Iraqi Journal of Industrial Research, Vol. 10, No. 1 (2023)

Open Access

Iraqi Journal of Industrial Research (IJOIR)

Journal homepage: http://ijoir.gov.iq

Naturally Occurring Radioactive Materials in the Soil of Near Basra Oil Company Fields

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Article information

Article history:
Received: January, 03, 2023
Accepted: May, 19, 2023
Available online: June, 14, 2023

Keywords:
Natural radioactive elements,
Norm,
External absorbed dose rate,
Oil industry

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DOI:
https://doi.org/10.53523/ijoirVol10I1ID299

Abstract

The present study aimed to determine the levels of contamination with Natural Occurring Radioactive Materials NORM in one of the south oil company fields. The external gamma absorbed dose rate Dγ measured in units of µSv/h was caused by gamma rays of Radium 222, Radium 228 (Thorium 232), and Potassium 40, respectively. The largest value is 9.220 µSv/h. It was found that the highest specific activity (concentration) for Radium 226 is 1136 Bq/kg and the lowest is 0.06 and the highest specific activity for Radium 228 is 721 Bq/kg and the lowest is 0.02 Bq/kg. As for Radium 224, its highest specific activity is 631 Bq/kg and the lowest is 0.02 Bq/kg. Radium-228 is higher than that of Radium-224 or Radium-226, as the Radium-224 is from the Uranium-232 series, and the Radium-228 and Radium-224 are from the Thorium-232 series, meaning that the percentage of the daughters of the Thorium chain is lower than the percentage of the daughters of the Thorium chain Uranium, because the half-life of Radium-226 is 1600 years, which is greater than the half-life of Radium-228, which is 5.75 years. A comparison was made between the local results with the results of soil in Amman. Methods of treating pollution with natural radioactive materials in the oil industry were also discussed.

1. Introduction

The oil and gas industry is one of the industries that pollute the environment in which the workers and the population in the oil extraction area are exposed to the danger of natural radioactive substances that are concentrated as a result of this industry. Natural radioactive materials accompany oil extracted from the ground, to be placed on the inner walls of pipelines and oil separation or storage warehouses. These deposits have either scaly sediment or sludge form. The arrangement of these stores is due to physical (changes in temperature and weight) and chemical variables amid the stages of fluid partition (oil and going with water) that diminish the disintegration of particles when they reach the surface of the soil and hence crystallize within the frame of numerous salts, such as barium sulfate and strontium. Since of the comparable chemical properties of Radium, Calcium, Barium, and Strontium, they accelerate together, taking part within the arrangement of carbonate or sulfate compound salts, which gather within the frame of stores in channels and gear over the surface of the ground. The sum of amassed dregs depends on a few components, counting the pumping rate, the time of entry

of the provided or created water from the bowl to the surface, the topographical structure of the bowl, and the nature of the infusion water [1].

Study location: South Oil Company - Central Station Iraqi Standards for Contaminated Soils: The third part of NORM waste produced by the Iraqi oil and gas industry is contaminated soil. In general, about 3200,000 m of contaminated soil is reported. 226Ra has reached a value higher than 100 Bq/g in the hot spots. Table (1) shows some values in different locations it was noted that the pollution was concentrated in the surface layer of the soil to a depth of 50 cm in some areas.

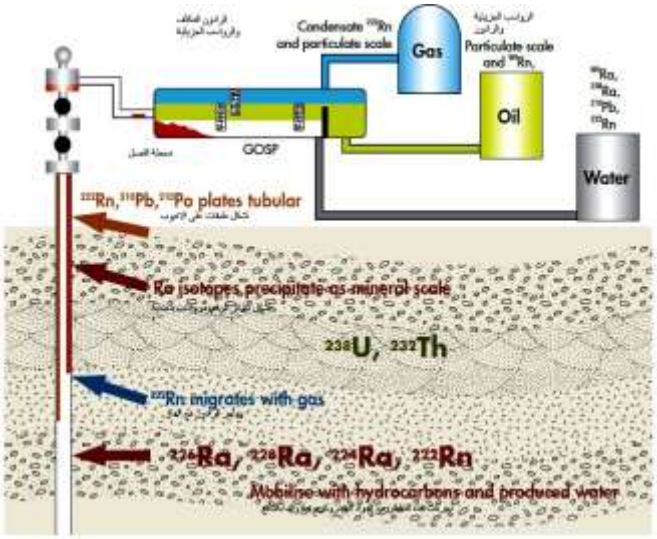

Figure (1). The origin of NORM, showing NORM backlogs in restore operations.

The Iraqi standards for disposal and cleaning of contaminated soil have been determined according to [2-5].

1. Contaminated soils containing less than 0.15 Bq/g of Ra226 do not need any treatment.
2. Contaminated areas containing the specific activity of Ra226 between 0.15 Bq/g and 5.2 Bq/g need special on-site treatment to reduce exposure to a value below 100 µSv/y, and no standards have been set for the disposal of hard and soft sediments and contaminated equipment.
3. Soils containing specific activity of Ra226 higher than 5.2 Bq/g are considered radioactive waste.

The specific activity of the soil may not exceed 100 µSv/y or not exceed 10 Bq/g i.e. 270 nCi/kg, which the IAEA suggests not to exceed, where the natural radiation background is 0.07 µGy/h. [6].

The previously observed criteria are based on the following considerations: The mass or surface specific activity of some isotopic activity such as Ra226 or the overall specific activity of the site can be used. A quantitative risk assessment study was conducted. This study presented a relationship between the rate of specific activity of 226Ra in polluted sites and the dose received by members of the public Figure (2) and Table (4). It was found that the external exposure is the most dominant, but it is greater by at least one rank in value than the other paths. It was also noted that the linear relationship remains dominant as long as the specific activity is under 15 Bq/g and the specific activity is 2260.15 Bq/g Ra. These values give a dose of 1mSv for an individual who is a permanent resident of the site while in the case of specific activity of Radium 226Ra was (5.2 Bq/g) giving a dose of 20 mSv assuming that the residence is 100%. The current residence in all contaminated sites is much less than 10%.

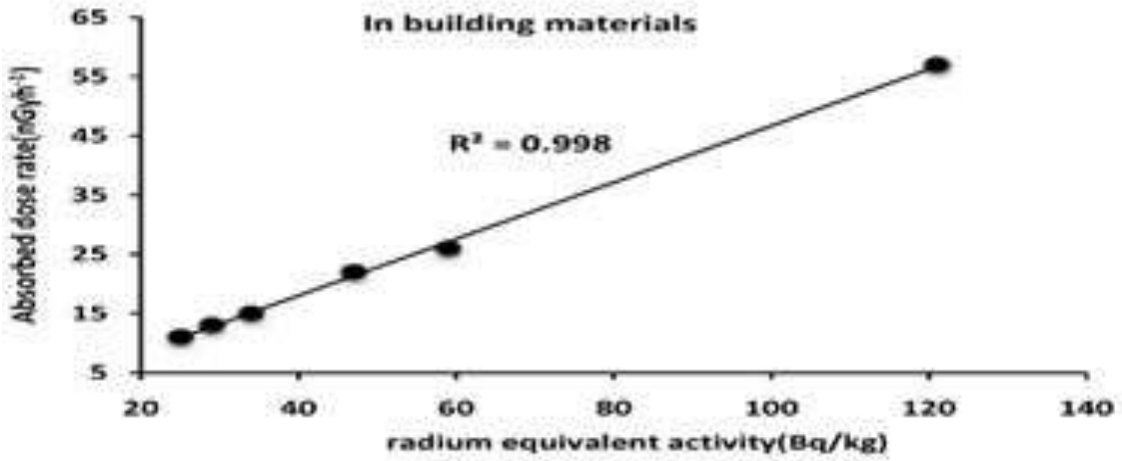

Figure (2). The relationship between the annual dose received by workers and the specific activity of 226Ra.

2. Material And Methods

Sample collection: 16 surface soil samples were collected from different places from the central treatment plant in Al-Omar field, which is expected to have a percentage of contamination in some places. The surface soil samples were collected at a depth of approximately 5 cm and the equivalent of 1200 to 1400 grams of soil. **Radiation surveys:** Preliminary radiological surveys were conducted for different places in the central treatment plant A lump device (gamma counter) equipped with a sodium iodide detector shown in Figure (3) is used. The readings were taken at a height of 1 meter from the surface of the soil and based on the preliminary radiological surveys of the areas where it was believed that it is polluted by oil activity nearby. a number of soil samples were collected. Table (1) shows the results of radiological surveys of these places and their locations.

Figure (3). Ludlum device (gamma counter) equipped with a sodium iodide detector.

Table (1). The results of radiological surveys in the central processing plant.

Code number	Location	Location of the measurement result in Ludlum
S1	Next to the pump for water output 402-P	50.3
S2	Next to the water pump output 408-P	127
S3	309 Next To oil tank-T	160
S4	400 Next To Water Pump Output-P	50.0
S5	404 Next To Water Pump Output -P	50.2
S6	Next To Water Pump Output 36104-P	67.5
S7	Next To Water Pump Output 36122-P	158
S8	Next To Water Pump Output 36407-P	56.6
S9	Next To Water Pump Output 502-P	50.5
S10	Next To Water Pump Output 357-P	50.0

Radiological analysis of samples: The laboratory analysis for the determination of isotopes of Radium in the soil includes two stages: the first stage is the preparation of soil samples, and then the measurement by gamma spectrometers, as follows:

Soil samples preparation stage: Soil samples were prepared for measurement by gamma spectroscopy by removing the stones completely by passing the samples through a 39 Mi sieve and then dried in the oven for 22 hours at 90°C. After drying and grinding to reach the greatest degree of homogeneity between the grains of the material forming the sample. A quantity of soil was filled in the approved measuring containers, where the sample amount was 1111 grams and that quantity is sufficient to determine the activity of Radium isotopes with an uncertainty of less than 5% (± **σ ???**) and with a reliability degree of 55% for the time of examining one sample by a count of 51 thousand seconds, which is equivalent to 13 hours 53 minutes (49980 seconds). If the samples have high activity, it is possible to reduce the enumeration time, HPGe after the soil samples were stored and preserved for one month in order to balance the isotopes of Radium with their offspring. The measurement was carried out in cooperation with the spectroscopy laboratories of the Iraqi Atomic Energy Commission.

3. Results and Discussion

Radiometric sampling: In the current study, a gamma HpGe spectrometer with high separating capacity was used, equipped with a highly purified P-Type germanium detector, with a relative detection efficiency of 80%, and a separation ability at 1332 KeV, which is 1.85 KeV, to determine the activity of Radium isotopes. The spectral line at 186.2 KeV with a relative intensity of 3.51% was used to determine the activity of Radium-226. The references indicate that its concentrations are less than 0.1 Becquerel/gram [7].

In addition, the activity of Radium (Ra-226) was also measured by measuring the activity of its daughters Lead (214Pb) and Bismuth (214Bi). 214Pb has two energies, 351.9 KeV and 295.2 KeV, while 214Bi has two energies, 609 KeV and 1120 KeV, after closing the sample tightly and keeping it for a month to ensure the balance of Radium and its Insert here in terms of radioactive dissociation. The activity of Radium 224Ra was determined by spectroscopic lines of its daughters Lead (212-Pb), Bi (212-Bi), and Thallium (Tl208), while the activity of Radium 228 was determined by in terms of its daughter Ac 228 (Table 2).

Table (2). shows the results of measuring the radioactivity in the soil of the central treatment plant.

S_5(Bq/kg)	S_4(Bq/kg)	S_3(Bq/kg)	S_2(Bq/kg)	S_1(Bq/kg)	sample number
			16.6±11.2		Th234
0.007±0.0006	0.002±0.0005	2.6±0.7	7.7±0.6		Cs137
0.3±0.01	0.2±0.01	148.9±10.7	257±11.5	0.3±0.04	K40
		253.6±8.3	252.9±6.4		Tl208
		233±45.3	338.6±13.4		Bi212
		336.6±12	318.2±8.3		Pb212
0.60±0.020	0.04±0.003	531.5±16.6	433.1±9.7	1.1±0.04	Ac228
		756.2±23.7	532.4±18.5		Bi214
		856.9±28.1	698.2±17		Pb214
0.30±0.008	0.02±0.003			0.5±0.02	Ra224
1.1±0.04	0.1±0.02	1136±59	900.8±40.6	2.1±0.1	Ra226
S_{10}(Bq/kg)	**S_9(Bq/kg)**	**S_8(Bq/kg)**	**S_7(Bq/kg)**	**S_6(Bq/kg)**	**sample number**
					Th234
0.005±0.001	0.003±0.0004	1.1±0.5	7.2±0.7	6.8±0.5	Cs137
0.2±0.02	0.2±0.01	92±10	212.8±12	278.6±13	K40
			142.1±5.4	111.8±4	Tl208
			134.9±30.9	140.5±8.8	Bi212
			179.4±9.3	136.9±5.2	Pb212
0.03±0.003	0.05±0.003	11±2	620.7±21.4	232.5±7.4	Ac228
			134.9±30.9	267.9±9.4	Bi214
			840.7±36.4	301.4±10.4	Pb214
0.03±0.002	0.04±0.003	12±1.2			Ra224
0.1±0.02	0.1±0.02	47±10	1117±74.1	479±26.9	Ra226

Table (3). Specific activity (becquerels per kilogram) of the surrounding soil.

Gamma Probability	Energy (KeV) Gamma Rays	Parent	Radionuclide
11.26	833	Ac-228	Ra-228
26.6	911.2		
43.5	239	Pb-212	Ra-224
6.64	727.3	Bi-212	
30.58	583.2	Tl-208	
3.51	186.2	Ra-226	Ra-226
18.2 35.1	295.2 351.9	Pb-214	
44.6 14.7	609 1120	Bi-214	
4.05	46.5	Pb-210	Pb-210

As shown in Table (3), the specific activities of Radium-226 are very variable and ranged from 0.1 Bq/kg to 1136 Bq/kg, and the highest activity was discovered as (1011, 7111, 1136) for samples 13-7-3, respectively. Sample 3 was taken from the side of a 405-T oil tank, sample 7 from the pump side of the output water, and sample 13 from the 309-T tank side. The specific activity of Radium-224 on the other hand ranged between 721 Bq/kg-0.02 Bq/kg and for Radium-228 it was higher than for Radium-224. The following Figure (4) represents the spectrum of S7 sample.

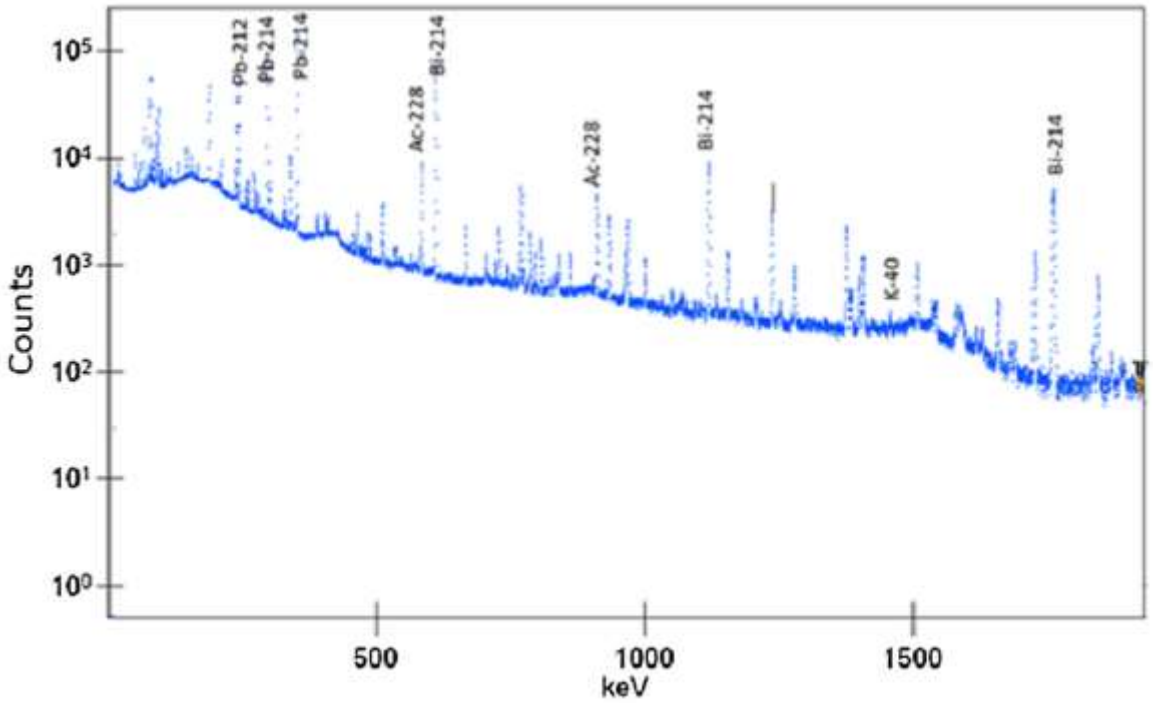

Figure (4). The spectrum of a contaminated soil sample (S7)

To calculate the external gamma absorbed dose rate for each of the samples, the following relationship was used:

$D\gamma = (0.417\ CRa + 0.604\ CTh + 0.0417\ CK) \times 10^{-3}$ [12]

where Thorium has been replaced by Radium -228 because according to [15] the main contribution of the Thorium chain is the daughter 228Ra:

$D\gamma = (0.417\ CRa + 0.604\ CRa\text{-}228(Th) + 0.0417\ CK) \times 10^{-3}$

where CRa, CTh, and CK are the specific activity of Radium-222, Radium-222 (Thorium-232), and Potassium 40, respectively, in Bq/kg and Dγ in μSv/h due to gamma radiation, which is the reference dose in the air outside the building at a height of 1 m. The equation is proposed by the United Nations Scientific Committee on the effect of nuclear radiation [15]. The constants in the equation represent the contribution of these elements to the total dose produced

Estimated by the effect of gamma rays, assuming that the rest of the doses of 137Cs, 90Sr, 235U are negligible and do not give a significant effect on the total dose of the background radiation, as it was assumed that 379 Bq/kg of Radium 226Ra or 250 Bq/kg of Thorium 232Th or 2219 Bq/kg of Potassium 40K all yield the same gamma dose rate.

Table (4). represents the dose rate for each of the samples.

location	Dose rate(μSv/h)	location	(Dose rate μSv/h)
soil 1	0.00155261	soil 9	0.00008024
soil 2	0.6479429	soil 10	0.00006816
soil 3	0.8009471	soil 11	0.5345098
soil 4	0.0000742	soil 12	0.00004544
soil 5	0.0008336	soil 13	0.7023512
soil 6	0.35179062	soil 14	0.04130067
soil 7	0.84956556	soil 15	0.03543297
soil 8	0.0300794	soil 16	0.8405995

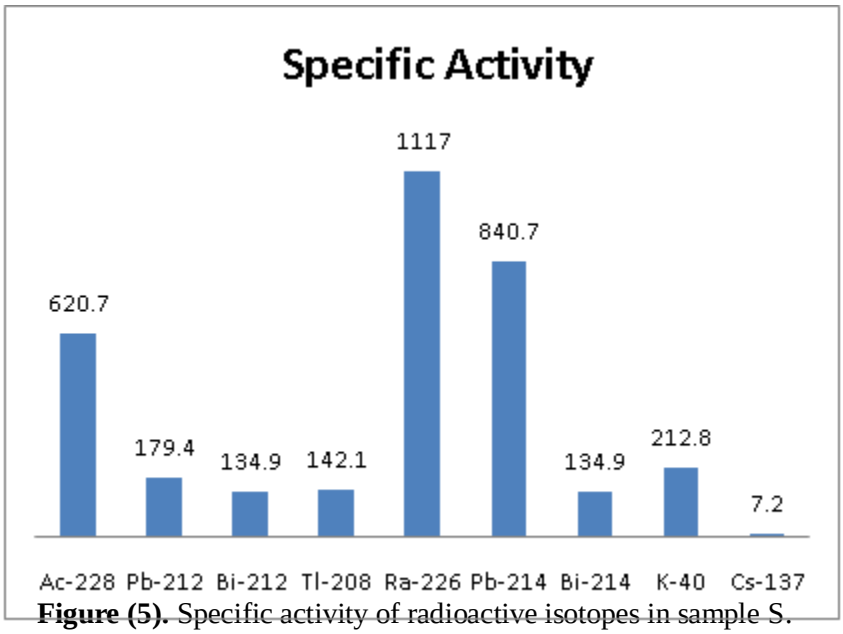

Figure (5). Specific activity of radioactive isotopes in sample S.

It is noted from the following figure (5) that Radium-226 has the highest specific activity in this sample and that Cesium has the lowest specific activity. Also, Bismuth-212 and Bismuth-214 have equal values of specific activities.

4. Conclusions

The measured concentrations of radium isotopes in the soil have been found to be significantly higher than the values reported both globally and in a country that is geographically close to Iraq, such as Oman. Soil samples were taken from a station employing workers and it was contaminated, which calls for the use of more efficient systems in the station's work and control, and some protocols applied by foreign companies were inaccurate and unsafe, such as the method of spreading radioactive soil and distributing it in the environment. This study aimed to determine the levels of contamination with Natural Occurring Radioactive Materials NORM in one of the South oil company fields. Results showed that gamma absorbed dose Dγ measured in units of Sv/h was caused by gamma rays of Radium 222, Radium 228 (Thorium 232), and Potassium 40, with the highest concentration being 1136 Bq/kg and the lowest being 0.02 Bq/kg. Methods of treating pollution with natural radioactive materials in the oil industry were discussed.

References

[1] F. Djahanguiri; G. M. Reimer; R. Holub; C. S. Aliev; T. Zolotovitskaya, "Radioactive Pollution at the Oil Fields of the Apsheron Peninsula, Caspian Sea, Azerbaijan, "*Paper presented at the SPE/EPA Exploration and Production Environmental Conference*, Dallas, Texas, Paper Number: SPE-38389-MS, 1997.

[2] I. Othman, M. S. Al-Masri, H. Suman, "Public And Regulatory Acceptability Of NORM Contaminated Soil Disposal: The SYRIAN EXPERINCE, "*Atomic Energy Commission of Syria, Damascus*. (2008)

[3] I. Othman, M. S. Al-Masri "Characterization of NORM Contaminated Sites At The Syrian Oil Field: Sampling, Analysis And Data Management. "*Atomic Energy Commission of Syria. Damascus*. (2008).

[4] M. S. Al-Masri & H. Suman , "NORM Waste Management in the Oil and Gas Industry: The Syrian Experience, "*Journal of Radioanalytical and Nuclear Chemistry*, vol. 256, pp. 159–162, 2003.

[5] Guidelines for the management of Naturally Occurring Radioactive Material (NORM) in the oil & gas industry, IOGP Report No. 412, September 2008.

[6] Gesell, Thomas F. "Occupational Radiation Exposure Due to 222Rn in Natural Gas and Natural Gas Products, "*Health Physics*, Vol. 29. PP 681-687, November 1975.

[7] Bulletin on Management of Naturally Occurring Radioactive Materials (NORM) in Oil and Gas Production. American Petroleum Institute, Washington, 11 API, 1992.

[8] P.R.Gray, "NORM Contamination in the Petroleum Industry. "*J Pet Technol* . vol. 45, pp. 12–16, 1993.

[9] Zhuravel, N. E, "Ukrainian Oil Field NORM Contaminated Examined, "*Oil Gas J*, vol. 97, pp. 103-105, 1999.

[10] Mohamed Al-Hilal, Mosa Aissa, "Characterizing the natural radiation levels throughout the main geological units of Sabkhat al Jabboul area northern Syria: Technical Reports. "*J Environ Radioact*, vol. 140, pp. 1-10, 2015.

[11] Bou-Rabee F. , Al-Zamel A. Z. , Al-Fares R. A. , and Bem H, "Technologically enhanced naturally occurring radioactive materials in the oil industry (TENORM).A review, *Institute of Nuclear Chemistry and Technology*, vol. 54, No. 1, 2018.

[12] Al-Farsi, Afkar Nadhim, "Radiological aspects of petroleum exploration and production in the sultanate of Oman. PhD thesis, Queensland University of Technology, 2008.

[13] Al-Masri, M.S., Suman, H., Aba, A., Mukalalati, H, "NORM Levels at Der Ezzor Petroleum Company (DEZPC) Oilfield in Der Ezzor Area, Syrian Arab Republic, 2021.

[14] Sabina Yasmin, Bijoy Sonker Barua, Masud Kamal, and Md. Abdur Rashid, "Prasong Kessaratikoon and Susaira Awaekechi Natural Radioactivity Measurement In Soil Samples Collected From Municipal Area Of Hat Yai District In Songkhla Province, "*KMITL Science Technology Journal, Thailand*, vol. 8, No. 2, 2021.