SCIENTIFIC PAPERS
OF THE UNIVERSITY OF PARDUBICE
Series A
Faculty of Chemical Technology
12 (2006)

EXTRACTION METHOD FOR DETERMINATION OF Na, K AND Mg IN FEED MIXTURES

Anna KREJČOVÁ[1], Tomáš ČERNOHORSKÝ and Miloslav POUZAR
Department of Environmental Protection,
The University of Pardubice, CZ–532 10 Pardubice

Received January 2, 2006

In order to improve the speed at low of cost, a method of analysis of animal feed for sodium, potassium and magnesium by ultrasound-assisted leaching with a mixture of strong acids prior to analysis by flame atomic absorption spectrometry (FAAS) has been proposed. The influence of acid mixture composition, time, temperature and sample mass on the leaching recovery was examined in order to find the most suitable working conditions. The ultrasound-assisted leaching carried out at optimized conditions (mixture of HNO_3, HCl, H_2O_2, ultrasound bath, 60 °C, 30 min) was established to be a fast and quantitative method for the feed mixture sample preparation before determination of Na and K. Mg was found to be only partially extractable. The microwave ashing procedure was utilized as the standard for comparison of alternative methods of sample preparation. The leaching method followed by FAAS was applied to analyze feed mixtures and the results indicate that it is appropriate for determination of Na, K content in feed mixtures (recovery for Na 97-101 %, K 97-102 %).

---

[1] To whom correspondence should be addressed.

Introduction

Determination of macro-elements (Na, K, Mg, Ca, P) in raw materials and final products tends to be an important step in quality control of fodder and production control of feed mixtures. The knowledge of content of the above-mentioned elements in feeds is an important piece of information for planning the optimum composition on animal nutrition. A correct content of these elements in feed can affect the body weight increases achieved in livestock [1-4].

The elemental analysis of feed, food products and raw materials can be done using a variety of analytical procedures. With respect to the technique used for determination of nutritionally important elements in feed mixtures, atomic absorption spectrometry (AAS) has been the most popular in all their alternatives [5,6]. Flame atomic absorption spectrometry (FAAS) is very easy to use [7-9]. But commercial equipments for sample decomposition and the possibility of automated systems and multi-elemental analysis have made inductively coupled plasma emission spectrometry (ICP-AES) wide expanded, especially in terms of versatility and wide dynamic range [7,10-17]. The preparation of samples for these methods usually involves some way of digestion in order to destroy organic matter and to transform solid sample to liquid one, by dry or wet ashing.

A microwave-assisted digestion was adopted, e.g., for ICP-AES and GF-AAS (graphite furnace atomic absorption spectrometry) analysis of nutritional elements and contaminants in sheep feed [10], for ICP-OES and ICP-MS (inductively coupled plasma mass spectrometry) analysis of samples of Soya meal [11], for ICP-AES and ICP-MS analysis of plant tissues [15]. Wet digestion with nitric acid was used for analysis of fodder samples by ICP-MS method [18] and for the ICP-AES and ICP-MS analysis of plant tissues [15]. Dry ashing in a muffle oven was adopted for FAAS analysis of matrix and trace elements in papaya samples [7]. Dry ashing was used before AAS and spectrophotometric analysis of minerals and phosphorus in soy-based, whey-based, and enteral formulae [19]. Borkowska-Burnecka *et al.* [16] compared three digestion procedures (high-pressure microwave digestion, conventional wet digestion and dry ashing for ICP-AES analysis of matrix and trace elements in samples of vegetables and cereals). In analysis of feed mixtures, it is often very difficult to carry out a complete decomposition of the sample because these samples usually have a complex and considerably variable composition of their matrix. At present, control laboratories of feed factories and producers of feed mixtures widely adopt FAAS analysis. For analysis of samples with organic matrix, the rentgenfluorescence spectrometry (XRF) is suitable. The XRF spectrometry is not suitable for determination of Na, K and Mg in feed mixtures. This technique was applied for the determination of matrix and trace elements in plant tissues [20-22]. Černohorský *et al.* [23] proposed a fast and cheap method for determination of P and Ca in feed mixtures by means of XRF spectrometry.

Since Na, K and Mg are easily extractable elements, it is possible to overcome decomposition step in analytical procedure with leaching of sample [24]. The present paper describes a cheap method for determination of Na, K and Mg content in feed mixtures by a flame atomic absorption spectrometry after extraction with a mixture of HNO_3, HCl and H_2O_2 in ultrasonic bath. The recommended procedure is a supplement to the XRF method for determination of P, Ca and microelements in mixtures and raw materials [23].

Material and Methods

Reagents and Materials

Samples of raw materials — meat and bone meal, fish meal, rape fodder plant, sunflower scrap, rape seed were used (all VKS Pardubice, Czech Republic). Feed mixtures for pigs (A1-1886, A2-1691, CDP-1799, KPB-1803, KPK-1848, KA-1913), for poultry (K1-1997, NT1-1882, BR1-1808, BR2-1755, KR-1911) and for cattle (COTB-1906, DOUP-2, DOB-1907, DOPS-1889, SKOTB-1501) were used (all ZZN Semily, Czech Republic). Feed mixtures differ depending on the animals fed but they usually contain fodder grains, soyabean, rape seed and sunflower extracted meal, rape cakes, wheat pollard, peas, vegetable oil, pulverized limestone, dicalcium phosphate, salt, biofactors, vitamins.

The following chemicals were used for the microwave digestion and leaching: 65 % (w/v) HNO_3 (Lachema, Brno, Czech Republic), 35 % (w/v) HCl (Lachema, Brno, Czech Republic), 30 % (w/v) H_2O_2 (Penta, Chrudim, Czech Republic), NH_4Cl (Lachema, Brno, Czech Republic).

The calibration standards for the FAAS determination were prepared by diluting the standard solutions of Na, K and Mg (1.000 ± 0.002 g l^{-1}; Analytica, Prague, Czech Republic). The concentrations of calibration standards were 0.01; 0.5; 1.0; 2.0; 2.5; 5.0 mg l^{-1} for Na, 0.05; 0.5; 1.0; 2.0; 2.5; 5.0 mg l^{-1} for K and 0.01; 0.5; 1.0; 1.5; 2.0; 2.5 mg l^{-1} for Mg. For the preparation of solutions and dilution of samples and standards, we used demineralized water obtained from MILLI Q^+ apparatus (Millipore, Bedford, USA).

The method was validated using the NIST Standard Reference Material 1577b Bovine liver, Chinese reference material GBW 07603 Bush twigs and leaves (Langfang, China), GBW 07604 poplar leaves (Langfang, China), GBW 07605 Tea (Langfang, China) and control inland samples (clover, cocoa).

Sample Preparation

Prior to analysis, all samples were homogenized in a grinding mill VM-4 (OPS, Přerov, Czech Republic). The extractions of Na, K and Mg were carried out using the ultrasonic bath Powersonic UCC 1 (Czech Republic) or the rotation shaker RSR 01, Labio a.s. (Czech Republic). Alternatively, samples were decomposed by microwave-assisted digestion.

Microwave-Assisted Decomposition

The samples were decomposed in the microwave decomposition apparatus BM 1 S/2 (Plazmatronika, Poznan, Poland). A 0.2-0.5 g homogenized sample was weighed into an acid washed 150 ml PTFE digestion tube. Seven ml concentrated nitric acid (65 % (w/v)) was added and the tube was heated in a microwave oven at the power setting at 70 % for 5 min, at 90 % for 5 min and at 100 % for 10 min. The maximum output of the microwave generator was 700 W (the minimum pressure 24×10^5 Pa, the maximum pressure 25×10^5 Pa). The digest was transferred into a 50-mL acid washed volumetric flask, filled up with demineralized water and stored in polypropylene flasks. For each sample, six replicates and one acid blank were run.

Leaching

To avoid microwave digestion prior to analysis, selected leaching agents (water, acids and their mixtures) were used as extraction media for feed mixtures: H_2O, 2 mol l^{-1} HCl, 2 mol l^{-1} HNO_3, 1 mol l^{-1} HCl + 2 mol l^{-1} HNO_3, 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3, 3 mol l^{-1} HCl + 1 mol l^{-1} HNO_3, 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 + H_2O_2. As described in "Results and Discussion" and Table III, 1 or 2 ml H_2O_2 was used together with total volume 20 ml of extraction agents. For extraction, common laboratory equipment, e.g., a rotator shaker and an ultrasonic bath were used. In order to describe the influence of various parameters on the sonification process, the extraction efficiency was evaluated for the extraction temperature 25-75 °C sonification time 5-60 min and sample weight 0.1-2 g at the same volume of extracting mixture. A sample was weighed in 100 ml polyethylene bottle. The sample was treated with leaching agent (20 ml) in the ultrasound bath or in the horizontal shaker (at 10 rpm). After finishing the extraction, the obtained suspension was filtrated and made up to 50 ml volume by addition of demineralized water.

Analytical method

The determination of Na, K and Mg in samples of feed mixtures was carried out with the flame atomic absorption spectrometer 906 AA Elite (GBC, Dandenong, Australia) after sample decomposition or extraction. The working wavelengths were 589.0 nm for Na, 766.5 nm for K and 285.2 nm for Mg. The air/acetylene flame was used for all the elements.

Results and Discussion

Analytical characteristics of the process

Limits of detection (m g^{-1}) for solution were calculated as the 3-fold standard deviation of repeated measurements ($n = 10$) of the blanks. The procedural limits of detection (mg kg^{-1}) in extracted and 200-fold diluted sample are summarized in Table I. Running repetitive microwave digestion and leaching of reference materials tested the accuracy and efficiency of the method. The external standards used for calibration were regularly reinserted after every ten samples to reveal a potential shift of initial calibration. The uncertainty of estimates (relative standard deviation) was based on repeated analyses of leached sample DOB-1907 (ten replicates).

Table I Analytical characteristics of proposed method for determination of Na, K and Mg in feed mixtures using FAAS after ultrasonic-assisted leaching

Parameter	Na	K	Mg
3σ, μg l^{-1}]	0.0156	0.147	0.021
LOD, μg g^{-1}	3.12	29.4	4.04
Precision, *RSD*, %	8.26	10.3	9.11

Microwave digestion

The total element concentration in unknown samples was determined before optimizing the leaching procedure. The FAAS analysis after microwave decomposition was used as the control technique for the determination of Na, K and Mg. For the raw materials and feed mixtures analyzed, the total amounts of Na, K and Mg are presented in Table II as the minimum and maximum values found. Samples of rape seed, sunflower scrap, rape fodder plant and clover hay represented vegetal raw materials that are generally characterized by low Na content. In the case of analytical method used, Na was not detected. The Mg

content varied in tenths of percent (w/w) and the K content did not exceed 2 %. In analysed animal raw materials (three brands of meat and bone meal and two brands of fish meal) and feed mixtures (five samples of feed mixtures for cattle, five for poultry and five for pigs), Mg also ranged in tenths of percent and K roughly from 0.5 to 2 %. In this case, Na was determined in tenths of percent.

The reference materials NIST SRM 1577b, GBW 07603, GBW 07604 and GBW 07605 and the inland control samples cocoa and clover were included in as a control for digestion and FAAS analysis. These results are summarized in Table II. For reference material analysed, the statistical analysis proved that the content of Na, K and Mg found tallied with the declared amount.

Table II Determination of Na, K and Mg in feed mixtures, raw materials and reference materials using FAAS after microwave-assisted decomposition

		Na, $\mu g\ g^{-1}$	K, $\mu g\ g^{-1}$	Mg, $\mu g\ g^{-1}$
Raw materials	Vegetal	< *LOD*	8678-19726	2913-9108
	Animal	5321 – 11809	5028-18515	2095-3577
Feed mixtures	Cattle	1345 – 7761	6713-14214	1627-5336
	Poultry	2211 – 2382	6564-7273	1763-2119
	Pigs	2156 – 3471	6277-10629	1918-3032
GBW 07603	Found	18821 ± 983	9445 ± 566	4609 ± 330
	Declared	19600 ± 1000	9200 ± 600	4800 ± 300
GBW 07604	Found	208 ± 11	13915 ± 670	6841 ±312
	Declared	200 ± 10	13800 ± 400	6500 ± 300
GBW 07605	Found	42 ± 2	16536 ± 421	1726 ± 99
	Declared	44 ± 4	16600 ± 600	1700 ± 100
NIST SRM 1577b	Found	2383 ± 110	9998 ±512	619 ± 29
	Declared	2420 ± 60	9940 ± 20	601 ± 28
Cocoa (in-home	Found	< *LOD*	15400 ± 600	4460 ± 187
control sample)	Declared	< *LOD*	16600 ± 400	5130 ± 110
Clover (in-home	Found	189 ± 15	37801 ±1191	2108 ± 523
control sample)	Declared	200 ± 11	40480 ± 1350	2330 ± 112

Leaching

In order to describe the influence of various parameters on the leaching process, the ground feed mixture DOB-1907 (for cattle), sunflower scrap (vegetal material) and meat-and-bone meal (animal material) and the reference materials GBW

07603 and GBW 07604 were examined. Water, nitric acid, hydrochloric acid, and their mixtures (in one case with H_2O_2) were employed for the extraction experiment.

After having established the initial conditions (the mass of sample 0.5 g, the temperature of 25 °C and sonification time 45 min), the influence of leaching agent present was evaluated. For each sample tested, five extractions were carried out. The results are expressed as recovery of analysis (R [%], R = leached amount/total amount ×100) and summarized in Table III. The extraction efficiencies strongly depend on the leaching mixture used. The lowest efficiencies were observed for water. The presence of acids as well as hydrogen peroxide improved leaching process. The best results were reached for the mixture of 1 mol l^{-1} HCl, 3 mol l^{-1} HNO_3 and H_2O_2 (mixture "G").

Considering the results of the leaching process, the influence of temperature in the presence of the best leaching mixture (20 ml of mixture 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 1 ml 30 % v/v H_2O_2) was evaluated for 0.5 g samples of feed mixture DOB 1907 and 45 min sonification in the range of 25-75 °C. The results are shown in Fig. 1. The increase in temperature led to more effective extraction of Na, K and Mg. The leaching recoveries were practically constant above 60 °C.

For the 0.5 g of feed mixture DOB 1907 and the leaching mixture 20 mL of 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 1 ml H_2O_2 at 60 °C, the influence of sonification time was evaluated for 5-60 min. A prolonged time improved extraction recovery of Na, K and Mg. For all elements, sonification time of 30-40 min ensures constant recovery.

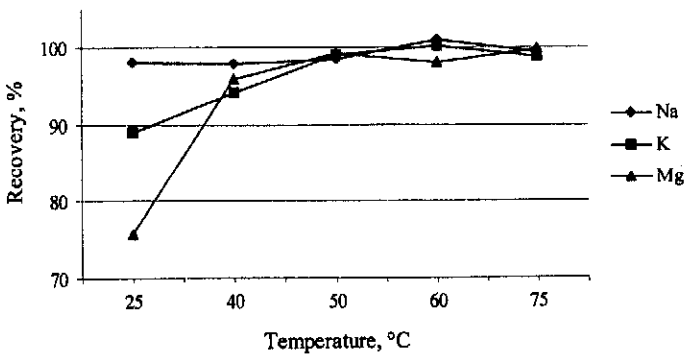

Fig. 1 The influence of the ultrasonic bath temperature on the extraction efficiency

In the next step, a similar extraction technique was tested using a rotator shaker. The mass 0.5 g of feed mixture DOB-1907 was treated with boiling 20 ml of 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 1 ml H_2O_2 and shaken with the rotator shaker at 10 rpm. The influence of leaching time on recovery of analysis was observed (Fig. 2). Na, K and is completely extracted after four hours.

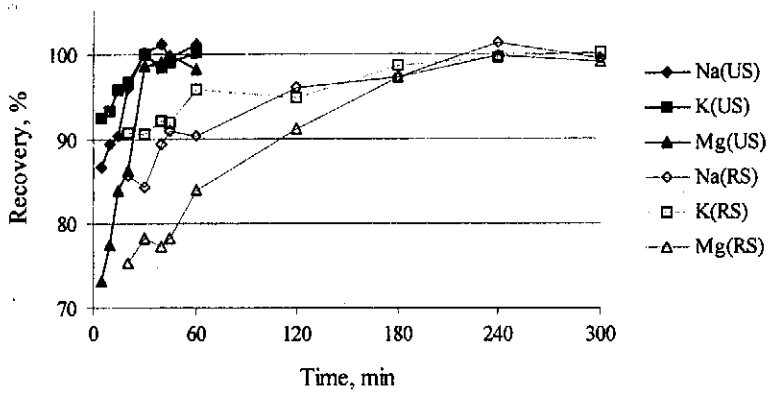

Fig. 2 The influence of leaching time on the extraction efficiency for the ultrasonic bath (US) and the rotator shaker (RS)

Table III The extraction efficiency of tested agents

	Item	R, % for the leaching mixture						
	Sample	A	B	C	D	E	F	G
Na	DOB-1907	45.5	89.6	80.5	98.7	100.2	89.6	98.0
	Meat-and-bone meal	39.2	33.3	96.1	98.1	98.1	86.8	98.3
	GBW 07603	47.8	78.9	82.6	95.4	101.0	92.3	100.3
	GBW 07604	42.8	85.6	84.2	92.6	99.5	94.6	98.7
K	DOB-1907	32.4	80.3	73.9	79.6	88.7	81.7	88.9
	Sunflower scrap	45.7	95.4	82.7	78.7	95.9	96.4	95.2
	Meat-and-bone meal	32.0	32.0	72.0	64.0	51.4	76.0	89.0
	GBW 07603	44.6	94.2	87.6	80.3	96.1	97.9	99.6
	BW 07604	42.1	91.8	78.9	79.2	97.2	98.7	101.3
Mg	DOB-1907	38948	96.2	90.6	90.6	90.6	96.2	97.2
	Sunflower scrap	38728	50.5	52.7	58.2	53.8	59.3	75.8
	Meat and bone meal	38731	38741	70.0	80.0	60.0	65.0	82.6
	GBW 07603	38938	48.6	57.9	65.2	78.2	69.5	90.6
	GBW 07604	38847	51.4	52.3	59.7	76.6	70.9	92.1

A – 20 ml H_2O
B – 20 ml 2 mol l^{-1} HCl
C – 20 ml 2 mol l^{-1} HNO_3
D – 20 ml 1 mo l^{-1} HCl + 2 mol l^{-1} HNO_3
E – 20 ml 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3
F – 20 ml 3 mol l^{-1} HCl + 1 mol l^{-1} HNO_3
G – 20 ml 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 1 ml 30 % v/v H_2O_2

The extraction process using the rotatory shaker is time-consuming and non-effective.

After establishing standard extraction condition (sonification 30 min and temperature 60 °C), the dependence of recovery on leached sample mass (0.1-2 g) was studied for two total volumes. The first was 20 ml 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 1 ml H_2O_2. The second volume was 40 ml 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 2 ml H_2O_2 (Fig. 3). For all studied elements, satisfactory recoveries were obtained with the mass of sample up to 0.5 g and the first leaching volume (20 ml acid + 1 ml H_2O_2). In the case of the second volume (40 ml acid + 2 ml H_2O_2), acceptable recoveries were found for the mass of sample up to 1 g. For a higher extracted sample mass, decreasing recoveries were observed.

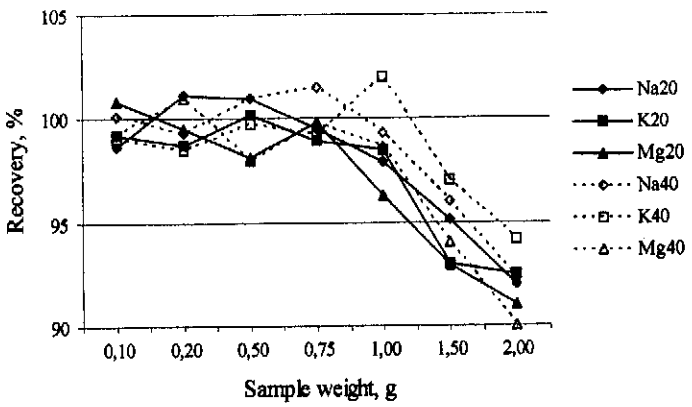

Fig. 3 The influence of the sample mass and the volume of leaching mixture on the extraction efficiency: Na, K, Mg20 – the recovery for 20 ml 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 1 ml H_2O_2; Na, K, Mg40 – the recovery for 40 ml 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 2 ml H_2O_2

The optimized ultrasound-assisted leaching followed by FAAS was employed to analysis of the feed mixtures, raw and reference materials: a weight about 0.3 g, 20 ml of mixture 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 1 ml 30 % H_2O_2, the ultrasonic bath, 60 °C, 30 min. All samples were leached ten-times. These results were compared with the result of mineralized samples analysis (digested six-times). The results are summarized in Table IV (mean value ± standard deviation). Considering the results of the leaching process, sodium and potassium are easily extractable. Magnesium was found to be partially extractable. Especially in case of magnesium, the extraction efficiencies (Figure IV) strongly depend on the kind of analysed material. In the case of Na and K, the extraction efficiencies were similar and the statistical test proved that recoveries approached 100 %.

Table IV The results of FAAS analysis of extracted and digested samples

Feed mixtures	Na, $\mu g.g^{-1}$		K, $\mu g.g^{-1}$		Mg, $\mu g.g^{-1}$	
Raw materials	Extraction	Digestion	Extraction	Digestion	Extraction	Digestion
Meat-and-bone meal	5180 ± 171	5280 ± 250	4920 ± 270	4990 ± 260	1730 ± 80	2040 ± 100
Fish meal	11400 ± 500	11500 ± 470	17900 ± 620	18500 ± 666	243 ± 120	2820 ± 110
Sunflower scrap	< *LOD*	< *LOD*	19400 ± 760	19700 ± 780	8220 ± 330	9130 ± 310
Rape fodder	< *LOD*	< *LOD*	8660 ± 320	8610 ±320	2290 ± 120	2950 ± 710
DOB-1907	1880 ± 90	1870 ± 90	14200 ± 560	14300 ±600	5210 ± 170	5280 ± 120
KA-1913	2710 ± 90	2690 ± 90	7030 ± 340	7050 ± 310	2390 ± 90	2410 ± 90
Cocoa (inland CS)	< *LOD*	< *LOD*	14300 ± 700	15400 ± 620	3880 ± 220	4470 ± 190
Clover (inland CS)	187 ± 20	189 ± 15	37900± 160	37800±120	1900 ± 230	2110 ± 220
GBW 07603	18600 ± 500	19000 ± 530	8920 ± 440	8740 ± 370	2990 ± 190	3800 ± 170
GBW 07605	46 ± 3	45 ± 1	15300 ± 790	16000 ± 740	1090 ± 72	1370 ± 59

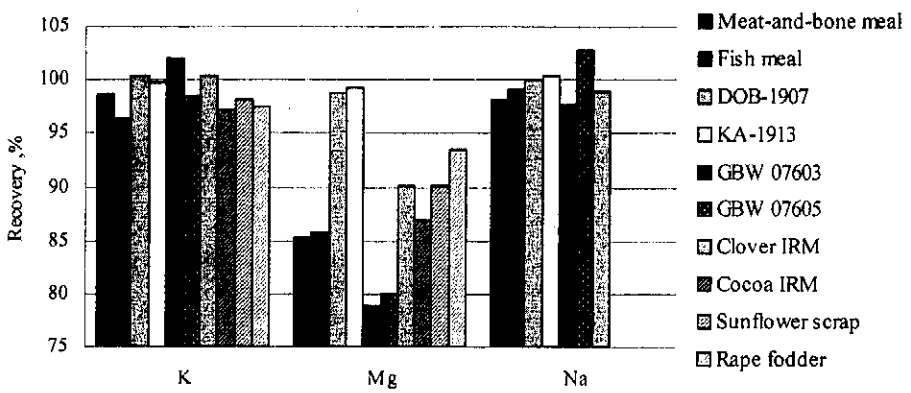

Fig. 4 The extraction efficiency for optimized leaching method

Conclusion

Ultrasound-assisted leaching as a sample preparation procedure prior to FAAS was tested to be suitable for routine determination of Na, K and Mg in samples of feed mixtures. The optimizing of extraction mixture composition, sonification time, and temperature and sample mass was carried out and led to the proposed method: a 0.2-0.5 g sample with 20 ml 1 mol l^{-1} HCl + 3 mol l^{-1} HNO_3 and 1 ml H_2O_2 is leached in the ultrasonic bath at 60 °C for 30 min. The accuracy of the study was validated with the reference materials GBW 07603, GBW 07604, GBW 07605, and the inland control samples of cocoa and clover.

For sodium and potassium, the leaching recoveries approached 100 %. In the case of magnesium, it was found that it is partially extractable. The extraction recovery ranged from 80 to 100 % and significantly depended on the kind of material analysed.

The proposed procedure of analysis of animal feed for sodium and potassium by ultrasound-assisted leaching with the acids mixture prior to analysis by flame atomic absorption spectroscopy is simple, quick and cheap and can be alternatively used to a microwave-assisted digestion or another time-consuming and more expensive total acid digestion. Considering the procedure detection limits (3.1 mg kg^{-1} for Na, 29 mg kg^{-1} for K and 4.0 mg kg^{-1} for Mg) and total amount of element in feed mixtures (Na 1.3-7.8 g kg^{-1}, K 6.3-14.2 g kg^{-1}, Mg 1.6-5.3 g kg^{-1}), the above-mentioned method is convenient. The recommended procedure is a supplement to the XRF method for determination of P, Ca and microelements in feed mixtures [23]. Use of a large-capacity ultrasound bath ensures a high sample throughput (approximately 30 samples at once) in agricultural laboratory at low cost.

Acknowledgements

Financial support from the Ministry of Education, Youth and Sports of the Czech Republic within project MSM 0021627502 used in this work is greatly appreciated.

References

[1] Clum N.J., Fitzpatrick M.P., Dierenfeld E.S.: Zoo Biol. **15**, 525 (1996).
[2] Pollard G.V., Richardson C.R., Karnezos T.P.: Anim. Feed Sc. Tech. **98**, 121 (2002).
[3] Smith A., Rose S.P., Wells R.G., Pirgozliev V.: Brit. Poulry Sci. **41**, 598 (2000).
[4] Dy Peñaflorida V.: Aquaculture **172**, 281 (1999).
[5] Seiler H.G., Sigel A., Sigel H.: *Handbook on Metals in Clinical and Analytical Chemistry,* C.V. Mosby Company, St. Louis, Washington DC, Toronto, 1987.
[6] Van Loon J.C.: *Selected Methods of Trace Metal Analysis. Biological and Environmental Samples*, Wiley, New York, 1985.
[7] Hardisson A., Rubio C., Baez A., Martin M.M., Alvarez R.: Eu. Food Res. Tech. **212**, 175 (2001).
[8] Puong T.D., Chuong P.V., Khiem D.T., Kokot S.: Analyst **124**, 553 (1999).
[9] Kiliç Z., Acar O., Ulasan U., Ilim M.: Food Chem. **76**, 107 (2002).

[10] Chyi P., de la Fuente M., Barrado E., Vega M., Philips C.: Fresenius J. Anal. Chem. **361**, 343 (1998).
[11] Fingerová H., Koplík R.: Fresenius J. Anal. Chem. **363**, 545 (1999).
[12] Gundersen V., McCall D., Bechmann, I.E.:. J. Agr. Food Chem. **49**, 3808 (2001).
[13] Hokura A., Matsuka H., Katsuki F., Haraguchi H.: Anal. Sci. **16**, 1161 (2000).
[14] Lavado R.S., Porcelli C.A., Alvarez R.: Soil Till. Res. **62**, 55 (2001).
[15] Rodushkin I., Ruth T., Huhtasaari A.: Anal. Chim. Acta **378**, 191 (1999).
[16] Borkowska-Burnecka J., Miazga W., Zyrnicki W.: Chem. Anal. (Warsaw) **45**, 429 (2000).
[17] Hua K.M., Kay M., Indyk H.E.: Food Chem. **68**, 463 2000).
[18] Mc Guire J.S., Hite D.A.: J. AOAC Int. **81**, 923 (1998).
[19] Cook K.K.: J. AOAC Int. **80**, 834 (1997).
[20] Nas S., Gokalp H.Y., Sahin Y.: Z. Lebensm. Unters. Forch. **196**, 32 (1999).
[21] Omote J., Hisayuki K., Toda K.: Anal. Chim. Acta **307**, 117 (1995).
[22] Richardson D.H.S., Shore M., Hartree R., Richardson R.M.: Sci. Total Environ. **176**, 97 (1995).
[23] Černohorský T., Pouzar M., Krejčová A.: Chem. Anal. (Warsaw) – in press.
[24] Borkowska-Burnecka J., Wisz J., Zyrnicki W.: Chem. Anal. (Warsaw) **48**, 115 (2003).