DECONTAMINATION OF RADIOACTIVE PHOSPHOGYPSUM

Vacariu Vintila Teodoru[1], Filip Gheorghe[1], Manuela Cazana[2]

[1]Research and Design Institute for Rare and Radioactive Metals.
[2]Ministry of Industries, National Committee for Materials Recycling.

INTRODUCTION

Phosphate fertilizer industry is a source of concern for environment mostly by phosphogypsum resulted as waste in the sulfuric acid attack of phosphate rock. Phosphogypsum is obtained in large amounts and is found in deposits located around fertilizer plants. There are four plants in Romania and the same time four phosphogypsum deposits of approximately 8 millions tons resulted in the dihydrate process of sulfuric acid attack of phosphate rock. The phosphogypsum acumulated all over the world is very large perhaps more than 1 billion tons.

The main problem consists of phosphogypsum radioactivity where all ^{226}Ra from natural sedimentary rock is present being a serious hazard radioactivity source. The radioactivity of romanian phosphogypsum fluctuate between 7 and 30 pCi/g ^{226}Ra.

Due to its radioactivity, restrictions in phosphogypsum use in construction industry or in agriculture have been imposed in many countries. The environmental contamination by phosphogypsum was in attention of many scientists and various studies have been carried out in view of eliminating ^{226}Ra and creating the conditions of its use in various fields (1-4). This study is an atempt to solve this problem.

EXPERIMENTAL

In this work three hypothesis were taken into consideration:

- radium removal from phosphogypsum by treatment with ammonium salt solutions which increase radium sulfate solubility;
- physical concentration of radium with hydrocyclones since experimental data have shown that ^{226}Ra is concentrated in fine grains of phosphogypsum;
- phosphogypsum (calcium sulfate) conversion to sodium or ammonium salts by treatment with sodium hydroxide, sodium carbonate or ammonium carbonate and radium retention on ion exchangers from the solution resulted by dissolution in hydrochloric acid of phosphogypsum conversion residue.

The research work was carried out at laboratory scale and in pilot plant. Phosphogypsum was obtained from SOFERT S.A. Bacau and had an average radioactivity of 17 pCi/g. Technical reagents have been used. Laboratory experiments were carried out in usual glass equipment. The experiments from pilot plant were made in suitable equipment acid and base resistant. ^{226}Ra determination was carried out by emanometric method.

RESULTS AND DISCUSSION

Phosphogypsum treatment by ammonium salts solutions gave no significant results, and the radium rate removal from phosphogypsum was very low. The requirement of unitary washing operations being over hundred.

The hydrocyclone separation using a unit of 100 mm diameter led to two granulometric fractions of phosphogypsum:+45 with radioactivity of 7.8 pCi/g and -45μ with radioactivity 27.9 pCi/g. The +45μ fraction, representing approx.40% weight may be used according to some literature data in the material of

construction industry.

In order to convert calcium sulfate (phoshogypsum) to sodium or ammonium sulphates, the chemical treatment is carried out and the conversion yields obtained are given:90-95% with 700 kg NaOH/t phosphogypsum or 600 kg ammonium carbonate/t phosphogypsum, consumption.

The hydrochloric solution containing radium and calcium, obtained from dissolution of residue resulted in the conversion process of phosphogypsum, by hydrochloric acid with dissolution yield of 98.9% and acid consumption of 600 kg/t phosphogypsum.

The efficiency of various ion exchangers for radium retention from hydrochloric solution was also studied. The best results were obtained with ion exchanger VIONIT CS-34S made by VIROMET S.A.-Victoria (Romania). The loading curve with ^{226}Ra of this resin is given in Fig.1, where experimental conditions are mentioned.

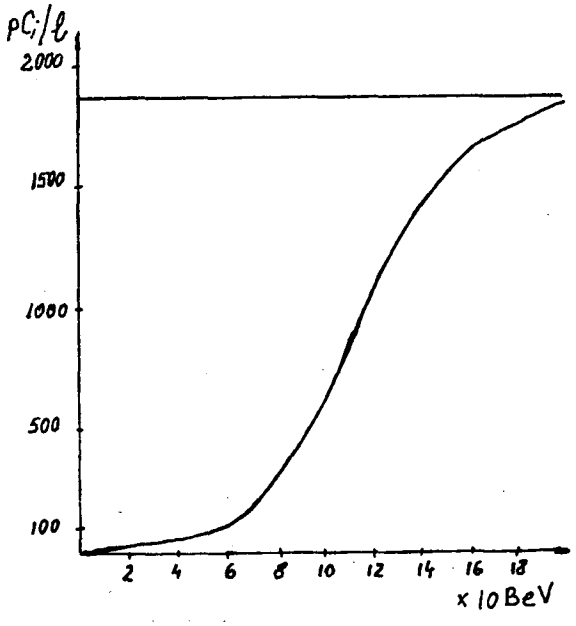

Fig.1. SORPTION ISOTHERM OF ^{226}Ra ON VIONIT CS-34S RESIN.

The pilot plant experimental data have confirmed the laboratory results and led to the flowsheet of the process for radioactive elimination and phosphogypsum conversion as given in Fig.2.

BIBLIOGRAPHY

1. Beretka J.Mathew P.J. Health Pys.1985.1985, 48(1), 87-95.
2. Krisyuk E.M. Parkhomenko v.1., Rep.S.A.S. DDR, 1979, SAAS-250, 1978, 199-204.
3. Moisset J.Proc.Int. Symp.Int.Phosphogypsum, 2 nd, 1986(publ. 1988) 1, 303-17.
4. Lange Paul H.Jr., U.S.A. 146, 568 (cl.423-170; G 21 F 9/28) 27 mar 1979, Appl.821026, 01 aug.1977.

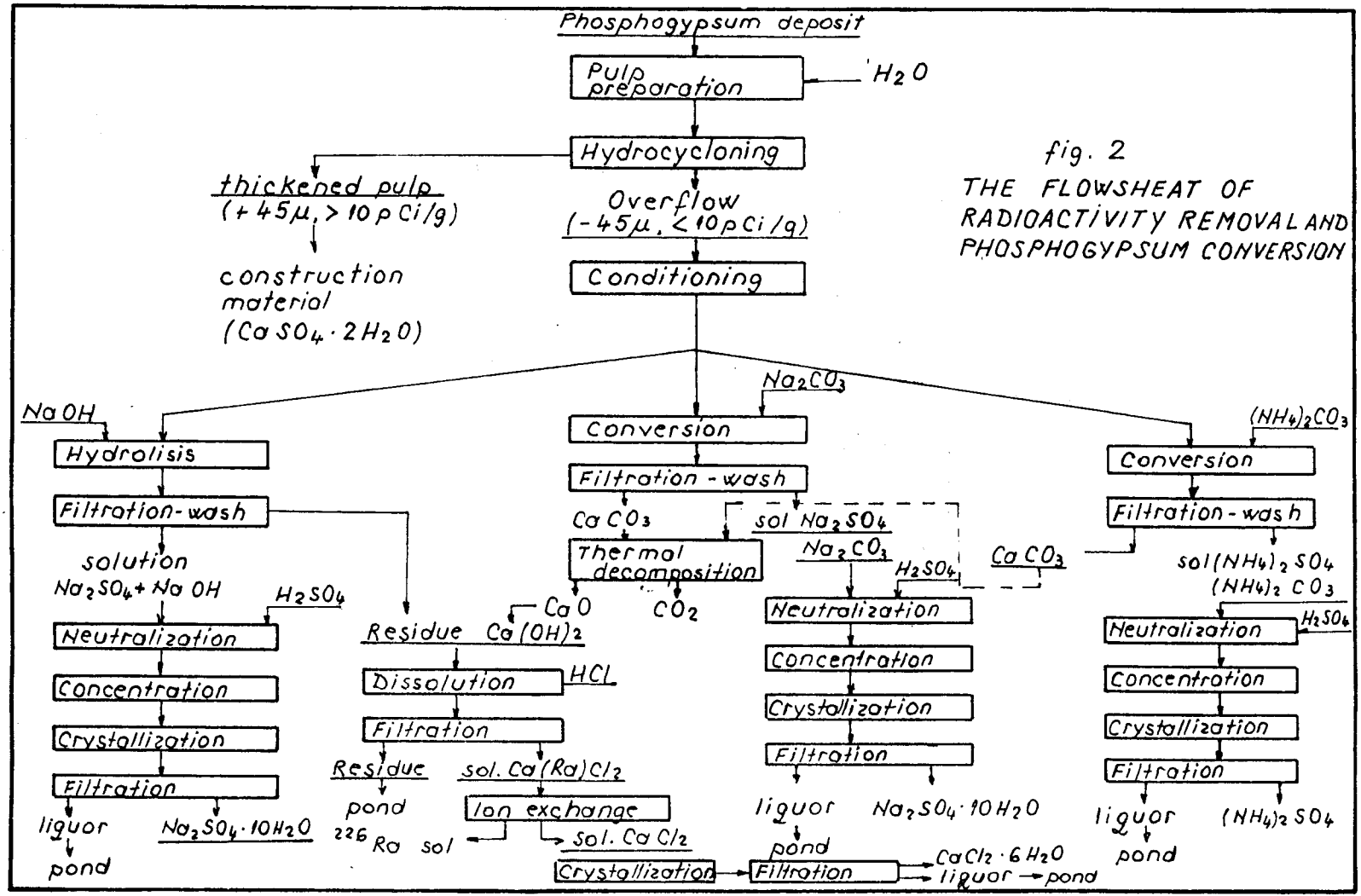

fig. 2
THE FLOWSHEAT OF RADIOACTIVITY REMOVAL AND PHOSPHOGYPSUM CONVERSION