EGE UNIVERSITY
GRADUATE SCHOOL OF NATURAL AND APPLIED SCIENCES

(MASTER OF SCIENCE DEGREE THESIS)

INVESTIGATION OF ENRICHMENT POSSIBILITY OF NATURAL OXYGEN-18 ISOTOPE RATIO IN WATER USING MICROWAVE IRRADIATION TECHNIQUE AND INVESTIGATION OF ITS COMMERCIAL APPLICATION POTENTIAL

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Discipline Code: 405.04.01 (Nuclear Chemistry)
Date of Presentation: 07.02.2012

Bornova-IZMIR

2012

Selen EKİM tarafından Yüksek Lisans Tezi olarak sunulan "Investigation of Enrichment Possibility of Natural Oxygen-18 Isotope Ratio in Water Using Microwave Irradiation Technique and Investigation of its Commercial Application Potential" başlıklı bu çalışma, E. Ü. Lisansüstü Eğitim ve Öğretim Yönetmeliği ile E. Ü. Fen Bilimleri Enstitüsü Eğitim ve Öğretim Yönergesi'nin ilgili hükümleri uyarınca, tarafımızdan değerlendirilerek savunmaya değer bulunmuş ve 07 / 02 / 2012 tarihinde yapılan tez savunma sınavında aday oybirliği ile BAŞARILI bulunmuştur.

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ÖZET

MİKRODALGA IŞINLAMALARI İLE SUDAKİ DOĞAL OKSİJEN-18 İZOTOPUNUN ZENGİNLEŞTİRİLME İMKANININ İNCELENMESİ VE BUNUN TİCARİ AÇIDAN KULLANILABİLİRLİĞİNİN ARAŞTIRILMASI

Selen EKİM

Yüksek Lisans Tezi, Kimya Bölümü
Tez Danışmanı: Prof. Dr. Turan ÜNAK
Şubat 2012, 84 sayfa

Doğal Oksijen-18 (^{18}O) izotopu açısından zenginleştirilmiş su, Medikal Siklotron'larda radyoaktif Fluor-18 (^{18}F) üretimi için hedef madde olarak kullanılmaktadır. ^{18}F Nükleer Tıp'ta sıklıkla kullanılan, etkin bir görüntüleme ajanı olan ^{18}F ile işaretli 2-Fluoro-2-deoksi-D-Glukoz'un (^{18}FDG) sentezlenmesinde kullanılan son derece önemli bir radyoizotoptur. Medikal siklotronlarda ^{18}F üretmek üzere, ^{18}O (p, n) ^{18}F nükleer reaksiyonunun gerçekleştirilmesi için hedef madde olarak kullanılan "^{18}O'li Ağır Su"yun zenginlik oranının % 97'ler düzeyinde olması gerekmektedir. Bu durum, "^{18}O'li Ağır Su" üretiminin önemini ortaya koymaktadır. ^{18}FDG, başta kanser olmak üzere, birçok hastalığın erken dönemde teşhis edilmesini sağlayan, önemli bir görüntüleme ajanıdır. Fakat, üretimi sırasında gerek Medikal Siklotron'larda yürütülen işlemler, gerekse hedef madde olarak kullanılan "^{18}O'li Ağır Su"yun üretimi için, endüstride en uygun teknik olarak uygulanan Fraksiyonlu Destilasyon tekniği ile, suyun ^{18}O açısından zenginleştirilmesi, oldukça yüksek maliyeti olan bir uygulamadır. Bu nedenle de, sağlık sektöründe ^{18}FDG oldukça pahalı bir görüntüleme ajanıdır. Eğer, "^{18}O'li Ağır Su"yun maliyetinin düşürülebilmesi halinde, ^{18}FDG'nin maliyeti de büyük ölçüde düşecektir. Bunun için de, ^{18}O açısından zenginleştirilmiş su üretimi için daha etkin ve daha ucuz bir izotop zenginleştirme tekniğinin geliştirilmesine imkan sağlayacak çalışmalar büyük önem taşımaktadır. İşte bu çalışmada, mikrodalga ışınlamaları kullanılmak suretiyle, doğal suyun ışınlanarak ^{18}O izotopu açısından bir zenginleşme sağlanıp sağlanamayacağının ortaya çıkarılması ve olumlu sonuçlar alınması halinde ise, bunun ekonomik açıdan bir değerlendirmesinin yapılması amaçlanmış bulunmaktadır.

Çalışmada, 20,000 mL saf su aşama aşama 5 mL kalana dek mikrodalga ışınlaması altında buharlaşmaya tabi tutulmuştur. Her aşamada elde edilen su örneklerinin ^{18}O ölçümleri, Isotope Ratio – Mass Spectrometry (IR-MS) kullanılarak yapılmıştır. Bu ölçümler, kararlı izotop ölçümleri alanında Dünya genelinde tanınan ve bir İngiliz firması olan "Iso-Analytical Ltd." tarafından gerçekleştirilmiştir. Önemli bir sonuç olarak belitmek gerekir ki doğal su olarak kullanılan saf suyun başlangıçta % 0.19874 doğal ^{18}O zenginlik oranının, ışınlamaların son aşamasında elde edilen su örneklerinde % 0.21959'e kadar yükseldiği gözlenmiştir. Bu da, başlangıçtaki zenginlik oranının % 10'lar mertebesine ulaşmış olması anlamını taşımaktadır.

Aynı buharlaştırma işlemleri paralel olarak hot-plate üzerinde ısıtma yapılmak suretiyle de tekrarlanmıştır. Bu işlemler sonucunda elde edilen zenginleşme oranı ise % 9'lar mertebesine bulunmuştur ki bu da, açık şekilde mikrodalga ışınlamalarının sudaki ^{18}O zenginleşmesini arttırdığını ortaya koymaktadır.

Bunun yanında, zenginleşme faktörleri ise, mikrodalga ışınlamaları ve termal ısıtma için sırasıyla 1.1049 ve 1.0976 olarak hesaplanmıştır. Bu sonuçlar da, mikrodalga ışınlamalarının sudaki ^{18}O izotopunun zenginleştirilmesi üzerine olumlu etkisini daha belirgin bir şekilde ortaya koymaktadır.

Bu çalışmadan çıkarılabilecek genel değerlendirme ise, mikrodalga ışınlamalarının en azından, sudaki ^{18}O izotopunun zenginleştirilmesinde alternatif bir zenginleştirme yöntemi olarak kullanılabileceğinin kanıtlanmış olmasıdır. Buradan hareketle, diğer kararlı izotopların zenginleştirilmesi üzerinde de, benzer çalışmaların yapılması önem kazanmış bulunmaktadır.

Anahtar sözcükler: İzotop, Zenginleştirme, Mikrodalga, Işınlama, Kararlı izotop, Oksijen-18, ^{18}O, Doğal abondans, İzotop oranları, Kütle spektrometresi, IR-MS, 2-Fluoro-2-deoksi-D-Glukoz, ^{18}FDG, ^{18}F, ^{18}O'li Ağır Su, Medikal Siklotron.

ABSTRACT

INVESTIGATION OF ENRICHMENT POSSIBILITY OF NATURAL OXYGEN-18 ISOTOPE RATIO IN WATER USING MICROWAVE IRRADIATION TECHNIQUE AND INVESTIGATION OF ITS COMMERCIAL APPLICATION POTENTIAL

Selen EKİM

MSc in Chemistry
Supervisor: Turan ÜNAK, Professor of Nuclear Chemistry
February 2012, 84 pages

Heavy water containing the Oxygen-18 isotope has been used as a target material in medical cyclotrons for production of radioactive Fluorine-18 isotope, which is used for labelling of glucose. ^{18}F-labelled 2-Fluoro-2-Dexy-D-Glucose (^{18}FDG) is an effective imaging agent in routine nuclear medicine applications and is generally used for early detection of cancer in the human body, especially in the brain. For this reason, ^{18}O heavy water is a very important precursor for production of ^{18}F and consequently for the synthesis of ^{18}FDG. ^{18}O heavy water should have at least 97 % enrichment for ^{18}O isotopic ratio for these applications. This results in a high cost for ^{18}O heavy water and this also affects the cost of ^{18}FDG. In the case of development of more practical and cheaper techniques for enrichment of ^{18}O in water, the cost of ^{18}FDG applications in nuclear medicine will surely decrease. In this study, the enrichment possibility of ^{18}O isotopic ratio in natural water was investigated using microwave irradiation technique of water and the results obtained were evaluated for economical application potential of this technique in industry.

About 20,000 mL of natural pure water was evaporated up to about 5 mL by microwave irradiation step by step. ^{18}O isotopic ratios of these water samples were measured by the use of isotope ratio mass spectrometry (IR-MS) technique. These measurements were realised by a competent company in the UK that is well known for stable isotope measurements. For this, the samples were sent by an international parcel service to the UK in the same time intervals. As an important result, it was observed that while the natural ^{18}O isotopic ratios of natural pure water was measured to be 0.19874 %, that of water samples obtained at the last evaporation

steps were measured to be 0.21959 %. This means that the natural ^{18}O isotopic ratio of pure water was enriched to be about 10 % as a result of microwave irradiation of water.

The same evaporation operations were also repeated using conventional heating on a hot plate. In this case, the enrichment level was found to be about 9 %. This has clearly demonstrated the microwave irradiation effect on the ^{18}O enrichment in water.

On the other hand, the enrichment factors, which mean the rates of final and initial ^{18}O enrichment ratios as atom percent, corresponding to microwave irradiation and conventional heating were found to be 1.1049 and 1.0976, respectively. This also clearly indicates the positive microwave effect on the ^{18}O enrichment ratio of water.

The final conclusion of this study may be summarised as follows: the results obtained in this study have verified that the microwave irradiation process may be used as an alternative potential process, at least for ^{18}O enrichment of water. Starting from this study, the enrichment of other stable isotopes in different natural samples may also be studied using microwave irradiation technique.

Key words: Isotope, Enrichment, Microwave, Irradiation, Stable isotopes, Oxygen-18, ^{18}O, Natural abundance, Isotopic ratio, Mass spectrometry, IR-MS, 2-Fluoro-2-deoxyi-D-glucose, ^{18}FDG, ^{18}F, ^{18}O Heavy water, Medical cyclotron.

ACKNOWLEDGEMENT

I would like to express my sincere gratitude to my supervisor **Turan ÜNAK**, **Professor of Nuclear Chemistry** for his continuous support during all my studies and all my educational activities. His valuable academic guidance will help me for directing my future academic life.

My sincere thanks also goes to my colleagues **Çiğdem ÇETIN**, **Gözde SARICAN**, and **Gülcan ÜNAK** for their material and moral supports during my studies.

Also, I would like to thank the main authorities of the "**21st Century Research and Development Company**" for their permissions to be used their research laboratory facilities at the **KOSGEB** building at the **Ege University** Campus area, and for their financial support, which was offered for helping to cover the required expenses related to these studies.

In addition, I would like also to thank the **Research Funding Office of Ege University,** which partially funded my research project (Project code: BAB Projesi, FEN-2010, 069) and the **Iso-Analytical Ltd.** (UK) for their attention on Oxygen-18 measurements in this study.

My deepest gratitude is to all of my family members who supported me at each step of my life including my graduate and undergraduate studies and to all of my friends for their patience and spiritual supports.

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1. INTRODUCTION

1.1 General Information about the Stable Isotopes and ^{18}O

1.1.1 Atomic nuclei and isotopes

1.1.1.1 The structure of an atomic nucleus

Atomic nuclei are roughly consisted of particles that are called protons and neutrons. Protons and neutrons are both called as 'nucleons', because they cannot be distinguished in the nuclei. Around the nucleus there are also electrons which revolve in circular motion on a definite orbitals. Each orbital represents a specific energy level that depends on its distance from the nucleus.

Protons and neutrons are not the fundamental particles on the subatomic scale. Nucleons are composed of smaller particles that are called quarks and leptons. The energy that is required to produce a quark is much higher than the energy required to split the nucleus.

Protons are positively charged particles but in the nucleus, they can live together despite of the Coulomb repulsion between them. Neutrons are neutral particles though they do not have any charge. So, what is the force that holds the neutrons and protons together in a nucleus? Obviously, there are effective forces which maintain the integrity of the nucleus and prevent it from its disintegration by Coulomb repulsion. It is responsible for balancing the repulsive electric force between the protons. The potential that represents the energy of holding the protons and the neutrons in the nucleus was modelled by a Japanese scientist Yukawa. According to his theory, there are mesons between protons and neutrons. Mesons have both negatively and positively charged types. If a negative meson frees from a neutron, it forms a meson bridge between a proton and the positively charged part that remained from the neutron. So, the Coulomb repulsion between nucleons is precluded and the particles hold together strongly by the help of meson bridge. But, it is thought that these electronic forces is not enough to hold the nucleons together so strongly (Ünak, 2006).

Another type of forces which are effective in a nucleus, is magnetic interactions between nucleons. In the nucleus each nucleon rotates around its own axis. So, spin-spin interactions occur in a nucleus and it is known as magnetic

interaction. It is important to remember that this interaction shows itself between the particles which are so closed, as a distance of 10^{-13}cm between the nucleons. That is why the magnetic forces are so effective in an atomic nucleus. Finally, it does not wrong to say that the magnetic forces are the main type of forces that hold the nucleons together to form an atomic nucleus (Ünak, 2006).

1.1.1.2 The stability of an atomic nucleus

The stability of nuclei is characterized by some important rules. One of them is the symmetry rule, which states that in a stable nucleus with low atomic number, the neutron-to-proton ratio, *N / Z*, is approximately equal to 1. In stable nuclei, the *N / Z* ratio is always greater than '1', with a maximum value of about 1.5 for the heaviest stable nuclei. The electrostatic Coulomb repulsion between positively charged protons is rapidly increased with increasing *Z*, in order to maintain the stability in the nuclei, more neutrons than protons are incorporated into the nucleus (Figure 1.1.1.2) (Hoefs, 2009).

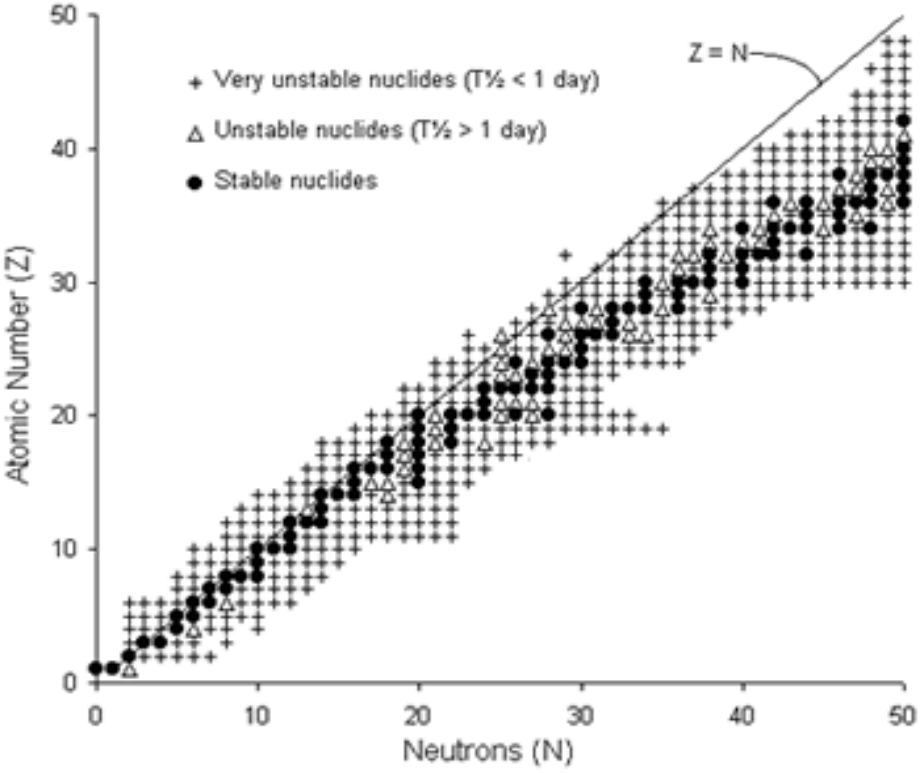


Figure 1.1.1.2 Plot between numbers of protons (Z) and neutrons (N) of stable and unstable nuclides.

The other rule is the *Oddo–Harkins* rule, and it states that nuclei of even atomic numbers are more abundant than the ones with odd numbers. As shown in

Table 1.1.1.2, the most common of the four possible combinations is even–even (proton-neutron) and the least common is odd–odd (proton-neutron) (Hoefs, 2009).

Table 1.1.1.2 Odd-Even factor in nuclear stability.

Number of Protons – Neutrons in the nucleus	**Number of Stable Natural Nuclei**
Even – even	160
Even – odd	56
Odd – even	50
Odd – odd	5

Nuclei in which either the number of protons or the number of neutrons is equal to one of the following numbers, "2, 8, 20, 28, 50, 82, and 126", which are corresponded to the number of nucleons of fully filled nuclear shells, are highly stable. These numbers are called "magic" numbers and the numbers of protons, neutrons or both of them in the most stable and abundant nuclei are corresponded to these magic numbers. Some examples are ^{4}He, ^{16}O, ^{40}Ca, and ^{208}Pb (ChemPaths, 2011).

1.1.1.3 Stable and radioactive isotopes

Isotopes are atoms of the same element which have different number of neutrons. The proton numbers of atoms are their elemental characteristics, but the number of neutrons can only differ their isotopic characters. The difference in the number of neutrons between the various isotopes of an element means that the isotopes have different masses of the same element. All isotopes of an element have practically identical chemical properties and this means that it is difficult to separate isotopes from each other by conventional chemical processes. However, the physical properties of the isotopes, such as their masses, boiling points, and freezing points are different and isotopes can be separated, if even difficultly, from each other by using these physical differences. Elements can naturally have one or more isotopes. Some natural isotopes of certain elements are radioactive (Hoefs, 2009).

Stable isotopes do not undergo radioactive decay within our detection limits; but, they can be produced by the decay of radioactive isotopes. Radioactive isotopes undergo radioactive decay and are transformed to other isotopes. This means that they are unstable. Depending on the detection limits of radioactive decay speed, the term stable is relative (Hoefs, 2009).

It is important to outline that some theoretical considerations show that also protons are not really stable particles and should be decayed with a life-time of about $10^{28 - 30}$ years. So, the isotopes which are considered to be practically stable, should be decayed like radioactive isotopes forming the lighter isotopes in the universal time scale (Ünak, 2006).

1.1.1.4 Isotopic structures of natural elements

Natural elements often have one or more isotopes. There are only 21 elements that have only one stable isotope in nature, are known as isotopically pure elements (Hoefs, 2009). All other elements are exist in the nature as mixtures of at least two isotopes. The relative abundance of different isotopes of the same element may be different.

Stable isotopes are occurred in nature; but, the majority of radioactive isotopes are not naturally occurred because of their short half-lives. Radioactive isotopes which have enough long half-lives (about 10^8 years or longer) can be naturally occurred, if even, with very low naturally abundances. The natural abundance of an isotope indicates its amount found in the nature. As an example, Uranium has ^{235}U and ^{238}U naturally occurring isotopes and both of them are radioactive. Their half-lives are enough long (^{235}U $t_{1/2}$ = $7.13.10^8$ years, ^{238}U $t_{1/2}$= $4.51.10^9$ years). Thus, they could reach today from their production epoch, which means from Big-Bang of about 13.7 billion years earlier. The natural abundances of these isotopes are 0.720 % for ^{235}U and 99.2745 % for ^{238}U. For example, if 100,000 uranium atoms are analyzed, one would expect to find approximately 99,275 ^{238}U isotope and only 720 ^{235}U isotope.

Thorium has only one natural radioactive isotope, ^{232}Th, and of course, its natural abundance is 100 %. Its half-life is $1.405.10^{10}$ years. This means that everywhere, for example in $Th(NO_3)_4$ x $5.H_2O$ salt has only ^{232}Th isotope. Only naturally occurring isotope of Iodine is ^{127}I with abundance, of course, 100 %.

Other Iodine isotopes that total numbers are about 30, are radioactive and not exist in nature.

Six Hydrogen isotopes are principally known: ^{1}H (Protium), ^{2}H (Deuterium), ^{3}H (Ttritium), ^{4}H, ^{5}H, and ^{6}H, ^{7}H, but only, ^{1}H and ^{2}H are stable and naturally occurred. ^{3}H is a radioactive isotope with a half-life of 12.33 year, of course, it is not possible to coincide this isotope in nature. Other ^{4}H, ^{5}H, ^{6}H, ^{7}H Hydrogen isotopes have extremely short half-lives and decay by exotic ways, for example, ^{6}H decays through triple neutron emission and has a half-life of 3×10^{-22} seconds (Gurov et al., 2004).

1.1.1.5 Stable isotopes of some natural elements

Hydrogen is the most abundant element in the universe and ^{1}H has an abundance of 99.985 %. Helium follows Hydrogen and has two stable isotopes, ^{3}He and ^{4}He. ^{4}He is more common than ^{3}He. While the natural abundances of ^{4}He is 99.999863 %, that of ^{3}He is only 0.000137 %. In Table 1.1.1.5, some important natural elements and their stable isotopes are given with their natural abundances.

Table 1.1.1.5 Natural abundances of some natural elements.

Element	Isotopes	Abundances (%)
Hydrogen	^{1}H ^{2}H	99.985 0.015
Oxygen	^{16}O ^{17}O ^{18}O	99.759 0.037 0.204
Carbon	^{12}C ^{13}C	98.890 1.110
Nitrogen	^{14}N ^{15}N	99.630 0.370
Sulphur	^{32}S ^{33}S ^{34}S ^{36}S	95.000 0.760 4.220 0.014

Relative abundances of an element can be measured by Isotope Ratio Mass - Spectroscopy (IR – MS) technique. This technique can be used also to determine the enrichment levels of whatever isotope of an element in small amounts of samples.

Natural isotopic abundance ratios show significant and characteristic variations when they are measured very precisely. Isotopic ratio measurements are useful in many applications as metabolic studies using isotopically enriched elements as tracers; climate studies using measurements of temperature-dependent oxygen and carbon isotope ratios in foraminifers; rock age dating using radiogenic isotopes of elements such as lead, neodymium or strontium; and source determinations using carbon isotopic ratios (American Society for Mass Spectroscopy, 2011).

1.1.2 Oxygen-18

1.1.2.1 Oxygen and its stable and radioactive isotopes

Oxygen has 15 isotopes and 3 of them are stable. The naturally occurring isotopes of Oxygen are stable ^{16}O, ^{17}O, and ^{18}O isotopes with abundances 99.759 %, 0.037 %, and 0.204 %, respectively (Table of Nuclides, 2011).

$^{12 - 15}O$, and $^{19 - 26}O$, isotopes of Oxygen are known and radioactive. Within these isotopes ^{15}O has the longest half-life of about 122.24 seconds. Other radioactive isotopes of Oxygen have extremely shorter half-lives to be about in the range of seconds or milliseconds (Table of Nuclides, 2011).

1.1.2.2 Naturally occuring Oxygen isotopes

As it was noted above, ^{18}O has an abundance of about 0.204 %. This means that if 100,000 H_2O molecules are analyzed, it would be determined that 204 water molecules contain ^{18}O isotopes and the molecular weight of these water molecules ($H_2{}^{18}O$) are 20. $H_2{}^{18}O$ is known as "^{18}O enriched heavy water" and used as the target material for production of radioactive ^{18}F isotope in medical cyclotrons by the use of ^{18}O (p, n) ^{18}F nuclear reactions. Thus, ^{18}O enriched heavy water has become a very important material for todays's nuclear medical applications.

1.1.2.3 Natural abundances of stable Oxygen isotopes

The relative and absolute abundance of ^{16}O is high, because it is a principal product of stellar evolution and because it is a primary isotope, meaning it can be made by stars that were initially made exclusively of Hydrogen. Most ^{16}O is synthesized at the end of the Helium fusion process in stars and in addition to this, the triple-alpha reaction creates ^{12}C, which captures a ^{4}He to make ^{16}O. The neon burning process creates additional ^{16}O (Meyer, 2005).

Both ^{17}O and ^{18}O are secondary isotopes, meaning that their nucleosynthesis requires seed nuclei. ^{17}O is primarily made by the burning of hydrogen into Helium during the C – N – O cycle; thus, it is a common isotope in the Hydrogen burning zones of stars. Most of the ^{18}O is produced when ^{14}N (made abundant from C – N – O burning) captures a ^{4}He nucleus, making ^{18}O common in the Helium-rich zones of stars (Meyer, 2005). Approximately, for two Oxygen nuclei to undergo nuclear fusion to form the heavier nucleus of Sulphur a billion degrees Celsius is required (Emsley, 2001).

1.1.2.4 Natural abundance of O^{18} and its natural enrichment

In the universe the abundance of ^{18}O is about 0.204 %. It is known that 1,000 mL water contains approximately 2.04 mL of $H_2{}^{18}O$; but, this ratio can differ in some cases. It is known that plant leave waters and some fruit waters have been enriched by ^{18}O having higher ^{18}O ratio than 0.204 %, because of the continuous evaporation on the surfaces of plant leaves and fruits (Craig and Gordon, 1965; Dongmann et al., 1974; Farquhar et al., 1989).

The relative abundances of stable isotopes of an element may be changed by some processes and this is known as isotopic fractionation (Barrie et al. 1989). ^{18}O can be found in all samples which consist of Oxygen; but, in some cases, as it was noted above paragraph, the relative abundance of ^{18}O may be higher than 0.204 %. Seawater can be given as another good example. Because of evaporation, seawater may be enriched for ^{18}O. So, during the long evaporation periods the lighter isotope, ^{16}O, leaves first the seawater surface and ^{18}O may be enriched (Craig and Gordon, 1965).

Gamo et al., (1988) states that stratosphere of the atmosphere is enriched by 2 - 3 ‰ in ^{18}O compared to the troposphere of the atmosphere.

Inequality in the ratio of the heavy isotope to the lighter, ^{18}O / ^{16}O, which are measured in the atmosphere and seawater, is known as "Dole Effect" (Dole 1936, Morita 1935). In 1935, it was reported that the air contained higher ^{18}O ratio than the seawater and this was measured as 23.5 ‰ (Kroopnick and Craig, 1972). This inequality is because of respiration in plants and in animals. Respiration removes the lighter ^{16}O in preference to ^{18}O. So, the relative amount of ^{18}O in the atmosphere increases. This inequality is balanced by photosynthesis (Guy et al., 1989).

Koziet et al., (1994) reported some studies for the analyses of fruit and vegetable juices on ^{18}O and ^{2}H determinations and measured 0.32 ‰ and 0.43 ‰ enrichments in ^{18}O, respectively.

It is known that climatic conditions can affect the isotopic contents of rainwaters and fruit juice waters. In the warmer climate, ^{18}O abundance of water increases (Friedman, 1953).

When the rainwater or the irrigation water absorbed by a plant, fractionation by evapotranspiration occurs and the water is enriched in the heavy isotopes. (Farquhar and Cernusak, 2005).

Relative abundances of an element can be measured by using the Isotopic Ratio - Mass Spectroscopy technique (IR - MS). This technique can be also used for determination of isotopic enrichments.

1.1.2.5 Measurement of ^{18}O ratios in various samples

IR - MS is a type of mass spectrometry technique in which mass spectrometric principles are used for measuring the relative isotopic abundance of an element in a sample (Stellaard and Elzinga, 2005). The IR - MS technique works by introducing a pure gas into an ionisation chamber which is held under high vacuum. Electron is stripped from the gas molecule thus an ion is formed in ionisation chamber. The ions are accelerated out of the chamber by a high voltage electric field into a magnetic field. The ions subjected to an effectively centrifugal force, which is dependent on its velocity and is normal to its direction of travel. This force provides the ionic molecule to follow a circular path. Heavier molecules will be deflected less than light ones due to their weight and ion beams of different mass will be separated as they exit the magnetic field. After

separation, the ions are collected and measured using a simple detector. In order to prevent the colliding of the ions travel through the IR - MS with other molecules, high vacuum is used (Iso-Analytical Ltd., 2011).

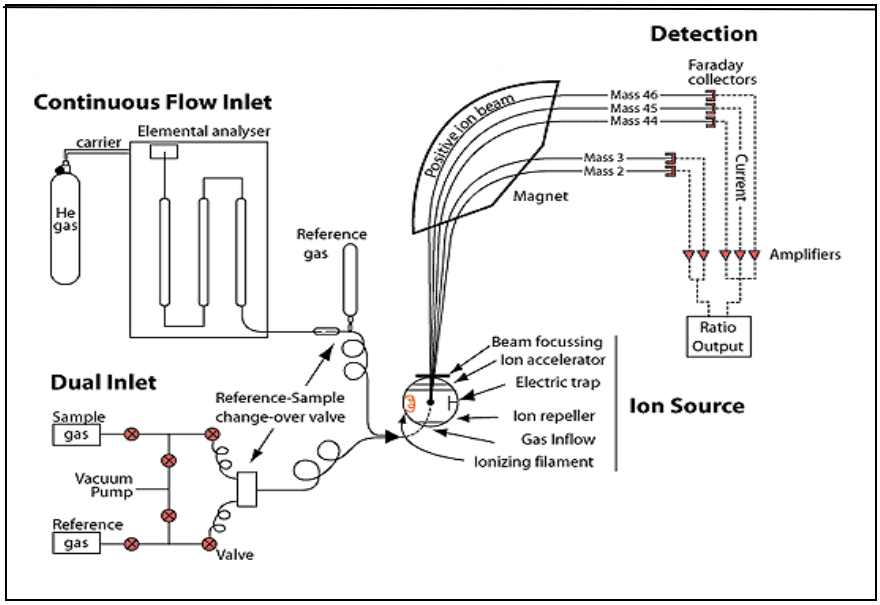


Figure 1.1.2.5 The scheme of IR-MS measurement technique.

In a water sample, after determining the isotopic ratio (^{18}O / ^{16}O) by using IR-MS technique, the sample's isotopic ratio can be compared to a specific reference in order to understand, if the sample water is enriched or depleted in ^{18}O. The ratio of ^{18}O to ^{16}O (or $\delta^{18}O$) in the sample is compared to the Vienna Standard Mean Ocean Water's isotopic ratios (VSMOW), using the following equation:

$$\delta^{18}O = \left(\frac{\left(\frac{^{18}O}{^{16}O}\right)_{sample}}{\left(\frac{^{18}O}{^{16}O}\right)_{standard}} - 1 \right) * 1000\,‰$$

$\delta^{18}O$ values are reported in per mil (‰). The differences between samples and the standard may appear very low, a difference of even 1 per mil is quite significant.

VSMOW is a standard water sample which defines the isotopic composition of whatever sample of water. Despite the ocean water, VSMOW does not include

any salt or other substances usually found in seawater and refers to pure water with a particular composition of isotopes. VSMOW is used as a reference standard for comparing Hydrogen and Oxygen isotope ratios, mostly in water samples. In order to make high accuracy measurement of water's physical properties and for defining laboratory standards, high purity VSMOW water is also used (American Society of Limnology and Oceanography, 1995).

1.1.3 Uses and applications of ^{18}O

^{18}O is commonly used in hydrologic studies. It is known that isotopes undergo fractionations according to mass differences during phase changes. By recognizing the isotopic variations of source waters, they can be distinguished from each other using analytical tools such as hydrograph separation. Stable isotopes can be used within a mine system by giving information about precipitation inputs into the subsurface mine workings (University of Colorado and United States Environmental Protection Agency, 2011).

^{18}O is also used as a tracer during the medical and biological researches in order to study life processes of the brain according to the analysis of metabolic products of biogenic acids in the blood, urine, cerebrospinal fluid under the supervision, and treatment of patients with schizophrenia, manic depression, Parkinson's disease, or the study of oxygen exchange in the lung alveoli in the diagnosis of pulmonary diseases. In these cases, by simple inhalation of molecular Oxygen ^{18}O can be introduced into the body (Centre of Molecular Research, 2011).

The development of various methods of isotopic analysis leaded up the use of stable isotope - labelled compounds possible in many environmental studies. Experts note that the use stable isotopes of Oxygen contributes significantly to the overall environmental program as the use of Sulfur Dioxide, labelled by ^{18}O makes it possible to identify the source of acid pollution after moving clouds for hundreds of kilometers (Centre of Molecular Research, 2011).

^{18}O is also used to produce therapeutic cancer drugs. It is majorly used for production of an important PET radioisotope, ^{18}F, in cyclotrons via ^{18}O (p, n) ^{18}F nuclear reaction. ^{18}F has a half-life of 110 minutes and decays by β^+ emission. Following the decay of a ^{18}F nucleus, β^+ particle which emitted from the nucleus, meets an electron on its pathway and they form a Positronium atom together.

According to the spins of β^+ and e^- two kinds of Positronium atoms may be formed being orto-Positronium and para-Positronium. Their half-lives are considerably different to be about 125 picoseconds and femto seconds, respectively. As a result of its stability because of their spin directions, orto-Positronium is most frequently formed with factor of about 1,115. At the end of this life-time, an orto-Positronium atom is annihilated and two γ photons are created having exactly an equal energy which is 511 keV of each. Consequently, a total energy of 1.022 MeV is created by the annihilation of a Positronium atom either orto or para form. Para-Positronium atom also creates the same total energy, resulting three γ photons emission with different energies. This total energy corresponds to the annihilation of a positron – electron pair, i.e. β^+ and e^- (Green and Lee, 1964; Ünak, 1973).

^{18}F which is produced in cyclotrons by using ^{18}O enriched water, is used for synthesizing of radiofluorinated 2-Fluoro-2-deoxy-D-glucose (^{18}FDG) which is used as an important radiopharmaceutical in nuclear medical applications. ^{18}FDG is able mostly to be cumulated in the brain cells following its injection into body. γ Photons emitted from ^{18}FDG which is cumulated inside the cells, can be used for imaging of tissues and organs. This is known as "Positron Emission Tomography" technique (PET) (Di Chiro et al, 1988)

PET is rapidly developed in recent years as a modern technique for clinical diagnosis and biochemical researches in nuclear medicine and is commonly used with combination of Computed Tomography technique (PET / CT). This technological development allows to investigate the processes of cerebral blood flow, glucose metabolism in the central nervous and cardiovascular systems, to evaluate the impact of drugs on neuroreceptors, etc. (Centre of Molecular Research, 2011).

1.1.4 ^{18}O enrichment techniques

Small differences between the mass numbers of isotopes affects only fairly their basic physical and chemical properties and therefore, Oxygen isotope compositions of various natural compounds are approximately found to be nearly equal. The contents of ^{18}O in atmospheric air, fresh water, and ocean water are 0.204 %, 0.198 %, and 0.200 %, respectively, and various minerals varies by no more than 0.008 %. Therefore, the enrichment of ^{18}O for production of $H_2{}^{18}O$

atmospheric air or natural water as a raw material, is preferentially used (Centre of Molecular Research, 2011).

^{18}O Enrichment of whatever material in an industrial scale, the rectification of liquid Oxygen, Nitrogen Oxyde, or water; the chemical isotope exchange techniques are commonly used. For example in Russia, the rectification of water technique is preferred considering the economical technical feasibilities. Moreover, the most convenient form of ^{18}O to store, to transport, and to use is its form of ^{18}O enriched water (Centre of Molecular Research, 2011).

Rectification method is well studied and widely used for purification of substances in the chemical industry and for the separation of substances with different boiling points (Centre of Molecular Research, 2011).

Natural water can be regarded as a mixture of low-boiling component $H_2{}^{16}O$ with a boiling point at normal pressure of 100 ^{o}C and high-boiling component $H_2{}^{18}O$ with boiling point 100.15 ^{o}C. Separation of $H_2{}^{16}O$ and $H_2{}^{18}O$ occurs in distillation columns. Each column consists of a cube-evaporator, the actual columns filled nozzle, condenser, and the upper reservoir (Centre of Molecular Research, 2011).

In a cube-evaporator there is a continuous evaporation of water from the column. Steam rising up the column interacts with the water flowing as a film on the nozzle - mass transfer occurs. In this case, steam is depleted and the flowing water is enriched with high boiling component $H_2{}^{18}O$ (Centre of Molecular Research, 2011).

At the top of the column steam entered into the condenser, cooled by water from a water recycling system. In the condenser there is a continuous condensation of depleted ^{18}O water vapour. Gravity condensate enters the upper reservoir and mixed with water located there. Then, this water is fed to the tower of the column. The consumption of water corresponds to the mass flow of steam. During this operation process, ^{18}O is transferred from the upper reservoir to the cube-evaporator: Water in cube-evaporator is enriched with ^{18}O and water in the upper reservoir is depleted of this isotope (Centre of Molecular Research, 2011).

1.1.4.1 General information about ^{18}O enriched water

According to the relative abundances of Oxygen isotopes, it is expected the isotopic fractionation in a natural water sample would be as ^{16}O (99.759 %) ^{17}O (0.037 %), and ^{18}O (0.204 %); but, as it was outlined in recent paragraphs, because of some natural processes these isotopic ratios may be different in various samples of water.

Enrichment of ^{18}O in water means the increase of relative abundance of ^{18}O isotopic ratios higher than 0.204 %; for example, 95 % ^{18}O enriched water may be obtained. This kind of water is known as "^{18}O-Heavy Water", "^{18}O Enriched Heavy Water" or "^{18}O Enriched Water". The density of ^{18}O enriched water is higher than that of normal light water, but the density of ^{18}O enriched water are avoided depending on its ^{18}O enrichment level.

1.1.4.2 The commercial potential of ^{18}O enriched water

^{18}O has many different applications in science and technology rather than the nuclear medical applications which were outlined in recent sections. In this context, ^{18}O enriched water has an important commercial potential, as it was notified in recent paragraphs, it is a cyclotron target material for ^{18}F production. For this reason, it is very important to develop new economical and applicable techniques for ^{18}O enrichment of water.

1.2 Basic Information about the Isotopic Enrichment Methods

1.2.1 Isotopic enrichment of Uranium isotopes

Uranium is a natural element which has two natural isotopes, ^{235}U and ^{238}U, and the basic material of nuclear technology. The relative abundances of these isotopes are 0.711 % for ^{235}U and 99.284 % for ^{238}U. ^{235}U is the only nuclide in nature that gives fission reaction with thermal neutrons. In nuclear reactors, heat energy of ^{235}U. "Critical Mass" is as an important factor for producing the chain reactions of fission. The critical mass is the smallest amount of fissional material which is needed for continuation of chain reactions of fission. For this reason, enrichment of ^{235}U is an important process to reach the critical mass, because ^{235}U's relative abundance is very low and the majority of natural Uranium is consisted of ^{238}U isotope. In case of ^{235}U enrichment ratio is high, the possibility

of start of chain reactions meeting a thermal neutron will, of course, increase (Rhodes, 1986). Different techniques may be applied for enrichment of ^{235}U isotope. These are listed below:

- Diffusion techniques
  - Gaseous diffusion
  - Thermal diffusion
- Centrifuge techniques
  - Gas centrifuge
  - Zippe centrifuge
- Laser techniques
  - Atomic vapour laser isotope separation (AVLIS)
  - Molecular laser isotope separation (MLIS)
  - Separation of isotopes by laser excitation (SILEX)
- Other techniques
  - Aerodynamic processes
  - Electromagnetic isotope separation
  - Chemical techniques
  - Plasma separation

Gaseous diffusion and gas centrifuge techniques are the most popular techniques for enrichment of ^{235}U isotope in industrial scale. The main principles of these techniques are shortly notified below:

1.2.1.1 Gaseous diffusion

^{238}U and ^{235}U have different diffusion rates in gas phases, because of the molecular mass differences between their chemical compounds used for this process. As is known well, gaseous diffusion is based on Graham's law, which states that the rate of diffusion of a gas is inversely proportional to the square root of its molecular mass. Gaseous diffusion is a technique which is commonly applied in nuclear technology to produce ^{235}U enriched materials. In this technique a gaseous compound at NTP of Uranium, UF_6, is passed through semi-permeable membrane containers. UF_6 molecules which contain ^{235}U isotopes will pass out of the container more rapidly than the heavier molecules that contain ^{238}U isotopes. The gas leaving the container is enriched in the lighter molecules and this means the depletion of residual gas for ^{235}U isotopes (Goldsworthy, 2008). Gaseous diffusion is one of the most popular Uranium enrichment technique

applied in nuclear technology. This technique is used for production of 33 % of ^{235}U enriched nuclear materials in today's World (Goldsworthy, 2008).

1.2.1.2 Gas centrifuge

Gas centrifuge technique is also commonly used for isotope separation of gases. A centrifuge relies on the principles of centripetal force and can separate different isotopic molecules in a gradient along the radius of a rotating container, because of the difference between isotopic masses. In nuclear technology, gas centrifuges are used for separation of ^{235}U from ^{238}U. Many centrifuges unites are arranged in a cascade system in order to obtain high level of separation of these isotopes. This process yields higher ^{235}U enrichment levels respecting to same ^{235}U enrichment levels obtained by gaseous diffusion process, while using significantly less energy (Olander, 1981; Maier- Komor, 2009).

1.2.2 Enrichment of stable isotopes

The enrichment techniques of stable isotopes are, in principles, similar the techniques applied for Uranium, but according the chemical properties and isotopic masses, the techniques which may be applied, are summarized as follows:

These techniques are:

- Those based directly on the atomic weight of the isotope,
- Those based on the small differences in chemical reaction rates produced by different atomic weights,
- Those based on the properties not directly connected to atomic weight, but also, for example, the effects related to nuclear resonances capabilities.

In this context, Gaseous diffusion, Centrifugal effect, Electromagnetic separation, Laser separation, Chemical methods, and Gravity related techniques may be used in stable isotope enrichment processes in industry.

Electromagnetic separation relies on the principle of mass spectroscopy. Charged particles are deflected in a magnetic field and the amount of deflection depends upon the particle's mass. Chemical methods rely on the fact that reaction

rates are very slightly affected by atomic mass. This technique is used for light atoms and especially in enrichment of Hydrogen isotopes (Kouzmine et al., 1997; Greenland, 1990). In Gravity technique, isotopes of Carbon, Oxygen, and Nitrogen can be purified by chilling these gases or compounds nearly to their liquification temperature in very tall columns (70 to 200 m). The heavier isotopes sink and the lighter isotopes rise, where they are easily collected. This process is also called as "Cryogenic distillation" (Rhodes, 1986).

1.2.2.1 Enrichment cascades

All large-scale isotope separation techniques employ a number of similar stages which produce successively higher enrichment levels of the desired isotope. Each stage enriches the product of the previous step further before being sent to the next stage. Similarly, the tailings from each stage are returned to the previous stage for further processing. This creates a sequential enriching system called a cascade (Buchachenko and Berdinsky, 1999).

1.2.3 Enrichment of Deuterium

Deuterated heavy water, D_2O, is a kind of water in which both light Hydrogen isotopes are replaced with Deuterium isotopes, Deuterium (D) isotope contains one proton and one neutron and D_2O is present in natural water, but in very only quantity being less than 1 part in 5,000. D_2O is one of the two principal moderators which allow a nuclear reactor to operate with natural uranium as its fuel. The other moderator is reactor-grade graphite (graphite containing less than 5 ppm Boron and with a density exceeding (1.50 gm / cm^3). The first nuclear reactor built in 1942 used graphite as the moderator; German efforts during World War II concentrated on using heavy water to moderate a reactor using natural Uranium (Federation of American Scientists, 2011).

The importance of D_2O in nuclear technology is closely related to its use for production of Plutonium for nuclear weapons, entirely bypassing the Uranium enrichment process and all of the related technological infrastructures. In addition, heavy-water-moderated reactors may be also used to produce Tritium (^3H) which is used in fusion weapons (Hydrogen bomb) (Federation of American Scientists, 2011).

Although one speaks of making heavy water, Deuterium is not made in the process; rather, molecules of heavy water are separated from the vast quantity of light water which also contains D_2O or HDO (singly deuterated water). Alternatively, the water may be electrolyzed to produce Oxygen and Hydrogen containing Deuterium gas. Then, Hydrogen gas is liquefied and distilled for separating light Hydrogen and Deuterium gases. Finally, the resulting Deuterium gas is reacted with Oxygen to form D_2O. (Federation of American Scientists, 2011; Ayres and Trilling, 1971).

The production of heavy water in significant amounts requires a technical infrastructure, but one which has similarities to ammonia production, alcohol distillation, and other common industrial processes. One may separate heavy water directly from natural water or first enrich the deuterium content in Hydrogen gas. It is possible to take advantage of the different boiling points of heavy water (101.4 °C) and normal light water (100 °C). However, because of the low abundance of Deuterium, an enormous amount of water would have to be boiled to obtain useful amounts of Deuterium. Because, the evaporation of water consumes high energy and would use enormous quantities of fuel or electricity. Practical facilities which exploit chemical differences use processes requiring much smaller amounts of energy. Separation methods include distillation of liquid Hydrogen and various chemical exchange processes which exploit the differing affinities of Deuterium and Hydrogen for various compounds. These include the Ammonia / Hydrogen system, which uses Potassium Amide as the catalyst, and the Hydrogen Sulfide / Water system (Girdler Sulfide process) (Federation of American Scientists, 2011).

Separation factors per step are significantly larger for Deuterium enrichment than that of Uranium enrichment, because of the larger relative mass difference between Deuterium and Uranium. While ^{235}U is 0.72 % of natural uranium, and must be enriched to 90 % of the product, Deuterium is only 0.015 % of the Hydrogen in water and must be enriched to greater than 99 %. If the input stream has at least 5 % heavy water, vacuum distillation is a preferred way to separate heavy from normal water (Federation of American Scientists, 2011).

This process is virtually identical to that used to distil brandy from wine. The principal visible difference is the use of a phosphor-bronze packing that has been chemically treated to improve wettability for the distillation column rather than a copper packing. Most organic liquids are non-polar and wet virtually any

metal, while water, being a highly polar molecule with a high surface tension, wets very few metals. The process works best at low temperatures where water flows are small, so wetting the packing in the column is of particular importance (Federation of American Scientists, 2011).

Heavy water is produced in Argentina, Canada, India, and Norway. Presumably, all five declared nuclear weapons states can produce the material. The first commercial heavy water plant was the Norsk Hydro facility in Norway (built 1934, capacity 12 metric tons per year); this is the plant which was attacked by the Allies to deny heavy water to Germany. As stated above, the largest plant, is the Bruce Plant in Canada (1979; 700 metric tons / year). India's apparent capacity is very high, but its program has been troubled. Accidents and shutdowns have led to effective limitations on production (Federation of American Scientists, 2011).

The Bruce Heavy Water Plant in Ontario, Canada, is the world's largest producer of D_2O. It uses the Girdler Sulfide (GS) process which incorporates a double cascade in each step. In the upper (cold, 30-40 °C) section, Deuterium from Hydrogen Sulfide preferentially migrates into water. In the lower (hot, 120-140 °C) section, Deuterium preferentially migrates from water into Hydrogen Sulfide. An appropriate cascade arrangement actually accomplishes enrichment. In the first stage the gas is enriched from 0.015 % Deuterium to 0.07 %. The second column enriches this to 0.35 %, and the third column achieves an enrichment between 10 % and 30 % Deuterium. This product is sent to a distillation unit for finishing to 99.75 % "reactor-grade" heavy water. Only, about one-fifth of the Deuterium in the plant feed water becomes heavy water product. The production of a single pound of heavy water requires 340,000 pounds of feed water. (Federation of American Scientists, 2011)

1.2.4 Isotope enrichment by using electromagnetic radiations

Laser isotope enrichment technique is the most important enrichment method by using electromagnetic radiations. Marling, (1975) reported a successful result on the enriched dissociation products, mainly formaldehyde and carbon monoxide in a desired isotope of Carbon, Oxygen, or Hydrogen are obtained by the selective photodissociation of glyoxal. Glyoxal was subjected to electromagnetic radiation of a predetermined wavelength which excites and

induces dissociation of only those molecules of glyoxal containing the desired isotope.

1.2.4.1 Isotope enrichment by using laser

A laser source is a device that emits light electromagnetic radiation through a process of optical amplification based on the stimulated emission of photons. The term "laser" originated as an wettability for Light Amplification by Stimulated Emission of Radiation (Gould, 1959).

None of the laser enrichment methods have attained sufficient proof of principal status to be economically attractive to pursue commercially. Some of the authors have succeeded in separating Sulfur isotopes using a rather new and different method, know as Condensation repression. In this scheme, a gas of the selected isotopes for enrichment, is irradiated with a laser at a particular wavelength that would excite only one of the isotopes. The entire gas is subjected to low temperatures sufficient to cause condensation on a cold surface or coagulation in the gas. Those molecules in the gas that the laser excited are not as likely to condense or dimerize (coagulate into a double molecule, called a dimer) as unexcited molecules. Hence, in cold-wall condensation, gas drawn out of the system is enriched in the isotope that was laser-excited (Eerkens et al., 2006).

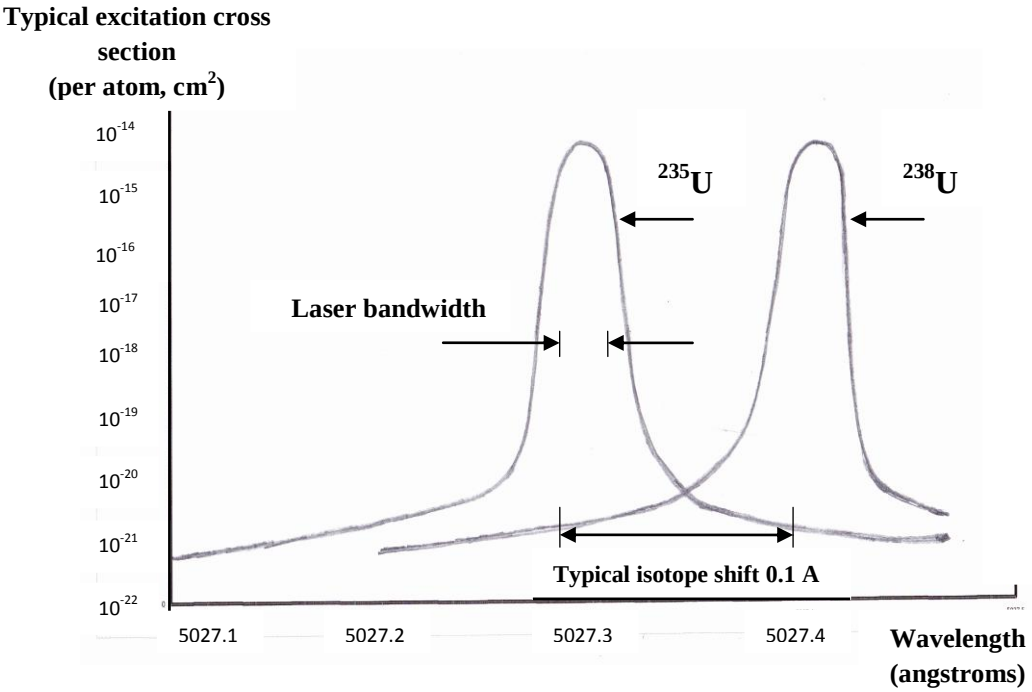


Figure 1.2.4.1 Excitation cross sections for the ^{235}U and ^{238}U isotopes at different laser wavelengths.

Figure 1.2.4.1 (Ragheb, 2011) shows the separation of Uranium isotopes by different laser wavelengths.

1.2.4.2. Isotope enrichment by microwave radiations

Isotope enrichment possibility by the use of microwaves was examined by some scientists. Okazaki et al., (1988) reported that microwave modulation of the product yield can be exploited as a method for the isotopic separation, because in most cases isotopes have different nuclear magnetic moments. It was demonstrated by these authors for the first time that the isotopic enrichment can be achieved by a microwave field through the ESR transitions of the intermediate radical pair in the spin trapping reaction of the photoreduction of menadione in a micellar solution made whit a mixture of ordinary Sodium Dodecyl Sulfate [SDS(H)] and its perdeuteriated isomer [SDS(D)] (Okazaki, 1988).

1.3. Basic Information about the Microwaves and its Applications

1.3.1 Basic information about the electromagnetic waves

Electromagnetic waves or in general electromagnetic radiations consist of an oscillating electrical and magnetic field which are at right angles to each other and spaced at a particular wavelength. Electromagnetic waves travels through the space at the speed of light (2.997924 10^8 m/s). James Clerk Maxwell first formally postulated electromagnetic waves. He was able to create equations that explained the relationship between light and electromagnetic waves. Electromagnetic radiation exhibits both wave properties and corpuscular properties at the same time. Both wave and corpuscular characteristics of electromagnetic radiations were confirmed by a large number of experimental observations. Wave characteristics are more apparent when electromagnetic radiation is measured over relatively large time scales and over large distances; but, particular characteristics are more evident when measuring short time scales and distances (Jefimenko, 2004; Tipler, 2004).

Wavelengths of the electromagnetic radiations spectrum is varied from very long radio waves to very short gamma rays. Their frequencies are inversely proportional to wavelength, according to the following equation:

$$\mathbf{c = \nu\ \lambda}$$

Where, $\boldsymbol{c}$ is the speed of electromagnetic radiation in the vacuum, $\boldsymbol{\nu}$ is the frequency of electromagnetic radiation, and $\boldsymbol{\lambda}$ is the wavelength of electromagnetic radiation.

The energy of an electromagnetic wave that has a specific frequency is given by the following equation:

$$\mathbf{E} = \boldsymbol{\nu}\ \mathbf{h}$$

Here, **E** is the energy of the electromagnetic wave and **h** is the universal Planck's constant and equals to $6.6260689633 \times 10^{-34}$ J or $4.1356673310 \times 10^{-15}$ eV.

According to their energies, electromagnetic radiations can be ionising or non-ionising. X-rays and **γ**-rays are ionising radiations with high frequencies and energies. It means that they can remove electrons from the atoms or molecules. Non-ionising radiations do not have the sufficient energy to ionize atoms or molecules, they can only excite atoms or molecules which means the movement of electrons to higher energy states (Ünak, 2006).

Generally, electromagnetic radiations are classified depending on their wavelengths as radio waves, microwaves, infrared, visible, ultraviolet, X-rays, and **γ**-rays. As it is known well, the electromagnetic radiations in the visible region is perceived as light. In this context, the behaviour of electromagnetic radiations depends on their wavelengths. When an electromagnetic radiation interacts with single atom or molecule, its behaviour also strongly depends on its energy. The energy of an electromagnetic wave is quantumized. The energy quantum is known as “photon” in the quantum mechanical considerations. Photon represents the corpuscular characteristic of electromagnetic radiations (Jefimenko, 2004; Tipler, 2004). The categories of electromagnetic radiations according to their energies, wavelengths, and frequencies, which means the electromagnetic spectrum is represented schematicly in Figure 1.3.1 (MicroWorlds, 2011).

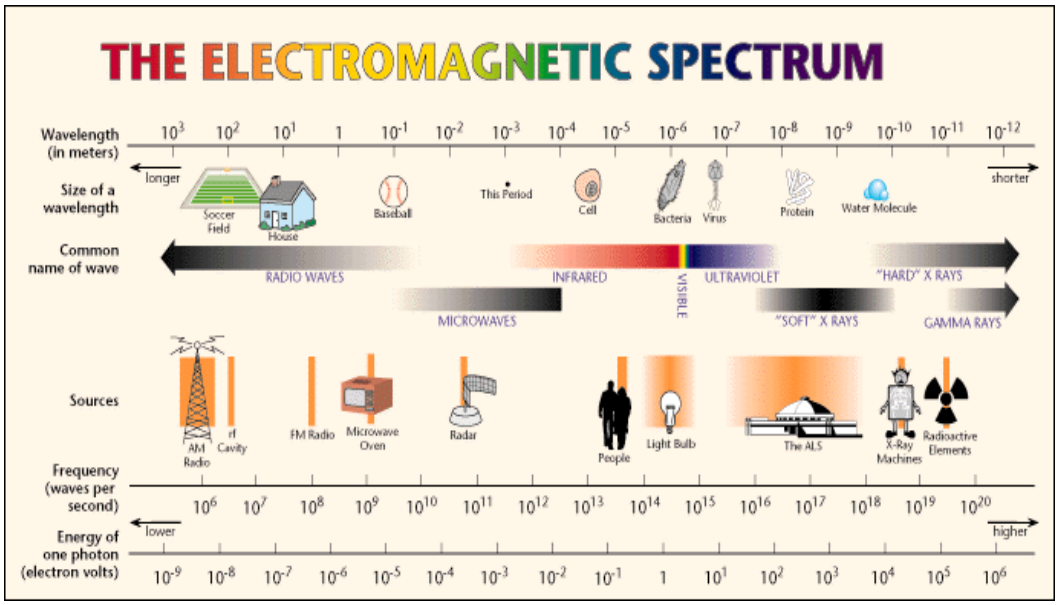


Figure 1.3.1. The electromagnetic radiations spectrum.

1.3.2. Microwaves in the electromagnetic spectrum

Microwaves takes place in a category of electromagnetic radiations spectrum with their wavelengths ranging from as long as 1 m to as short as 1 mm, or their frequencies ranging from 0.3 GHz and 300 GHz. Microwaves takes particularly place between the regions of high energy radiowaves and low energy infrared radiations. Their energy range is between 10^{-5} and 10^{-2} eV, which can only result plasma oscillations and molecular rotations, when they interacts with matter.

1.3.3. Producing heat energy from the microwaves

While the microwave frequencies range from 0.3 GHz to 300 GHz, the most domestic ovens and chemical microwave reactors are operated at a frequency of 2.45 GHz in order to avoid interference with telecommunication frequencies and possible and unpredictable health effects on the public (Boye, 2005).

Some substances absorb energy from the microwaves in a process called “Dielectric Heating” This means that the absorption of microwaves by some substances results the production of heat and consequently, the heating of these substances (Baghurst and Mingos, 1992). Microwave heating relies on two principal mechanisms as is notified below:

1.3.3.1. Dipolar polarisation

The electric field component of microwave irradiations causes dipoles and ions to align, and as the applied electric field oscillates, the dipole - ion field is forced to re-align itself. In this process, energy is lost as heat, through both molecular friction and dielectric loss (Lidström et al., 2001).

It is well known that water molecules have characteristic electric dipoles. The orientation of the dipoles changes while direction of the electric field of microwaves change overtime. Polar molecules, partially or completely adjust themselves in the direction of the field by rotation (Lidström et al., 2011).

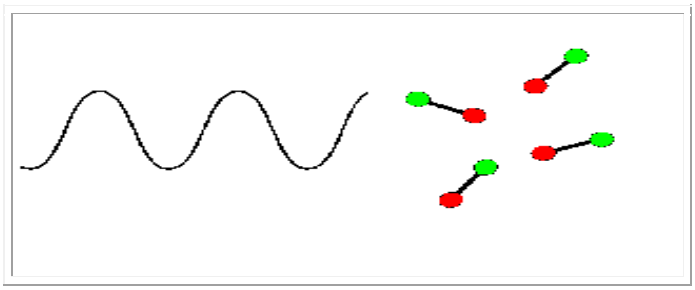


Figure 1.3.3.1 Dipolar polarisation.

1.3.3.2 Conduction mechanism

Where the irradiated sample with microwaves becomes an electrical conductor, the charge carriers (electrons, ions, etc.) are moved through the material under the influence of the electric field, **E**, resulting in a polarisation, **P**. These induced currents will cause heating in the sample due to any electrical resistance. For a very good conductor, complete polarisation may be achieved in approximately 10^{-18} s, indicating that under the influence of a 2.45 GHz microwave, the conducting electrons move precisely in phase with the field (Whittaker, 1997).

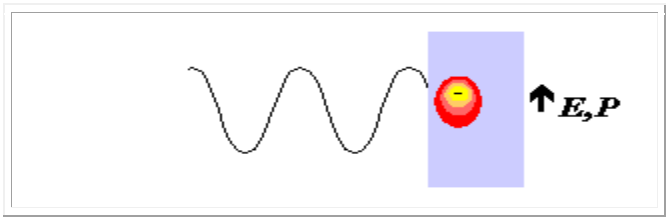


Figure 1.3.3.2 Conduction mechanism.

If the substance is too conducting, such as a metal, most of the microwave energy does not penetrate the surface of the material, but it is reflected. However, the colossal surface voltages which may still be induced are responsible for the arcing that is observed from metals under microwave radiation. Thus, if pure water is heated in a microwave oven, where a variant of the polarisation mechanism dominates, the heating rate is significantly less than the same volume salt added water. In the latter case, both mechanisms occur and contribute to the heating effect (Whittaker, 1997).

In gases or liquids, molecules can rotate with field frequencies of 1 MHz and higher (Gabriel et al., 1998). The rotating molecules hit other molecules and put them into motion; so, the energy is lost as heat during these processes.

1.4. Microwaves and its Applications in Chemistry

1.4.1 Basic information about the microwave applications

In 1945, the heating effect of microwaves was discovered and in 1947, first commercial microwave oven was built. The first household microwave ovens were built in 1967. After that time, microwaves have constantly been used for heating foods and drinks in today's life.

Microwaves can also be used as an energy source for chemical reactions and processes. Through dielectric heating, reaction mixtures are homogenously heated and reaction times are significantly reduced compared to conventionally (thermally) heated systems and acceptable yields and selectivities maintained. The disadvantage of the chemical reactions and processes occurred in the microwave field is that they depend more on the employed devices and substances than in the case of thermal heating. Many organic chemical reactions and processes need some energy to occur. Most often, thermal heating processes are used to provide the energy to a specific reaction. On the other hand, electromagnetic electromagnetic radiations in some frequencies have a great influence on the energy input into chemical reactions (Internet Chemistry, 2011; Haswell and Kingston, 1997).

Photochemistry studies the chemical reactions occurred by non-ionizing radiations and these reactions called "Photoreactions". UV / Vis radiations have been known and used for a long time, but in 1980's microwaves started to be

commonly used in laboratories and industrial establishments. The first organic reaction occured by using microwave energy is reported in 1986 (Gedye et al., 1986, Giguere et al., 1986).

A field in which microwaves are already used in chemical laboratories is the extraction (microwave - assisted extraction, MAE) of organic pollutants for the isolation and preparation of natural products. MAE becomes an alternative to conventional Soxhlet extractions, which usually involve long extraction times and high amounts of solvents. Advantages of MAE are associated with the increased boiling point of the used extracting solvent due to pressure increase (Ganzler et al., 1990; Lopez-Avila et al., 1995).

Kawamura et al., (1999) applied microwave digestion as pre-treatment of environmental samples for the determination of Lead isotopes by ICP-MS. Microwave digestion decomposed the samples easily and rapidly without lead contamination.

Lucchesi et al., (2004) tried solvent - free microwave extraction (SFME) of essential oil from aromatic herbs. Experiments were performed at atmospheric pressure without solvent or water.

Microwaves were also used in labelling some organic compounds by radioisotopes or stable isotopes in recent years (Guo et al., 2008).

1.4.2 The importance of microwave chemistry

The application of microwave irradiations to chemical reactions has been created a new application field which is known as “Microwave Chemistry”. Many reactions and processes need energy adding to occur. Microwave irradiation, offering a clean, cheap, and convenient method of heating is becoming a popular method of heating samples in laboratories. It often results in higher yields and shorter reaction times (Tripathy et al., 2010).

It takes much longer to achieve the target temperature by conventional heating and it usually requires the use of a furnace or oil bath, which heats the walls of the reactor. Microwave heating, in theory producing more uniform heating, is able to heat the target compounds without heating the furnace or oil

bath, which saves time and energy. It is also able to heat sufficiently thin objects throughout their volume (Loupy, 2006).

Microwave energy is converted to heat as different amounts of energy bydifferent compounds and this results the differently heating of some different parts of the same objects (Bukhariya et al., 2011).

As it seems, there are many advantages of applying microwaves to chemical reactions; so, the importance of Microwave Chemistry has become prominent in recent years.

Electromagnetic waves and especially lasers are being used in isotope enrichment processes (Karlov and Prokhoro, 1976). In this context, it is important to investigate the microwave effects on the isotope enrichment processes.

1.5 Main Goals of this Study

^{18}O enriched water ($H_2{}^{18}O$) with about % 97 – 99 enrichment ratio is commonly used as irradiation target material in medical cyclotrons for ^{18}F production. This means that $H_2{}^{18}O$ has become an important material in nuclear medical applications. In general, $H_2{}^{18}O$ can be produced by successive distillations of natural water and this is its commercial production technique in industry.

The main goal of this study is to try preliminarily testing of a new enrichment technique of ^{18}O in natural water hoping to be more practical and cheaper. In this context, the microwave irradiations of water have been subjected to be the main experimental procedure of this study. Also, the commercial evaluation of microwave irradiation technique was aimed.

2. EXPERIMENTAL

2.1 Specifications of Materials and Equipments

2.2.1 Purchasing of natural water which was used for evaporations

Natural water which was used in this study was purchased from a water purification company (Çiçekçi Ticaret, in Izmir). Analytical characteristics of this water is given in Table 2.2.1.

Table 2.2.1 Cationic and anionic ingredients of pure water used in this study for evaporations.

Cationic and anionic ingredients	Measurement Method	Measured values (mg/L)
Copper (Cu)	ICP-MS	0.003
Zinc (Zn)	ICP-MS	0.005
Iron (Fe)	ICP-MS	5.000
Fluoride (F^-)	IC	0.030
Calcium (Ca)	IC	1.000
Chloride (Cl^-)	IC	1.000
Lead (Pb)	ICP-MS	2.000
Magnesium (Mg)	IC	1.000
Nitrate (NO_3^-)	IC	1.000
Potassium (K)	IC	0.300
Sodium (Na)	IC	0.800
Total Hardness	Titrimetry	0.200
Nitrite (NO_2)	IC	0.100
Ammonia (NH_3)	IC	0.200

.

As it is seen in Table 2.1.1, water used for enrichment processes in this study was purified natural water and its ^{18}O natural ratio was measured to be 0.19874 ± 0.00003 % by Iso-Analytical Ltd. in the UK (Table 3.1.3).

2.1.2 Purchasing of ^{18}O enriched water

$H_2{}^{18}O$ with enrichment ratios of 97 % and 10 % which were used in this study for testing of reliability of ^{18}O measurements carried out by Iso-Analytical Ltd. in the UK, were purchased from Shangai Research Institute of Chemical Industry in China. Analytical characteristics of $H_2{}^{18}O$ are given in Tables 2.1.2.1. and 2.1.2.2.

Table 2.1.2.1 Analytical characteristics of 97 % enriched $H_2{}^{18}O$.

Isotopic Analyses	Units	Measured Values
^{18}O	Atom %	97.5
^{17}O	Atom %	0.7
^{16}O	Atom %	1.8
Chemical Purity		99.99 % +
Electrical conductivity	µS / cm	0.52
Pyrogen	EU / mL	0.12
Sterility		Passed
pH		7.0
Total organic carbon	mg / L	0.2
Fluoride (F)	mg / L	0.002
Chloride (Cl)	mg / L	0.002
Bromide (Br)	mg / L	0.002
Iodide (I)	mg / L	0.002
Calcium (Ca)	mg / L	0.02
Magnesium (Mh)	mg / L	0.02
Sodium (Na)	mg / L	0.2
Potassium (K)	mg / L	0.02
Copper (Cu)	mg / L	0.01
Iron (Fe)	mg / L	0.02
Phosphate	mg / L	0.01
Nitrate	mg / L	0,01

Table 2.1.2.2 Analytical characteristics of 10 % enriched $H_2{}^{18}O$.

Isotopic Analyses	**Units**	**Measured Values**
^{18}O	Atom %	10.5
^{17}O	Atom %	0.06
^{16}O	Atom %	89.44
Chemical Purity		99.99 % +
Electrical conductivity	μS / cm	1.2
pH		6.9
Total organic carbon	mg / L	0.3
Fluoride (F)	mg / L	0.002
Chloride (Cl)	mg / L	0.002
Bromide (Br)	mg / L	0.002
Iodide (I)	mg / L	0.002
Calcium (Ca)	mg / L	0.03
Magnesium (Mh)	mg / L	0.01
Sodium (Na)	mg / L	0.1
Potassium (K)	mg / L	0.02
Copper (Cu)	mg / L	0.02
Iron (Fe)	mg / L	0.02
Phosphate	mg / L	0.01
Nitrate	mg / L	0,01

2.1.3 Microwave oven and its modification for continuous irradiations

In this study, a microwave oven which was designed for kitchen applications has been used for continuous irradiations of natural water; but, this microwave oven was first considerably modified for its adaptation to this kind of irradiations. Shi et al. (2006) also modified a domestic microwave oven for their experiments, saponification of oils and fats under normal pressure. In this study, in water evaporation operations by microwave irradiations a modified domestic microwave oven was used (Figure 2.1.3., Ünak et al., 2011). The initial technical specifications of this microwave oven are given below:

Mark: Tesco; Model: MM08, 230-240V~50 Hz; Input Power: 1150 W
Microwave Output: 700 W, 2,45 GHz frequency

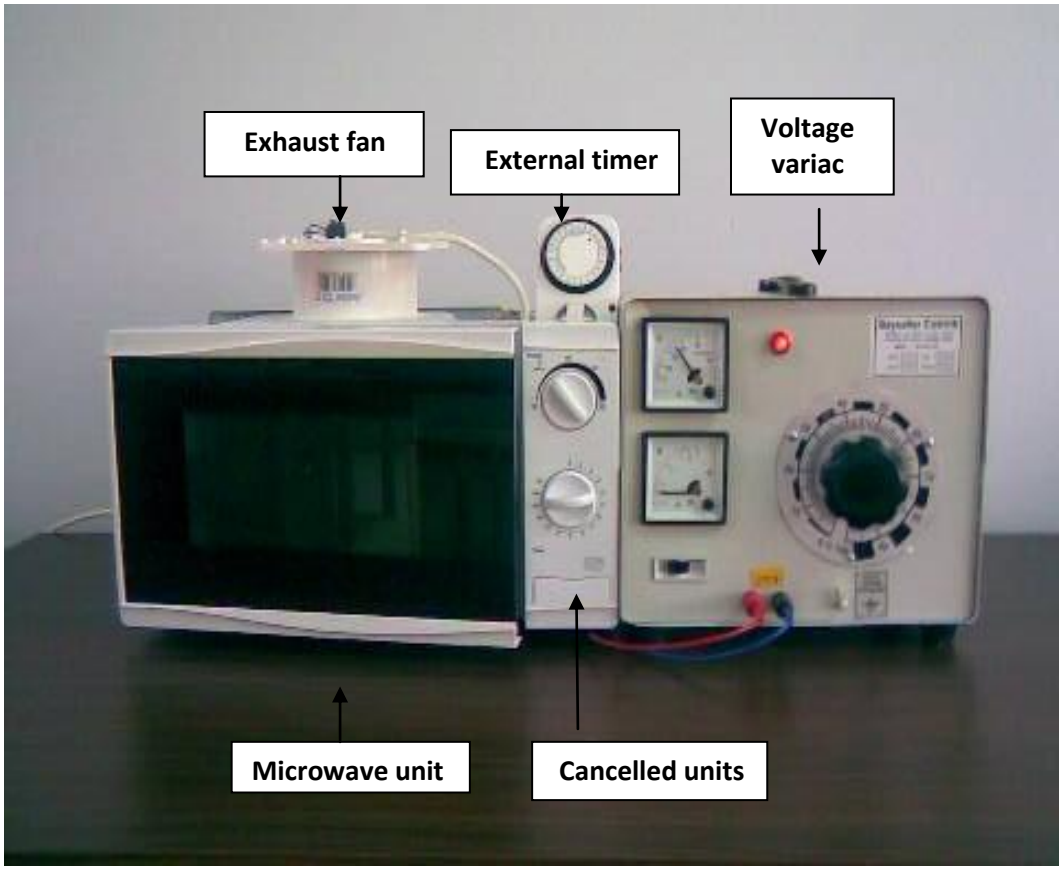


Figure 2.1.3 A view of the modified microwave kitchen oven which was used in this study as a microwave irradiation source.

A simple domestic microwave oven was not convenient for required irradiations which were expected in this study. For this reason, a serious modification was necessary for being able to adjust a domestic microwave oven to this kind of irradiations. As it is known that a simple domestic microwave oven works under the control of a thermostat which regulates the internal temperature of the oven. This means that the objects placed inside of the oven are irradiated in dashed time intervals by microwave radiation. In addition, the maximal irradiation periode of such an oven is about 1 h. On the other hand, the water vapor accumulated in the oven during the long term evaporations of water have created serious problems for safely evaporation operations. Briefly, the following modifications were done on a simple domestic microwave oven:

1) The original timer and the thermostat units of the microwave oven were deactivated. Instead of them an external timer and a voltage variac were assembled as indicated below.

2) An exhaust fan with a capacity of ventilation of about 150 m^3 / h was mounted on the top panel of oven making a hole and it was connected to the electric circuit of the microwave oven in order to build up a strong ventilation system; thus, when the oven starts to work, the fan starts in parallel. In some cases of evaporations, 1,250 mL water was irradiated and consequently, high amounts of water vapour were produced inside the small volume of microwave oven and this could not easily be exhausted from the microwave oven.

3) A voltage variac was introduced into the main electric circuit of oven, which guarantees the power control of the oven. Thus, the time and the speed of water evaporations could appropriately be arranged.

4) An external timer which was capable to work during 24 h and adjustable in 15 min intervals, was introduced into the main electric circuit of oven. This was used for being automatically controlled the work of oven.

2.1.4 Hot-plate used for conventional water evaporations

In order to determine the real microwave absorption effect on the enrichment of ^{18}O in water, it was necessary to know in parallel the conventional heating effect on the same enrichment procedure. For this reason, a hot-plate was used for conventional water evaporation operations. Technical specifications of this hot-plate are as follows: Mark: Esco; Type: EHP401; 230 V, 50 Hz; Power: 1000 W; Current: 4.3 A.

2.1.5 Other tools

2.1.5.1 Balance

In order to determine with precision of evaporated quantities of water a toploading balance was used. Technical specifications of this balance are as follows: OHAUS marked, Adventurer Pro AV2101 model, d = 0.1 g.

2.1.5.2 Electric power counter

In order to determine the power consumed per unit evaporation time and to evaluate the economical aspect of enrichment procedure applied in this study, an

electrical power counter was used. Technical specifications of this counter are as follows: Viko, VEM - M580DB0 Electronic Monophase Watt Hour Meter.

2.2 Water Evaporation Operations

2.2.1 Using microwave power equipment

It was emphasized in a recent paragraph that the main goal of this study is the investigation of enrichment possibility of natural ^{18}O isotope ratio in water by the use of microwave irradiations. Techniques for ^{18}O enrichment in water were recently outlined and it is well known that the "fractional distillation" technique of water is commonly used in industry for ^{18}O enrichment processes.

During the evaporation of water, H_2O molecules which contain lighter ^{16}O isotopes, are first left from the water surface and the molecules which contain ^{18}O isotopes may be enriched in the water remained (Craig and Gordon, 1965). This general principle of ^{18}O enrichment effect provides a basic principle for the evaporation operations applied in this study for enrichment of ^{18}O in natural water by the use of microwave irradiations. Microwaves are widely used in today's life everywhere as an energy source. Especially, it widely used in microwave chemistry applications (Loupy, 2006). In this context, "microwave heating" was used in this study for evaporation operations of water comparing with the conventional heating in order to examine the microwave irradiation effect, if any, on the ^{18}O enrichment in water.

In the experiments, the modified microwave oven as described above, which emits microwaves in 2.45 GHz frequency, was used as a microwave irradiation source.

2.2.1.1 <u>Evaporations details</u>

The evaporation steps of water have been summarized in Scheme 2.2.1.1. As it is seen, 20,000 mL water was definitively evaporated up to 5 mL. For this, 1,250 mL water was used as a starting volume of water for each evaporation operation which has basically three steps. This means that 1,250 mL water was evaporated up to 80 mL at the first step and this step was repeated 16 times. All these waters of 80 mL were mixed and obtained another 1,250 (= 80 x 16) mL water which was evaporated again up to 80 mL at the second step. At third step,

80 mL water remained from the second step was evaporated up to 5 mL. Of course, it was impossible to irradiate 20,000 mL natural water in one step, because of the very limited size (44 cm x 25.8 cm x 34,5 cm) of the simple domestic microwave oven which was used in this study. A sample from starting natural pure water and two other samples from the waters non-evaporated at the end of each step were collected. Finally, two samples of each evaporation operation were sent for ^{18}O ratio measurements to Iso-Analytical Ltd. in the UK.

20,000 mL (= 16 x 1,250 mL) (Natural water)
⇩ ⇩
1,250 mL (= 16 x 80 mL) (Remained 1,250 mL water)
⇩
80 mL (Remained 80 mL water)
⇩
5 mL (Remained 5 mL water)

Scheme 2.2.1.1 Evaporation steps of enrichment operations of natural water.

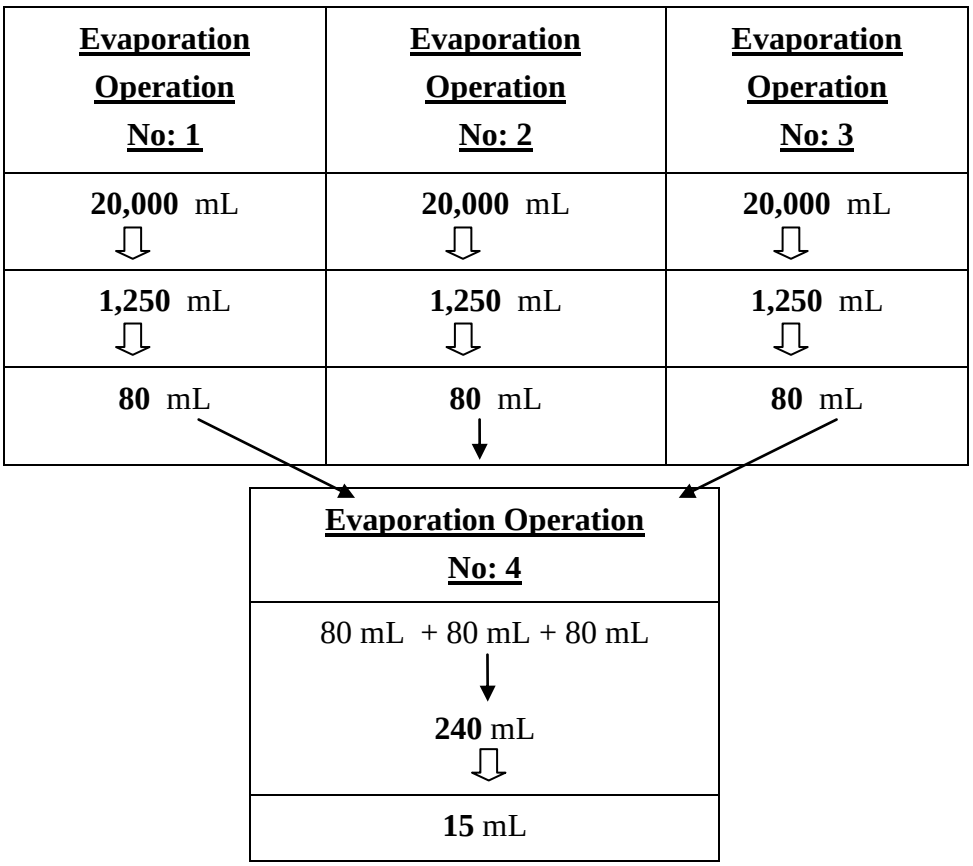
For these irradiations 1,250 mL natural water was irradiated in a glass vessel of about 2 L having a form nearly semi-conical for facilitating the evaporation of water and is well suitable for microwave irradiations. The evaporation of 1,250 mL water to a volume of 80 mL at the first step has needed about 2 h and 50 min. Each 80 mL water portions obtained at the ends of 16 evaporation operations (80 mL x 16) were mixed and this means that a new 1,250 mL enriched water as the first step enrichment product was obtained. A sample of about 1.5 mL taken from this water was used for ^{18}O measurement.

Again, the final 1,250 mL enriched water was similarly evaporated up to 80 mL and a new sample of about 1.5 mL as the second step enrichment product was used for ^{18}O measurement.

Evaporation operations as described above, which have 16 repetitions with 2 steps each were repeated 3 times for obtaining at least triple data for reliability of the experimental results.

As it is seen in the Scheme 2.2.1.2., Evaporation Operation No: 4, was started from 240 mL enriched water and reached 15 mL which means that its

evaporation ratio is nearly equal to that of No's 1 – 3 (1,250 / 80 ≈ 240 / 15 = 80 / 5).

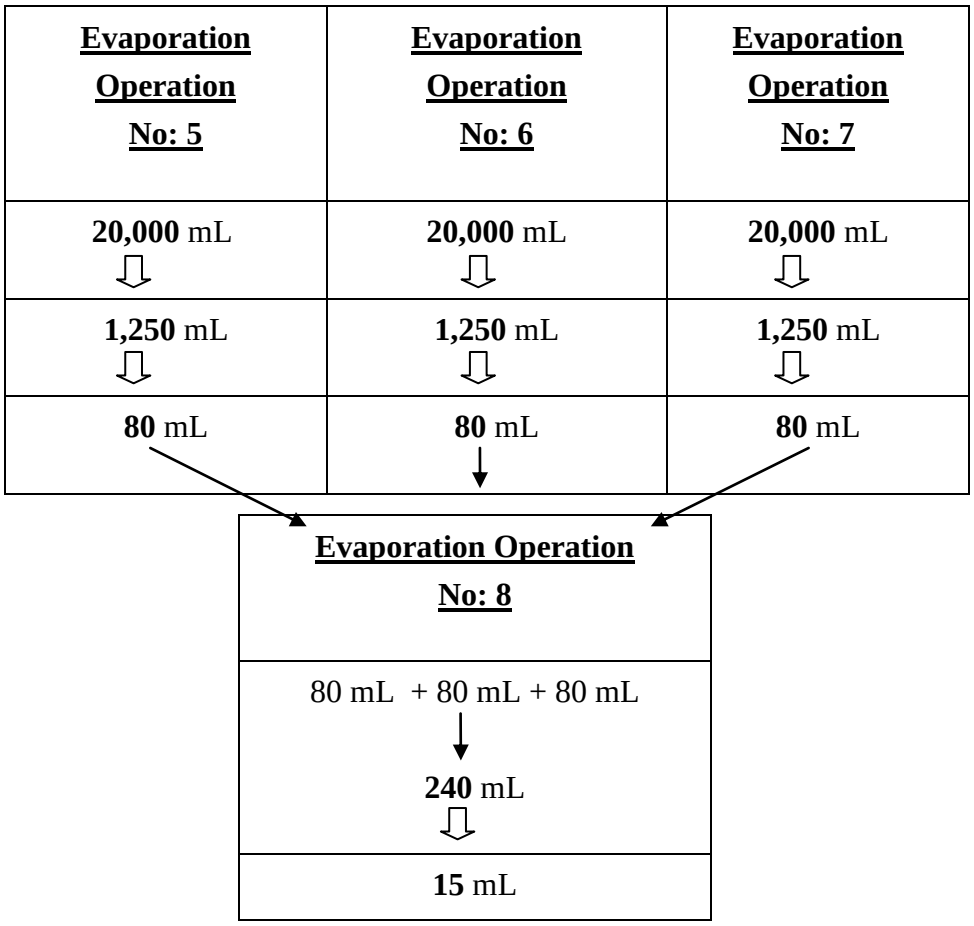


Scheme 2.2.1.2 Schematic representation of evaporation operations by the use of microwave irradiations.

It is also important to underline that; in all steps, same quality glass vessels were used for evaporation of water in microwave oven; thus, the magnitude of experimental errors has provided to be minimized.

2.2.2 Using conventional heating on hot-plate

As it was emphasized in recent paragraphs, for clearly verification of microwave irradiation effect on the enrichment of ^{18}O ratio in water, the effect of conventional thermal heating would also be considered. For this reason, a hot-plate was used for conventional water evaporation operations. All evaporation operations which were described in detail above by the use of microwave irradiations were also equivalently repeated by the use of conventional heating on a hot-plate (Scheme 2.2.2).



Scheme 2.2.2 Schematic representation of evaporation operations by the use of conventional heating on hot-plate.

2.2.3 Details of water samples analysed for ^{18}O enrichment

All water samples taken from each enrichment steps of evaporation operations were sent to Iso-Analytical Ltd, in the UK for ^{18}O ratio measurements expecting the observation of increasing of ^{18}O enrichment. Isotopic Ratio - Mass Spectrometry (IR-MS) technique was applied by this professional expert company on the stable isotopes analyses.

2.3 Measurement of ^{18}O Ratios in Water Samples

2.3.1 Preparation of samples for ^{18}O analyses

About 1.5 mL water samples which were taken from each evaporation steps were stored in a refrigerator until their transfers to Iso-Analytical Ltd. in the UK in polypropylene Eppendorf tubes of 2 mL clung with parafilm.

Table 2.3.1 Water samples used for ^{18}O measurements.

Operation No.	Sample No.	Sample Description
1	1	20,000 mL → 1,250 mL
	2	1,250 mL → 80 mL
2	3	20,000 mL → 1,250 mL
	4	1,250 mL → 80 mL
3	5	20,000 mL → 1,250 mL
	6	1,250 mL → 80 mL
4	7	80 mL → 5 mL
5	8	20,000 mL → 1,250 mL
	9	1,250 mL → 80 mL
6	10	20,000 mL → 1,250 mL
	11	1,250 mL → 80 mL
7	12	20,000 mL → 1,250 mL
	13	1,250 mL → 80 mL
8	14	80 mL → 5 mL
Other	15	Natural water
	16	Natural water
	17	5 % Enriched ^{18}O water
	18	1 % Enriched ^{18}O water
	19	Irradiation of NaCl solution
	20	Irradiation of $K_3[Fe(CN)_6]$ solution

It was expected to protect these samples from open air contact; because of probable isotopic exchanges between ^{18}O of enriched water molecules and ^{18}O of open air oxygen molecules. According to an earlier study, this may be resulted considerable decrease on the ^{18}O ratios of enriched water samples (Ekim et al., 2011).

2.3.2 Transfer of samples to Iso-Analytical Ltd, a company in the UK for ^{18}O measurements

As it was emphasized in recent paragraphs, isotopic ratio of ^{18}O of whatever sample can be measured by Isotopic Ratio - Mass Spectrometry (IR-MS) technique. In the original country of this study, in Turkey, any private or governmental laboratory could not be found for being carried out the

measurement of ^{18}O ratios in water samples, especially for considerably high number of samples. Consequently, all samples collected in this study to be about 20 were sent to professional expert company on the stable isotope ratios measurement in the UK (Iso-Analytical) in order to measure their ^{18}O ratios. Samples were carefully packaged before their handling by an international parcel service. Each parcel which was sent on different dates, had about 5 - 10 water samples and the results of all these measurements were received in about 8 - 10 days following their delivering.

2.3.3 Test of the validity of ^{18}O measurement technique

The validity of ^{18}O measurement technique applied for the water sample measurements in this study was tested by two reference water samples which were prepared with known quantity and known percentage of ^{18}O enrichment to be about 1 % and 5 %. They were prepared by using 97 % ^{18}O enriched water purchased from Shangai Research Institute of Chemical Industry in China. For dilution, natural pure water (^{18}O ratio: % 0,19874) was used. The ^{18}O percentages of reference water samples which have about 1 % and 5 % ^{18}O enrichments, were measured by Iso-Analytical Ltd. to be 1,07860 % and 5,18435 %, respectively. These results have clearly exhibited the validity of the ^{18}O measurement technique applied in this study.

2.4 Economical Evaluation of ^{18}O Enrichment Procedure Applied

2.4.1 Determination of total electrical power consumption

An electric powermeter as Watt per hour (Wh) was connected to the external electrical circuit of the modified microwave irradiation equipment to determine the electrical power consumption per unit evaporation time. Electrical power (kW) consumption for per hour of evaporation time is given in the Table 2.4.1.1.

Table 2.4.1.1 Electrical power consumption of the modified microwave irradiation equipment per hour of evaporation time.

Time (per hour)	Consumed Electrical Power (kW)
1st Measurement	1.007
2nd Measurement	1.007
3rd Measurement	1.008
Average	1.007 ± 0.001

It should be also notified that in order to test the validity of electrical power determination in this study by the use of an electric powermeter as Watt per hour (Wh) was also tested using an electrical lamp of 500 Wh as a reference equipment which consumes known electrical power per h, on which an approval label of the manifacturer company was stamped. The lamp was connected to the electric power meter and digital counts were read for some 1 h intervals. The electrical power consumption for 1 per h of the reference lamp was averagely found to be 0.502 ± 0.001 kWh and this result was reasonable for safely using the electric powermeter for energy consumption measurements and effective cost calculations.

Table. 2.4.1.2 Total irradiation time for 3 evaporation operations which were carried out using modified microwave irradiation equipment.

	Irradiation Times		
Evaporation Steps	**Evaporation Operation No: 1**	**Evaporation Operation No: 2**	**Evaporation Operation No: 3**
20,000 mL → 1,250 mL or 16 x (1,250 mL → 80 mL)	16 x 170 min = 2720 min	16 x 170 min = 2720 min	16 x 170 min = 2720 min
1,250 mL → 80 mL	170 min.	170 min.	170 min
Sub total irradiation time for an evaporation operation	**2,890 min**	**2,890 min**	**2,890 min**

As it is given in Table 2.4.1.1, electrical power consumption per hour of evaporation time is 1.007 ± 0.001 kWh and this was used to calculate total electrical power consumption for evaporation operations. Tables 2.4.1.2 and 2.4.1.3, summarizes the irradiation times for evaporation operations.

As it is seen in Tables 2.4.1.2 and 2.4.1.3, the total irradiation time for all evaporation operations is which was carried out using modified microwave irradiation equipment is about 145.17 h.

Table. 2.4.1.3 Total Irradiation time for Evaporation Operation No. 4 and total irradiation time for all irradiations which was carried out using modified microwave irradiation equipment.

Evaporation Step	**Irradiation Time for Evaporation Operation No: 4**
240 mL → 15 mL	**40 min**
TOTAL	**8,710 min = 145.17 h**

.

As it was given above, the electrical power consumption was measured to be 1.007 kW / h and this means that total electrical power consumption for all evaporation operations which were carried out using modified microwave irradiation equipment is about 146.18 kW (1.007 kW / h x 145.17 h).

2.4.2 Calculation of cost effective of microwave irradiations

. In the original country of this study, in Turkey the electric consumption costs differently for domestic or industrial users and the prices are changed time by time. In spite of these, the average cost of energy consumption per kWh including all taxes and other payments which is shown on the invoices as total amount due to payment can be considered to be about 0.25 TL. Starting from this price total cost of all microwave irradiations which were carried out in this study may be calculated to be 36.5 TL. According to the data given on Scheme 2.2.1.2., total volume of water which was finally evaporated at the end of all evaporation operations is about 60 L. This means that the final effective cost of water

evaporation operations using the modified microwave irradiation equipment used in this study can be found to be about 0.60 TL / L.

3. RESULTS AND DISCUSSION

3.1 Results Related to the Enrichment of ^{18}O Ratio in Water

3.1.1 Results obtained by microwave irradiation process

As it was given in the Experimental Section, 16 times 1,250 mL natural water were evaporated by microwave irradiation up to 80 mL and these evaporations were repeated three time. These evaporation series were mentioned to be Evaporation Operations No: 1, 2, and 3. Totally, 6 enriched water samples were collected from these Evaporation Operations and used for ^{18}O measurements. In addition, 3 enriched water of 80 mL were mixed and obtained 240 mL water having an identical volumic evaporation ratio of that of above Evaporation Operation series and this was identified to be Evaporation Operation No: 4. Including two new samples obtained from this operation series and a sample of starting natural water, the ^{18}O enrichment ratios of totally 9 samples were measured. The results obtained were tabulated in the following tables (Tables 3.1.1.1,.2,.3, and .4).

Table 3.1.1.1 ^{18}O Measurements of the Evaporation Operation No: 1.

Evaporation Operation No: 1 (Microwave irradiation)	**^{18}O Enrichment Ratio (Measured to be Atom %)**
20,000 mL (Natural water) ⇩	0.19874 ± 0.00003
1,250 mL ⇩	0.20552 ± 0.00003
80 mL	0.21246 ± 0.00003

As it is seen clearly from these measurements given in these tables, the natural ^{18}O isotopic ratios, 0.19874, has been considerably increased up to about 0.21959 at the end of the Evaporation Operation No: 4. This means clearly that microwave irradiations resulted significant ^{18}O enrichment in water. The enrichment ratio has been considered to be about % 10.4911 (0.21959 - 0.19874 = 0.02085 x 100 / 0.19874). Surely, this is a very successful enrichment level.

Table 3.1.1.2 ^{18}O measurements of the Evaporation Operation No: 2.

Evaporation Operation No: 2 (Microwave irradiation)	**^{18}O Enrichment Ratio (Measured to be Atom %)**
20,000 mL (Natural water) ⇩	0.19874 ± 0.00003
1,250 mL ⇩	0.20544 ± 0.00003
80 mL	0.21215± 0.00003

Table 3.1.1.3 ^{18}O measurements of the Evaporation Operation No: 3.

Evaporation Operation No: 3 (Microwave irradiation)	**^{18}O Enrichment Ratio (Measured to be Atom %)**
20,000 mL (Natural water) ⇩	0.19874 ± 0.00003
1,250 mL ⇩	0.20560 ± 0.00003
80 mL	0.21242 ± 0.00003

Table 3.1.1.4 ^{18}O measurements of the Evaporation Operation No: 4.

Evaporation Operation No: 4 (Microwave irradiation)	**^{18}O Enrichment Ratio (Measured to be Atom %)**
80 mL (From No. 1) 80 mL (From No. 2) + 80 mL (From No. 3) 240 mL or 80 mL ⇩ ⇩	0.21234 ± 0.00003
15 mL 5 mL	0.21959 ± 0.00003

3.1.2 Results obtained by conventional heating

The enrichment ratio of about % 10.4911 which is given in above paragraph may be a total result of two individual effects: The first one may be the conventional heating effect of microwaves and the second may be an additional enrichment effect of microwaves. For verification of this estimation, it was necessary to carry out the identical evaporation operations using conventional heating. Starting from this consideration, as it was described in the Experimental Section, Additional Evaporation Operations were identically repeated using a conventional heating equipment, a hot-plate. Similarly, 16 times 1,250 mL natural water was evaporated by the use of a hot-plate up to 80 mL and these evaporations were repeated three time. These evaporation series were mentioned to be Evaporation Operations No: 5, 6, and 7. Totally, 6 enriched water samples were collected from these Evaporation Operations and used for ^{18}O measurements. In addition, 3 enriched water of 80 mL were mixed and obtained 240 mL water having an identical volumic evaporation ratio of that of above Evaporation Operation series and this was identified to be Evaporation Operation No: 8. Including of two new samples obtained from this operation serial, the ^{18}O enrichment ratios of totally 8 samples were measured. The results obtained were tabulated in the following tables (Tables 3.1.2.1,.2,.3, and.4).

Table 3.1.2.1 ^{18}O measurements of the Evaporation Operation No: 5.

Evaporation Operation No: 5 (Conventional heating)	**^{18}O Enrichment Ratio (Measured to be Atom %)**
20,000 mL (Natural Water) ⇩	0.19874 ± 0.00003
1,250 mL ⇩	0.20512 ± 0.00003
80 mL	0.21176 ± 0.00003

As it is seen clearly from these measurements given in these tables, the natural ^{18}O isotopic ratios, 0.19874, has been also increased up to about 0.21813 at the end of the Evaporation Operation No: 8. Also, this shows that conventional heating resulted ^{18}O enrichment in water. The enrichment ratio has been considered to be about % 9.7565 (0.21813 - 0.19874 = 0.01939 x 100 / 0.19874). Surely, this is a very successful enrichment level.

Table 3.1.2.2 ^{18}O measurements of the Evaporation Operation No: 6.

Evaporation Operation No: 6 (Conventional heating)	**^{18}O Enrichment Ratio (Measured to be Atom %)**
20,000 mL (Natural Water) ⇩	0.19874 ± 0.00003
1,250 mL ⇩	0.20504 ± 0.00003
80 mL	0.21130 ± 0.00003

Table 3.1.2.3 ^{18}O Measurements of the Evaporation Operation No: 7.

Evaporation Operation No: 7 (Conventional heating)	**^{18}O Enrichment Ratio (Measured to be Atom %)**
20,000 mL (Natural Water) ⇩	0.19874 ± 0.00003
1,250 mL ⇩	0.20489 ± 0.00003
80 mL	0.21125 ± 0.00003

Table 3.1.2.4 ^{18}O Measurements of the Evaporation Operation No: 8.

Evaporation Operation No: 8 (Conventional heating)	**^{18}O Enrichment Ratio (Measured to be Atom %)**
80 mL (From No. 5) 80 mL (From No. 6) + 80 mL (From No. 7) 240 mL or 80 mL	0.21144 ± 0.00003
⇩ ⇩ 15 mL 5 mL	0.21813 ± 0.00003

3.1.3 Standard deviations of measured ^{18}O enrichment ratios

A technical report of Iso-Analytical Ltd. on the ^{18}O enrichment ratios measurements of our samples is given in Table 3.1.3.

Table 3.1.3 A technical report of Iso-Analytical Ltd. on the ^{18}O enrichment ratio measurements of a party of our water samples.

^{2}H ^{13}C ^{15}N ^{18}O ^{34}S ISO ANALYTICAL EXPERTISE IN STABLE ISOTOPE ANALYSIS		
LABORATORY REPORT: Results Files		
Client Details		
Name:	EGE University	
Contact:	Turan Unak	
Sample Details		
Number:	5	
Material:	Water	
Sample Tracking		
IA Reference No.:	120118-1	
Date of Arrival:	18.01.2012	
Analysis Details		
Isotope :	Oxygen-18	
Method:	Equilibration-IRMS	
Report Date:	24.01.2012	
Oxygen-18 Analysis Results		

Sample Identification	**Measured ^{18}O enrichment (Atom%)**	**Sample Description**
1	0,19968	Plant leaf water # 1
2	0,20481	Plant leaf water # 2
3	0,20638	0.035 M NaCl solution
4	0,20765	0.035 M K salt solution
5	0,19869	Natural pure water

NB. The samples did not require dilution prior to measurement because of their low enrichment.

Quality Control Reference Standards

Reference Standard	**Result $\delta\ ^{18}O_{V\text{-}SMOW}$ (‰)**	**Result ^{18}O (atom %)**
IA R055	109,00	0,22188
"	108,74	0,22183
"	108,74	0,22183
mean=	108,83	0,22185
1sd=	**0,15**	**0,00003**
accepted=	108,63	0,22181
n	3	3

3.1.4 Difference between conventional heating and microwave irradiation processes

The enrichment ratios corresponding to two individual effects being microwave irradiation and conventional heating are obtained to be % 10.4911 and % 9.7565, respectively. At first view, the influence of microwave irradiation is considerably poor to be about % 0.7347, but this is an important result; because, this is a first experimental observation of microwave effect in this field on the stable isotope enrichment technology. In the literature, the enrichment methods attempted by the use of electromagnetic radiations are only related to some laser and UV irradiations (Olivares and Roja, 2002; Letokhov and Moore, 1976; Mahata and Bhattacharya, 2007). Table 3.1.3 summarizes the comparable ^{18}O measurements corresponding to the identical evaporation operations.

Table 3.1.3 Comparison of the ^{18}O enrichment ratios corresponding to microwave irradiation and conventional heating processes.

Water Samples	**^{18}O Enrichment Ratio (Measured to be Atom %) (Microwave irradiation)**	**^{18}O Enrichment Ratio (Measured to be Atom %) (Conventional heating)**
20,000 mL ⇩	0.19874 ± 0.00003	0.19874 ± 0.00003
1,250 mL ⇩	0.20552 ± 0.00003	0.20502 ± 0.00003
80 mL ⇩	0.21234 ± 0.00003	0.21144 ± 0.00003
5 mL (240 mL➔ 15 mL)	0.21959 ± 0.00003	0.21813 ± 0.00003

Briefly, as it is seen from these results, the evaporation of 20,000 mL water up to 5 mL either by microwave irradiation and conventional heating may result ^{18}O enrichment step by step. Also, the enrichment factors which mean the rates of final and initial ^{18}O enrichment ratios as atom percent, are determined to be 1.1049 and 1.0976, for microwave irradiation and conventional heating, respectively. These factors also exhibit the effectiveness of microwave irradiation on ^{18}O enrichment in water.

3.1.5 Evaluation of the Role of Microwaves on the Enrichment of ^{18}O Ratio in Water

Microwave irradiation is a means of rapidly introducing energy into a chemical system in a manner different from the traditional methods of thermal heating (Loupy, 2006).

According to the report given by Kappe et al., (2009), to the thermal - kinetic effects, microwave effects that are caused by the uniqueness of the microwave dielectric heating mechanisms must also be considered:

"These effects should be termed "specific microwave effects" and shall be defined as accelerations of chemical transformations in microwave field that cannot be achieved or duplicated by conventional heating; but, essentially these are still thermal effects. The question of the boiling point of a liquid undergoing microwave irradiation is one of the basic problems facing microwave heating. Several groups have established that the enthalpy of vaporization is the same under both microwave and conventional heating. These studies have also shown that the rate of evaporation, as well as the temperature of both vapour and liquid at the interface, depends strongly on the experimental conditions. It was established that microwave - heated liquids boil at temperature above the equilibrium boiling point at atmospheric pressure. For several solvents, the superheating temperature can be up to 40 °C above the classical boiling point. "

Superheating effect of microwaves may cause higher ^{18}O isotope enrichment in water than conventional heating. By the superheating effect boiling starts above 100 °C and highly heated water molecules that energized and water vapors may give an isotopic exchange reaction more rapidly and those exchange reactions may cause higher enrichment in ^{18}O in water.

$$H_2{}^{16}O_{(l)} + H_2{}^{18}O_{(g)} \leftrightarrow H_2{}^{18}O_{(l)} + H_2{}^{16}O_{(g)}$$

3.2 Statistical Evaluation of Enrichment Steps

Figure 3.2 represents the increase of the ^{18}O enrichment ratios as a function of initial water volume. Starting from this enrichment function, it can be concluded that the required water volume, which to be initiated for obtaining such an enrichment ratio; for example, to be about 60 % / L or 98 % / mL, may be determined. But unfortunately, the statistical evaluation of these results, which are used to form the enrichment function given in Figure 3.2 indicates enormous water volumes for being initiated for considerably high enrichment ratios. For practical applications, of course, this is not an encouraging result, but in any case it is well known that isotopic enrichment processes are generally necessiated high

number successive repetitions of applied unit operations (Van Hook et al., 1991; Kihara and Kambe, 2011).

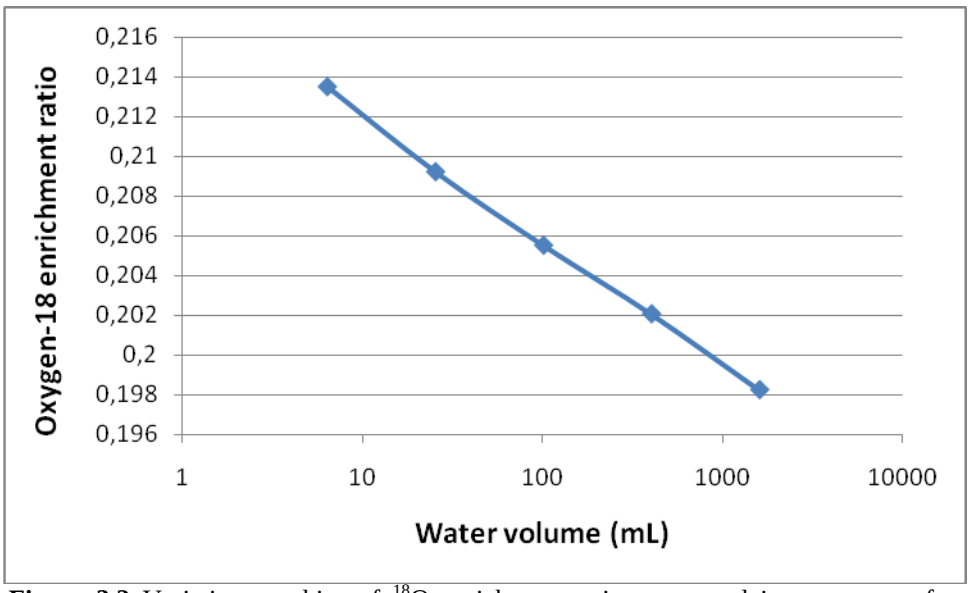


Figure 3.2 Variation graphics of ^{18}O enrichment ratios measured in water as a function of starting water volume.

According to the experimental results obtained in this study as the measured ^{18}O enrichment ratios, the following mathematical relations can be suggested for enrichment steps of water by the use of microwave irradiations for evaporation operations:

$$X_1 \qquad\qquad k_1 = 0.1982300$$

$$X_2 = X_1 / 250 \qquad k_2 = 1.0702214992\ k_1$$

$$X_3 = X_2 / 250 \qquad k_3 = 1.0702214992\ k_2$$

$$X_4 = X_3 / 250 \qquad k_4 = 1.0702214992\ k_3$$

$$\downarrow \qquad\qquad\qquad \downarrow$$

$$1\ mL = X_n = X_{n-1} / 250 \qquad k_n = 1.0702214992\ k_{n-1} = 100.0000000$$

$$1\ mL = X_{n-1} / 250 \qquad 1.0702214992\ k_{n-1} = 100.0000000$$

$$1\ mL = X_n \qquad\qquad k_n = 100.0000000$$

Where k_i represents the ^{18}O enrichment ratios in which k_1 and k_n represent the ^{18}O enrichment ratio of natural water and highest enrichment ratio being theoretically 100 %, respectively. Starting from these statistics following equations are derived:

$$k_n = k_1(1.0702214992)^{n-1}$$

$$1.0702214992^{n-1} = k_n / k_1$$

$$X_1 = X_n (250)^{n-1}$$

$$250^{n-1} = X_1 / X_n$$

$$\downarrow$$

$$(233.204137^{n-1}) (1.0702214992^{n-1}) = X_1 / X_n$$

$$(233.204137^{n-1}) (k_n / k_1) = X_1 / X_n$$

Where, k_1 represents measured ^{18}O enrichment of starting water, k_n represents ^{18}O ratio after "n" enrichment operation step, X_1 is starting volume of water, and X_n is the volume of water remained after "n" enrichment operation step.

3.2.1 Mathematical formulation of enrichment steps

Microsoft Excel 2003 (Version 11) computational program gives the following equation as a mathematical function of the experimental variation steps of ^{18}O enrichment ratio measurements given in Figure 3.2.

$$\mathbf{y = 0.2186\ x^{-0.0133}}$$

Required water volumes should be calculated using this mathematical equation for a given ^{18}O enrichment level in water; but unfortunately, the results obtained in this way are not reasonable and encouraging at this experimental step.

3.3 Evaluation of the Results Obtained as to the Commercial Application Potential

3.3.1 Monetarily evaluation of enrichment method applied in this study

According to the today's marketing price, the cost of ^{18}O enriched water per mL varies depending on the original country, commercial image of the company, techniques applied for enrichment procedures, etc. Some prices have been shown and compared in Table 3.3.1. As it seen from this table, 1 mL of ^{18}O enriched water per mL with the same enrichment level may vary from 30 $ to 150 $. Of

course, this does not reflect a logical economy, because the highest cost is about 5 folder of the lowest cost. This means that the global market is not yet established and saturated for ^{18}O enriched water. Today, this complicates the monetarily evaluation of a newly applicable technique for production of ^{18}O enriched water which will be competed in the global market. Nevertheless, it seems very difficult already now to make an economical statement, when the microwave irradiation technique will be able to be used in the future for commercial production of ^{18}O enriched water, will be economical or not or compatible with other competitor companies.

Table 3.3.1 Some prices for ^{18}O enriched water in the international market.

^{18}O- Water Suppliers	**Price**
A company from USA, % 97 ^{18}O water, 1 g	191 €
A company from China, % 97 ^{18}O water, 1g	35 USD
A company from USA, % 98 ^{18}O water, 1g	150 €

3.3.2 Comparison of this technique with that other classical techniques applying

Kim et al., (2009) reported following statements on the fractional distillation technique which is commonly used for ^{18}O enrichment of water:

"Distillation, which uses the different boiling points (same as the equilibrium vapour pressure) of the given materials, is known as the most effective process for separation of the light isotopes such as Hydrogen, Carbon, and Oxygen, etc. Membrane distillation uses the equilibrium vapour pressure of the materials as well as their differentiated pore diffusion characteristics, while fractional distillation and cold distillation use only the vapour pressure difference of the materials. Cold distillation for separation of the Oxygen stable isotopes uses the equilibrium vapor pressure differences between isotopic NO (Nitric Oxide) whose boiling point is very low. It is advantageous, because its stage separation factor is relatively high (separation factor for ^{18}O, $\alpha \approx 1.03$) and also, it can produce the other isotopes, ^{14}N and ^{15}N, as byproducts. However, the separation system should be built by the resistive materials due to corrosive nature of NO and it should be handled carefully due to its toxicity. It is also required to use thermal insulation to operate the system long period since the process requires the cold temperature as low as 77 K. More importantly, it is not economic, because the additional material conversion system is necessary to transfer the initial material nitric acid to water which is used in the cyclotron as a target. Fractional distillation of water, however, is more efficient than cold distillation, because capital and operation costs are lower than cold distillation since no heavy thermal insulation is necessary due to its relatively high process temperature (330 K). In addition, the product of the process is directly used in a cyclotron to produce the radioisotope ^{18}F. Although, fractional distillation has many advantageous merits

compared to cold distillation, it is still an expensive process due to its huge distillation towers and long equilibrium time caused by relatively low separation factor (α ≈ 1.0037). ”

The aim of this study was the investigation of enrichment possibility of natural ^{18}O isotope ratio in water using microwave irradiation technique and the investigation of its commercially application potential. For this reason, the evaluation of advantages and disadvantages of the processes applied in this study has great importance. As it is clearly observed that ^{18}O enrichment ratio measurements of samples collected at the ends of evaporation operations, microwave irradiations have resulted higher ^{18}O enrichment ratios in water respecting to the corresponding ratios when conventional heating was used. Starting from this definitive result it may be concluded that because of “Superheating effect” light water molecules which means water molecules having only ^{16}O isotopes can gain higher energies than that of higher water molecules having ^{18}O isotope. Consequently, microwave irradiations have resulted higher ^{18}O enrichment ratios in water.

In this study enrichment cascades were not designed as industry style enrichment processes. In fractional distillation method or other isotope enrichment methods used for ^{18}O enrichment in industry, enriched water goes forward to next stage and depleted water goes back to the previous stage so there is no need to feed the system again and again. But in this study, depleted portion was faded as water vapor and was not used in previous stages. This caused highly consumption of water and this was not economical.

3.4 Evaporation Rates of some Ionic Aqueous Solutions under Microwave Irradiation

3.4.1 Variation of evaporation rates versus molar concentrations of ionic solutions

According to another study which was recently realized at our laboratory, the evaporation rates of aqueous solution of different chemical salts at different concentrations under microwave irradiations were determined and very interesting results were observed (Ünak et al., 2011). According to this study, the evaporation rates aqueous solution of different chemical salts at different concentrations under microwave irradiations were observed to be considerably variable depending on the ionic or non-ionic characters of salts and on the number of ions solute in water. In this context, the highest evaporation rates were observed for 0.035 M

NaCl and $K_3[Fe(CN)_6]$ aqueous solutions. This means that in case of microwave irradiation is carried out using the aqueous solution of these chemical salts may be the same ^{18}O enrichment evaporation operations will be able realized in shorter time and this will result the decrease of evaporation costs which was calculated in the recent sections. Starting from this consideration, 1,250 mL 0.035 M NaCl and $K_3[Fe(CN)_6]$ aqueous solutions were separately evaporated up to 80 mL under the identical conditions of water evaporation operations which were applied for microwave irradiations of water. Finally, the evaporation durations were observed to be shorter than that of corresponding water evaporation operations as given in the Table 3.4.1.

Table 3.4.1 Evaporation durations of naturel water and aqueous solutions corresponding to the evaporation of 1,250 mL volumes up to 80 mL.

Evaporation Durations		
Naturel water	**0.035 M NaCl**	**0.035 M $K_3[Fe(CN)_6]$**
170 min	160 min	150 min

The evaporated aqueous solutions of these chemical salts were distilled up to dryness and all distilled water was collected. The samples taken from distilled water were used for ^{18}O measurements to test, if any effect is exist on the ^{18}O enrichment ratio of water.

. 3.4.1.1 Results obtained with NaCl solution

Table 3.4.1.1 compares the ^{18}O enrichment ratios of water and 0.035 M NaCl solution.

Table 3.4.1.1 Comparison of the ^{18}O enrichment ratios of natural water and 0.035 M NaCl solution.

Samples Description	**^{18}O Enrichment Ratio (Measured to be Atom %) (Microwave irradiation)**
Natural water	0.19874 ± 0.00003
0.035 M aqueous solution of NaCl	0.20638 ± 0.00003

3.4.1.2 Results obtained with $K_3[Fe(CN)_6]$ solution

Table 3.4.1.2. compares the ^{18}O enrichment ratios of water and 0.035 M $K_3[Fe(CN)_6]$ solution.

Table 3.4.1.2 Comparison of the ^{18}O enrichment ratios of natural water and 0.035 M $K_3[Fe(CN)_6]$ solution.

Samples	**^{18}O Enrichment Ratio (Measured to be Atom %) (Microwave irradiation)**
Natural water	0.19874 ± 0.00003
0.035 M aqueous solution of $K_3[Fe(CN)_6]$	0.20765 ± 0.00003

3.5 Comparison of ^{18}O Enrichment Parameters of Pure Water and Ionic Solutions

3.5.1 Influence of dissolved ions on ^{18}O enrichment of water using microwave irradiation

As it was notified in recent paragraphs, 1,250 mL water with natural ^{18}O ratio of about 0.19874 was enriched to ^{18}O ratio of about 0.20552 (average values of 3 different measurements which are corresponding to same evaporation factors) when it was evaporated to 80 mL. This means 3.4115 % enrichment in ^{18}O was occurred after these evaporation operations. As it is seen in Tables 3.4.1.1 and 3.4.1.2, measured ^{18}O enrichment ratios for ionic solutions of NaCl and $K_3[Fe(CN)_6]$ are 0.20638 % and 0.20765 %, respectively. Comparison of these results is given in 3.5.1. As it is clearly seen from this comparison, ionic species found in water leads the increase of ^{18}O enrichment ratio in water. As it was emphasized in recent paragraphs, microwave irradiations cause a strong "dielectric heating" in ionic solutions and during the evaporation, water molecules that contain ^{18}O isotope may more easily get free from intermolecular attraction by superheating effect and leave the water surface. The results given about for ionic solutions are corresponding well with this consideration related to microwave absorption processes.

Table 3.5.1 Comparison of the ionic effect on the ^{18}O enrichment ratios of water.

Evaporation with a factor of about 16	**Pure Water**	**0.035 M NaCl Solution**	**0.035 M $K_3[Fe(CN)_6]$ Solution**
1,250 mL ⇩	0.19874 ± 0.00003	0.19874 ± 0.00003	0.19874 ± 0.00003
80 mL	0.20552 ± 0.00003	0.20638 ± 0.00003	0.20765 ± 0.00003
Additional enrichment ratio	3.4115 % **0.0000 %**	3.8442 % **0.4327 %**	4.4832 % **1.0717 %**

In addition, 0.035 M NaCl solution contains 0.070 mol (0.035 mol x 2) ion per Litter, but 0.035 M $K_3[Fe(CN)_6]$ solution contains 0.14 mol (0.035 mol x 4) ion per Litter. Higher ion concentration in $K_3[Fe(CN)_6]$ solution induces more powerful superheating effect during the irradiation. This causes higher ^{18}O enrichment in $K_3[Fe(CN)_6]$ solution than NaCl solution which has the same Molarity as it is seen in Table 3.5.1.

3.5.2 Economical evaluation of ^{18}O enrichment process of water using dissolved ions

The studies showed that irradiation of ionic solutions may be used as an alternative technique for ^{18}O enrichment in water. The method provides higher ^{18}O enrichment per unit evaporation time. Disadvantage of the method is the requirement of complete distillation after enrichment operations in order to gain pure water. Separation factor is higher in this method; so, ^{18}O enrichment time is shorter and this may result more economical applications.

4. CONCLUSIONS

The experimental results obtained in this study have clearly exhibited the positive effect of microwave irradiations on ^{18}O enrichment ratio of water. 2.45 GHz frequency microwaves were used and this frequency seems to be ideal for evaporation of water; in spite of this, the increasing of intensity of microwaves may be to result much more effective and quicker enrichment processes. It is important to conclude that at the following steps of these kinds of investigations, different microwaves with longer or shorter wavelength should be examined. The design of evaporation cascades may also improve the ^{18}O enrichment process of water.

On the other hand, starting from the results obtained with ionic solutions, it may be concluded that the use of ionic solutions instead of pure water will be much more advantageous for cheaper and quicker ^{18}O enrichment process of water using microwave irradiations.

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