PHOSPHOR AND NITROGEN ELEMENTS ANALYSIS IN TAPIOCA WASTE PRODUCTS BY NEUTRON BEAM ACTIVATION

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ABSTRAK

ANALISIS UNSUR P DAN N DALAM LIMBAH TAPIOKA DENGAN METODA AKTIVASI BERKAS NEUTRON CEPAT. Telah dilakukan analisis unsur P dan N dalam cuplikan limbah tapioka dengan menggunakan metoda aktivasi neutron cepat 14,3 MeV yang dihasilkan oleh Generator Neutron (Merek Sames, tipe J.25-150kV) dari reaksi inti $^{3}H(d,n)^{4}He$. Dari pabrik tapioka dihasilkan limbah yang mengandung P dan N yang dapat dimanfaatkan untuk bahan baku pupuk. Untuk menentukan persentase kandungan P dan N digunakan analisis kuantitatif secara relatif. Kadar P total di limbah tapioka lapisan atas , lapisan tengah dan lapisan bawah masing-masing besarnya adalah 1,11-1,50%, 1,38-2,15 % dan 1,58-2,45%. Sementara kadar N total di limbah tapioka lapisan atas , lapisan tengah dan lapisan bawah masing-masing besarnya adalah 0,92-1,15%, 1,12-1,45 % dan 1,14-1,69%. Limbah tapioka yang mengandung unsur N dan P dapat digunakan sebagai bahan baku pupuk.

ABSTRACT

PHOSPHOR AND NITROGEN ELEMENTS ANALYSIS IN TAPIOCA WASTE PRODUCTS BY NEUTRON BEAM ACTIVATION. The analysis of phospor and nitrogen elements in tapioca waste products by neutron beam activation analysis have been done. The 14,3 MeV fast neutron beam was produced by Neutron Generator (Sames trade-mark, type J.25-150 KeV) from $^{3}H(d,n)^{4}He$ nuclear reaction.The side products of tapioca industry was tapioca waste with P and N contents. The tapioca waste was used for raw material of fertilizer. The quantitative analysis by the relative method was used for the determination of the percentage of phosphor element concentration. It was found that the total phosphor element concentration in the upper, middle and lower layers of tapioca waste products were 1.11-1.50%, 1.38-2.15 % and 1.58-2.45% of the samples weight respectively. Afterward, the total nitrogen element concentration in the upper, middle and lower layers of tapioca waste products were 0.92-1.15%, 1.12-1.45 % and 1.14-1.69% of the samples weight respectively. The tapioca waste products which content of P and N elements can be used for fertilizer raw material.

INTRODUCTION

The tapioca industrial area was spread in Central Java, for instance in Karanganyar, Wonogiri, Batang, Pemalang , Demak, Wangon and Banyumas.[1] The tapioca industrial wastes were accumulated in the wastes storage and not to be used yet. The main feed of industrial tapioca is cassava tuber (*Manihot utilissima*). The waste material products of tapioca industries were cassava tuber mud, skin tuber and other materials. The cassava tuber contents about 0.4-0.6% of phosphor element and 1-2% of nitrogen element.[2] Of course, the crisis era makes it difficult to buy the raw material for ferlilizer from abroad. The use of tapioca wastes as a local raw materials for fertlizer production is important to develop in Indonesia, especially for self support in fertilizer need.

Fast neutron beam (14.3 MeV) was produced by neutron generator. Neutron generator is an accelerator, because there are ion (electron) acceleration processes to be collided with tritium target. Those process produced 14.3 MeV fast neutron. Fast neutron activation analysis used for light elements, i.e. N,P, K, S etc.[3] Nuclear reactor (Kartini) is not an accelerator, because there are not ion acceleration processes. Neutron thermal with range energy of 0.25-0.5 eV was produced by nuclear reactor. Thermal neutron activation analysis used for heavy elements, i.e. As, Cu, Zn, Hg Cd etc.

Fast neutron beam activation method can be used for determination of P and N elements in tapioca wastes. Fast neutron beam of 14.3 MeV energy was produced by $^{3}H(d,n)^{4}He$ nuclear reaction. It will interact with ^{31}P of tapioca waste samples and produce ^{28}Al isotope by $^{31}P(n,\alpha)^{28}Al$ nuclear reaction. The radiosotope of ^{28}Al emits gamma rays with energy $E\gamma$=1.778 MeV and half life 2.30 minutes. Fast neutron beam of 14.3 MeV energy will interact with ^{14}N of tapioca waste samples and produced ^{13}N isotope by $^{14}N(n,2n)^{13}N$ reaction. The radiosotope of ^{13}N was

emitted gamma rays with energy Eγ=511 KeV and half life 9.96 minutes.

EXPERIMENTAL PROCEDURE

Samples collection

Tapioca waste samples for the P and N elements analysis were collected from 2 locations in Karanganyar district, 2 locations from Batang district and 2 locations from Wonogiri district on August 1997. From the pile up of tapioca wastes storage was differentiated between top layer (0 - 0.50 m) , middle layer (0.50-1.00 m) and bottom layer (> 1.00 m). Sample of about 1 kg was filled in a plastic bag and brought to laboratory. The fresh cassava tuber samples were used as a control to compare with the tapioca wastes samples.

Samples preparation

Tapioca wastes samples were dried at temperature 50-60 °C . The samples were ground in mortar porcelain and filtered with 100 mesh fine filter. Each samples weighing about 100 mg, was placed in a polyethylene vial (10 mm x 30mm) and identified. The vials were heat-sealed. Standard for P element was prepared by mixing the cellulose and P_2O_5 in variation of P element concentration. Standard for N was prepared by mixing the cellulose and D. Leucine in variation of N element concentration.

Irradiation and Counting

All fast neutron irradiations of samples were carried out in the accelerator laboratory by the neutron generator. Irradiation time was 30 minutes. During irradiation processes the neutron generator was operated in a high voltage of 110 kV and 500 mA of the current. The samples was cooled about 1 minute after irradiation. Samples and standards both were counted by NaI(Tl) detector for 600 seconds. The gamma spectrometer was provided by high voltage source, preamplifier, amplifier, accuspec and computer for MCA

RESULTS AND DISCUSSION

In general, nuclides of ^{31}P and ^{14}N are found in the tapioka wastes. Afer fast neutron irradiation, there were the nuclear reaction of ^{31}P and ^{14}N nuclides as shown in the Table 1.

Table 1. Nuclear reaction and nuclear data of nuclides ^{31}P and ^{14}N after fast neutron irradiation. [4,5]

Nuclear rections	Abundance (%)	Cross section (mb)	Half life (minutes)	Eγ (MeV)
$^{31}P(n,\alpha)^{28}Al$	100	150	2.31	1.78
$^{31}P(n,p)^{31}Si$	100	83	152.20	1.26
$^{31}P(n,2n)^{30}P$	100	14	2.31	0.51
$^{14}N(n,2n)^{13}N$	90.63	5.7	9.96	0.511

Base on the peaks energy 1.78 MeV of P element and 0.511 MeV of N element, the quantitative analysis of those elements can be done. Evaluation of peaks energy 1.78 MeV of P element were interferences by ^{27}Al and ^{28}Si nuclides [see Table 2]. So, the correction of P element analysis result were needed.

Table 2. Nuclear reaction and nuclear data of nuclides ^{27}Al and ^{28}Si that interferences of peaks energy 1.78 MeV of P element.[4,5]

Nuclear rections	Abundance (%)	Cross section (mb)	Half life (minutes)	Eγ (MeV)
$^{28}Si(n,p)^{28}Al$	92.27	2.35	2.31	1.78
$^{27}Al(n,\gamma)^{28}Al$	100	0.50	2.31	1.78

Concentration of N and P elements in the tapioca wastes was calculated using formula

$$Cps_0 = Cps_t \cdot e^{0.693\, t/T}$$

and

$$W_{samples} = \frac{Cps_{samples}}{Cps_{standards}} \times W_{standards} \quad (1)$$

where

Cps_0 = the counting rate at the time taken out from the neutron generator

Cps_t = the initial counting rate

t = delay time

T = half life of ^{13}N or ^{28}Al

$W_{samples}$ = Weight of N or P in the samples

$W_{standards}$ = Weight of N or P in the standards

$Cps_{samples}$ = counts per seconds of ^{13}N or ^{28}Al in the samples

$Cps_{standards}$ = counts per seconds of ^{13}N or ^{28}Al in the standards

The energy calibration of the machine was done using the standard before counting the samples. Calibration yield of standard is given in Table 3.

Table 3. The energy calibration with some types of isotopes.

No. of channels	Isotope types	E-γ (keV)	E-γ calibration (keV)
1461	Na-22	511	510.88
1881	Cs-137	662	662.14
3271	Co-60	1,170	1,170.93
3547	Na-22	1,276	1,273.44
3703	Co-60	1,330	1,331.61

It was shown that energy calibration results of isotope Na-22, Cs-137, Co-60, Na-22, and Co-60 were 510.88 keV, 662.14 keV, 1,170.93 keV, 1,273.44 keV, and 1,331.61 keV. E-γ calibration result was not significant by the different from the E-γ isototopes of the table. The qualitative analyses of P and N elements in tapioca waste is given in Table 4.

Table 4. The qualitative analyses of P and N elements in tapioca waste

Sampling locations	No. of channels	E-γ (keV)
Upper layer	1435	511.12
	2347	844.05
	4761	1,784.10
Middle layer	1435	510.44
	2347	843.15
	4761	1,783.59
Bottom layer	1435	511.05
	2347	843.10
	4761	1,782.75

After neutron activation samples, the E-γ spectrum emitted from the radionuclides was the same as the E-γ callibration.. The E-γ of 510.44-511.12 keV come from some types of radionuclides , i.e. ^{13}N, ^{31}P, ^{22}Na, ^{54}Cu, ^{65}Zn dan ^{37}Ni. Base on the nuclear data, cross section and the probabilities of nuclear reaction, the E-γ of 510.44-511.12 keV came from $^{14}N(n,2n)^{13}N$ reaction. It was assumed that tapioca waste contents of ^{14}N isotopes. E-γ of 843.10-844.05 keV was emitted from some types of isotopes ^{27}Mg, ^{72}Ga and ^{54}Mn. Based on nuclear data, cross section and the probabilities of nuclear reaction, the E-γ of 843.10-844.05 keV was emitted by ^{27}Mg isotope and come from $^{17}Al(n,p)^{28}Mg$ reaction. It was assumed that ^{27}Al isotope is present in the tapioca waste samples. After tapioca waste samples were irradiated with 14.3 MeV of neutron, the Eγ of 1,782.75-1,784.10 keV was emitted by ^{28}Al isotope coming from $^{31}P(n,\alpha)^{28}Al$ reaction. It was assumed that ^{31}P isotope is present in the tapioca waste samples. The quantitative analyses of P and N elements in tapioca waste from Karanganyar, Batang and Wonogiri districts are given in Table 5, 6, and 7.

It was shown that P and N elements were found in the storage pile up of tapioca wastes in Karanganyar district (Table 5). There were 1.3-1.5% of P element and 0.92-1.10% of N element in upper layer, 1.8-2.15% of P element and 1.12-1.13% of N element in middle layer, 2.25-2.44% of P element and 1.14-1.17% of N element in bottom layer of tapioca wastes. Meanwhile, the controls of tapioca waste were 1.11% content of P element and 0.89% content of N element. In general the tapioca waste were containing P and N elements higher than tapioca controls.

It was shown that P and N elements were found in the storage pile up of tapioca wastes in Batang district (Table 6). There were 1.12-1.24% of P

Table 5. The quantitative analyses of P and N elements in tapioca waste from Karanganyar district by neutron beam activation

Sampling districts	Sampling locations	P concent. (%)	N concent. (%)
Karanganyar-1	Upper layer Middle layer Bottom layer	1.5± 0.41 1.8±0.62 2.44 ±0.35	0.92±0.11 1.13±0.10 1.17±0.23
Karanganyar-2	Upper layer Middle layer Bottom layer	1.3±0.51 2.15±0.24 2.25±0.19	1.10±0.08 1.12±0.11 1.14±0.20
Controls	-	1.11±0,47	0.89±0.18

element and 1.12-1.15% of N element in upper layer, 1.38-1.47% of P element and 1.18-1.24% of N element in middle layer, 1.94-2.45% of P element and 1.38-1.45% of N element in bottom layer of tapioca wastes. Meanwhile, the controls of tapioca waste were 1.09% content of P element and 0.99% content of N element. In general the tapioca waste were containing P and N elements higher than tapioca controls.

Table 6. The quantitative analyses of P and N elements in tapioca waste samples from Batang district by neutron beam activation

Sampling districts	Sampling locations	P concent. (%)	N concent. (%)
Batang-1	Upper layer Middle layer Bottom layer	1.12±0.08 1.38±0.09 2.45±0.75	1.10±0.09 1.18±0.29 1.38±0.30
Batang-2	Upper layer Middle layer Bottom layer	1.24±0.32 1.47±0.16 1.94±0.37	1.15±0.09 1.24±0.19 1.45±0.6
Controls	-	1.09	0.99

Table 7. The quantitative analyses of P and N elements in tapioca waste samples from Wonogiri district by neutron beam activation

Sampling districts	Sampling locations	P concent. (%)	N concent. (%)
Wonogiri-1	Upper layer Middle layer Bottom layer	1.11±0.07 1.92±0.63 2.16±0.44	0.98±0.06 1.45±0.31 1.69±0.41
Wonogiri-2	Upper layer Middle layer Bottom layer	1.23±0.12 1.87±0.061 1.98±0.07	1.12±0.9 1.38±0.21 1.65±0.47
Controls	-	1.00±0.08	1.06±0.11

It was shown that P and N elements were found in the storage pile up of tapioca wastes in Batang district (Table 7). There were 1.12-1.23% of P element and 0.08-1.65% of N element in upper layer, 1.87-1.92% of P element and 1.38-1.45% of N element in middle layer, 1.98-2.16% of P element and 1.65-1.69% of N element in bottom layer of tapioca wastes. Meanwhile, the controls of tapioca waste samples were 1.00% content of P element and 1.65% content of N element. In general the tapioca waste samples were containing P and N elements higher than tapioca controls.

CONCLUSIONS

1. In general, the bottom layer of tapioca waste pile up storage contents P and N elements were higher than other pile up layers of tapioca wastes samples. Meanwhile the concentration of P and N elements in the upper layer was the same with the controls.
2. The mean concentration of P element in upper layer, middle layer and bottom layer of tapioca wastes pile up storage were 1.11-1.50%, 1.38-2.15% and 1.58-2.45%.
3. The mean concentration of N element in upper layer, middle layer and bottom layer of tapioca waste pile up storage were 0.92-1.15%, 1.12-1.45 % and 1.14-1.69%.
4. The mean concentration P element in tapioca waste samples products in Karanganyar, Batang and Wonogiri districts were 1.11-2.44% , 1.12-2.45% and 1.11-2.16%. Although, the mean concentration of N element in tapioca waste samples products in Karanganyar, Batang and Wonogiri districts were 0.92-1.17%, 1.10-1.45% and 0.98-1.69%,
5. The tapioca wastes samples which containing of P and N elements can be used for raw materials in fertilizer production.

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TANYA JAWAB

Suharni H.

- Bagaimana prosedurnya jika kita ingin melakukan penelitian akselerator di BATAN ?. Apa cukup terjangkau dari segi biaya ?
- Apakah bahan hasil mutasi dengan akselerator cukup aman bagi manusia ?

Muryono

- *Membuat surat resmi ke kepala P3TM BATAN, Jl. Babarsari Kotak Pos 1008 Yogyakarta. Apabila judul penelitiannya mendukung program BATAN maka biaya penelitian akan terjangkau karena akan diberikan subsidi dari P3TM BATAN. Besar dan jenis subsidi yang diberikan akan ditentukan olehkepala P3TM.*
- *Hasil mutasi dengan akselerator tidak berbahaya karena berkas neutron cepat dengan energi 14,3 MeV hanya akan mengaktifkan unsur-unsur ringan (N,P,K,S) dengan umur paroh yang ordenya menit. Padahal pertumbuhan biji sampai panen ordenya hari sampai bulan . Dengan demikian unsur-unsur ringan yang radioaktif sudah habis meluruh dan menjadi unsur yang stabil.*

Sutiyah

- Kalau memang Bapak dapat menghasilkan pupuk yang relatif baik dan murah, kenapa tidak diproduksi pupuk tersebut/dipasarkan ?

Muryono

- *Arahnya memang untuk mendapatkan pupuk N dengan bahan lokal yang relatif lebih murah daripada pupuk N buatan pabrik. Sebagai lembaga penelitian P3TM BATAN bertugas menghasilkan penelitian dasar dan terapan yang bermanfaat bagi masyarakat. Dalam hal ini, P3TM BATAN tidak akan mendirikan pabrik pupuk dan sekaligus memasarkannya. Dipersilahkan pada pihak ketiga untuk mendirikan pabrik pupuk N yang murah dengan menerapkan hasil penelitian tersebut.*

Riyatun

- Mohon dijelaskan secara garis besar analisis yang dilakukan sehingga diperoleh kadar P dan kadar N?

Muryono

Limbah tapioka dikeringkan pada suhu sekitar 50°C, dihaluskan dan disaring dengan ukuran butir 100 mesh. Standar P dibuat dengan campuran

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selulose dan P_2O_5 sedangkan standar N dibuat dengan campuran selulose dan D.Leusine. Iradiasi cuplikan dan standar dilakukan selama 30 menit dengan neutron cepat (energi 14,3 MeV). Pencacahan dilakukan dengan detektor NaI(Tl) selama 600 detik Dari cacah yang diperoleh dapat dihitung kadar N dan P di dalam limbah tapioka dengan menggunakan rumus (1).