



## A Novel Approach for Improving Wrinkle Resistance and Flame Retardancy Properties of Linen Fabrics

Ş.S. Uğur<sup>1\*</sup>, M. Bilgiç<sup>1</sup>

**Abstract:** Flax is an important fibre for textile industry not least because of its excellent properties. However, in its untreated form, linen fabrics possess poor wrinkle resistance and burn easily with a high flame velocity. Linen fabrics functional finishing researchs have been mainly focused on reducing these problems to achieve the desired fabric property for using its as wearing apparel, household textile and composite material. This study was undertaken to investigate the novel durable wrinkle resistant and flame retardant finishing of the linen by using 1,2,3,4-Butanetetracarboxylic acid, Nano polyurethane for cross-linking and Al<sup>2</sup>O<sup>3</sup> nanoparticles for catalyst in padding process. Attenuated Total Reflectance Fourier Transform Infrared Spectroscopy and Scanning Electron Microscopy with Energy Dispersive X-ray analysis were used to characterization. Colour difference, whiteness and yellowness analysis was performed on the linen fabrics before and after the chemical treatment. Tensile strength tests of the warp and weft yarns were performed to evaluate the effect of finishing treatment. With the chemical treatment, linen fabrics wrinkle resistant and flame retardant properties enhanced. The durability of the functional properties were also tested after 5 washing cycle.

**Keywords:** Flax fiber, wrinkle-resistance, flame retardancy, BTCA, Nano PU, Al<sub>2</sub>O<sub>3</sub> nanoparticles.

### 1. Introduction

In the recent decades, demands for natural fibers than its synthetic counterparts increased day by day because of their eco-friendly character. Among these natural fibers, flax is widely used for wearing apparel, household textiles and industrial composite materials. Flax fibers main chemical components are alpha and hemi celluloses. When flax fibers used in a textile material process, it is named as linen. The reasons of preference for linen textiles, they shows excellent tensile properties, comfort, high tenacity and appearance. Nevertheless, linen fabrics have high stiffness, low resilience, low wrinkle recovery angle disadvantages (Mccall et al., 2001; Kim and Csiszar 2005; Lam et al., 2010; Vasile et al., 2012). Chemical modification of linen fabrics for improving the undesired properties has been studied for attaining a higher quality and technical performance rather than by using a high cost fibre (Choi et al., 1994; Tzanov and Cavaco-Paulo 2006; Holme. 2007).

In the present time, wrinkling has an increasing effect on the overall quality of the linen fabrics used as apparel and household textiles. Wrinkles are defined as fabric deformation based on its viscoelastic properties. Linen fabrics have a very poor resistance to creasing due to the high orientation of cellulose content in the fibre. Wrinkle free finishing is also known by consumers as 'Easy Care' and 'Wrinkle-resistant'. Nowadays, chemical agents which can be used for wrinkle-resistant of cotton fabrics, can't be used for linen fabrics efficiently because of the flax fiber tendency to crease easily. The main problem with these chemical agents in use, while wrinkle recovery angle increases, the breaking strength of the fabric decreases. So, a new chemical finishing process which improves Easy Care properties, without damaging tensile properties, is still an unsolved problem. Flame retardancy properties of the linen fabrics is an important property when will be used as industrial composite material. But the same tensile properties damaging problem occurs with the flame retardant chemical agents, too

<sup>1</sup>Süleyman Demirel University, Textile Engineering, 32260,Isparta, Turkey.

<sup>2</sup>Süleyman Demirel University, Graduated School of Natural And Applied Sciences, Textile Engineering, 32260,Isparta, Turkey.

\*Corresponding author (iletisim yazarı): [suleugur@sdu.edu.tr](mailto:suleugur@sdu.edu.tr)

Citation (Atf): Uğur, Ş.S., Bilgiç, M. (2017). A Novel Approach for Improving Wrinkle Resistance and Flame Retardancy Properties of Linen Fabrics. *Bilge International Journal of Science and Technology Research*, 1 (2): 79-86.

(Mccall et al., 2001; Kim et al., 2005; Vasile et al., 2012).

The improvement of a finishing process which improves easy care properties, without influencing strength characteristics, has been a long-term aim for researchers, is still remains to be satisfactorily solved. (Vasile et al., 2012). The ester crosslinking of cotton by 1,2,3,4-Butanetetracarboxylic acid (BTCA), catalyzed with sodium hypophosphite (SHP), has been found to be superior for wrinkle-resistant property of cotton fabric with increase in strength properties (Choi et al., 1994; Tzanov et al., 2006; Holme. 2007; Lam et al., 2010).

Using a co-catalyst like titanium dioxide (TiO<sub>2</sub>) has been found feasible to enhance the crease recovery performance with minimum side effects (Wang and Chen, 2005; Chen and Wang, 2006; Liu et al., 2008).

Durable functional finishing systems that does not lose too much of its mechanical strength for linen fabrics are needed. The objective of this work is to investigate the novel multifunctional chemical finishing of linen fabric by using Nano Polyurethane (Nano PU), BTCA for cross-linking in padding process. Al<sub>2</sub>O<sub>3</sub> nanoparticles used as co-reactants in the chemical formulation to enhance the performance of chemical finishing and minimize the side effects. Fourier Transform Infrared Spectroscopy with Attenuated Total Reflection mode (FTIR-ATR) and Scanning Electron Microscopy with Energy Dispersive X-ray analysis (SEM-EDX) were used for characterization. Flax fabrics wrinkle-recovery and the finishing treatments on the some physical properties (tensile strength, colour change, whiteness and yellowness values) of the linen fabric has also been studied.

## 2. Material and Method

### 2.1. Materials

1,2,3,4-Butanetetracarboxylic acid (BTCA), Sodium hypophosphite (SHP) and Aluminum oxide nanoparticles (Al<sub>2</sub>O<sub>3</sub> nanoparticles, particle size <50 nm, specific surface area 40 m<sup>2</sup>/g) were purchased from Aldrich. Nano polyurethane (Baypret Nano PU, particle size <100 nm) anionic dispersion was purchased from Tanatex Chemicals. 1,2% Al<sub>2</sub>O<sub>3</sub> nanoparticle suspensions were prepared at 40 Watt for 1 hour by Sonics Vibra-Cell Ultrasonic Homogenizer.

The bleached linen woven fabric was used as substrate. The fabric properties: plain weaved, 250.2 g/m<sup>2</sup>, 17 threads/cm warp and 12 threads/cm weft. The fabric was cut into approximately 35 cm x 70 cm pieces before padding.

### 2.2. Impregnation process

For finishing process, a laboratory type padding machine was used and 7 recipes were applied with 75% pick-up value to the linen fabric as shown in Table 1. A and B receipts were used as control receipts for obtaining the chemical's effect solely to the functional properties. For the other receipts pH values were evaluated. After the impregnating processes, all the samples were dried at 100 °C and cured at 150 °C for 5 min.

**Table 1.** Applied receipts to linen fabrics

Samples	Receipt	pH values
A	5% BTCA 10% SHP	-
B	20% Nano PU	-
C	5% BTCA 10% SHP 1% Al <sub>2</sub> O <sub>3</sub>	3.5-4
D	5% BTCA 10% SHP 2% Al <sub>2</sub> O <sub>3</sub>	3.5-4
E	5% BTCA 10% SHP 20% Nano PU	5
F	5% BTCA 10% SHP 40% Nano PU	5
G	5% BTCA 10% SHP 1% Al <sub>2</sub> O <sub>3</sub> 20% Nano PU	4

### 2.3. Evaluation Methods

#### Fourier transform infrared attenuated total reflectance (FTIR-ATR) measurements

Perkin Elmer Spectrum BX spectrometer was used to obtain the infrared spectra of surfaces using an ATR sampler. The spectra were taken over a wave number range of 4000–400 cm<sup>-1</sup> with a resolution of 2 cm<sup>-1</sup> at room conditions.

### Scanning electron microscopy (SEM-EDX)

FEI QUANTA 250 FEG high resolution scanning electron microscope (SEM) equipped with energy dispersive spectroscopy (EDX-Oxford Aztec) Microanalysis system was used to examine the surfaces of woven cotton samples at an acceleration voltage of 10 kv. The cotton fabric samples were coated with 10 nm Au/Pd prior to SEM observation.

### Whiteness, yellowness and colour difference value measurement

Minolta 3600d spectrophotometer was used to obtain the colour difference, whiteness and yellowness (Stensby index) values of the untreated and functional finishing applied linen fabrics, which is using D 65 light source to examine the chemical effect on the yellowing properties of the fabrics.

### Tensile strength measurement

The mechanical tests were performed on a Lloyd LR5K Plus electronic tensile strength machine according to EN ISO 13934-1 Standard- Textiles- Tensile properties of fabrics- Part 1: Determination of maximum force and elongation at maximum force using the strip method. The breaking strength of the untreated and functional finished fabrics was tested at fracture. Linen fabrics were kept for 24 h at standard conditions (20°C and 65% Relative Humidity) before the mechanical tests. All the sample tensile strength analysis was repeated after 5 washing cycles.

### Flame retardancy measurement

Flame retardant properties of the fabrics were measured according to DIN 543352 Standard (45 ° law by burning the ignition testing textile fabric side of the combustion characteristics) by Govmark 45 ° combustion tester.

### Wrinkle-Resistance

The crease recovery angle test was carried out according to the TSE TS 390 EN 22313-1996 (Textile Fabrics-Determination Of The Recovery From Creasing Of A Horizontally Folded Specimen By Measuring The Angle Of Recovery) Standard in a crease recovery angle tester. Fabric samples were cut in 40mm x 15mm dimensions in both weft and warp directions. Then samples were

horizontally folded and kept under 10N forces for 60 min. When the time ran out, the load was removed and the angle between folded parts was measured by using a crease recovery angle tester after 0.5, 30 and 60 min.

### Washing procedure

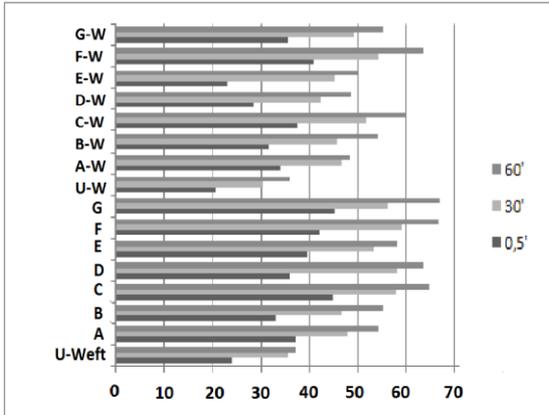
Functional properties of functional finished linen fabrics such as flame retardant and wrinkle resistance functions were analyzed after the padding process. For determining the durability of these functional properties, linen fabric samples were washed 5 times at 40 °C for 30 min. with laboratory type washing machine Gyrowash.

### 3. Results

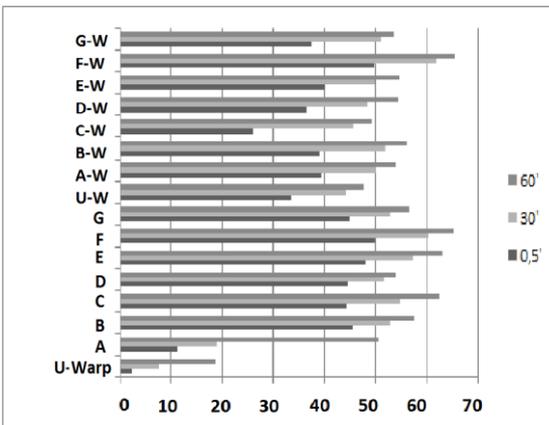
The wrinkling which is the major disadvantage of linen fabric may handle the situation by crease-resistant finishing processes. Figure 1 and 2 shows the results of the untreated, treated and washed linen fabrics, weft and warp directions wrinkle resistance test results. The test results proved that the applied finishing processes, especially with Nano PU content are very effective in improving the wrinkling properties. For the treated fabrics on weft directions receipt F and G shows the best improvement for wrinkle angle after 60 minutes. On the warp direction test results, it can be clearly seen receipt G gives the best results.

After 5 washing cycles, significant increase in wrinkle resistance still remains. According to the weft and warp direction wrinkle resistance test results before and after the washing cycles, it can be said that Receipt F is the optimum chemical composition.

A strong adsorption band with a maximum of 1030  $\text{cm}^{-1}$  is a result of the overlapping bands attributed to the functional groups of linen fibres cellulose groups, namely the C-C, C-O and C-O-C stretches vibrations. After finishing processes, this band shows increase in intensity when the C-O groups occupied with  $\text{Al}_2\text{O}_3$  nanoparticles for samples C, D and G. The FTIR-KBr spectra of  $\text{Al}_2\text{O}_3$  nanoparticles is shown in Figure 3.

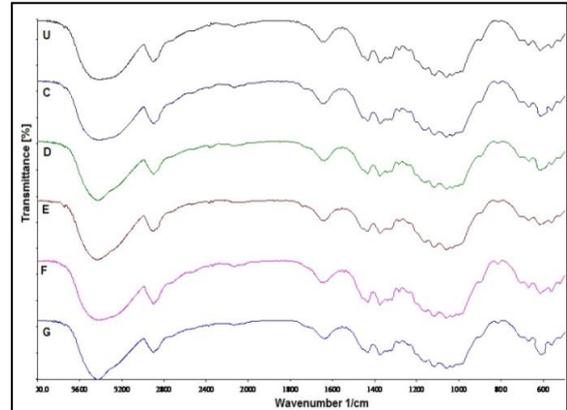


**Figure 1.** Weft direction wrinkle resistance test results of the linen fabrics after 0.5, 30 and 60 minutes.



**Figure 2.** Warp direction wrinkle resistance test results of the linen fabrics after 0.5, 30 and 60 minutes.

Al<sub>2</sub>O<sub>3</sub> nanoparticles indicate a strong metal–oxygen (M–O) adsorption band between 830 and 550 cm<sup>-1</sup> wavenumbers (Uğur et al. 2011). An absorption peak around 550 cm<sup>-1</sup> on the C, D and G fabrics FTIR-ATR spectra can be attributed to Al<sub>2</sub>O<sub>3</sub> nanoparticle according to the FTIR-KBr spectra of Al<sub>2</sub>O<sub>3</sub> nanoparticles. E, F and G sample spectra shows a vibration band between 3200-3400 cm<sup>-1</sup> wavenumbers according to the N-H groups of the Nano PU content. The same intensity increases can be seen on the bands 2800-3000 cm<sup>-1</sup> (CH-CH) and 1500 cm<sup>-1</sup> (H-N-C=O) due to the amide groups of the Nano PU. For all the treated samples, a strong band near 1700 cm<sup>-1</sup> is assigned to carboxyl acid groups (-COOH) introduced by the BTCA content.



**Figure 3.** FTIR-ATR spectra for untreated and treated flax fabrics.

Colour, whiteness and yellowness values of all the samples are presented in Table 2. It can be observed that the performed treatment in the present of Nano PU totally resulted in worst whiteness and yellowing values. This means that with the increasing concentration of the Nano PU causes significant deterioration on the fabric appearance. All the test results bearing in the mind that it can be seen finishing treatment except F shows close colour, whiteness and yellowness values.

**Table 2.** Colour (L, a, b and C), whiteness and yellowness values of untreated and treated flax fabrics

Samples	Colour Values				Whiteness Values	Yellowness Values
	L	a	b	C		
U	93.72	-	-	1.4	85.37	-1.68
		1.23	0.67			
		ΔE			Whiteness Values	Yellowness Values
					Difference	Difference
A		0.87			-5.78	3.09
B		1.94			-7.11	5.18
C		0.83			-4.23	3.15
D		0.1			-2.11	1.81
E		1.55			-5.9	4.47
F		3.57			-11.35	6.51
G		1.52			-6.69	4.37

The flame retardancy values of the untreated, treated samples before and after 5 laundering cycles is calculated and reported in Table 3 to evaluate the laundering stability of the finishing. Flame retardant properties of the samples enhanced with the applied receipts approximately 80% and bigger. It can be realized that the concentration of both Nano PU and  $Al_2O_3$  nanoparticle are important determining factor in

the flame retardancy property. On top of that, increasing Nano PU and  $Al_2O_3$  nanoparticle concentrations have greatly affected the resulted values. The best results were taken from the samples treated with F and G receipts. Furthermore, it can reach a conclusion that the flame retardancy property of treated linen fabrics can withstand against laundering for at least 5 washing cycles.

**Table 3.** Flame retardancy test results of untreated and treated flax fabrics after treatment and washing cycles

Samples	Flame retardancy (s)	% Difference	Flame retardancy after washing cycle (s)	% Difference
U	29.025	-	-	-
A	47.7	64.34	35.1	20.93
B	41.725	43.76	31.4	8.18
C	52.175	79.76	50.2	72.95
D	52.825	81.99	50.6	74.33
E	52.325	80.28	48.9	68.47
F	55.075	89.75	51.4	77.09
G	54.875	89.06	51.2	76.40

Table 4 shows the warp and weft breaking strength test results of the untreated and treated flax fabrics and the difference (%) of the treated fabrics values according to the untreated sample. With the A and B control samples, it can be clearly seen BTCA and Nano PU contents quite a little affected breaking strength of the fabrics in the negative direction. With  $Al_2O_3$  nanoparticle

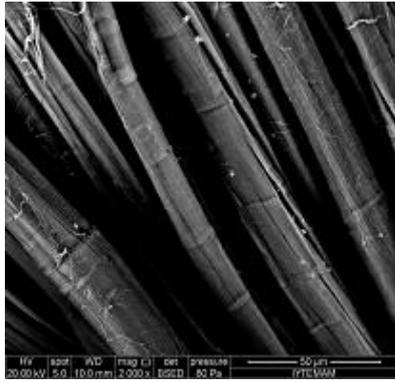
addition in the receipts this negative direction changed to the positive side. Especially for the G sample in the warp samples breaking strength values increased approximately 8%. It is concluded that the resulted treated samples breaking strength properties did not affect in a negative way.

**Table 4.** Breaking strength of the untreated and treated flax fabrics

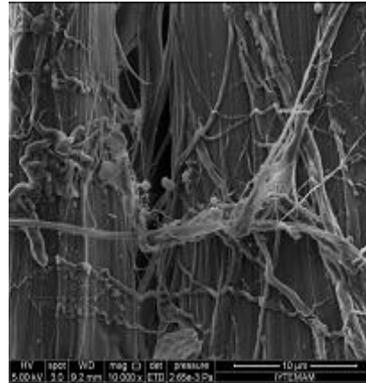
Samples	Breaking strength (warp, N)	Difference (%)	Breaking strength (weft, N)	Difference (%)
U	606.648	-	513.742	-
A	599.856	- 1.12	503.21	- 2.05
B	611.698	0.83	512.232	- 0.29
C	629.486	3.76	514.238	0.09
D	635.08	4.69	515.646	0.37
E	614.256	1.25	509.864	- 0.76
F	619.982	2.19	511.223	- 0.49
G	655.854	8.11	515.439	0.33

Scanning electron microscopic analysis examined the surface topology of untreated and treated flax fabrics shows in Figure 4. The SEM photographs expose that the effect of crosslinking is visible as the definition of fine structural detail in the

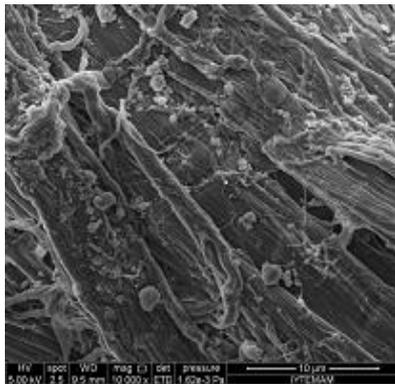
surface regions of flax fibers. With  $Al_2O_3$  nanoparticle addition in the receipts, some agglomerations on the fibers surfaces were seen. The surface topography of the fibers is entirely modified after the treatments.



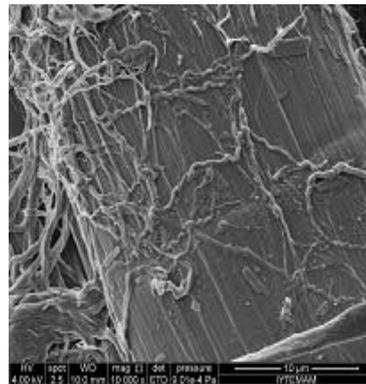
U



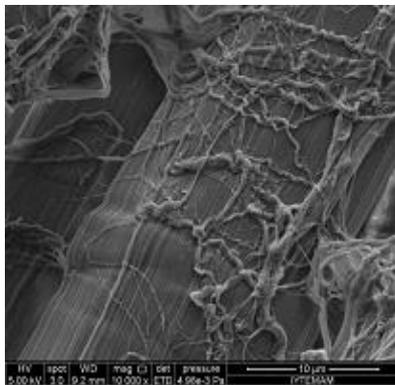
C



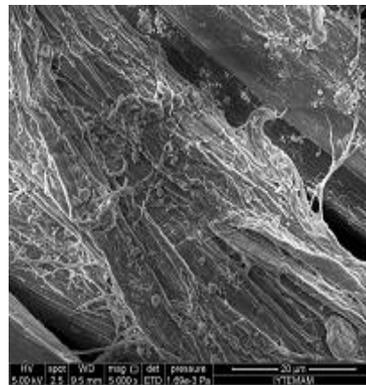
D



E

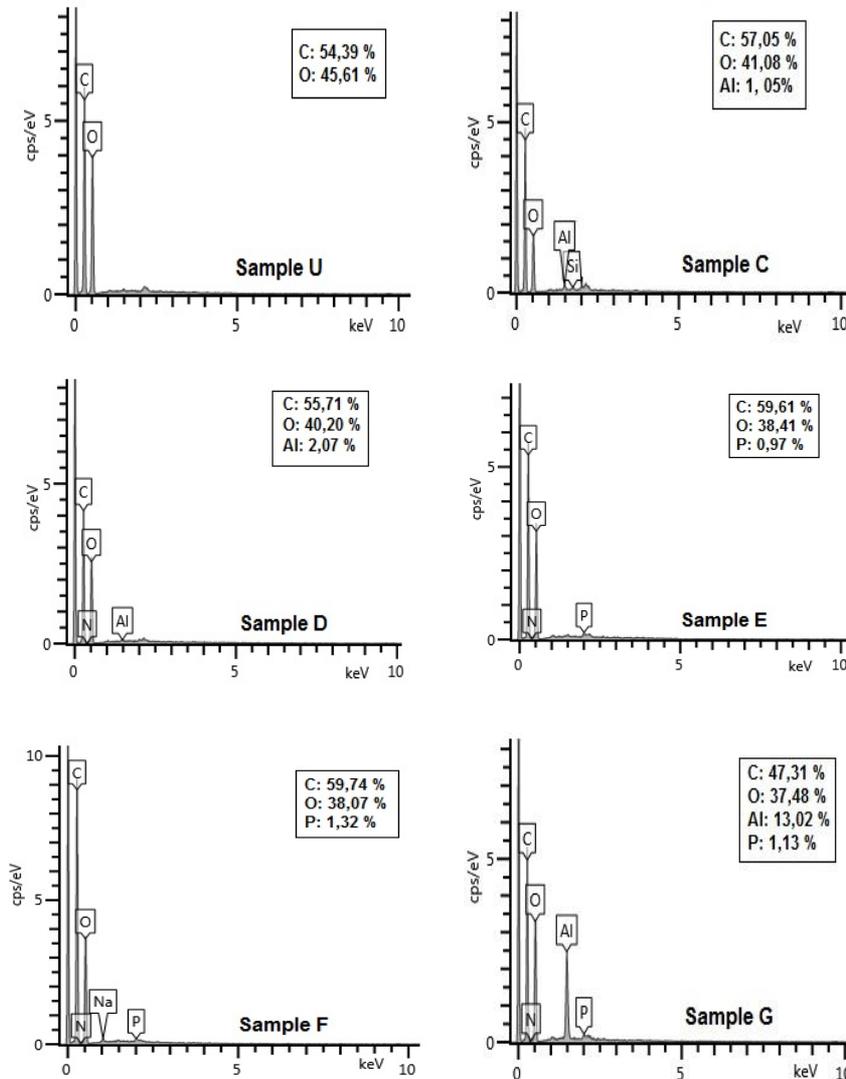


F



G

**Figure 4.** SEM spectra of untreated and treated flax fabrics.



**Figure 5.** EDS spectra of untreated and treated flax fabrics

SEM-EDS analysis was also performed to verify the elemental composition of the treated flax fabric surfaces. Figure 5 shows EDS survey spectra of untreated and treated flax fabrics. The aluminium amount is determined as 1.05% for C, 2,07% for D and 13.02% for G samples. In the use of Nano PU and BTCA as crosslinking agent in the G receipt the aluminium peak shows an increase. These results promoted by the SEM photographs.

#### 4. Discussion and Conclusions

In conclusion, it is demonstrated and characterized the possible durable multifunctional finishing process of linen fabrics. ATR-FTIR and SEM verified the presence of the Al<sub>2</sub>O<sub>3</sub> nanoparticles and Nano PU deposited on the flax fibres. The flax

fabrics applied with the Al<sub>2</sub>O<sub>3</sub> nanoparticles exhibited attractive wrinkle resistant and flame retardant properties. With the receipt G can be seen as the optimum values of the contents according to functional and physical properties. Al<sub>2</sub>O<sub>3</sub> nanoparticles used as co-reactant into the chemical formulation enhanced the performance of chemical finishing and minimize side effects.

#### References

- Chen C. C., Wang C.C. (2006). Crosslinking of cotton cellulose with succinic acid in the presence of titanium dioxide nano-catalyst under UV irradiation. *J Sol Gel Sci Technol* 40(1):31-38.

- Choi H.M., Welch C.M., Morris N.M. (1994). Nonphosphorus catalysts for formaldehyde-free DP finishing of cotton with 1,2,3,4-butanetetracarboxylic acid. *Textil de Res J* 64(9):501-507.
- Holme I. (2007). Innovative Technologies for high performance textiles. *Color Technol* 123:59-73.
- Kim E., Csiszar E. (2005). Chemical Finishing of Linen and Ramie Fabrics, *journal of Natural Fibers*, Vol. 2(3), 39-52.
- Lam Y. L., Kan C.W., Yuen C. W: M. (2010). Wrinkle-resistant finishing of cotton fabric with BTCA-the effect of cocatalyst. *Textile Res J*. Doi:10.1177/0040517510380777.
- Liu X., Hu J., Babu M.K., Wang S. (2008). Elasticity and Shape Memory Effect of Shape Memory Fabrics. *Textile Research Journal*, 78: 1048-1056.
- Mccall R. D., Kernaghan K. J., Sharma, H. S. S. (2001). Analysis of a Crease-Resisting Finish on Linen Fabrics Using Fourier Transform Infrared Spectroscopy and Visible and Near-Infrared Spectroscopy, *Journal of Applied Polymer Science*, Vol. 82, 1886-1896.
- Tzanov T., Cavaco-Paulo A. (2006). Surface modification of cellulose fibers with hydrolases and kinases. Modified fibers with medical and speciality applications, 59-180.
- Uğur Ş.S., Sarıışık M., Aktaş A.H. (2011). Nano-Al<sub>2</sub>O<sub>3</sub> Multilayer Film Deposition on Cotton Fabrics by Layer-by Layer Deposition Method, *Materials Research Bulletin*, 46, 1202-1206.
- Vasile S., Ciesielska-Wrobel I. L., Langenhove L. V. (2012). Wrinkle Recovery of Flax Fabrics with embedded Suparelastic Shape Memory Alloys Wires, *FIBRES & TeXTİLES in Eastern Europe*, 20, 4(93): 56-61.
- Wang C.C., Chen C.C. (2005). Physical properties of crosslinked cellulose catalyzed with nano titanium dioxide. *J Appl Polym Sci* 97(6):2450-2456.
- Yuen C.W. M., Ku S.K.A., Kan C.W., Cheng Y.F., Choi P. S.R., Lam Y.L. (2007). Using nano TiO<sub>2</sub> as co-catalyst for improving wrinkle-resistant of cotton fabric. *Surf Rev Lett* 14(4):571-575.