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Kausar, Pervez

Food Processing Laboratory, Faculty of Agriculture, Kyushu University

Nomura, Danji

Food Processing Laboratory, Faculty of Agriculture, Kyushu University

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A New Approach to Pectin Manufacture by Copper Method

Part 1. Preparation of Pectin Pomace, Pectin Extraction and Concentration by Copper Salt

Pervez Kausar and Danji Nomura

Food Processing Laboratory, Faculty of Agriculture,
Kyushu University 46-09, Fukuoka 812

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This study was undertaken purely from processing point of view, hence the conclusions made were also, which better fitted in the process, judged by the experience. It was found that unleached peel can not be dried satisfactorily because of its sticky nature and development of undesirable dark reddish brown color, and that cold leaching seemed more practical than hot leaching when compared on the basis of economy and merits in return. Quality of pectin in citrus material was adversely affected by drying temperature. whereas. moisture contents of the dry pomace and the temperature of peel in its final stage of drying controlled the quality of pectin in the pomace. Yields of cold and hot leached pomace were 11.2 % and 9.6 % respectively and that 7-10 % moisture contained pomace did not show any promising undesirable change during one year of storage when packed in the polythene bags and kept in a dry place. Total extractable pectin yield increased about 3 % and 5 % by cold and hot leaching respectively (due to concentration of pectin in the peel by leaching out other soluble solids), this difference was more promising when yield calculated on qualitative basis. An extract suitable for Cu^{2+} precipitation (0.50-o. 55 % pectin) was obtained by extracting one part weight of pomace with 40 parts of 0.13 % HCl solution (extract pH 1.7) for 45 minutes at 95°C. Best Cu-pectin precipitates were obtained when 20mg of Cu^{2+} as CuCl_2 was added in 100 ml of 0.50-o. 55% pectin contained extract and then pH was adjusted at 2.8 with conc. NH_4OH , after cooling the mixture to about 20°C.

INTRODUCTION

Annual production of Unshu citrus in Japan in the year 1978-79 was about 35.4×10^6 tons, out of which 8×10^6 was processed and the remaining consumed as fresh. Processing leaves behind about 50 % of its weight as waste in the form of peel, which at present is dried in the direct fire-rotary dryer after mixing with calcium oxide and in turn is used as animal feed. At present drying cost of peel is higher than the price fetched from the sale of dry peel as a feed, this burden on the orange juice industry is increasing day by day due to gradual increase in the energy cost. In such circumstances, processor of citrus fruit is left with no choice but to utilize this peel to produce other useful products out of it, and this way cut the cost of its disposal as a feed.

Unshu citrus peel contains about 15 % pectin on dry basis (D. B.) which

shows Japan has a potential to produce enough pectin, which will meet not only the domestic needs but can be exported as well, whereas, at present all the pectin used in various food industries is imported one. Keeping in view these facts this study is undertaken as a first step towards pectin production.

To handle the bulk volume of peel and to run the pectin industry throughout the year, there is no other way but to preserve and stabilize peel by drying (pomace manufacture). Rouse (1967, 1977) and Rouse and Crandall (1978) had reported data on the quality of pectin extracted from the commercially dried citrus pomace. Shepherd and Graham (1952) studied the effect of drying on the quality of pectin by using two temperatures in two stage drying in a forced draft oven. Crandall et al. (1978) published results on the pectin pomace manufacture by using a direct fire rotary dryer at different inlet air temperatures.

To extract pectin from the pomace, which exists as insoluble protopectin within the pomace, it is necessary to convert it into soluble pectin by acid or enzymatic treatment, if it is required to prepare powdered pectin. Up to now almost all the mineral acids have been tried to extract pectin (Huang, 1973). Recently, Rouse and Crandall (1978) extracted pectin from lime and lemon peel by nitric acid. Acid extraction is actually a compromise between pH, time and temperature, to extract maximum pectin by least affecting its quality, keeping in view the handling problem, equipment available and economy etc. Kaseem (1969) used ionizing radiation for hydrolysis of pectin and studied their effect on the properties of extracted pectin. Commercially hydrochloric acid, sulfurous acid and sulfuric acid are being used for extraction of pectin depending upon the producer's choice and modification of final product. Sakai et al. (1978) used protopectinase to hydrolyze protopectin.

After extraction and clarification of pectin solution it needs to be concentrated, which is one of the most expensive step within the pectin manufacture process. 3-4% pectin concentrate can be efficiently and economically precipitated in the alcohol, being insoluble in the organic solvent, whereas concentration of pectin in the extract is extremely low. Up to date concentration on commercial scale is done by vacuum evaporation and aluminum precipitation method, throughout the world, depending upon the choice and economical position of the producer. In this study concentration of pectin extract will be done by making Cu-pectin complex initiated by Nomura and Naito (1953, 1955), which seemed to have following advantages over aluminum method :

a) Quantity of Cu^{2+} required to form 3-4 % concentrate is extremely low as compared to aluminum.

b) Cu^{2+} can combine with pectin even in very dilute pectin extract.

c) Cu-pectin precipitate is very sensitive to pH and temperature and can easily be changed into solution, which might favor the next demetaling step.

MATERIALS AND METHODS

A. Preparation of pectin pomace

Unshu citrus peel was obtained from Fukuoka-ken Engeiren Amagi Mikan Juice Factory during 1977-78 and 1978-79 at optimum harvest seasons (mid of Nov. to the end of Dec.) after juice extraction by inline juice extractor. Peel was divided into three parts, two parts were cold and hot leached according to the following method.

Cold leaching : One part of the peel was mixed with three parts of tap water (v/v) for 20 minutes in a batch process.

Hot leaching : One part of peel was mixed with 3 parts of hot water at 75°C and mixed for 15 minutes.

In both the cases cold and hot leached peels were drained on a screen and excess water was removed by a filter type centrifuge. All the three parts of peel were dried according to the scheme shown in Fig. 1 using a forced draft air oven by evenly spreading peel on a wire screen and drying pomace to 7-10 % and 3-5 % moisture for a predetermined time.

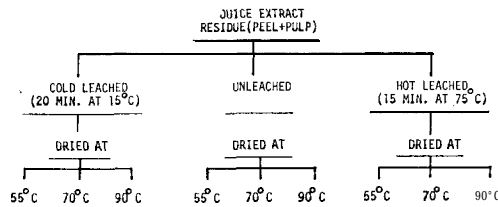


Fig. 1. Experimental layout.

Pectin was extracted from the pomace with 40 times by weight of acidified water (0.13% HCl) for 45 minutes at 85°C.

Chemical and qualitative analyses :

- 1) Pectin yield was reported as dry alcohol precipitate.
- 2) Jelly was prepared according to the method described by IFT Committee on Pectin Standardization (1959).
- 3) Jelly rupture point was measured by curd meter.
- 4) 150 grade pectin yield was calculated by the

$$\text{formula} = \frac{\text{Extractable pectin} \times \text{Rupture point of sample Jelly}}{\text{Rupture point of 150 grade pectin}} .$$

5) Relative consistancy was measured by Ostwald pipette with a discharge time of 68 sec. for distilled water at 30°C according to the formula = t_s/t_0 , where t_s represents discharge time for 0.5 % pectin solution and t_0 discharge time for distilled water.

6) Apparent molecular weight was calculated by the formula = $\frac{6 ((t_s/t_0)^{1/6} - 1)}{4.7 \times 10^{-5} C}$ described by Kim *et al.* (1978).

7) All other analyses were made according to AOAC (1960) and methods used at West Regional Research Laboratory, California; for extraction and analysis of pectic materials (Owens et al., 1952).

B. Extraction of pectin from pectin pomace

Pectin was extracted with 0.1-O. 25 % HCl for 30-60 minutes at 85-100°C, and yield was reported as dry alcohol precipitate. Pectin pomace and acidified water ratio from 1 : 30 to 1 : 60 were tried.

C. Preparation of Cu-pectin complex

Different precipitation trials were run to find out the best precipitation conditions by adding varying amounts of Cu²⁺ using CuCl₂•2H₂O solution in a known volume of pectin extract cooled to 20°C, and then pH was adjusted by drop wise adding conc. NH₄OH solution till precipitate appeared (pH 2. S), during alkali addition solution was constantly mixed at slow speed.

RESULTS

Pectin pomace manufacture

Comparison of proximate composition (Table 1) showed that crude protein and pectin contents were higher and crude fat and ash contents were lower in the year 1977-78 than 1978-79. Pectin contents, crude fiber and pentose increased, whereas crude fat and crude protein decreased after cold leaching.

Table 1. Proximate composition of fresh and cold leached peel (fresh basis). Average of at least three batches.

Sampling year	Treatment	Moisture (%)	Crude fiber (%)	Crude fat (%)	Protein N x 6.25 (%)	Ash (%)	N F E		
							Pectin (a)	Pentose (b)	NFE-(a+b)
1977-78	fresh	84.5	2.61	0.21	2.38	0.58	2.32	2.53	4.87
1978-79	fresh	33.6		0.30	2.26	0.62	2.30	2.61	
1977-78	cold leached	85.2	2.72	0.12	1.97	0.48	3.0	2.82	3.68

Table 2. Percent yield of pectin pomace from fresh peel in the years 1977-78 and 1978-79 (dry basis). Average of more than 300 kg in each case.

Untreated (kg)	Cold leached			Hot leached		
	Yield (kg)	Soluble solids (%)	solids of drain (%)	Yield (kg)	Soluble solids (%)	solids of drain (%)
17.1	11.2		1.5	9.63		2.0

Pomace yields of unleached, cold and hot leached peels are shown in Table 2, which showed that hot leached drain had higher soluble solids (2.0 %) than cold leached drain (1.5 %). Yields of cold and hot leached peel pomace were almost 65.1% and 56.3 % of the unleached pomace.

Effect of leaching and drying on the total extractable pectin is shown in

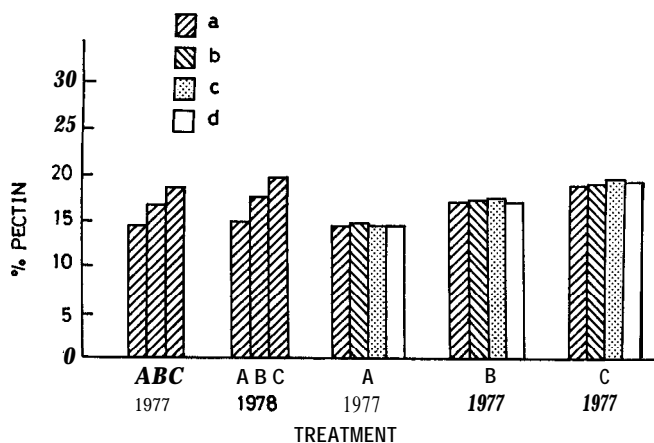


Fig. 2. Effect of leaching and drying on the total extractable pectin (dry basis). A : Unleached, B : Cold leached, C : Hot leached.

- a : 80-85 % moisture (fresh).
- b : 7-10 % moisture (dried at 55°C).
- c : 7-10 % moisture (dried at 70°C).
- d : 3-5 % moisture (dried at 90°C).

Fig. 2. Results revealed that drying at 55°C, 70°C and 90°C to a moisture content of 7-10 % and 3-5 % did not affect at all, the extractability of pectin in all the cases (unleached, cold and hot leached pomace), whereas amount of extractable pectin increased to about 2.5 % and 5.0% after cold and hot leaching respectively. When amount of extractable pectin calculated on 150 grade pectin basis (Fig. 3), result showed that pectin extracted from unleached pomace dried at 90°C was most affected and yield decreased 20 Sd 2 % when dried to 3-5 % moisture and 10 Sd 2.9 % when dried to 7-10 % moisture at 55°C, as compared to fresh peel. In case of cold leaching loss on the basis of total extractable pectin was maximum 26.6 Sd 2.3% when dried to 3-5 % moisture at 90°C and minimum 16.6 Sd 1.2 % when dried to 7-10 % moisture at 55°C, as compared to fresh leached peel, whereas, in case of hot leached pomace loss was maximum, 34.2 Sd 2.6 % when dried to 3-5 % moisture at 90°C and minimum 19.6 Sd 3 % in case of pomace dried at 55°C to 7-10 % moisture. Comparison of consistency of 0.5 % pectin solutions made from pectins extracted from differently treated and dried pomace are shown in Fig. 4, results showed that consistency dropped to almost 1/2 on drying. Qualitative analyses of pectins prepared from differently treated and dried pomace are reported in Table 3. Data showed that purity of pectins prepared from pomace when judged on the basis of anhydrogalacturonic acid ranged between 85-91 Sd 2-4 %, and equivalent weights ranged between 930 and 1162.76 Sd 0.1%. Methoxyl contents of pectins showed that Unshu citrus peel pectin falls under high methoxyl pectin category (methoxyl contents more than 7.0%) having a degree of esterification between 45.6 and 54.5 Sd 1.2 %. Apparent molecular weight

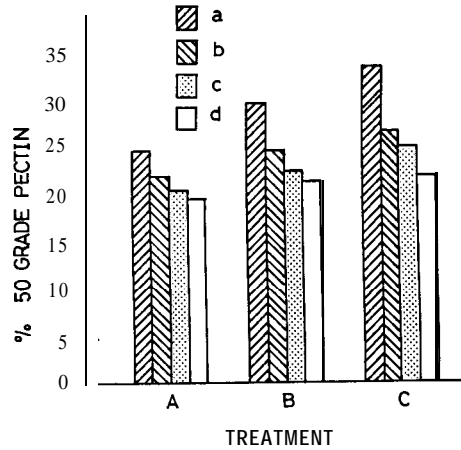


Fig. 3. Effect of leaching and drying on the qualitative yield (dry basis).

A : Unleached, B : Cold leached, C : Hot leached.

a : 80-85 % moisture (fresh).

b : 7-10 % moisture (dried at 55°C).

c : 7-10 % moisture (dried at 70°C).

d : 3-5 % moisture (dried at 90°C).

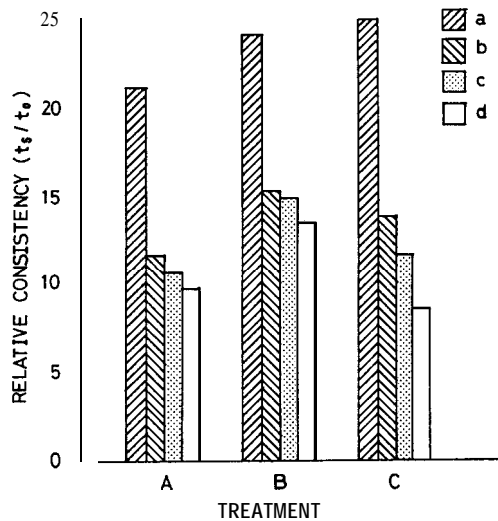


Fig. 4. Comparison of the consistency of 0.5 % pectin solutions made from pectins extracted from differently treated and dried pomace.

t_s : discharge time for 0.5 % pectin solution.

t_0 : discharge time for distilled water.

Symbols used are same as in Figs. 2 and 3.

Table 3. Qualitative analysis of pectins prepared from differently treated and dried pomace (dry basis).

Treatment	Drying temp. (°C)	AGA ¹⁾	Equivalent weight	Methoxyl contents (%)	Degree of esterification (%)	Apparent molecular weight (kg mole-r)	Jelly rupture point ²⁾ (g/cm ²)
Unleached	—						50
	55	90.36	1163.66	8.46	51.47	162.3	45
	70						42
Cold leached	90	90.75 90.75	1098.90 1074.57	8.18 7.83	50.12 47.98	157.7 148.3	40
	—	88.25	1161.30	8.45	51.78	194.6	52
	55	87.00	1070.57	7.57	47.61	147.3	43
Hot leached	70						40
	90	88.50 87.75	1007.60 992.99	7.63 7.77	46.75 46.38	142.7 131.7	37
	—	85.50	1090.45	8.54	51.33	211.4	53
Hot leached	55	86.00	974.17	7.89	46.75	163.2	42
	70						39
	90	88.75 91.00	934.31 930.09	7.745 63	45.65 44.47	146.9 129.8	35

¹⁾ Anhydrogalacturonic acid.

²⁾ Jelly rupture point of standard 150 grade pectin 30.0 g/cm².

in case of pectins prepared from unleached, cold and hot leached pomace ranged between 148.3 to 209.3, 131.7 to 194.6 and 129.8 to 211.4 kg mole⁻¹ respectively. Molecular weight decreased about 60,000–80,000 from maximum to minimum value in all the cases after drying. Force required to rupture test jellies measured by curd meter was between 35–50 g/cm² Sd 1–2 % in all the pectins. Results showed that higher molecular weight pectins formed more firm jellies.

Extraction of pectin

Number of pectin extraction trials were run to find out a suitable combination of time, temperature and pH, which will give a pectin extract of suitable concentration for copper method. Results are reported in Table 4. Results revealed that at 85°C only 85% of total extractable pectin was obtained even when extracted for a prolonged period of time (60min.) at pH 1.6, and when temperature was increased to 95°C for the same length of time and at same pH, value increased to 97.5 %, the next highest yield with minimum acid and energy input was obtained when extracted at 95°C for 45 minutes at pH 1.7. Concentration of pectin in the extract increased to double by decreasing acidified water to pomace ratio from 60 to 30, at the same time loss of pectin in the residue left after separation of extract, increased from 12-14% to 20-21 %.

Cu-pectin precipitation

The conditions of Cu-pectin precipitation are presented in Table 5, results showed that good and easily separatable precipitates by centrifugation were obtained when 100 ml of 0.56 % pectin solution was mixed with 20mg of Cu²⁺ and then mixture was cooled to 20°C and pH adjusted to 2.8 with concentrated NH₄OH. Yield of Cu-pectin precipitate and their purity are presented in Table 6. Results revealed that 100 g of cold leached pomace yielded 21.7 g of precip-

Table 4. Batch extraction of pectin from the pomace (cold leached). Results reported are at least average of three replicates.

Wt. acidified water/Wt. pomace	Extraction conditions			Pectin content of sol. (%)	Extract consistency (t_s/t_0)	Theor. total extractable pectin ¹⁾ (%)	Pectin loss in the residue ²⁾ (%)			
	Temp. (°C)	pH	Time (min.)							
60	85	1.9	20	0.22	6.4	53.6	12-14			
			30	0.25	6.9	60.9				
			60	0.28	6:5	68.3				
		1.7	20	0.25	6.5	60.9				
			30	0.25	6.6	65.8				
			60	0.30	7:2	73.2				
	1.6	60	0.35	a.1	85.3					
		95	1.9	20	0.28	7.1		68.3		
				30	0.31	7.2		75.6		
	60			0.36	6:8	87. a				
	1.7	60	20	0.32	7.0	78.0				
			30	0.32	7.8	85.3				
			60	0.38	7.5	92.6				
	1.6	60	60	0.4	8.6	97.5				
				100	1.9	20		0.34	5.3	82.9
						30		0.35	5.5	85.3
	60	0.38	5:2			92:6				
	1.7	60	20	0.35	5.3	85.3				
			30	0.37	5:2	90.2				
			60	0.40	5:2	97.5				
	1.6	60	30	0.41	4.5	100.03'				
			30	0.41	8.6					
			45	0.38	8.8	61.7				
	85	1.7	45	30	0.40	65.0				
45				0.42	7.2	65.0				
45				0.42	7.5	68.3				
40	1.9	45	30	8.3						
			45	0.48	8.4	78.0				
			30	0.52	7.5	84.5				
95	1.7	45	30	7.5	86.1					
			45	0.53	7.2	86.1				
			45	0.56	91.0	91.0				
30	95	1.7	30	0.72	12.1	87. a				
			45	0.74	12. a	90.2				
			45	0.79	9.6	96.3				
	100	1.7	45	0.79	9.6	96.3				

¹⁾ Calculated on the assumption that water retained in the residual peel after extraction of pectin, also contained the same amount of pectin as in the extract.

²⁾ On the basis of theoretical total extractable pectin.

³⁾ 100% theoretical value.

itates on dry basis of a purity 55.3 %. Recovery of pectin in the precipitates on the basis of extracted pectin was 85 %.

DISCUSSION

Total pectin contents of Unshu citrus peel are 15 %, compared to 21-22 % reported by Shepherd and Graham (1952) in other citrus, which mean Unshu

Table 5. Cu-pectin precipitation conditions.

wt. acidified water/Wt. pomace	Pectin content of sol. (%)	Precipitation conditions			Nature of precipitate	Remarks			
		pH	Temp. (°C)	Cu ²⁺ /100 ml extract (mg)					
30	0.74	2.8	60	15	viscous sol.	Incomplete precipitation			
				20	-do-	-do-			
				25	curd	Difficult to handle			
			20	40	2.8	20	20	very fine	Difficult to handle
							25	-do-	-do-
							20	easy to separate	Some ppt. reappear on addition of excess Cu ²⁺ in the drain
			25	-do-	Complete precipitation				
40	0.56	2.8	20	20	easy to separate	No ppt. on addition of excess Cu ²⁺ in the drain			
				15	viscous sol.	Incomplete precipitation			

Table 6. Cu-pectin complex yield and purity (% dry basis of cold leached pomace). Average of more than four replicates.

Pectin content of untreated dry peel (%)	Pectin content of cold leached pomace (%)	Pectin extracted (%)	Cu-pectin precipitate			
			Wt. (g)	Total pectin (%)	Purity ¹⁾ (%)	Recovery ²⁾ (%)
15.0	17.2	14.1	21.7	12.0	55.3	85

¹⁾ De-coppered pectin.

²⁾ On the basis of % pectin in the extract.

peel contains about 30% less pectin than peels being used for pectin manufacture. This difference was more prominent and increased to 50% and 38% when Unshu cold leached peel was compared with lime and lemon commercial cold leached peels, evaluated by Rouse and Crandall (1978).

As for quality of pectin is concerned Unshu citrus pomace is equally good as lime and lemon pomace, when compared on the basis of molecular weight, consistency and jelly grade.

Cold and hot leaching washed out 29% and 40% of total peel solids respectively, as compared to 20 % by commercial cold leaching. Braddock and Crandall (1978) reported that 30 % total peel solids were removed by applying twice, 1 : 2 peel water washing. Pectin contents increased 17 % and 33 % in case of cold and hot leaching, while Shepherd and Graham (1952) reported 17% increase after blanching and washing. Unleached peel behaved not well in the oven and turned into dark reddish brown color, similar observations by Crandall et al. (1978) made them to conclude that it is impossible to dry unleached peel as pectin pomace on commercial scale. They further reported that 25 % decrease in quality on basis of 150 grade pectin in case of lemon peel occurred when dried to 8-12% moisture in direct fire rotary dryer at temper-

ature of 370535°C in first stage and 134-176°C in the second stage of dryer, and that loss increased to 54% when further dried to 3-7 % moisture as compared to only 26 % in our experiment when dried to 3-5 % moisture at 90°C. These observations clearly indicate that peel should not be dried below 10 % moisture and that up to 10% moisture quick drying at high temperature have no much degradation effect on the quality of pectin in the pomace.

In case of extraction of pectin from the pomace, a temperature increment of 15°C, pH increment of 0.2 and time increment of 15 minutes in series of pectin extraction experiment showed that most suitable combinations of time, temperature and pH to get maximum pectin yield were, pH 1.6 at 95°C for 60 minutes (yield 97.5 %) and pH 1.7 at 95°C for 45 minutes (yield 91%). In similar experiments, Myers and Baker (1931), Gaddum (1934) and Rouse and Crandall (1978) tried time increments of 10, 20 and 5°C respectively. Rouse and Crandall (1978) reported best combination of pH 1.6 at 95°C for 30 minutes in case of lime and lemon pomace when HNO₃ was used for extraction.

Concentration of pectin extract during pectin manufacture is done by vacuum evaporation (Anonymous, 1967), and aluminum precipitation (Wilson, 1925; Joseph and Havighorst, 1952). Any one of these existing processes if applied as such to pectin manufacture from Unshu citrus pomace, will prove more expensive than imported pectin due to low pectin contents of peel as mentioned earlier. So there is no choice but either to find out a dry process to concentrate pectin in the peel by mechanically separating the pectin rich portion of peel (albedo) or to find out some cheaper methods of concentrating the pectin extract, so as to make it commercially possible to manufacture pectin from Unshu citrus peel. No literature up to now is cited concerning the first choice, whereas, second possibility to find out other economical concentration methods are actively being considered. Concentration of pectin solution by making Cu-pectin complex was tried by Nomura and Naito (1953, 1955) and they reported that yield of Cu-pectin complex was identical with the yield obtained by alcohol precipitation. They further reported that good precipitation results were obtained when equal amounts of pectin extract and CuCl₂ solutions (0.07-0.04%) were mixed and pH adjusted at 4.0, as compared to the precipitation conditions in this study using concentrated CuCl₂ solution (20 mg/100 ml pectin extract), good precipitates were obtained at pH 2.8. Amount of Cu²⁺ required to precipitate pectin in the extract in both the cases as compared to Al³⁺ was about 50% less, moreover, demetaling of precipitate by using IER (ion exchange resin) seems more economical because Cu-pectin complex can easily be changed into solution, which favors quick pick up of Cu²⁺ by IER. Decoppering of Cu-pectin complex by using IER will be reported in the next publication.

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