Evaluation of Rhododendron Luteum and Rhododendron Ponticum in Pulp and Paper Production

Procjena mogućnosti uporabe biljaka Rhododendron luteum i Rhododendron ponticum kao sirovine za proizvodnju celuloze i papira

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ABSTRACT • In this study, Rhododendron luteum and Rhododendron ponticum were evaluated as raw material for pulp and paper production. 12 different sodium borohydride (NaBH₄) added cooking trials were performed for each sample and kraft method was used for pulp production. Pulp properties, such as yield, kappa number and viscosity, and physical properties, such as breaking length and burst index, were determined for each trial. Besides, the effects of active alkali and NaBH₄ on the pulp and paper properties were also examined. Optimum cooking conditions were obtained by using 18 % active alkali for NaBH₄-free cooking experiments and 0.5 % NaBH₄ and 18 % active alkali for NaBH₄-added cooking experiments. In NaBH₄-added pulping condition, the screened yield, kappa number and viscosity of R. luteum were found to be 43.4 %, 40.1 and 949 cm³/g, respectively. The respective values for R. ponticum were 41.9 %, 44.5 and 885 cm³/g. The screened yields of R. luteum and R. ponticum increased by about 2.8 % and 5.3 %, respectively, with 5 % addition of NaBH₄ compared to NaBH₄-free cooking experiments. Furthermore, with the addition of NaBH₄, the kappa numbers decreased while the viscosity increased. The physical properties of the produced papers were also improved by using NaBH₄ in cooking liquor. According to the obtained results, it was found that R. luteum and R. ponticum species can be evaluated for pulp and paper production.

Keywords: R. luteum; R. ponticum; pulp; paper; NaBH₄

SAŽETAK • U ovom je radu istražena mogućnost uporabe biljaka Rhododendron luteum i Rhododendron ponticum kao sirovine za proizvodnju celuloze i papira. Za svaki uzorak provedeno je 12 različitih ispitivanja kuhanja s natrijevim borhidridom (NaBH₄), a celuloza je proizvedena kraft postupkom. Za svako ispitivanje određena su svojstva celuloze poput prinosa, kappa broja i viskoznosti, te fizička svojstva kao što su duljina lomljenja i indeks pucanja papira. Osim toga, ispitani su učinci aktivne lužine i NaBH₄ na svojstva celuloze i papira. Optimalni uvjeti kuhanja postignuti su upotrebom 18 % aktive lužine za eksperimentalno kuhanje bez NaBH₄ i upotrebom 0,5 % NaBH₄ i 18 % aktive lužine za eksperimentalno kuhanje s dodatkom NaBH₄. U proizvodnji celuloze iz biljke R. luteum s dodatkom NaBH₄ utvrđeno je da prinos prosijavanja iznosi 43,4 %, da je kappa broj 40,1, a viskoznost...
In the second half of the twentieth century, especially with the increase of world population and the increase in demand for forest products due to rapid developments in technology, the decrease in forest areas led the forest products industry to search for raw materials as an alternative to wood (Oner and Aslan, 2002; Tutus et al., 2011). Non-wood and new wood products are one of the best alternative raw materials (Kaldor, 1992; Comlekcioglu et al., 2016). Due to the rapid increase in paper consumption in recent years, alternative fibrous materials have gained importance in the pulp and paper industry (Daud and Law, 2011). The search for new wood raw materials still continues in the world.

Rhododendron species, a valuable ornamental shrub, have been cultivated since the 18th century. These species belong to the heath or Ericaceae family. Rhododendron, with about 1000 different species, extends from Southwest Africa to New Guinea. Some Rhododendron species are large species with a tree-type growth habit. It is evergreen or rarely deciduous. Turkish rhododendrons grow naturally up to 3000 m above the sea level. One of the most remarkable Rhododendron species in moist forest formations covering the northern coast of Turkey is R. luteum and the most common Rhododendron is R. ponticum. Fiber length, fiber width, lumen diameter and cell wall thicknesses of R luteum and R. ponticum are approximately 0.9-1.0 mm, 17-19 μm, 7-9 μm and 3-4 μm, respectively. Besides, these species have high holocellulose (77-80 %) and alpha cellulose (47-48 %) contents, which are important carbohydrates for pulp and paper industry (Birinci, 2008; Camlibel, 2008).

Although they appear on the northern slopes of the mountains on the Black Sea coast, there are more species in the Eastern Black Sea Region (Avci, 2004). In many studies, Rhododendron species have generally been evaluated for the production of medium density fiberboard (MDF) (Akgul et al., 2012; Akgul and Camlibel, 2008; Ayrimis et al., 2014). Besides, some studies have determined the chemical composition of some Rhododendron species (Birinci, 2008; Shrestha and Budhathoki, 2012).

The Kraft method is the most commonly used method for obtaining pulp suitable for papermaking. Increasing the pulp yield in pulp production is very important for the pulp and paper industry in terms of cost and economy. Modifying the Kraft cooking process, e.g. by adding NaBH₄, is a way of increasing the pulp yield (Courchene, 1998; Tutus and Eroglu, 2003; Tutus and Eroglu, 2004; Tutus and Usta 2004; Hafizoglu and Deniz, 2007; Istek and Gontek, 2009, Akgul et al., 2018). The end groups of carbohydrates are protected from peeling reactions by using NaBH₄ as a catalyst in cooking processes (Istek and Ozkan, 2008; Tutus and Ciclerk, 2016).

There is little information about the use of Rhododendron species in wood-based industries and there is no study on their use in pulp and paper production. The aim of this study was to evaluate Rhododendron luteum and Rhododendron ponticum stalks for pulp and paper production, using Kraft-NaBH₄ cooking methods.

R. luteum and R. ponticum species were obtained from Akdamar Village of Agkabat district of Trabzon province in Turkey. The chemical compositions of R. luteum and R. ponticum were determined in a previous study (Birinci, 2008) and are presented in Table 1 with some wood and non-wood species.

According to Table 1, the chemical composition of Rhododendron species is similar to that of hardwoods species. Rhododendron species show similarities among themselves. However, the lignin content of R. ponticum was higher than that of R. luteum. Due to their high holocellulose contents, it was concluded that Rhododendron species were considered suitable for pulp and paper production.

The samples were chopped and air dried. 12 cooking trials were applied on each species, using Kraft-NaBH₄ method given in Table 2 in order to determine optimum pulping conditions.

Cooking trials of the species were applied in a rotary digester with a 15-liter capacity and high pressure resistant. After cooking processes, the pulps were washed with tap water until the black liquor was removed, and the washed pulps were transferred to 0.15 mm slotted screen. Screened pulp and screen reject yields were calculated after the screening process. The kappa numbers and viscosity of the screened pulps were determined according to TAPPI T 236 om-13 and TAPPI T 230 om-08, respectively (Anonymous, 1998).

The pulps were beaten to 25 ±3 °SR (Schopper Riegler) freedom level in the holander beater according to TAPPI T 200 sp-96. Ten test papers were produced from the pulps obtained from each cooking trial. The breaking lengths and burst indices of the papers
were determined according to TAPPI T 494 om-01 and TAPPI T 403 om-91, respectively (Anonymous, 1998).
In order to determine the effects of active alkali and NaBH₄, the optimum cooking parameters were used.

Table 2 Pulping conditions of R. luteum and R. ponticum

| Pulping condition | Unit | Value |
|-------------------|------|-------|
| Active alkali | % | 18, 20, 22 |
| Sulfidity | % | 24 |
| NaBH₄ charge | % | 0, 0.3, 0.5, 0.7 |
| Cooking temperature | °C | 160 |
| Time to maximum temperature | min | 40 |
| Time at maximum temperature | min | 90 |
| Liquor to raw material ratio | l/kg | 4/1 |

Table 3 Screened yields, kappa numbers and viscosities of R. luteum and R. ponticum pulps

| Cooking No | Active alkali, % | NaBH₄, % | Screened yield, % | Kappa No | Viscosity, cm³/g |
|-------------|-----------------|---------|------------------|----------|-----------------|
| 1           | 18              | 0       | 42.15            | 45.74    | 895             |
| 2           | 20              | 0       | 41.78            | 42.16    | 860             |
| 3           | 22              | 0       | 41.05            | 41.84    | 838             |
| 4           | 20              | 0.3     | 43.03            | 42.06    | 928             |
| 5           | 22              | 0.3     | 42.55            | 39.60    | 912             |
| 6           | 22              | 0.3     | 41.83            | 40.10    | 866             |
| 7           | 22              | 0.5     | 43.35            | 40.10    | 949             |
| 8           | 20              | 0.5     | 42.91            | 38.88    | 924             |
| 9           | 22              | 0.5     | 42.00            | 36.56    | 902             |
| 10          | 18              | 0.7     | 43.78            | 38.98    | 988             |
| 11          | 20              | 0.7     | 43.25            | 37.10    | 939             |
| 12          | 22              | 0.7     | 42.60            | 41.23    | 918             |

3 RESULTS AND DISCUSSION
3. REZULTATI I RASPRAVA

The yield and chemical properties of R. luteum and R. ponticum was found lower than that of R. luteum by about 5.6 % and 15.5 %, respectively. The kappa numbers of R. ponticum and R. luteum were determined as 49.24 and 45.74. According to these results, R. luteum is more suitable for pulping production than R. ponticum. The effects of NaBH₄ on the properties of Rhododendron pulp are shown in Figure 1.

With the addition of NaBH₄ to the cooking liquor, the yield and viscosity of the pulps increased and kappa numbers decreased. NaBH₄ was effective on the screened yields as it had the ability to stop peeling reactions occurring in cellulose chains (Istek and Ozkan, 2008; Tutus and Cicekler, 2016). NaBH₄ removes the lignin from the pulp i.e. selective lignin delignification occurs, while increasing the viscosity by preserving
carbohydrates (Tutus, 2008). It also prevents shortening of cellulose and hemicellulose chains as it prevents damage to carbohydrates. In this way, the viscosity of the pulps is increased and strength pulps can be obtained. Recent studies have shown a decrease in the kappa numbers of the pulps obtained with the addition of NaBH4 to the cooking experiments (Copur and Tozuoglu, 2008; Gulsoy and Eroglu, 2011; Istek and Gonteki, 2009). With the addition of 0.5 % NaBH4, the screened pulp yields of R. luteum and R. ponticum pulps increased by approximately 2.8 % (from 42.15 % to 43.35 %) and 5.3 % (from 39.78 % to 41.87 %), while kappa numbers decreased by 12.3 % and 9.8 %, respectively. By keeping the active alkali high, the rate of depolymerization of carbohydrates during cooking increases. Therefore, both yield and viscosity decrease. The screened yields, kappa numbers and viscosity of R. luteum and R. ponticum papers decreased by increasing the active alkali from 18 % to 22 %. Many studies have reported that the increase of active alkali decreases the yield, kappa number and viscosity (Lopez et al., 2000; Yue et al., 2016; Zhai and Zhou, 2014).

The breaking lengths and burst indices of R. luteum and R. ponticum papers used in this study are given in Table 4.

According to Table 4, the breaking lengths and burst indices of the R. ponticum papers were found to be higher than those of R. luteum. It is clearly seen that the physical properties of the papers improve with the increase of active alkali and NaBH4 ratios (Figure 2).

Many studies have reported that boron compounds prevent peeling reaction during cooking, resulting in less damage to carbohydrates and therefore improved physical and optical properties of the papers.

### Table 4

| Cooking No | Active alkali, % | NaBH4, % | Breaking length, km | Burst index, kPa·m²·g⁻¹ |
|------------|-----------------|---------|---------------------|------------------------|
|            | Active alkali, % |         | R. luteum | R. ponticum | R. luteum | R. ponticum |
| 1          | 18              | 0       | 2.21      | 2.88        | 1.90      | 2.10        |
| 2          | 20              | 0       | 2.29      | 2.91        | 1.98      | 2.25        |
| 3          | 22              | 0       | 2.32      | 3.02        | 2.06      | 2.31        |
| 4          | 18              | 0.3     | 2.35      | 2.98        | 2.00      | 2.28        |
| 5          | 20              | 0.3     | 2.40      | 3.05        | 2.15      | 2.45        |
| 6          | 22              | 0.3     | 2.52      | 3.09        | 2.30      | 2.54        |
| 7          | 18              | 0.5     | 2.57      | 3.07        | 2.17      | 2.43        |
| 8          | 20              | 0.5     | 2.65      | 3.10        | 2.30      | 2.53        |
| 9          | 22              | 0.5     | 2.79      | 3.19        | 2.45      | 2.62        |
| 10         | 18              | 0.7     | 2.87      | 3.15        | 2.32      | 2.51        |
| 11         | 20              | 0.7     | 3.01      | 3.32        | 2.54      | 2.68        |
| 12         | 22              | 0.7     | 3.13      | 3.51        | 2.66      | 2.87        |
produced (Akgul et al., 2007; Copur and Tozluoglu, 2008; Istek and Ozkan, 2008; Istek and Gonteki, 2009; Gumuskaya et al., 2011; Erisir et al., 2015; Gulsoy et al., 2016). With the addition of 0.5 % NaBH₄, the breaking lengths of R. luteum and R. ponticum pulps increased by approximately 16.1 % and 6.4 %, and burst indices also increased by 14.2 % and 15.7 %, respectively. According to data in Tables 3 and 4, optimum cooking conditions for the two species in NaBH₄-added cooking experiments were determined by using 0.5 % NaBH₄ and 18 % active alkali (Cooking No: 7).

4 CONCLUSIONS
4. ZAKLJUČAK

In this study, pulp and papers of R. luteum and R. ponticum species were produced by using Kraft-NaBH₄ method and their properties were examined. As a result of this study, it was determined that the chemical properties of R. luteum and R. ponticum, such as yield, kappa, and viscosity, were found to be better for pulp and paper industry. Besides, when the physical properties of the produced papers are examined, it is seen that they are suitable for the production of the manufacture of many paper types. The short-fiber pulps obtained from R. luteum and R. ponticum can be mixed with long-fiber pulps in certain proportions and evaluated for the production of many paper types. Since 70 % of short-fiber raw materials are used for the production of writing-printing paper, the possibility of using R. luteum and R. ponticum species for the production of different types of quality paper is quite high.

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