

Wet Digestion Techniques for Determination of Chromium in Food Sample by Differential Pulse Stripping Voltammetry

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Currently, a large amount of harmful heavy metals is discharged into the soil environment, which will eventually affect the quality of food and harm human health through the food chain. Therefore, it is of great theoretical and practical significance to establish an analytical method for the determination of trace heavy metals in food. In this paper, a differential pulse stripping voltammetric method for the determination of chromium in food was established, and the sample was pretreated with a wet digestion method suitable for rapid detection in the field. Finally, this HNO₃-H₂O₂ wet digestion electrochemical determination method was evaluated.

Keywords: HNO₃-H₂O₂; Wet digestion; Electrochemistry; Chromium ions; Food sample

1. INTRODUCTION

Hexavalent chromium compounds contain chromium in the hexavalent state [1–4]. In the clinic, the harm of hexavalent chromium compounds to the human body is usually manifested in three aspects, namely, damage to the skin, damage to the respiratory system and damage to the digestive system. Long-term exposure to chromate can easily cause gastritis, gastric ulcers and intestinal ulcers. Excessive intake of hexavalent chromium can lead to serious renal failure and even cancer. Chromium compounds have been classified as a class of human occupational carcinogens by the International Cancer Research Institute [5–8]. In recent years, the problem of chromium pollution in soil and excessive chromium content in agricultural products has received great attention at home and abroad [9–12]. The safety of crops such as vegetables and grains cannot be ignored. Chromium in soil mainly comes from the discharge of chromium-containing wastewater, which mainly comes from leather tanning, dye synthesis, pharmaceutical saccharin organic synthesis, wood anticorrosion treatments, and the metal ferrochrome

industry. Plants are the main source of food for human beings, and chromium pollution can damage human health through the food chain. Chromium in plants mainly comes from water and soil and accumulates in plants through the absorption of elements during plant growth.

At present, the main methods of heavy metal detection and analysis are atomic absorption spectrometry, ultraviolet spectrophotometry, atomic fluorescence, chemiluminescence and electrochemistry [13–18]. Some studies also use X-ray fluorescence spectrometry. The advantage of X-ray fluorescence spectrometry is that it is nondestructive to the sample. Some studies use atomic absorption spectrometry, but such instruments can be expensive. The latest popular detection method is electrochemical stripping voltammetry, which is fast and accurate and can also be used for emergency detection in the field and other environments [19–26]. Among the electrochemical analysis methods, anodic stripping voltammetry combines potentiostatic electrolytic enrichment with voltammetric determination. This method can be used for the continuous and simultaneous determination of many metal ions and has high sensitivity.

The main component of food is organic matter. To accurately determine the heavy metal elements in food, it is necessary to destroy the organic matter in the sample. In food analysis, the food generally exists as a solid sample, so it is necessary to change the components to be tested into soluble chemical forms by digestion or extraction and then perform determination. The basic principles of trace analysis for sample pretreatment are to be able to separate the tested components from the sample without pollution and loss. Pretreatment cannot introduce the tested components or interfering substances. Pretreatment methods can be divided into wet digestion, dry ashing, high-temperature melting and microwave digestion. Wet digestion refers to a decomposition method that can aid in the direct determination of an analyte from a solid sample by chemical reaction under the conditions of heating with oxidants such as acid and hydrogen peroxide [27–31]. This method is widely used in trace analysis because of its strong adaptability, good reproducibility, low loss of volatile elements and convenience for simultaneous determination of multiple elements.

Attention to food safety is reflected in the detection of heavy metals in food. In the detection of heavy metal ions, there are many drawbacks, such as complex preparation steps, poor water solubility and serious interference of other ions. Chromium pollution will seriously impact nature because it is not easy to degrade and readily accumulates. Therefore, determining how to detect heavy metals in actual food samples is an urgent problem. At present, special electrochemical instrumentation for detecting heavy metals is mainly used for water quality monitoring, and it is not widely used in the detection of food and agricultural products. Therefore, it is of great significance to study special equipment for the low-cost and high-sensitivity electrochemical detection of heavy metals. Before the use of a heavy metal electrochemical detector, the sample must be pretreated. Therefore, it is very important to study a set of pretreatment technologies coupled with electrochemistry for the determination of heavy metals. In this paper, we modified a glassy carbon electrode by preplating mercury film. The influence of the mercury film on the peak current of the heavy metal chromium was determined. We also used $\text{HNO}_3\text{-H}_2\text{O}_2$ wet digestion technology to pretreat chromium in food. We studied the digestion scheme of chromium in food and determined the optimal digestion conditions. Finally, we established a $\text{HNO}_3\text{-H}_2\text{O}_2$ wet digestion electrochemical determination scheme.

2. EXPERIMENTAL

Potassium nitrate, potassium dichromate, mercuric chloride, chromium standard solution, anhydrous sodium acetate, diethylenetriaminepentaacetic acid (DPTA) and glacial acetic acid were purchased from Shanghai Aladdin Bio-Chem Technology Co., LTD. Distilled water was used throughout the experiments. A glassy carbon electrode was used as the working electrode. The electrode surface was modified with mercuric chloride solution to form a mercury film electrode for selective determination of chromium. A chromium standard stock solution was prepared by dissolving an accurately weighed amount of 1.4135 g of solid potassium dichromate in a 100 mL volumetric flask with secondary water and fixing the volume to obtain a chromium concentration of 1.0 mg/mL. A chromium standard solution was prepared by adding 1.0 mL of the chromium standard stock solution into a 100 mL volumetric flask and diluting with water to obtain a chromium concentration of 10 µg/mL. A buffer solution was prepared by accurately weighing 95.88 g of potassium nitrate, 14.33 g of anhydrous sodium acetate and 3.725 g of DTPA, placing them in a 500 mL volumetric flask, adding 310 µL of glacial acetic acid, and using distilled water to fix the volume.

Food samples should be pretreated before digestion. After removing the sundries from a dried grain sample, it was ground into powder in an agate mortar, passed through a 40 mesh standard inspection sieve, stored in a glass bottle, and refrigerated until use. Vegetables, fruits and other fresh samples with a high moisture content were homogenized by a homogenizer or chopped with a stainless steel knife to ensure that there were no large particles. These samples were stored in plastic bottles and kept in cold storage until use. For the wet digestion process, sample masses up to 5 g were transferred to a quartz vessel, mixed with 8 mL of 30% H₂O₂ and fixed on a microwave rotor. The microwave rotor was positioned inside a single reaction chamber cavity containing an HNO₃ solution. The microwave system was heated using a 20 min ramp and held for 10 min at temperatures ranging from 190 to 250 °C.

Differential pulse stripping voltammetry (DPSV) was applied to detect trace levels of chromium under optimized conditions. The DPSV parameters were optimized such that chromium was deposited for 120 s at -2.0 V.

3. RESULTS AND DISCUSSION

First, we investigated the effect of pulse width on the peak current of chromium in the range of 10 ~ 90 ms. Figure 1A shows the linear relationship between the peak current and pulse width. The results show that the peak current increases with increasing pulse width. When the pulse width is 60 ms, the peak current reaches its maximum, and when it is 60 ~ 90 ms, the peak current shows a downward trend. This result shows that the pulse width is the main factor affecting the peak current of chromium. Considering that the pulse width is shorter than its period, it is generally set to 40-80 ms. Obviously, the pulse width will affect the peak current generation. It can be seen from the experiment that the best pulse width is 60 ms.

The difference in step potential mainly affects the distance between the data points of the peak current curve, thus affecting the detection effect. The peak conditions were investigated in steps of 2, 5, 8, 10, 12, 14, 16, 18, and 20 mV. It can be seen from Figure 1B that the step size at 2-10 mV will have a certain impact on the peak current of the solution at the same concentration, but it tends to be stable at 10-20 mV. With increasing step size, the peak potential is shifted forward, so the step size is 10 mV.

The accumulation time on the surface of the working electrode has a direct effect on the peak current and the determination of the heavy metal chromium, and the equilibrium time will affect the peak pattern (Figure 1C and 1D). The effects of the concentration time in the range of 15-180 s and equilibrium time in the range of 1-20 s on the peak current of chromium were measured with other electrochemical parameters held constant. With increasing enrichment time, the peak current of chromium increases first, then decreases gradually, and reaches a maximum at 120 s. This result occurs because with increasing enrichment time, an increasing number of electroactive substances with small concentrations in the solution are concentrated on the surface of the working electrode, resulting in a reduction reaction [32,33]. However, after a certain enrichment time, the surface of the electrode is covered by an oxidized chromium complex, which changes the performance of the working electrode and leads to a decrease in the peak current. Therefore, 120 s is the best enrichment time. The influence of the equilibrium time on the peak current was investigated with the enrichment time fixed. Figure 1D shows that the equilibrium time has an effect on the peak current at 0 ~ 5 s. When the equilibrium time is 5 ~ 20 s, the peak current is basically unchanged. Therefore, the balance time is set to 5 s to save the experiment time.

Finally, we optimize the sampling period and set the sampling period to 0.1, 0.2, 0.3, 0.4, and 0.5 s. It can be seen from Figure 1E that with an increase in the sampling period, the peak height generally increases and then decreases. When the sampling period is 0.3 s, the peak height reaches its maximum value, so the sampling period is 0.3 s.

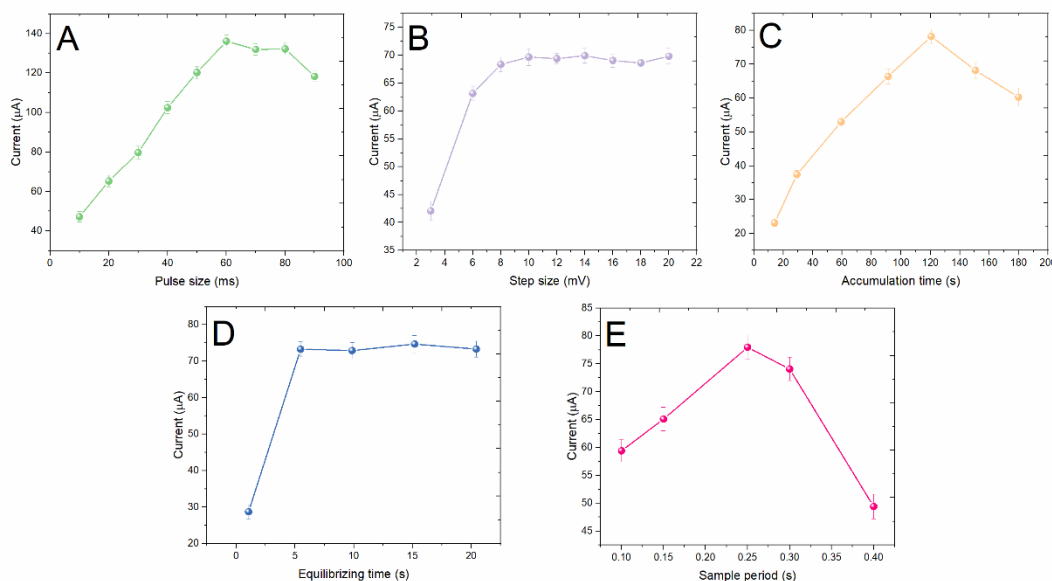


Figure 1. Effect of (A) pulse size, (B) step size, (C) accumulation time, (D) equilibration time and (E) sample period for Cr(VI) sensing. All experiments were conducted in buffer solution.

Mercury chloride solution is selected as the electrode treatment solution. The choice of HgCl_2 solution concentration has a great influence on the electrode sensitivity [34]. Therefore, the electrochemical behaviour of the coated electrode is investigated at different concentrations. The concentrations of HgCl_2 were 0.8, 0.4, 0.2, 0.1, 0.08, 0.05 and 0.025 g/L. The results show that the coating on the electrode surface is obviously uneven or cannot be coated at concentrations of 0.8 and 0.4 g/L. When the concentration of the HgCl_2 solution is varied from 0 ~ 0.2 g/L, the interactions between the active groups on the surface of the electrode and chromium ions in the solution are strongest at 0.08 g/L HgCl_2 , leading to the best separation and enrichment of chromium ions and greatly improving the sensitivity of the analysis. Therefore, the concentration of the electrode solution is set to 0.08 g/L.

The method to determine the dissolution peak involves adding a known reference material to the sample, detecting it by a chromium trace element analyser, and determining the quality when the peak height of a certain peak increases. A total of 0.4 mL of sample solution is added to a centrifuge tube containing 1.6 mL of buffer solution, and the electrochemical workstation of the trace element analyser is used for determination. Then, 10 μL of chromium standard solution is added, and the determination is continued. A change in peak height was observed. It is found that the peak height increases with the addition of the standard solution near the peak potential of -1.34 V, and the peak position of chromium is determined to be -1.34 V.

The selection of sensitivity has a great influence on the dissolution peak current of the elements to be measured. The single variable principle is used to investigate the influence of sensitivity factors. Generally, a relatively large sensitivity value is selected to ensure that the test current will not overflow, and the range of the reaction current can be roughly known after the test [35]. Then, the closest sensitivity is selected according to the reaction current range. Therefore, the general test is performed under the condition that the sensitivity is closest to the maximum value of the system.

The electrochemical determination of chromium involves measuring the relative electrode potential. In the primary battery system, the conductivity of the solution can be improved by adding supporting electrolyte to improve the measurement accuracy of the electrode potential. The main components in the bottom solution are KNO_3 , DTPA, and HAc-NaAc . In the process of determining the heavy metal chromium, the concentration of components may affect the dissolution peak sensitivity or peak type of chromium, so these factors need to be determined.

The solubility of the supporting electrolyte should be high enough, at least 50-100 times that of the electroactive substance. At the same time, the electrolyte should not be able to react with the solvent or the substance related to the electrode reaction in the system. KNO_3 and Na_2SO_4 are often used as supporting electrolytes in laboratory aqueous solutions. In this experiment, KNO_3 was used as the supporting electrolyte. Because the oxidation potential of KNO_3 is more positive than that of water and hydrogen peroxide, the reduction potential of potassium ions is more negative, and the potential window is larger than that of water. The optimal amount of the supporting electrolyte KNO_3 was quantified. Two millilitres of a 10 $\mu\text{g/mL}$ chromium standard solution and 30 mL of buffer solution are added to a 50 mL beaker, and the effects of KNO_3 concentrations of 0.2, 0.5, 1, 1.5, 2, 2.5 and 3 M on the oxidation peak current are tested, as shown in Figure 2A. Figure 3A shows that different concentrations of KNO_3 will affect the peak pattern of chromium. With increasing potassium nitrate concentration, the peak current shows an upward trend. When the concentration reaches 2 M, the peak height reaches its maximum

value, and the peak current of KNO_3 continues to increase, which is because the conductivity of the solution is determined by the concentration of KNO_3 . The higher the concentration of KNO_3 is, the greater the concentration of ionized ions in the solution is, and the higher the conductivity is; the peak current also increases with increasing KNO_3 concentration. However, when the solution reaches solution equilibrium, the concentration of KNO_3 will no longer affect the peak current. Therefore, we chose the concentration of KNO_3 to be 2 M.

DTPA, a chelating agent of metal ions, has a strong ability to chelate high-valent metal ions. We investigated the effect of the DTPA concentration on the peak current of chromium in the range of 1–8 g/L. It can be seen from Figure 2B that when the DTPA concentration increases from 1 g/L to 8 g/L, the chromium peak potential shifts negatively. The peak current of chromium increases with increasing DTPA concentration. When the DTPA concentration is 6–8 g/L, the peak current changes minimally, and when the DTPA concentration is 7 g/L, the peak current is the most stable.

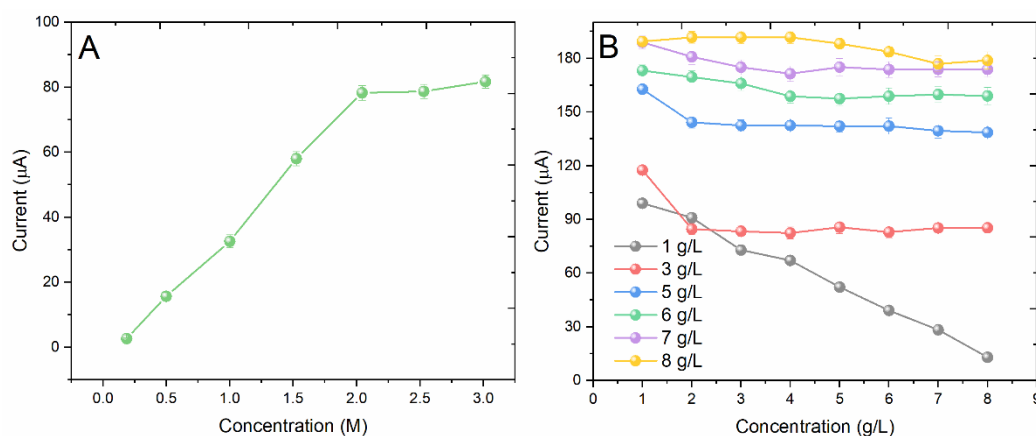


Figure 2. The effect of (A) the concentration of KNO_3 and (B) the amount of DTPA on the stripping peak current. All experiments were conducted in buffer solution with the optimized conditions indicated in Figure 1.

The selection of wet digestion conditions mainly includes the type of acid added, the amount of acid added, the digestion time and the digestion temperature. The boiling point of sulfuric acid is high, so it is easy to produce high-temperature carbonization when digesting, and explosions can easily occur when perchloric acid is mixed with organic matter. In this experiment, H_2O_2 and HNO_3 were selected as oxidants for digestion. The effect of the pH value on the chromium peak current is shown in Figure 3. It can be seen from the figure that there is no oxidation peak current when the solution has no colour change or is dark purple (a). When the solution just changed colour to red and lavender, the oxidation peak current reached its maximum (b), which is because the electrochemical determination of chromium needs to provide an HAc - NaAc buffer environment ($\text{pH} = 6$), and the oxidation peak current reaches its maximum in the presence of a peracid. The peracid or peralkaline digestion solution (c) will affect the buffer environment. The results show that the oxidation peak current can reach its maximum when the solution just changes colour, so it is the best when the solution just changes colour after titration.

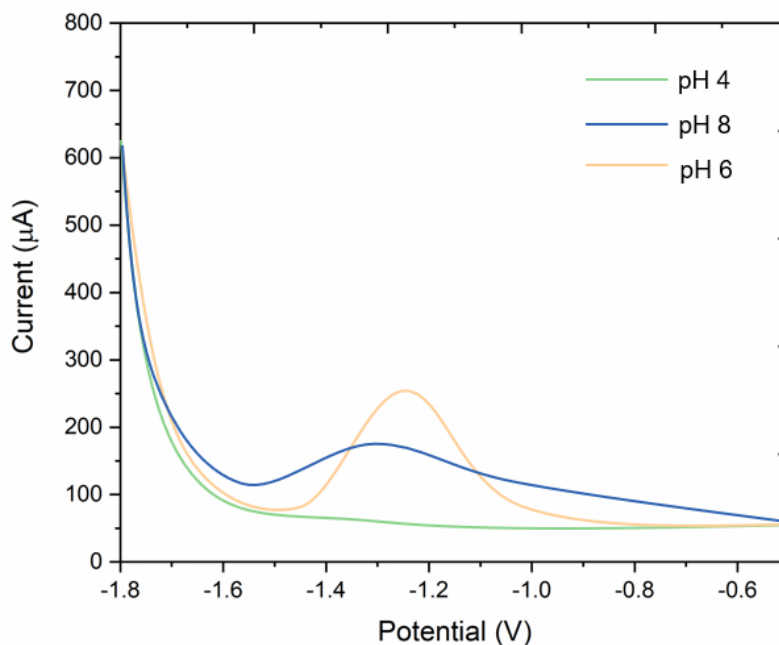


Figure 3. Effect of pH on the stripping peak current. All experiments were conducted in buffer solution with the optimized conditions indicated in Figure 1.

When the sample is placed at room temperature, it will be digested slowly, which will cause part of the organic matter to be digested. We investigated the effect of different temperatures (120 ~ 180 °C) on the oxidation peak current of millet: 1 g of the millet sample, 10 mL of nitric acid and 36 mL of hydrogen peroxide were added into a set of conical flasks, and digestion was performed at 120, 130, 140, 160, and 180 °C. The results are shown in Figure 4A and indicate that the concentration of chromium in the millet first increases and then decreases with increasing temperature. According to the literature, an increase in pressure occurs due to gases produced by the oxidation of the organic matrix and H_2O_2 decomposition [36]. At 130 °C, the concentration is the highest, and the digestion effect is the best. This result occurs because at relatively low temperatures, with increasing temperature, the reaction speed will be greatly accelerated. With an increase in temperature, rapid high-temperature heating will cause the solution to evaporate too fast and form excessive steam, resulting in loss of the sample solution and a low determined chromium concentration. In addition, an excessively high temperature will lead to a large amount of acid volatilization very rapidly, preventing the sample from being completely digested, which will also lead to low chromium concentration results. At 130 °C, the chromium concentration reaches its maximum, so 130 °C is selected as the best temperature. However, careful evaluation of the sample mass and H_2O_2 concentration should be performed to obtain a safe procedure [37]. Additionally, when comparing the pressures of the digestion systems using H_2O_2 and HNO_3 , a higher pressure was observed for H_2O_2 , probably due to increased pressure from H_2O_2 decomposition [38].

According to the literature, lactose (the major disaccharide present in milk) can be completely oxidized to formate in the presence of H_2O_2 at room temperature [39]. Regarding proteins, according to the literature, peptide bond cleavage is observed, and the subsequent oxidation of methionine, cysteine, tryptophan and histidine can also occur with H_2O_2 at alkaline pH or elevated temperature [40]. The

effects of the amount of nitric acid and hydrogen peroxide on the results of chromium determination were investigated. In a set of conical flasks, 1 g of millet was added, followed by 3, 5, 8, 10, 12, and 13 mL of nitric acid, separately; then, the samples were digested at 130 °C in the presence of 36 ml of hydrogen peroxide and were diluted to 10 ml for determination. See Figure 4B for the results. The results showed that when the amount of nitric acid added was from 3 to 13 mL, the concentration of chromium first increased and then decreased, reaching a maximum at 10 mL. The fixed amount of nitric acid was 10 mL, and 28, 32, 36, 38, 40, 42, and 44 mL of hydrogen peroxide was added. The effect of different amounts of H_2O_2 was then investigated; see Figure 5C for the determination results. The results showed that with increasing amounts of hydrogen peroxide, the concentration of chromium increased. When the H_2O_2 volume reached 38 mL, the concentration of chromium remained basically unchanged, indicating that the millet digestion was complete when the H_2O_2 amount was 38 mL. Therefore, 10 mL and 38 mL were selected as the best volumes of HNO_3 and H_2O_2 , respectively.

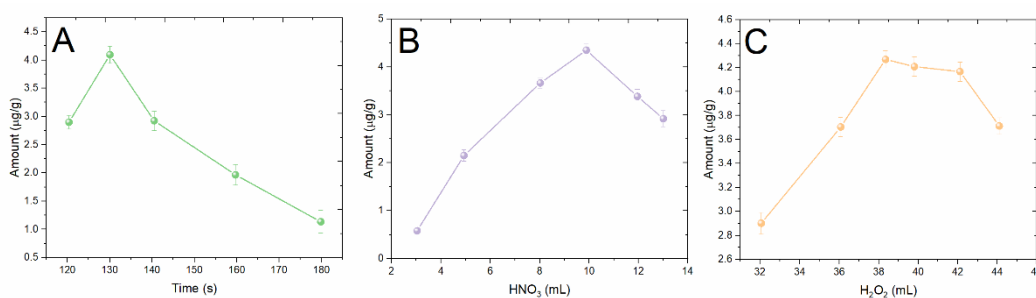


Figure 4. The effect of the (A) digestion temperature, (B) amount of HNO_3 and (C) amount of H_2O_2 on the stripping peak current. All experiments were conducted in buffer solution with the optimized conditions indicated in Figure 1.

The HNO_3 - H_2O_2 wet digestion electrochemical method was established. It is not difficult to carry out the electrochemical determination of chromium standard solutions. As long as a certain neutral buffer environment is provided, the electrochemical determination of chromium can be carried out. The determination results are shown in Figure 5. The sample after wet digestion showed a more distinct peak from chromium than that without digestion. The electrode was placed in the buffer solution of acetic acid and sodium acetate with a pH value of 6. The peak current of the chromium ion in millet was linear with its concentration in the range of 10-100 $\mu\text{g/L}$. The limit of detection was 2.5 ppb.

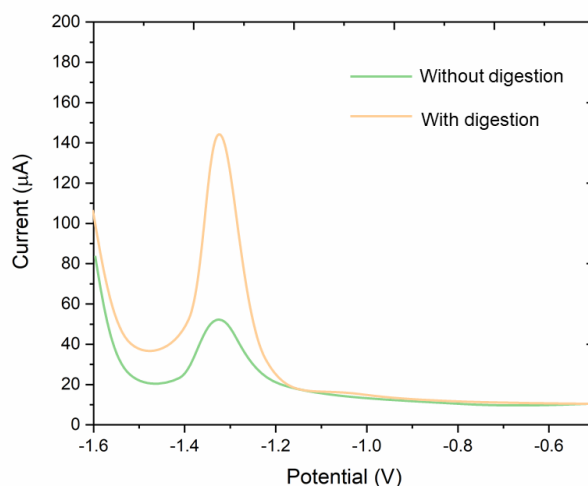


Figure 5. DPVs of chromium-containing samples with and without wet digestion. All experiments were conducted in buffer solution with the optimized conditions indicated in Figure 1.

A 1 g millet sample was used for digestion and determination, and a standard recovery experiment was carried out. The results are shown in Table 1, and the recovery is between 96% and 102%.

Table 1. Real millet test using the proposed $\text{HNO}_3\text{-H}_2\text{O}_2$ wet digestion electrochemical method.

Sample	Detection ($\mu\text{g/g}$)	Add ($\mu\text{g/g}$)	Detection ($\mu\text{g/g}$)	Recovery (%)
1	3.254	3	6.325	101.14
2	3.110	3	6.201	101.49
3	3.204	3	6.104	98.39
4	3.365	3	6.123	96.20
5	4.025	3	6.891	98.09

4. CONCLUSION

In this paper, the heavy metal chromium in food was studied and analysed by an electrochemical method combined with digestion technology. A low-cost and fast detection technology suitable for food detection was established. The pretreatment of chromium in food was carried out by nitric acid and hydrogen peroxide wet digestion technology. The optimal digestion scheme of chromium in food was studied. The experimental conditions were determined as follows: the predigestion time was 0 h, the digestion temperature was 130 °C, the volume of 65-68% HNO_3 was 10 mL, and the volume of 30% H_2O_2 was 38 mL. When the pH is slightly alkaline, the peak effect is the best; that is, when the digestion solution just turns red, the effect is the best. The reliability of the $\text{HNO}_3\text{-H}_2\text{O}_2$ wet digestion

electrochemical determination scheme was evaluated by comparison with the actual sample atomic absorption method.

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