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Available Online at www.achieverssciencejournal.org**Investigation of Micro, Macro and Heavy Elements in Some Selected Fruits and Vegetable Seeds to Assess Usefulness as Food Supplement**M. B. Okunade¹, D. A. Oyegoke^{1*}; O. D. Owolala¹ and S. M. Ajayi¹¹Department of Chemical Sciences, Achievers University Owo, Ondo State, Nigeria*E-mail address: mbokunade47@gmail.com/davidadeniran@gmail.com

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ABSTRACT

Ten different seed samples were obtained from Owo and Akure markets in Ondo State of Nigeria. They were dried in the oven between a temperature of 100 °C- 105 °C, then pulverized into powder and analysed for (20) twenty elements, of K , Ca, S, Si , P, Fe , Al , W, Mn ,Cu, Mg, Ni, Co, Pb, As, Ti, V, Cr, Rb and Ag using ED-X-ray Fluorescence (XRF) spectrophotometer. The concentration of elements listed above followed a decreasing order of K >Ca> S > Si > P> Fe > Al > W>Mn>Cu > Mg > Ni > Co >Pb> As > Ti > V > Cr >Rb> Ag. Five (5) macro elements, K, Ca, Mg, P, and S were detected in all the seeds samples at varying concentrations, also five (5) micro elements Zn, Cu, Fe, Si and Mn were detected in all the seeds samples also at varying concentrations. And eight (8) heavy elements, Cr, Co, Ni, As, Ag, Au, W and Pb were present in some of the seed samples. The following seeds were rich in Potassium (K), *P. dulcis*, *S. cannum*, *C. lanatus*, *C. papaya*, *C. manni* and *C. limon*. Phosphorus, (P) was present in concentration ranged between 0.2-1.6% and the following seeds:*C.lanatus*, *C.manni*, *C. olitorius* and *P. dulcis*were rich in phosphorus. Calcium (Ca) was detected between 0.1- 15.0% and the following seeds are rich in calcium: *C. olitorius*, *C. Lanatus*, *C. reticulata* and *P. dulcis*. Magnesium, (Mg) concentrations ranged between 0.2-0.4% and *P. dulcis*, *C. Lanatus*, *C. manni* and *C. olitorius*were rich in magnesium. Sulphur (S), concentrations ranged between 1.72-4.5% and *C. lanatus*, *C. sativus*, *C. olitorus*, *C. papaya*, *p. dulcis*, and *C. manni*were rich in sulphur. Micro-elements such as Fe (0.08-1.17%) were present in *P. dulcis*, *P. lanatus*, *C. manni*, *C. olitorus*, *C. limon*, *S. canum*, *T. occidentalis* and *C. reticulate seeds*. Zinc (Zn) was present in the range between 0.09-0.26% in seeds of *P. dulcis*, *P. lanatus*, *C. manni*, *C. olitorus*, *C. limon* and *S. canum*. For heavy metals, Lead (Pb) was detected in *C. papaya* with a concentration of 0.056% while Lead was present in other seed samples at very low concentration, 0.001-0.004%.

KEYWORDS: Heavy Metals; Micro-Elements; Macro-Elements; Percentage Concentrations**1. Introduction**

A variety of fruits and vegetables are consumed in Nigeria on a daily basis and they form an integral part of our diet. However, most times only the fleshy pulps of these fruits are consumed leaving the seed and the rind. Fruits and vegetables contain a high percentage of water averaging 85%, fats and protein in very small varying amounts, a fair proportion of carbohydrate present as cellulose, starch in small quantity and sugar (Shiundu 2004). Fruits and vegetables are also known for their high

micronutrient concentrations, including carotene or vitamin A, Vit K, Vit. C (ascorbic acid), riboflavin, iron, iodine and other mineral elements. (Shiundu 2004).

It is commonly acknowledged that the fruit processing sectors usually generate large amount of seeds as wastes. In the past, investigations were undertaken to explore their potential uses in food applications due to their high nutritional value (Okolie et al, 2012). For instance, papaya seeds have been used for decades as vermin-fungal agent as

well as a spicy flavoring substance. Also because of its abortive properties, the seeds were used to facilitate a good menstrual flow (Malacrida et al, 2011). Utilization of the seeds concurrently helps to deal with waste disposal problems which cause environmental pollution. For this reason, many fruit seeds have been evaluated for presence of micronutrients (Parry et al. 2008; Mabaleha et al, 2007), hence the main objective of this project.

Malnutrition is a major public health problem in most parts of the world. Many seeds contain micronutrients and macronutrients which can be used to prevent malnutrition and improve health status of humans generally but because deeper investigations have not been carried out on many seeds, hence their health benefits have not been confirmed, thus the importance of this project.

The micro and macro nutrients are dietary components, often referred to as vitamins and minerals, which although only required by the body in small amounts and are vital to development, disease prevention, and wellbeing of humans (IOM, 1997). Despite that it is required in trace amounts and micronutrient deficiencies are widespread, affecting approximately two billion people worldwide, the equivalent of a third of the

worlds population. At least half of children worldwide ages 6 months to 5 years suffer from one or more micronutrient deficiency, and globally more than 2 billion people are affected (IOM, 1997). The importance of this project is to investigate some micronutrients, macronutrients and heavy metals which are present in the selected seeds from Owo and Akure towns in Ondo State. The results of which can be used to arrest the local malnutrition problem, on further investigation.

2. Materials and methods

2.1 Materials

The seed samples were obtained from Owo (Oja-Ikoko market) in Owo Local government and Oja-Oba market in Akure. The samples were taken to Department of Biological Sciences, Achievers University for identification.

2.2 Methods

Each seed sample was washed with distilled water and with de-ionized water to remove any impurity, followed by drying in the oven at temperature of between 100– 105⁰C for 24hours to remove moisture.

Table 1: Samples and location of collection

S/N	Sample local name	Sample Botanical name	Point of collection	Location	State
1	Almond	<i>Prunus dulcis</i>	Market	Akure	Ondo
2	Lemon	<i>Citrus limon</i>	Market	Akure	Ondo
3	Tangerine	<i>Citrus reticulata</i>	Market	Owo	Ondo
4	Ewedu	<i>Corchorus solitorius</i>	Market	Owo	Ondo
5	Watermelon	<i>Citrullus lanatus</i>	Market	Akure	Ondo
6	Cucumber	<i>Cucumis sativus</i>	Market	Akure	Ondo
7	White melon	<i>Cucumeropsis mannii</i>	Market	Akure	Ondo
8	Garden egg	<i>Solanum incanum</i>	Market	Akure	Ondo
9	Pawpaw	<i>Carica papaya</i>	Market	Owo	Ondo
10	Ugwu	<i>Telfairia occidentalis</i>	Market	Owo	Ondo

The dried samples were pulverised using agate pestle and mortar, followed by sieving through 0.5mm mesh size sieve to obtain uniform particle size. Each sample was labelled and stored in a dry plastic container that had been pre-cleaned with concentrated Nitric acid to prevent heavy metal

contamination prior to analysis with X-ray fluorescence (XRF) spectrophotometer

3. Result and Discussions

In this study, the concentrations of eighteen elements were determined in each seed sample by

using XRF spectroscopy. The results showed various concentrations of five macro elements: K, Ca, Mg, P and S, five microelements: Zn, Cu, Fe, Si and Mn and eight heavy elements: Cr, Co, Ni, As, Ag, Au, W and Pb.

The concentrations (%) of Potassium (K), Calcium (Ca), Phosphorous (P), Sulphur (S), and Magnesium (Mg), were obtained at higher percentages as shown in the tables. They are all macro elements but Zinc (Zn), Copper (Cu), Iron (Fe), Silicon (Si) and Manganese (Mn) were detected at low concentrations as micro- elements. The heavy metals Lead (Pb), Chromium (Cr), Cobalt (Co), Silver (Ag), Gold (Au), were not detected in many seed samples as confirmed in Table of results.

Potassium – K

The percentage of potassium in the seed samples ranged between 3.0% - 15%, with *C.sativus* having the lowest percentage concentration of 3.0% and *S.canum* having the highest percentage of 14.96%. The percentage of potassium in the other seed samples were; *C. papaya* with 3.005%, *C.olitorius* with 3.59%, *C.mannii* with 3.6%, *C.limon* with 7.24%, citrus reticulate with 7.26%, *T.occidentalis* with 7.5%, *P.dulcis* with 8.51%, *C.lanatus* with 9.5%. Potassium (K) plays an important role in the body physiology and it is good for muscle movement. Its main role in the human body is to help maintain normal levels of fluids inside the body cell. (Vollestadet al., 1994) It is widely available in fruits and vegetables.

Calcium – Ca

The percentage of calcium in the seed samples ranged from 0.1% - 15%, with *T. occidentalis* having the lowest percentage of 0.13% and *C.olitorius* having the highest percentage of 14.94%. The percentages of calcium in the other seed samples were; *C.sativus* with 0.21%, *C. papayawith* 0.218%, *C.lanatus* with 2.51%, *C. reticulata* with 2.57%, *S. canum* with 0.28%, *C. manni* with 0.29%, *C. limon* with 3.95%, *P.dulcis* with 10.29%. It is used in the development and maintenance of bone structure. It functions in the clotting process, nerve transmission, hormone function and metabolism of vitamin D. (Gharibzahedi and Jafari, 2017).

Phosphorous – P

The percentage of phosphorous in the seed samples ranged from 0.2% - 1.6%, with *C.sativus* having the lowest percentage of 0.23% and *C.lanatus* having the highest percentage of 1.6%. The percentage of phosphorous in the other seed samples were; *C. papaya* with 0.67%, *Citrus reticulata* with 0.68%, *S.canum* with 0.71%, *C.limonwith* 0.73%, *T.occidentalis* with 0.77%, *C.mannii* with 1.05%, *C.olitorius* with 1.29%, *P.dulcis* with 1.45%. It plays a part in almost every chemical reaction within the body because it is present in every cell. It forms calcium phosphate with calcium in the bones & teeth in a 2-1 ratio. It is important in the utilization of carbohydrates, fats, and proteins for the growth and maintenance in the body. (IOM, 1997).

Silicon –Si

The percentage of silicon in the seed samples ranged from 0.1% - 1.8%, with *C.sativus* having the lowest percentage of 0.15% and *P.dulcis* having the highest percentage of 1.75%. The percentage of silicon in the other seed samples were; *C. papaya* with 0.23%, *C.reticulata* with 0.24%, *C.mannii* with 0.28%, *T.occidentaliswith* 0.32%, *C.limon* with 0.39%, *C.olitorius* with 0.56%, *S.canum* with 0.58%, *C.lanatus* with 0.79%. Silicon is necessary for the synthesis of collagen and elastin, it is also important for the health of connective tissues, bones, cartilages, tendons and joints.

Iron – Fe

The percentage of iron in the seed samples ranged from 0.08% - 1.2 %, with *C.papaya* having the lowest percentage of 0.08% and *P.dulcis* having the highest percentage of 1.16%. The percentage of iron in the other seed samples were; *C.reticulata* with 0.20%, *T.occientalis* with 0.21%, *C.mannii* with 0.31%, *C.olitorius* with 0.24%, *C.lanatus* with 0.25%, *S.canum* with 0.29%, *C. limon* with 0.32%, *C. sativus* with 0.36%. Iron is the most relevant ingredient of red dye of blood (hemoglobin), also essential in process of erythrocyte formation in bone marrow, iron bonds carbon dioxide in hemoglobin and transports it to the lungs, where it is removed. This element is also an ingredient of many enzymes and proteins participating in organism metabolism. It

takes part in DNA synthesis, is essential to correct build of skin, hair, nails, to appropriate functioning of immunity system. (Al-Fartusie and Mohssan, 2017).

Cobalt – Co

The percentage of cobalt in the seed samples ranged from 0% - 0.09%, with *C.olitorius*, *S.canum*, *C.limon*, *C.lanatus* having the lowest percentage of 0% i.e cobalt is detected in the seed samples but in very low concentration and *C. papaya* having the highest percentage of 0.09%. The percentage of cobalt in the other seed samples were; *C.reticulata* and *T.occidentalis* with 0.0002%, *C.mannii* with 0.002%, *C.sativus* with 0.005%, *P.dulcis* with 0.01%. Cobalt is an integral part of vitamin B12 and therefore essential for the functioning of the cell. It is involved in the production of antibacterial and antiviral compounds that prevent infections. (Al-Fartusie and Mohssan, 2017).

Aluminum – Al

The percentage of Aluminium in the seed samples ranged from 0.3% - 1.2%, with *C. papaya* having the lowest percentage of 0.35% and *P.dulcis* having the highest percentage of 1.12%. The percentage of Aluminium in the other seed samples were; *C.sativus* with 0.36%, *C.mannii* with 0.45%, *T.occidentalis* with 0.51%, *C.reticulata* with 0.54%, *C.limon* with 0.60%, *C.olitorius* with 0.65%, *C.lanatus* with 0.90%, *S.canum* with 0.98%. It is a metal that finds its way into food chains through anthropogenic activities.

Magnesium – Mg

The percentage of magnesium in the seed samples ranged from 0% - 0.40%, with *C.limon* with the lowest percentage of 0% i.e Magnesium was detected in the seed sample but in a very low concentration and *C.reticulata* having the highest percentage of 0.40%. The percentage of Magnesium in the other seed samples were; *C.olitorius* with 0.027%, *T.occidentalis* with 0.05%, *C.mannii* with 0.06%, *P.dulcis* with 0.28%, *C. papaya* with 0.29%, *S.canum* with 0.35%, *C.sativus* with 0.35% *C.lanatus* with 0.10%. In the body, Magnesium supply is located in the bones together with calcium and

phosphorus, while it is found in cellular fluids and some soft tissue. It is involved with energy production of glucose, protein and nucleic acid synthesis, the formation of urea, muscle impulse transmission and neurotransmission etc. (IOM, FNB, 1997)

Nickel – Ni

The percentage of Nickel in the seed samples ranged from 0.03% - 0.2%, with *C.lanatus* having the lowest percentage of 0.039% and *C. papaya* having the highest percentage of 0.198%. The percentage of Nickel in the other seed samples were; *S.canum* with 0.057%, *C.limon* with 0.063%, *P.dulcis* with 0.073%, *C.olitorius* with 0.074%, *C.reticulata* with 0.085%, *T.occidentalis* with 0.086%, *C.sativus* with 0.09%, *C.mannii* with 0.093%. It is a micronutrient element essential for proper functioning of the human body, as it increases hormonal activity and is involved in lipid metabolism. (Al-Fartusie and Mohssan, 2017).

Sulphur – S

The percentage of Sulphur in the seed samples ranged from 1.4% - 4.5%, with *T.occidentalis* having the lowest percentage of 1.43% and *C.lanatus* having the highest percentage of 4.5%. The percentage of Sulphur in the other seed samples were; *C.sativus* with 1.65%, *C.olitorius* with 1.71%, *C. papaya* with 1.74%, *C.reticulata* with 1.83%, *C.mannii* with 2.13%, *C.limon* with 2.23%, *S.canum* with 2.7%, *P.dulcis* with 3.38%. Sulphur is the eighth most abundant element in the human body and it plays an important role in the synthesis of certain essential amino acids. (Parcell, 2002)

Manganese – Mn

The percentage of Manganese in the seed samples ranged from 0% - 0.4%, with *C.reticulata* having the lowest percentage of 0%. Manganese was detected in the seed sample but at very low percentage. *C. papaya* having the highest percentage of 0.36%. The percentage of Manganese in the other seed samples were *C.limon* with 0.001%, *C.lanatus* and *S.canum* with 0.004%, *C.olitorius* with 0.008%, *T.occidentalis* with 0.0082%, *P.dulcis* with 0.01%, *C.sativus* with 0.015%, *C.mannii* with 0.02%. It

participates in build of enzymes metabolizing glucose and fatty acids, is a structural element of bones and skin. Manganese is very important in reproduction and proper functioning of central nervous system. (Merrell, 2016).

Zinc – Zn

The percentage of Zinc in the seed samples ranged from 0% - 0.3%, with *C. papaya* having the lowest percentage of 0%.and *P.dulcis* having the highest percentage of 0.26%. The percentage of Zinc in the other seed samples were; *C.sativus* with 0.09%, *S.canum* with 0.11%, *C.olitorius* with 0.12%, *C.reticulata* with 0.13%, *C.lanatus* with 0.14%, *T.occidentalis* with 0.14%, *C.limon* with 0.14%, *C.mannii* with 0.18%. Zinc is essential for the synthesis of DNA and RNA, proteins, insulin and sperm, essential for proper functioning of immunity system and for activation of over 80 enzymes. It takes part in metabolism of carbohydrates, fats, proteins and alcohol. It is necessary in protective process against free radicals, of taste and smell feeling, has influence on the appearance of hair and nails. (Mayo et al, 2014)

Tungsten – W

The percentage of Tungsten in the seed samples ranged from 0.09% - 0.45%, with *S.canum* having the lowest percentage of 0.09% and *P.dulcis* having the highest percentage of 0.45%. The percentage of Tungsten in the seed samples were; *C.lanatus* with 0.12%, *C.limon* with 0.14%, *C.reticulata* with 0.18%, *C.olitorius* with 0.18%, *T.occidentalis* with 0.19%, *C.mannii* with 0.21%, *C.sativus* with 0.26%, *C. papaya* with 0.28%. Tungsten leaves the human body rapidly as soon as it enters through the urine.

Copper – Cu

The percentage of copper in the seed samples ranged from 0.04 - 0.36%, with *C.olitorius* and *S.canum* having the lowest percentage of 0.04% and *C.sativus* having the highest percentage of 0.36%. The percentage of Copper in the other seed samples are ; *C.reticulata* with 0.06%, *C.lanatus* with 0.07%, *T.occidentalis* with 0.07%, *C.limon* with 0.08%, *C.mannii* with 0.08%, *P.dulcis* with 0.09%, *C. papaya* with 0.19%. Copper is essential for proper

functioning of organs. It takes part in erythrocyte formation, creation of bones and collagen, correct healing of wounds, absorption and transportation of iron, metabolism of fatty acids and RNA formation. It is an ingredient of some enzymes, fulfills some role in metabolism of central nervous system and dyes. (Uauy et al,1998).

Lead – Pb

The percentage of lead present in the seed samples ranged from 0% - 0.056%, with *C.mannii*, *C.sativus*, *S.canum*, *C.lanatus*, *C.reticulata* having the lowest percentage of 0% and *C. papaya* having the highest percentage of 0.056%. The percentage of Lead in the other seed samples were; *T.occidentalis* with 0.001%, *C.limon* with 0.004%, *P.dulcis* with 0.004%, *C.olitorius* with 0.007%. Lead is a cumulative toxicant that affect the body systems which is distributed to the brain, causing serious damages.

Arsenic – As

From the results obtained, the percentage of Arsenic in the seed samples were at zero concentration i.e 0%. It plays a role in the development of diabetes, cancer, vascular disease and lung disease. (Letizia and Andrea, 2013). It is highly toxic.

Chromium - Cr

From the results, the percentage of chromium in the seed samples was at zero concentration i.e 0%. It is the element regulating level of cholesterol and fatty acids. It participates in making cells sensitive to insulin (flattening of glycemic curve) and protein digestion. (Slawomir et al, 2014). Chromium is a toxic metal.

4. Conclusions and Recommendations

The investigation revealed that some seeds contained micro, macro and heavy elements. Those containing micro and macro elements may be recommended for consumption to supplements the nutrients present in them. While those containing heavy metals are not recommended for consumption at all. Seed containing high concentration of potassium K such as *P. dulcis*, *C. lanatus*, *C. manni*, *C. sativus*, *C. olitorius*, *C. limon*, *S. cannum*. *T. occidentallis*, *C. papaya* on further investigation

may be recommended as potassium supplement in addition to other sources of potassium. Calcium (Ca) Magnesium (Mg), Phosphorus (P) and Sulphur (S) found present in high concentration in *C. olitorius*, *C. lanatus*, *C. reticulate*, *P. dulcis* on further investigation could be recommended as nutrient supplements in addition to other sources already available in food industries. All seeds with heavy metal concentration 0.001-0.056% as investigated in this study are dangerous for consumption either at very low or moderately low concentrations because it is possible to bioaccumulate lead (pb) in human system. The study confirmed that micro elements, macro elements and heavy metals were present in some seeds. Those containing micro and macro element may be recommended as nutrient supplements on further investigation which will confirm their safety levels, while the seeds containing heavy metals are totally unacceptable for consumption because heavy metals are toxic and carcinogenic and bioaccumulate in human.

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Table 1: Table and graphical representation of elemental composition of tangerine seeds

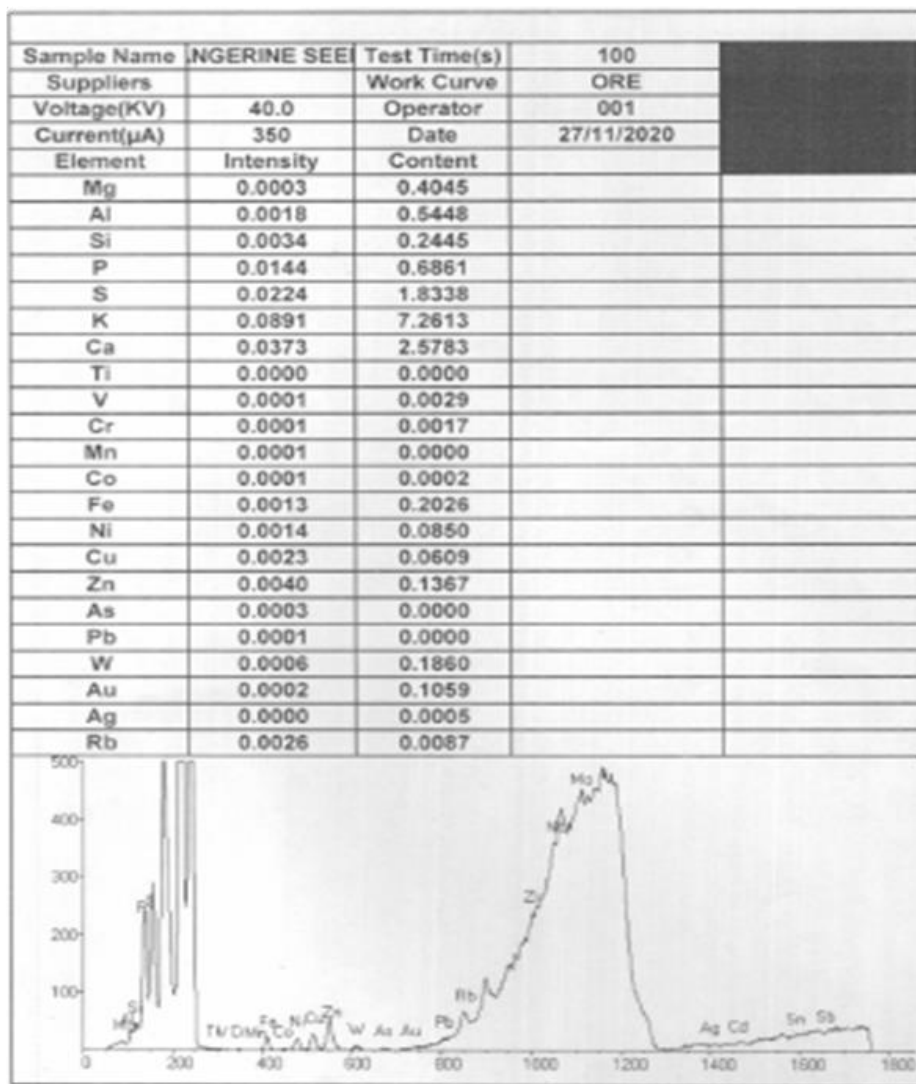


Table 2: Table and graphical representation of elemental composition of water melon seeds

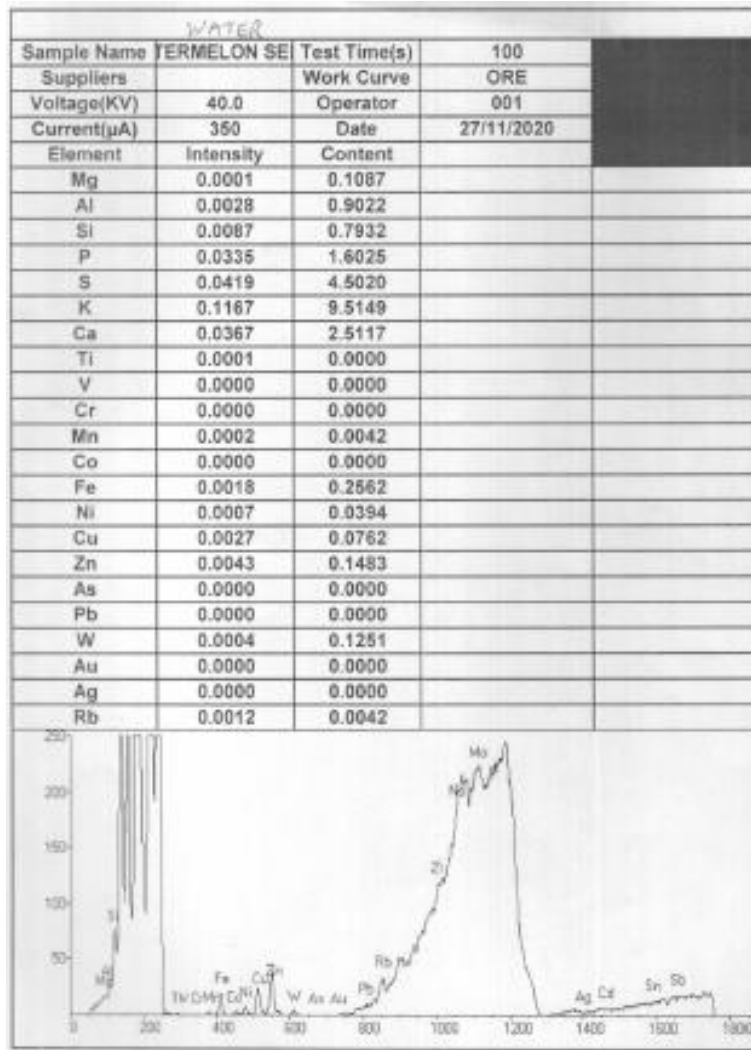


Table 3: Table and graphical representation of elemental composition of melon seeds

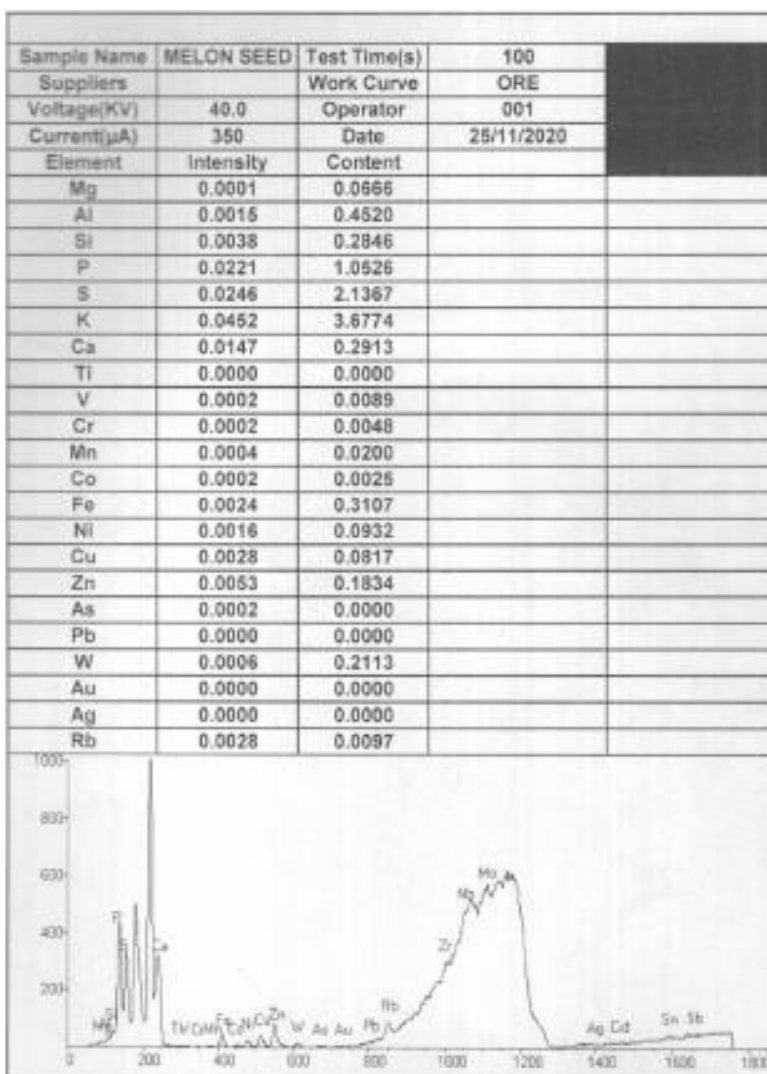


Table 4: Table and graphical representation of elemental composition of lemon seeds

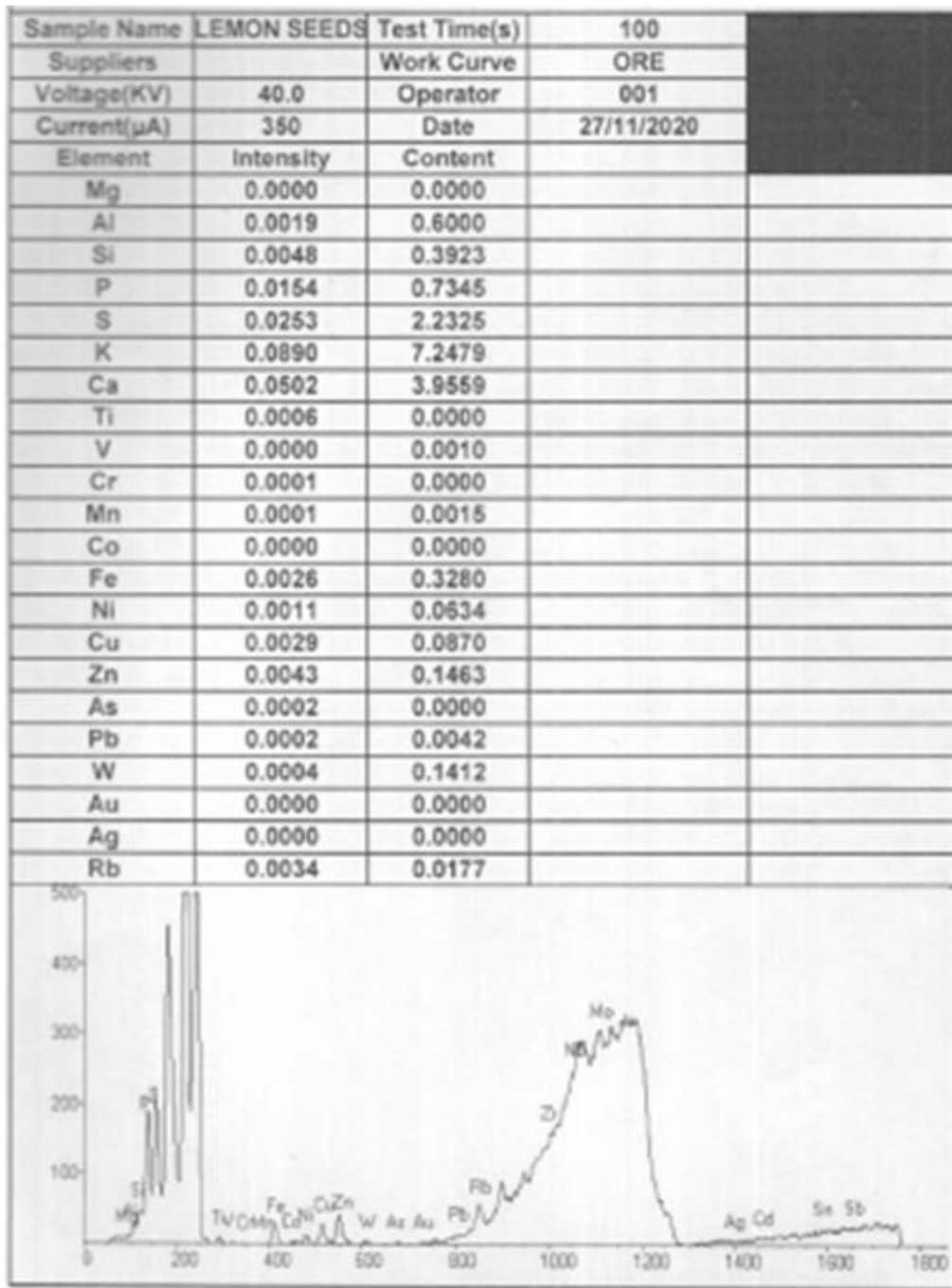


Table 5: Table and graphical representation of elemental composition of garden egg seeds

Sample Name	GARDEN EGG	Test Time(s)	100
Suppliers		Work Curve	ORE
Voltage(KV)	40.0	Operator	001
Current(μA)	350	Date	27/11/2020
Element	Intensity	Content	
Mg	0.0003	0.3541	
Al	0.0031	0.9899	
Si	0.0067	0.5850	
P	0.0150	0.7143	
S	0.0288	2.7027	
K	0.1836	14.9657	
Ca	0.0145	0.2878	
Ti	0.0001	0.0000	
V	0.0001	0.0040	
Cr	0.0001	0.0000	
Mn	0.0002	0.0041	
Co	0.0000	0.0000	
Fe	0.0022	0.2911	
Ni	0.0010	0.0575	
Cu	0.0018	0.0440	
Zn	0.0034	0.1142	
As	0.0001	0.0000	
Pb	0.0001	0.0000	
W	0.0003	0.0918	
Au	0.0000	0.0000	
Ag	0.0000	0.0000	
Rb	0.0054	0.0451	

The figure is an EDS spectrum showing intensity on the y-axis (0 to 500) and energy in keV on the x-axis (0 to 1800). The spectrum shows several sharp peaks at low energy (below 200 keV) corresponding to Mg, Al, Si, P, and S. A very large peak for Ca is visible around 280 keV. A broad peak for Fe is centered around 640 keV. A sharp peak for Zn is at approximately 860 keV. A large peak for Mo is centered around 1100 keV. A small peak for Rb is at approximately 1370 keV. Other elements like Ag, Cd, Sn, and Sb are indicated at higher energy values (around 1400-1800 keV).

Table 6: Table and graphical representation of elemental composition of ewedu seeds

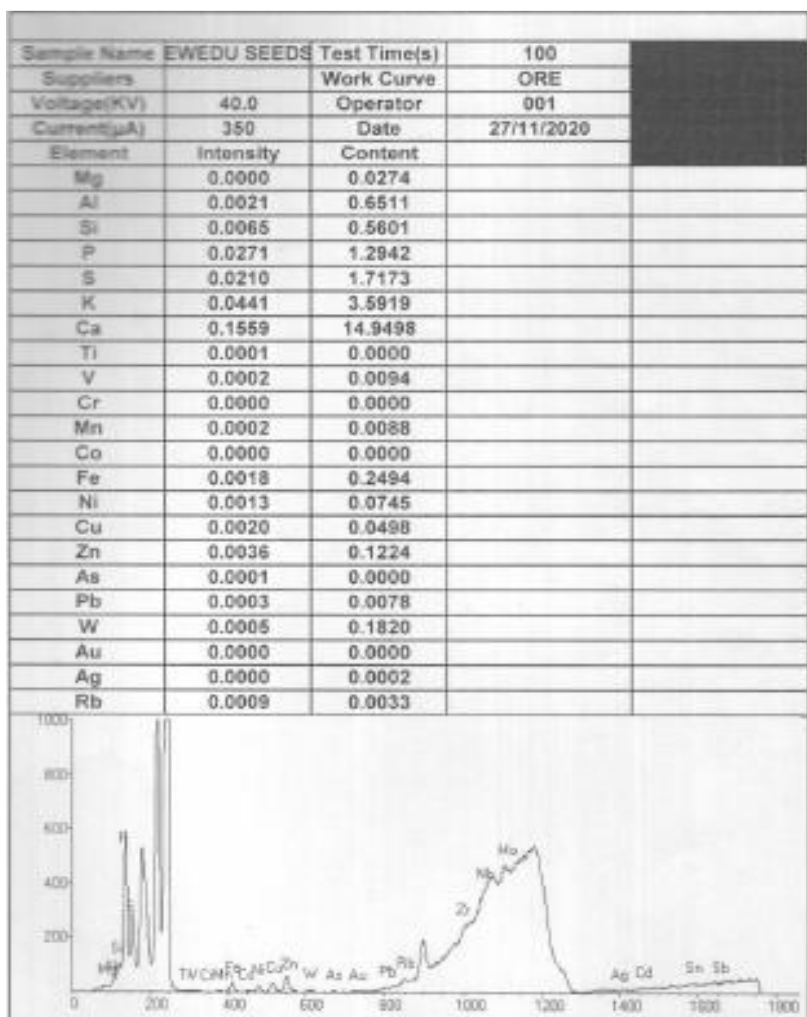


Table 7: Elemental composition of almond seeds

Sample Name	ALMOND SEED	Test Time(s)	100	
Suppliers		Work Curve	ORE	
Voltage(KV)	40.0	Operator	001	
Current(μ A)	350	Date	27/11/2020	
Element	Intensity	Content		
Mg	0.0002	0.2833		
Al	0.0035	1.1243		
Si	0.0181	1.7509		
P	0.0304	1.4521		
S	0.0338	3.3860		
K	0.1045	8.5197		
Ca	0.1095	10.2969		
Ti	0.0002	0.0000		
V	0.0001	0.0035		
Cr	0.0000	0.0000		
Mn	0.0003	0.0164		
Co	0.0005	0.0134		
Fe	0.0115	1.1698		
Ni	0.0013	0.0739		
Cu	0.0031	0.0948		
Zn	0.0080	0.2656		
As	0.0002	0.0000		
Pb	0.0002	0.0048		
W	0.0012	0.4573		
Au	0.0000	0.0000		
Ag	0.0000	0.0000		
Rb	0.0056	0.0487		