodate present to elemental iodine.

The liberated iodine was titrated with 0.004N freshly prepared sodium thiosulphate solution. Starch was used as the end point indicator.

The iodine value obtained by analysis immediately after the time of preparation was used as the starting or Time=0 concentration for all subsequent analyses of the same batch.

The results of the titration and neutron activation analysis were within  $\pm 5\%$  on actual salt samples. On carefully prepared iodine standards with minimum internal variability, the standard deviation between samples was between 1 and 2 %, indicating that the two techniques were interchangeable in the absence of interfering impurities.

## 4. RESULTS AND DISCUSSION

Iodine losses over the twelve month sampling period ranged from less than 10% to 100%. The rate of iodine loss was influenced by the salt's origin, the packaging material and the relative humidity during storage.

### 4.1. Relative Humidity

In all cases the samples stored at 100% RH lost iodine at a higher rate than those stored at 60%RH. Losses after six months storage at 60% RH were lower than expected based on the ICCIDD/WHO/UNICEF tables, ranging between near zero and 20%.

At high humidity the losses were more dramatic: in some samples effectively all of the iodine added to the sample disappeared within six months. At high humidity, the samples stored in containers which allowed contact with the air, lost two thirds or more of the added iodine within the year.

The results confirm the large effect of ambient humidity on iodine stability. While even under moist, tropical conditions it is unlikely that ambient relative humidity will remain at or near 100% for six months, once moisture is absorbed into the bag's contents, the high humidity will be retained. This is compounded by the fact that within bags exposed to sunlight, or in storage facilities heated by the sun, salt temperatures may readily rise to over 60°C. The typical effect of relative humidity is illustrated in Figure 1.

### 4.2. Packaging Material

Packaging affected the levels of moisture contained in the system. The LDPE film provided an excellent moisture barrier, and thus maintained the total water content in each bag approximately constant. Some moisture absorption was possible as the bags were not sealed sufficiently to prevent diffusion of air containing water and/or iodine in and out of the bags.

In open containers the absorbed and condensed moisture was retained and contributed to the instability of iodine. Woven HDPE bags, which also allowed the flow of air and moisture, behaved

**Effect of Relative Humidity**

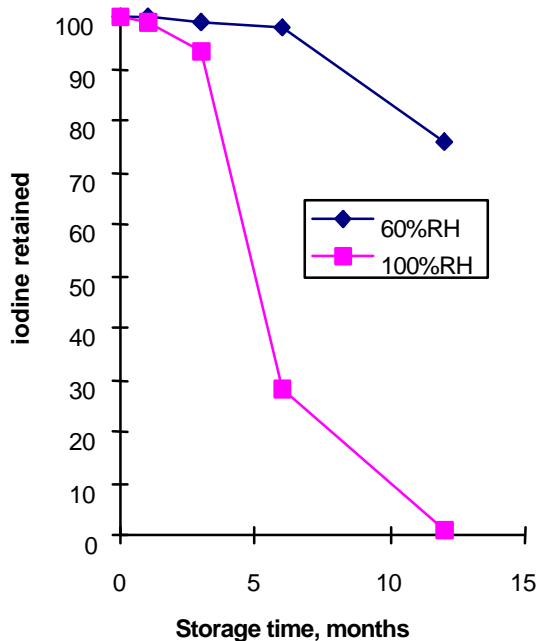


Figure 1. - Effect of relative humidity on iodine retention - Refined salt stored in HDPE.

similarly to the open containers at 60% RH. At high humidity the HDPE bags lost more iodine, as the condensed water leaked out of the bag, removing dissolved iodine.

**4.3. Country of origin**

The impurities, physical characteristics and the extent of processing at the source had a major effect on the stability of the salt. The salt samples received varied in colour from very bright white to dark gray or rusty brown. The samples had particle sizes ranging from ~100 µm to 15 mm, with great variability in the homogeneity of particle size.

Canadian salt, which was highly refined and stabilized lost less than 10% iodine during the first six month storage period, and less than 25% in a year at 60%RH. With 100% humidity the protection of the

**Effect of Packaging**

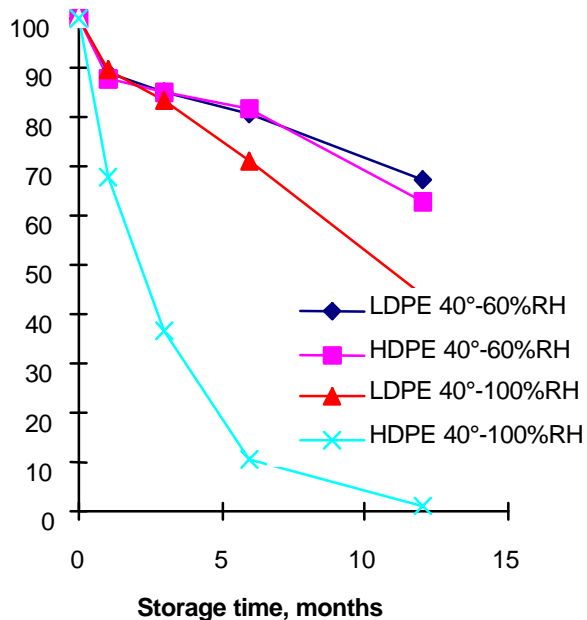


Figure 2. Effect of packaging on iodine retention.

polyethylene bag maintained iodine losses at less than 8%, while in the woven HDPE and open container, 63 and 95% iodine were lost.

Salts from Canada, India, the Philippines and Senegal lost less than 15% of the added iodine even at 100% relative humidity when stored in low density polyethylene bags. Salts from Tanzania, Indonesia and Ghana retained more than 70%, and Bolivia more than two thirds of the added iodine for the first six months, yet the Tanzanian salt dropped to only 22% iodine retention after a year under the same conditions.

At low humidity the salt from Ghana lost very little iodine. At high humidity the iodine content of Ghanaian salt remained high for the first three months, but dropped sharply during the next three months, ending up with 83% loss after a year.

## 5. CONCLUSIONS

We confirmed that moisture plays a critical role in the stability of iodine. In particular, when salt is stored at a temperature characteristic of storage and distribution conditions in many developing countries, moisture absorbed by hygroscopic impurities is the major contributor to the rapid loss of iodine.

Salt purification is the best technical means of preventing iodine loss; however, this would be prohibitively expensive in the short term for many developing countries.

By packaging salt in an effective moisture barrier such as solid low-density polyethylene bags, iodine losses can be significantly reduced. With solid LDPE packaging, losses of iodine from salt stored for up to six months can be kept in the range of 10-15%. Unfortunately woven bags are necessary for the bulk packing of salt to ensure adequate mechanical strength. Fitting woven HDPE bags with an impervious liner of HDPE or LDPE would appear to be an effective, low-cost method of improving iodine stability in iodized salt. As losses of iodine in all samples was significant after 12 months, efforts must be made to minimize the time required for distribution, retail and consumption to ensure efficient, effective use of the added iodine.

The results indicate that the control of moisture content in iodized salt throughout the manufacturing and distribution, by improved processing, better packaging and storage, is critical to the stability of the added iodine. In order to make allowances for the probable losses of iodine, countries must determine iodine losses from local iodized salt under local conditions, as these will be greatly affected by salt source, quality and processing technology.

## REFERENCES

- Anonymous - Official Methods of Analysis, 14<sup>th</sup> Edition Association of Official Analytical Chemists 33.147 Iodine in iodized salt, (1984)
- Anonymous "Recommended iodine levels in salt and guidelines for monitoring their adequacy and effectiveness" WHO /UNICEF/ ICCIDD booklet WHO/NUT/96.13, 1996
- Arroyave, G., Pineda, O., Scrimshaw, NS, "The stability of potassium iodate in crude table salt", Bull Wld Hlth Org 14, 183-85, 1956)

- Chauhan SA, Bhatt AM, Bhatt MP, Majeethia, KM, "Stability of iodized salt with respect to iodine content" R& Industry, 37, 38-41, 1992
- Diosady, L.L., Alberti, J.O., Venkatesh Mannar, M.G. and Stone, T.G. "Stability of Iodine in iodized salt used for the correction of iodine deficiency" .Food and Nutrition Bulletin 18:388-96. 1997
- Diosady, L.L., Alberti, J.O., Venkatesh Mannar, M.G. and FitzGerald, S. "Stability of Iodine in iodized salt used for the correction of iodine deficiency II" *ibid* 19:240-50. 1998
- Kelly F.C. - Studies on the stability of iodine compounds in iodized salt. Bull. World Health Org. **9**:217-230, 1953
- Kruger. P. - Principles of Activation Analysis - 1971, Wiley Interscience New York,
- Mannar, MG, JT Dunn, Salt iodization for the elimination of iodine deficiency ICCIDD, 1995
- Ranganathan S, Reddy V., Ramamoorthy, P. "Large scale production of salt fortified with iodine and iron" Food and Nutrition Bulletin 1996 17. 73-78
- Ranganathan S, Narasinga Rao BS "Stability of iodine in iodized salt", Indian Food Industry, 5 July/Sept 1986, 122-124
- Resolutions of the World Health Assembly WHA39.31 (1986) and WHA43.2 (1990)
- UNICEF - Small salt producers and universal salt iodization: Working paper, Nutrition section, UNICEF, 16 November, 1994
- WHO,- Iodine and Health: eliminating iodine deficiency disorders safely through salt iodization:, Geneva, August, 1994
- Zigong, 1992 "Experiment report on long-lasting iodized salt" Zigong Design and Research Institute for well and Rock Salt Industry, December 1992