

Improvement of Plywood Surface with Urethane Acrylate (M-1200) by UV Curing: Effect of Diluent Monomers and Surface Pretreatment on Physico-Mechanical and Degradable Properties

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Abstract

A set of formulations were prepared from oligomer urethane acrylate (M-1200). This oligomer is very sticky at room temperature and it was diluted with diluent monomer to reduce its viscosity so the formulated solutions could be easily coated on a substrate to prepare thin polymer film. The reactive diluents are a tri-functional monomer, trimethylolpropanetriacrylate (TMPTA) and 2-ethyl-2-(hydroxymethyl)-1,3-propanediolmethacrylate (EHMPTME). Photoinitiator Irgacure-369 was added to the formulations so that UV radiation could initiate free radical formation and ultimately these could start the overall polymerization process in the system. Thin polymer films were prepared on glass plate with these formulated solutions and finally applied on polished plywood surface to modify the surface, and both were cured under UV-radiation. The properties of UV-cured thin films were studied as a function of UV radiation intensities. Various characterization tests of the coated plywood substrates reveal that the properties of coated surfaces by the formulating solutions were terrifically increased. The raw plywood has 20% pendulum hardness (PH), but the samples treated by the coating T1 and T3 have 240% increase of surface hardness. Out of the two reactive monomers EHMPTME yielded overall better properties to the cured surface which increases surface gloss about 3000%. The adhesion strength and abrasion resistances are also higher for the coat prepared from this diluent. The surface of the plywood was also pretreated by applying a base coat. The pretreated cured surface only increases a small degree of the properties. The effect of simulating weather showed the cured surface yielded minimum loss of its properties after exposed to different weathering conditions.

Key words: Plywood; surface mutation; polymer coating; UV curing; urethane acrylate; physico-mechanical properties

Introduction

Modification of polymer surface is nowadays a quite common process to enhance polymer properties. Indeed, according to the nature of the grafted functional groups, properties such as biocompatibility, wetting with water, printability, and adhesion can be changed in a revolutionary way. The protective coatings on surfaces of various materials used today in outdoor and indoor applications are fabricated of solvent-based formulations, mainly thermosetting acrylates and unsaturated alkyd esters. UV-curable resins appear an alternative, as solvent-free formulation can be cured rapidly at ambient temperature to produce highly cross-linked polymers. These formulations offer several advantages as well for instance, curing occurs at ambient temperatures, which reduces reaction time; continuous operation; improved monomer stability; less atmospheric pollution; increased design flexibility through process control; and production of very high quality products [1-3]. The light stabilization of UV-curable coatings has been reported in several studies [4-7] but no major industrial openings have enlarged so far. One of the vital and intrinsic problems of this system is that the inner filler effect of the added UV absorber is slowing down the curing speed and is shortening the cure depth as well [5].

When a solution of initiator and monomer is irradiated on the substrate that must be modified, only one step is necessary. In a two-step setup, a solution of photoinitiator is first irradiated on the film surface to create initiating sites on this film. Then, a solution of monomer is added to this pretreated film and also irradiated. The monomer is thus grafted onto the film. Many experiments using

different monomers and different substrates such as polypropylene [8], polycarbonate [9], polyethylene [10], wood [11], partex, particle board [12], leather [13], hessian cloth (jute) [14], paperboard [15] etc. were previously reported in the literature. Recently, most of the work on UV-initiated photo polymerization was focused on a free-radical system based primarily on acrylate monomers. These monomers polymerize rapidly and are easily modified at the enter functionality, allowing materials with a variety of properties to be obtained.

In Bangladesh as in many other developing countries plywood has always been a construction material. It's a kind of board made by sticking thin layers of wood on top of each other. Its properties such as its higher strength relative to its weight, low heat conductivity made it advantageous for use in construction and decorative purposes. It usually used in doors, panels, windows, and furniture in Bangladesh. However, plywood has some drawbacks; all the inherent properties of plywood are not favorable for diversive uses; and some of them restrict its versatile and potential applications. Plywood is hygroscopic, and is dimensionally instable with variation of moisture content. This eventually results in weeping and uneven distortion. The nonabrasive character and the resistance to weather conditions of plywood are relatively low. Under some favorable conditions termites, bacteria, insects, microbes, and fungi can destroy plywood structure in a short period of time, and plywood is susceptible to combustion as well. The present work was undertaken to develop some formulations from M-1200 oligomer^[16]. This monomer is very sticky at room temperature so it needs diluents to reduce viscosity and thus the formulated solutions can be easily incorporated into the substrate surface to improve surface

through enhancing tensile strength, flexibility, abrasion resistance, adhesion strength, and surface gloss, and to reduce water absorption property. The reactive diluents used in this work are TMPTA (trimethylol propane triacrylate) and EHMPTME (2-ethyl-1-(hydroxymethyl)-1, 3-propanediol trimethacrylate). A comparison of the physico-mechanical properties of the cured surface by these two diluents was done. The effect of surface treatment by acetic acid and on the properties of modified surface was also investigated. The degradable properties of the modified surface due to simulating weathering test (for 300h) and soil degradation (for a period of 21 weeks) were also monitored.

Experimental

Materials

Urethane acrylate (M-1200) an Aronix product was procured from Toagosei Chemical Industry Co. Ltd. (Japan). This oligomer is almost solid and trimethylol propane triacrylate (TMPTA), a trifunctional monomer were procured from Radcure (Drogenbos, Belgium). Methyl Ethyl Ketone (MEK) was used as an antibubbling agent, and was obtained from Merck (Germany). Irgacure-369 (IRG-369) has an absorption band of 200–450 nm, and was used as photoinitiator during the UV radiation process to initiate photochemical reactions. It was procured from Ciba-Geigy. Plywood sample was collected from the local market of Bangladesh.

Methods

Different formulations were developed with urethane acrylate (M-1200) in combination with reactive monomer TMPTA (trimethylolpropanetricacrylate) and

EHMPTME (2-ethyl-1-(hydroxyl)-1, 3-propanedioltrimethacrylate. Irgacure 369(2-benzyl-2-dimethylamino-1(4-morpholinophenyl)-butanone) was added into the formulated solutions to initiate photochemical reaction for free radical polymerization. The compositions of formulations are shown in Table 1. The formulated solutions were applied on thin glass plate as well as on to the substrate surface that was smoothly polished by suitable sand paper (No. 0 and 1) before applying the coating. The substrate having the size 4"×4"×1/6" was coated with these formulated solutions using suitable bar coater (No.0.0028 for base coating and 0.0018 for top coating and on glass plate and these were procured from Abbey Chemicals Co. Australia). The thickness of the film was $36 \pm 3\mu\text{m}$. The coated glass plates and the samples were then exposed to a UV radiation source of 254–313 nm wave length and 2 kW power capacities (IST Technique Germany). The samples and the glass plates were passed to the radiation for several times to ensure full curing of the film through a conveyor belt attached to the machine. The speed of the conveyor was 4 m/min. After 24 h of application of UV radiation the coated glass plates and the substrates were subjected to various characterization tests.

Polymer film characterization

Polymer films prepared from different formulating solutions were characterized after 24 h of curing under UV radiation. Film hardness, an index of cross linking density at the surface of the film, was measured by using a pendulum hardness tester (model 5458, Byke Labotron). Gel content, amount of crosslink through the entire cured film, was determined by wrapping a known weight of the film in a

soxhlet, for extraction with hot acetone for 48 h. The difference of weight of the film before and after extraction determines the gel content. The extracted film was soaked in acetone for 24 h at room temperature. The difference of weight of the swollen film with that of the dry extracted film yields the swelling ratio. The cured films were carefully peeled off from the glass plates and these were cut with the help of a dumbbell cutter for tensile property measurement. The tensile properties of films was measured by using tensile strength machine (Instron, model-1011, UK) maintaining a gauge length of 0.50 inch at a crosshead speed of 0.01 in. /min. Tensile strength (TS) and elongation at break (E_b) were directly obtained with the help of a personal computer integrated with the machine.

Application on plywood surface

Plywood samples were cut into small pieces (size 4"×4"×1/6") and dried in an oven at 105°C to remove the free moisture until a constant weight is achieved by each sample. The surface of the plywood samples were polished smoothly with the help of a suitable sand paper. Then it was coated first with a base coat using a drawn down bar (No. 0.0028) and cured by a single pass under the UV lamp. It was polished again and coated with a top coat using the bar No. 0.0018, and finally the coating was cured under UV light at different intensities (number of passes). Surface hardness was measured by pendulum method as described above. The glossiness of the modified or muted surface was measured at two angles 60° and 20° using a micro gloss meter (Sheen-155) from Sheen Co., UK. Abrasion wear of the coated plywood surface was measured by the taber abrasion method using a taber abraser

(model 5130 of Richness Co., Germany). The lower taber index indicates better resistance to abrasion by the cured film. The adhesion ability of the UV-cured surface was determined by measuring the force that was applied to peel a certain portion of the cured film with the help of an adhesion tester (model 525) from Erichsen Co., Germany.

Pretreatment of the surface

1-5% acetic acid solution in water was used as pre-treating agents for plywood surface. In this respect acetic acid solution was poured into the plywood surface, after 5 minutes the acid solution was wiped out and washed with distilled water. It was then dried in oven for 10 hours to remove moisture. Finally the dried samples were coated by formulating solutions and cured under same UV radiation. The effect of pretreatment on different properties of the cured plywood surface was measured.

Degradability test

The coated plywood samples were subjected to severe weathering testing by simulating sunlight (4 h) using an UV lamp (313 nm) and condensation (2 h). The accelerated weathering testing was carried out for 300 h with the help of a weathering tester (Q-UV-26200, Q-Panel Co. USA). The loss of PH, gloss and adhesion properties as a result of simulated weathering test has been determined. The treated samples were also kept at 60° angle on the roof of the building (50 ft. above the plain), buried in soil and in a room at normal atmospheric conditions for 120 days to monitor the degradable nature of the coating. Underneath these conditions different properties of the coated surface were periodically monitored.

Results and Discussion

It is necessary to characterize UV-cured polymer films before applying on the surface of plywood. Most of the data presented in this report are averaged values of at least five different measurements.

Characterization of the UV cured polymer film

Film hardness

Surface hardness, an index of cross linking density at the surface of UV-cured thin polymer films prepared on a glass plate was determined by the pendulum method. Results of pendulum hardness (PH) of the films prepared at different UV doses represented by the number of passes are graphically shown in Figure 1. The PH increases with the UV dose, attains a maxima, and then decreases. The decrease of the PH could be caused by degradation of polymers cross linking at higher doses [17]. The exact mechanism of such degradation is not clearly known however, it could be the cleavage of the polymer chain due to overdose of UV radiation. The maximum PH values are obtained in between 12 and 14 passes of radiation doses for TMPTA series while it is at the 42nd passes in EHMPTME series. The formulation T3 containing 88% TMPTA yielded the highest PH (93.45%) after 12 passes of radiation followed by T2 in TMPTA series. We observed that increasing TMPTA concentration into the formulations increases the PH value to the same extent it is because TMPTA has trifunctional acrylated groups and thus has a branchlike behavior that creates more cross linking among the ingredients of the formulation [18]. While in EHMPTME series E2 containing 78% diluent yielded the highest PH after 42 passes

of UV radiation. This case PH increases with diluent concentration up to 78% but it decreases for more concentration monomer in the formulations. The most important observation in this experiment is EHMPTME needs more radiation doses to be cured than the other diluent TMPTA. The film cured with TMPTA showed higher surface hardness than the film prepared from EHMPTME.

Gel Content

The gel content is a representation of cross linking density through the entire films. Figure 2 shows the gel content of polymer films against the number of passes. The maximum gel content value (97.65%) is achieved by formulation T3 after 14 passes of radiation doses. This formulation contains higher TMPTA (88%) (Table I), and TMPTA has trifunctional acrylated groups that have a branching effect [17] and can yield more cross linking in the cured film. A multifunctional vinyl monomer promotes rapid free radical propagation reaction [18] leading to network (cross linking) structures. When TMPTA concentration is increased, the amount of residual unsaturation is also increased as a consequence of faster rate of formation of the three-dimensional network, causing restricted mobility at an early stage. The cross linking rate, especially during the early stage of radiation, is proportional to the TMPTA concentration [20]. The second highest gel content in the same series is given by the formulation E2 (95.7%) followed by E1. In the EHMPTME series the maximum gel is yielded by the film obtained from formulation E1 having the least amount of diluent followed by E2 and E3. In this case we observed the diluent concentration decreases the amount of cross linking. The maximum gel content

varies between 96 to 92% for this series indicates a sign of very good cross linking phenomena among the various constituents present in different formulations.

Tensile strength (TS)

Tensile property is very important in selecting diverse applications of polymer and polymer-coated flexible substrates. Results of tensile strength (kPSi) of UV cured polymer films are graphically shown in Figure 3 against the intensity of UV radiation. It is found that TS increases with radiation intensities and after maximum strength is achieved the TS then decreases. The maximum TS is obtained in between 12 to 14 passes for TMPTA diluent and after 39 passes for EHMPTME series and as usually the TS reduces after the attainment of maximum values. This may be caused by radiation deterioration of the polymer at higher doses [21]. However, the highest TS (8.5kPSi) are given by the formulation T2 in TMPTA series while the maximum TS (83-85kPSi) are yielded the formulation E1. In this case EHMPTME showed higher TS than that given by TMPTA.

Elongation at break (E_b)

Flexibility of a polymer is an essential parameter that is very actively considered during the application of a polymer in a certain area. The extent of elongation of these UV-cured films at the breaking point is shown in Figure 4. It is obvious that the film that showed higher TS doesn't necessarily showed higher elongation at break. For instance, in TMPTA series the highest elongation (22%) is achieved by the formulation T1, while the highest TS were observed for formulation T2. Similar

phenomena is observed for EHMPTME series as well hence, T1 showed the highest TS while formulation T2 yielded the highest elongation at break. It is also noticed that EHMPTME series gave higher elongation the TMPTA series.

Application of formulated solutions on plywood surface

After characterization of the UV-cured polymer films, different formulations were applied on the plywood surface. Formulation T1 in TMPTA series and formulation E1 in EHMPTME series were chosen as top coat because these two samples showed higher surface hardness in their respective diluent series. The other formulations T2, T3 and E2, E3 were applied as top coats. The base coats were applied on polished plywood surface and partially cured with UV radiation. Then the substrates were polished with suitable sandpaper. Top coat was applied and cured with UV radiation at different number of passes viz. 4-20 for TMPTA series and 30 to 45 for EHMPTME series. Various physical and mechanical parameters were determined as a function number of passes.

Pendulum hardness of the coated plywood surface

The surface hardness of the film coatings on plywood is measured by the pendulum technique. The results for both diluents series are graphically represented in Figure 5 versus the intensities of the UV radiation. The PH increases with an increase in the UV radiation doses; this reflects that there is an increase of the cross linking density in the polymer with the increase of the radiation intensities. The highest PH for TMPTA diluent's is usually given after 14 to 16 passes of UV

radiation on the coated surface while it needs about 40 passes of UV radiation to be cured completely for the diluent's EHMPTME. The figure also showed that relatively higher surface hardness was achieved by the sample coated with diluent TMPTMA. The highest PH (68.7%) was obtained for the sample coated with base coat T1 and top coat T3 and for the other diluent E1+E3 yielded maximum surface hardness.

Micro gloss property

The micro gloss property determines the attractiveness of the surface of the substrates and if the micro gloss values are higher this means it appears good to the human eyes. The gloss is generally measured at two angles (60° and 20°) of the plane of the modified surface. Results of surface gloss at 60° and 20° angles of the coated plain board substrates were graphically shown in Figure 6 and 7 against the doses of UV radiation. Gloss determined at 60° angles is generally higher than at 20° and the micro gloss properties is directly related to the extent of cross-linking and the surface gloss increases with an increase of cross-linking density [10]. It is observed that the formulations containing diluent EHMPTME showed predominantly higher surface gloss than the sample coated with the formulations having diluent TMPTA. It means that EHMPTME increases the glossiness of the surface while TMPTMA increases the hardness of the surface. We have observed from the figure that higher surface gloss both at 20° and 60° were achieved by the formulation coated with T1+T3.

Taber Abrasion

The coated (modified) plywood surface was abraded between two abrader revolving wheels applying a load of 100 g/cycle. The weight loss obtained by the sample due to such abrasion between the wheels in 100 cycles is related to the taber wear index. This means that the abrasion resistance is high, when the taber wear index is low. Results of taber wear index are shown in Figure 9. We observed that like wise glossiness of the surface the plywood surface coated with the formulations containing EHMPTME yielded higher resistivity towards abrasion resistance than that contained TMPTA. The lowest taber index value is given by the sample coated with the formulations T1+T3 it means this showed maximum abrasion resistance properties

Adhesion Strength

Adhesion is the force (N/mm^2) that is required to pull out the polymer coating from the plywood surface. If a coating is loosely bound to the substrate surface, then it can be easily peeled off by applying a small peeling force and vice versa. Results of adhesion strength (N/mm^2) are shown in Figure 10 versus UV radiation doses. We observed that EHMPTME containing polymer coatings on to the plywood surface is strongly bound than TMPTA coatings and there no chipping off area during crosscut technique for all sets of formulations. These results showed similar trends to gloss and abrasion as well. It is clear from these results that the presence of EHMPTME diluent to the prepolymer M-1200 gave higher cross link. We perceived that the plywood surface cured with the formulation E1+E3 yielded better physical

properties than that cured by T1+T3 although it requires higher UV radiation. We have presumed this formulation as optimized between these two diluents and for further experiments of this study only this condition will be applied.

Surface pretreatment

Plywood surface was pretreated with 1-5% acetic acid solution in water before applying the optimized formulations. In this respect acetic acid solution was poured into the plywood surface, after 5 minutes the acid solution was wiped out and washed with distilled water. It was then dried in oven for 10 hours to remove water and adhering moisture content. Finally the dried samples were coated by formulating solutions and cured under same UV light. The results of physico-mechanical properties (PH, surface gloss, adhesion and abrasion) are shown in Figure 10 with respect to acetic acid concentration. We observed that the pretreatment process do not effect much to the properties of the cured surface. It is also noticed that increasing acetic acid concentration only increases a little to the all properties.

Simulating weathering test

The plywood substrates coated by the formulation E1+E3 was subjected to severe weather treatment for a period of 300 h in the condition of simulating sunshine, dews and condensation at an alternating cycles. The effects of simulating weathering on various properties (PH, gloss, and adhesion) were assessed and the loss of these properties was graphically plotted in Fig. 11 against the time of weathering in hours. The change towards negative values clarifies that, the

properties initially increases to some extent and then decrease. The increase of the loss of properties may be consequence of the fact caused by further reaction of unused free radical for the period of initial curing method ^[16] (Ali et al. 1996). It is also remarked from the figure that the loss of the properties is very low after severe weathering treatment.

Degradable properties

The sample treated by the coating E1+E3 was kept in diverse environmental conditions like soil (containing approximately 3% water), in a room at normal temperature and pressure and also into the roof at 60° angle (about 60 ft above the ground level) in open air exposing to heavy rain fall, sunlight etc. for a period of 90 days (3 months) in order to study the effect of such environmental conditions on the degradability of the coated sample. The properties were monitored after every 20 days. It is obvious that the loss of each of these properties was more when the plain board samples were placed in soil containing 3% water followed by the sample exposed in to the roof of 60 ft building as it closely come in contact to different natural weathering conditions (like heavy rain fall, dews, sunlight etc.). The minimum losses in all cases were observed when the treated plywood samples were put at room temperature.

Conclusion

Plywood surface was coated with formulating solutions prepared from with oligomer M-1200. The reactive diluents are TMPTA and EHMPTME were used in

different proportions to the formulating solutions. Various characterization tests of the coated plywood substrates reveal that the properties of coated surfaces by the formulating solutions were terrifically increased. The raw plywood has PH of 20% but the samples treated by the coating T1+T3 increases the PH up to which means 240% increase of surface hardness. Out of the two reactive monomers EHMPTME yielded overall better properties to the cured surface it increases surface gloss about 3000%. The adhesion strength and abrasion resistance is also higher for the coat prepared from this diluent. The surface of the plywood was also pretreated applying base coat and top and the results of pretreated cured surface showed pretreatment increase the properties only to a small degree. The effect of simulating weather showed the cured surface yielded minimum loss of its properties after exposed to different weathering conditions.

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Figure Captions

- Fig.1** Pendulum hardness of UV-cured films on glass plate against UV radiation doses
- Fig.2** Gel content (% cross linking) of UV-cured films on glass plate at different radiation intensities
- Fig.3** Tensile strength (TS) (kPSi) of cured polymer films against no. of passes
- Fig.4** Elongation at break (E_b) (%) of UV-cured polymer films against no. of passes
- Fig.5** Surface hardness (%) of cured plywood after applying base coat and top coat against no. of passes.
- Fig.6** Micro gloss at 60° of UV-cured plywood surface at different radiation intensities.
- Fig.7** Micro gloss at 20° of UV-cured plywood surface at different radiation intensities.
- Fig.8** Abrasion resistance (taber wear index) of cured plywood surface versus UV doses.
- Fig.9** Adhesion strength of the coatings (N/mm^2) on cured plywood surface versus UV doses.
- Fig.10** Different properties of the plywood surface after pretreated with different concentration $KMnO_4$ under UV radiation.
- Fig.11** Loss of different physical properties due to accelerated weathering of UV cured plywood surface against the time of weathering in hours.

Fig. 1

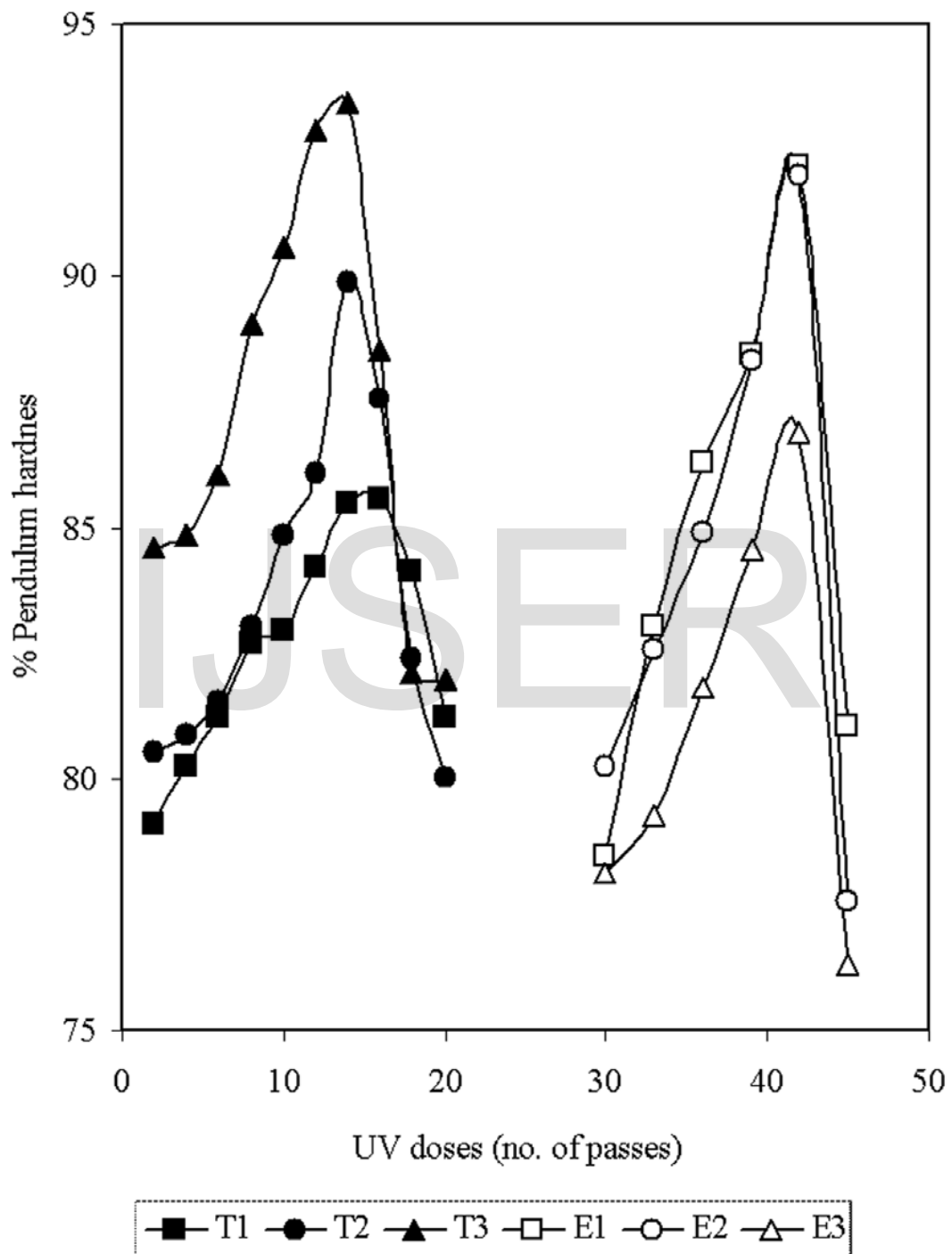


Fig. 2

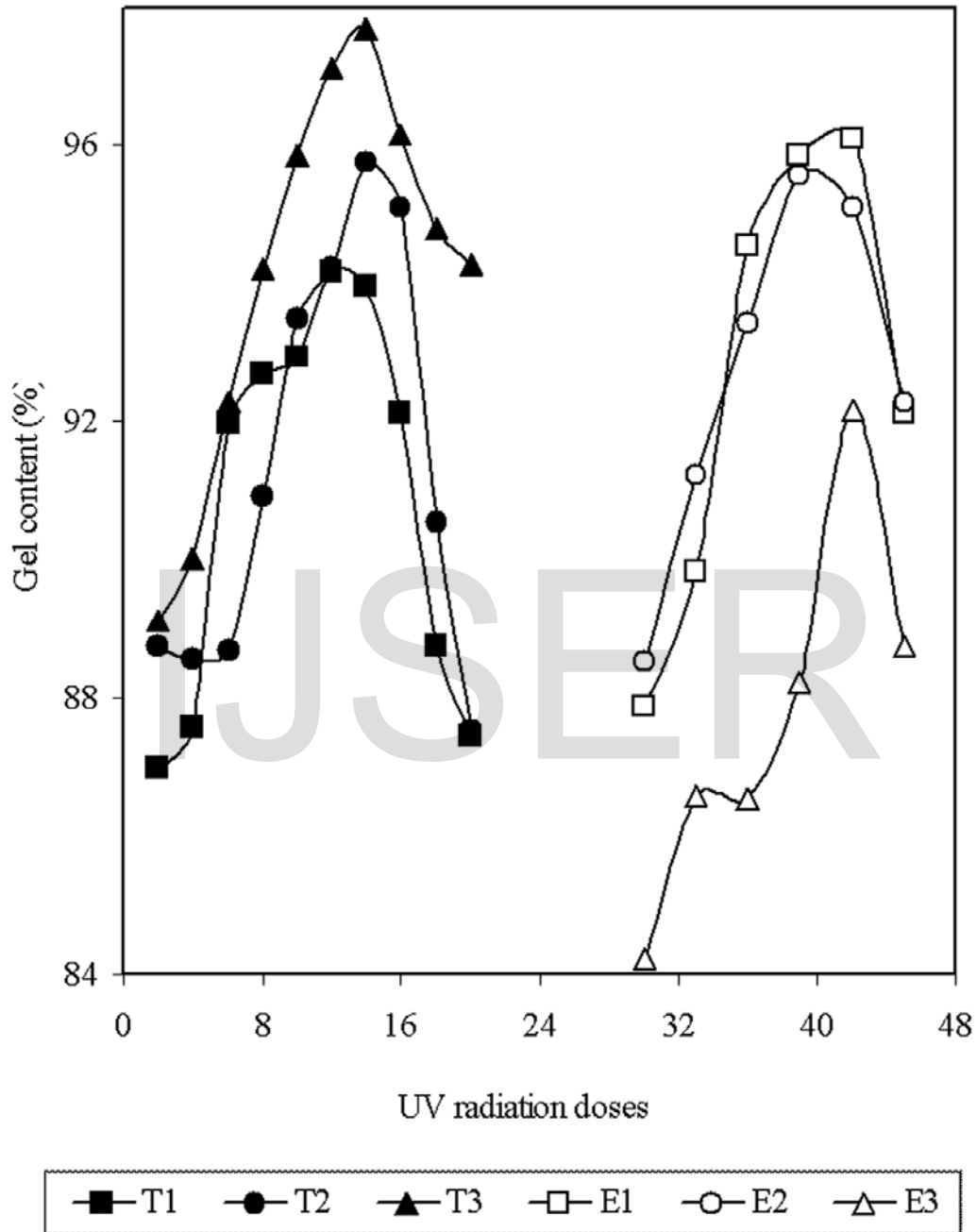


Fig. 3

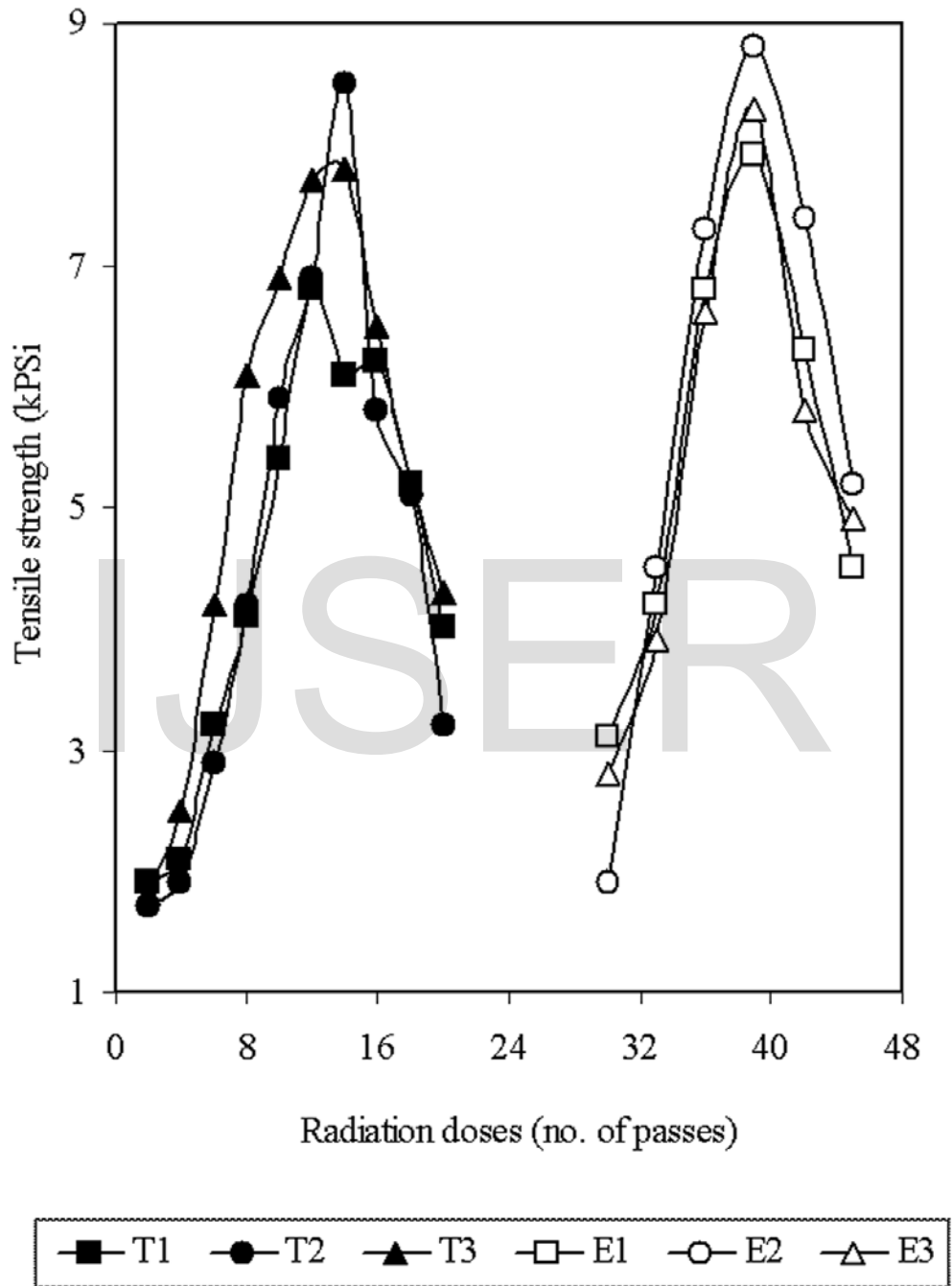


Fig. 4

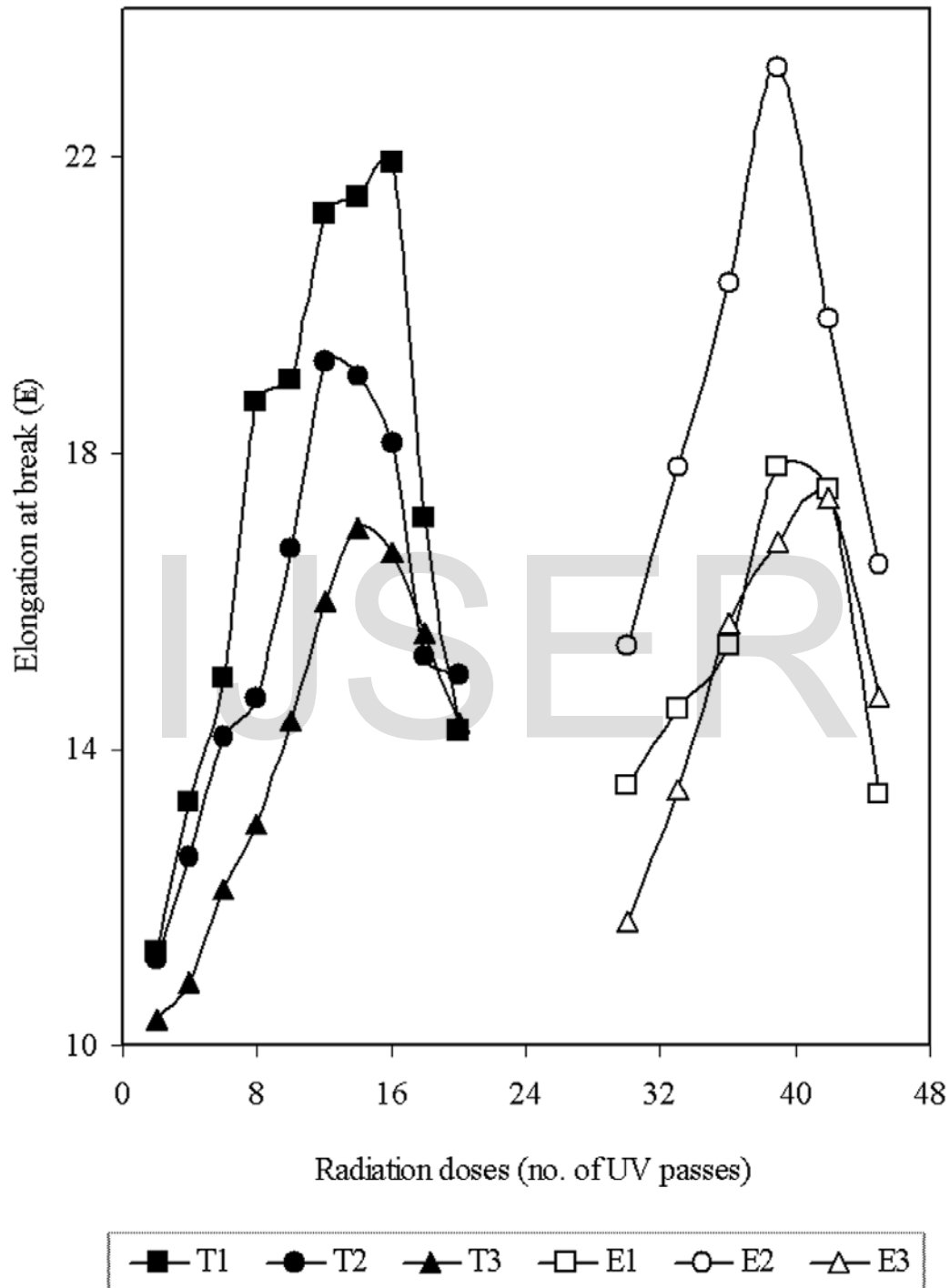


Fig. 5

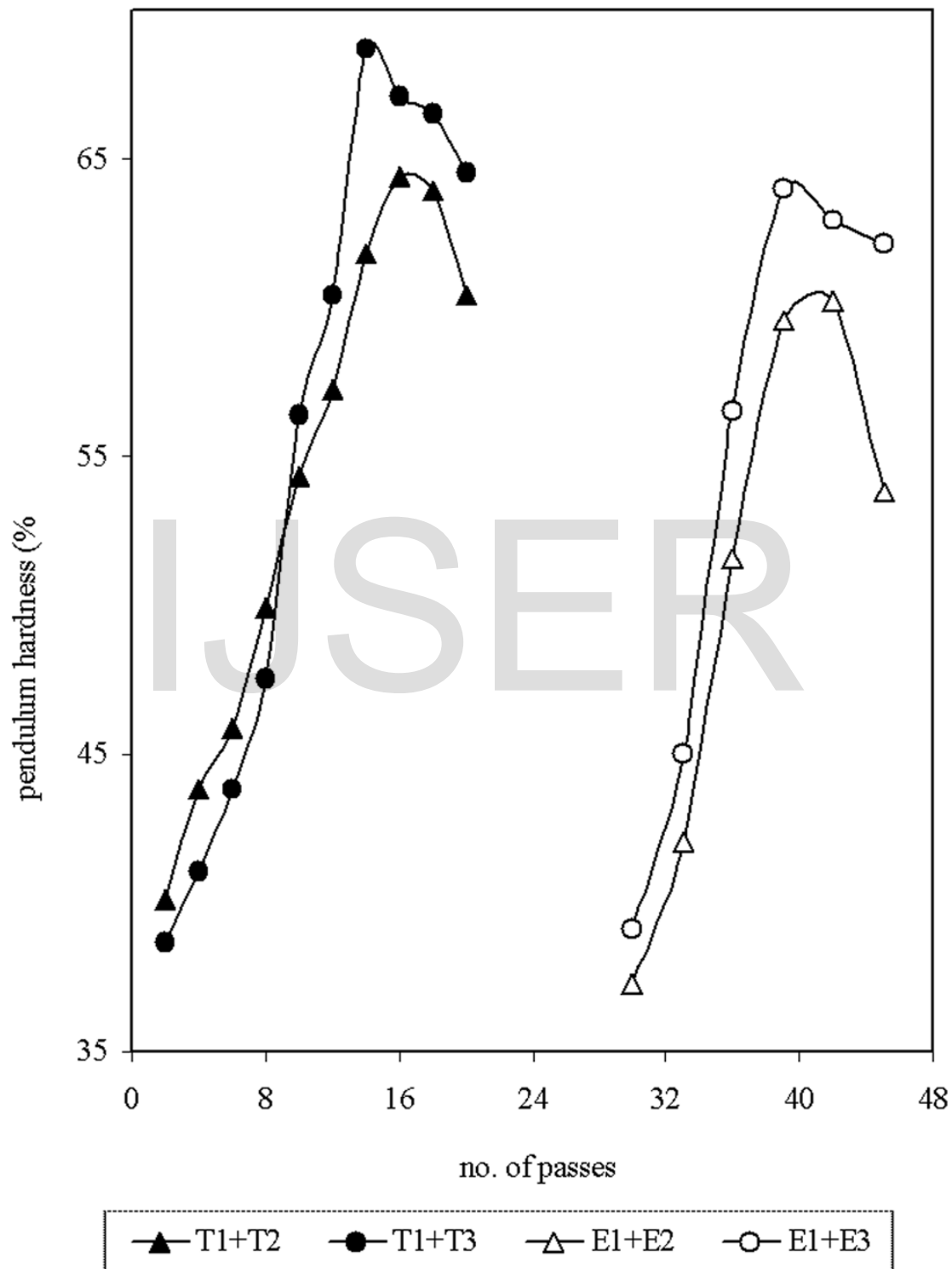


Fig. 6

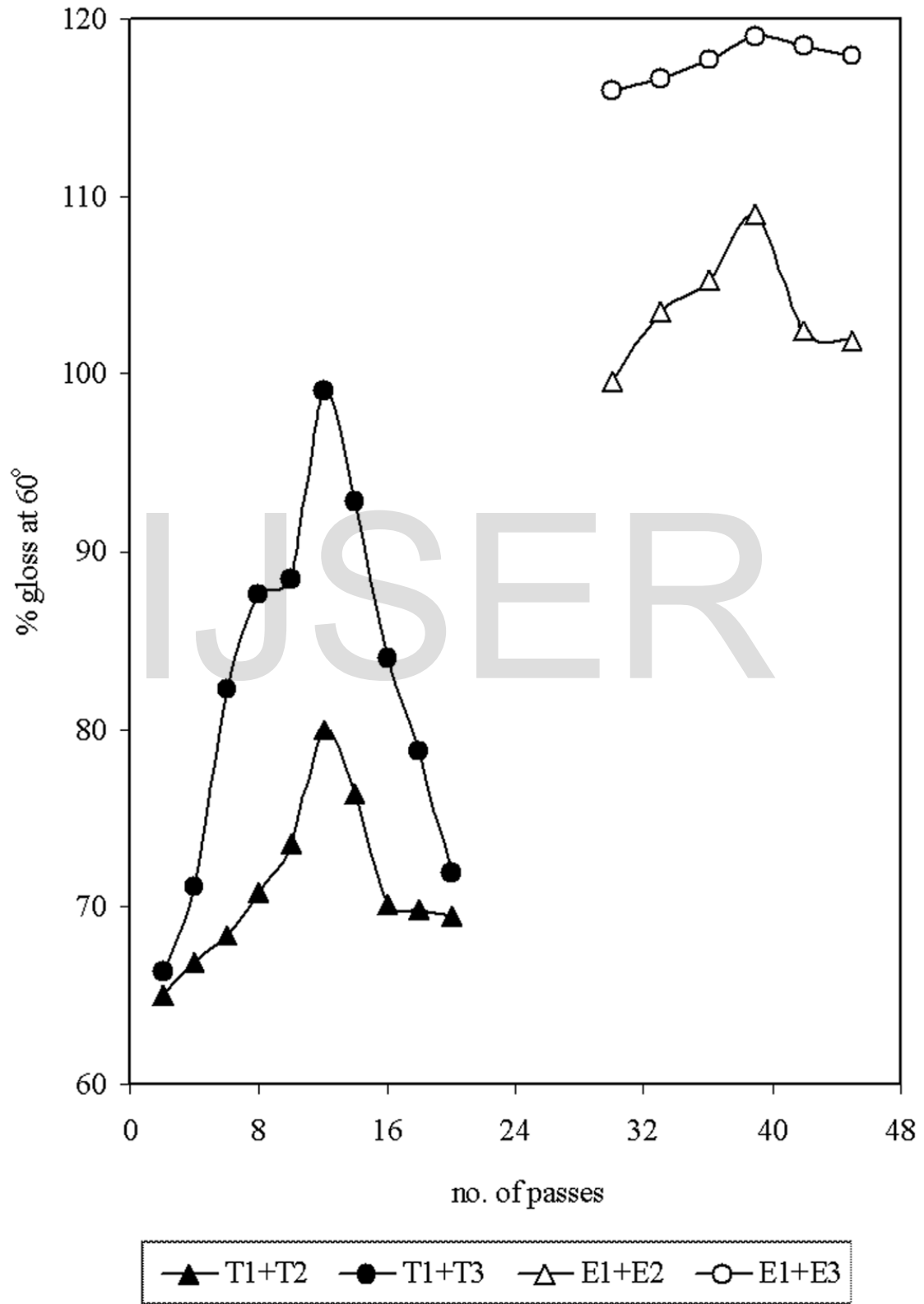


Fig. 7

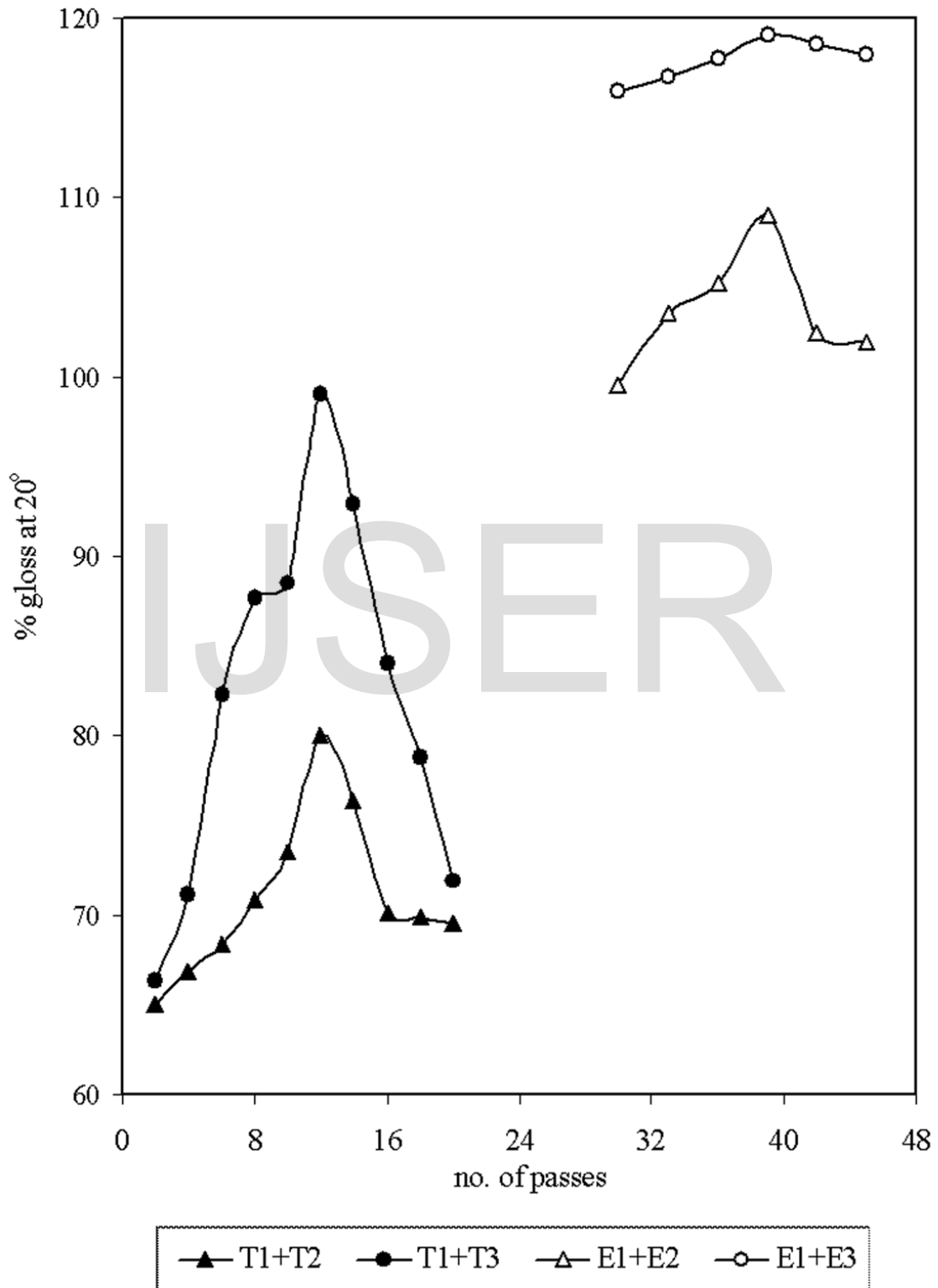


Fig. 8

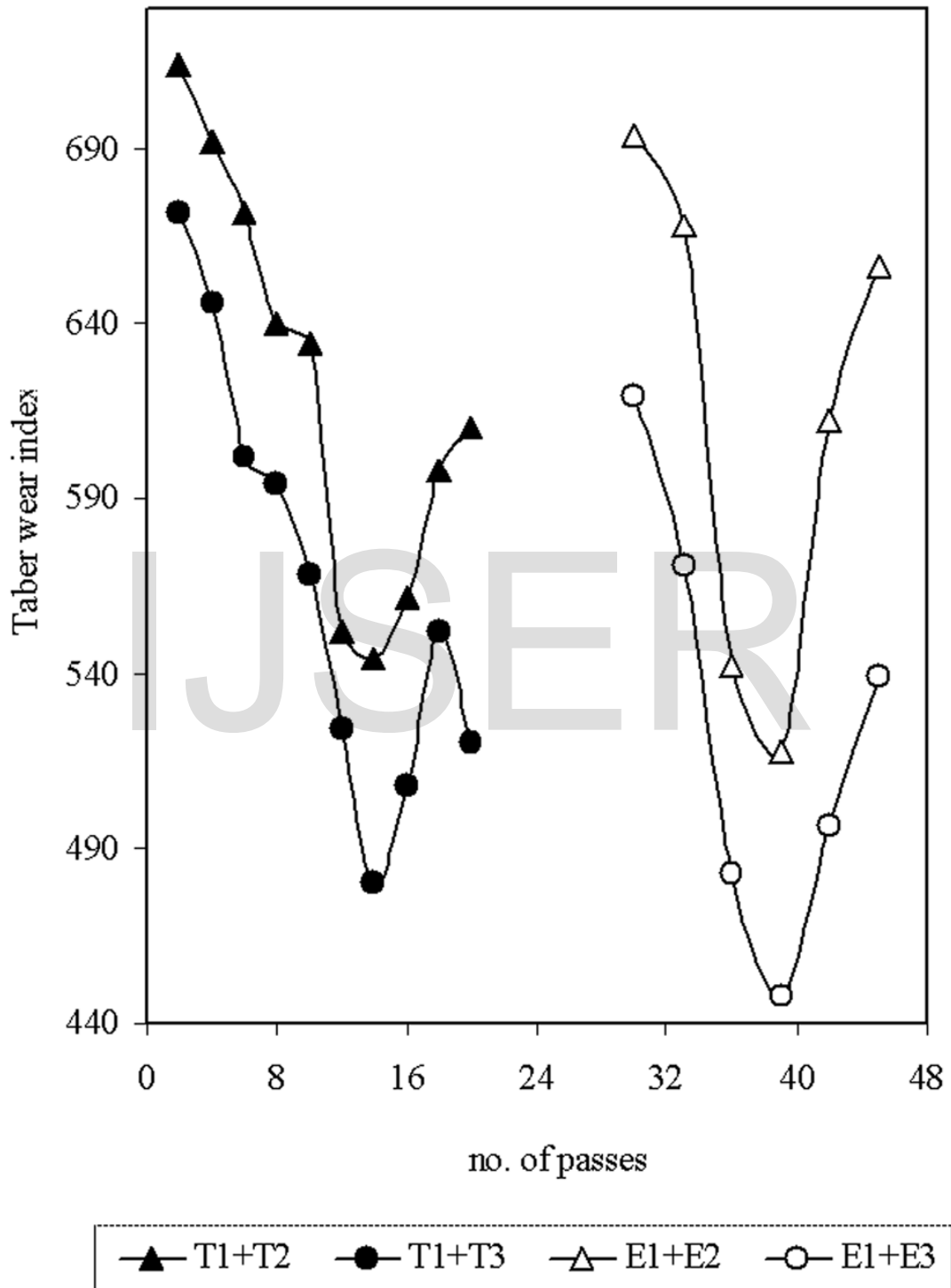


Fig. 9

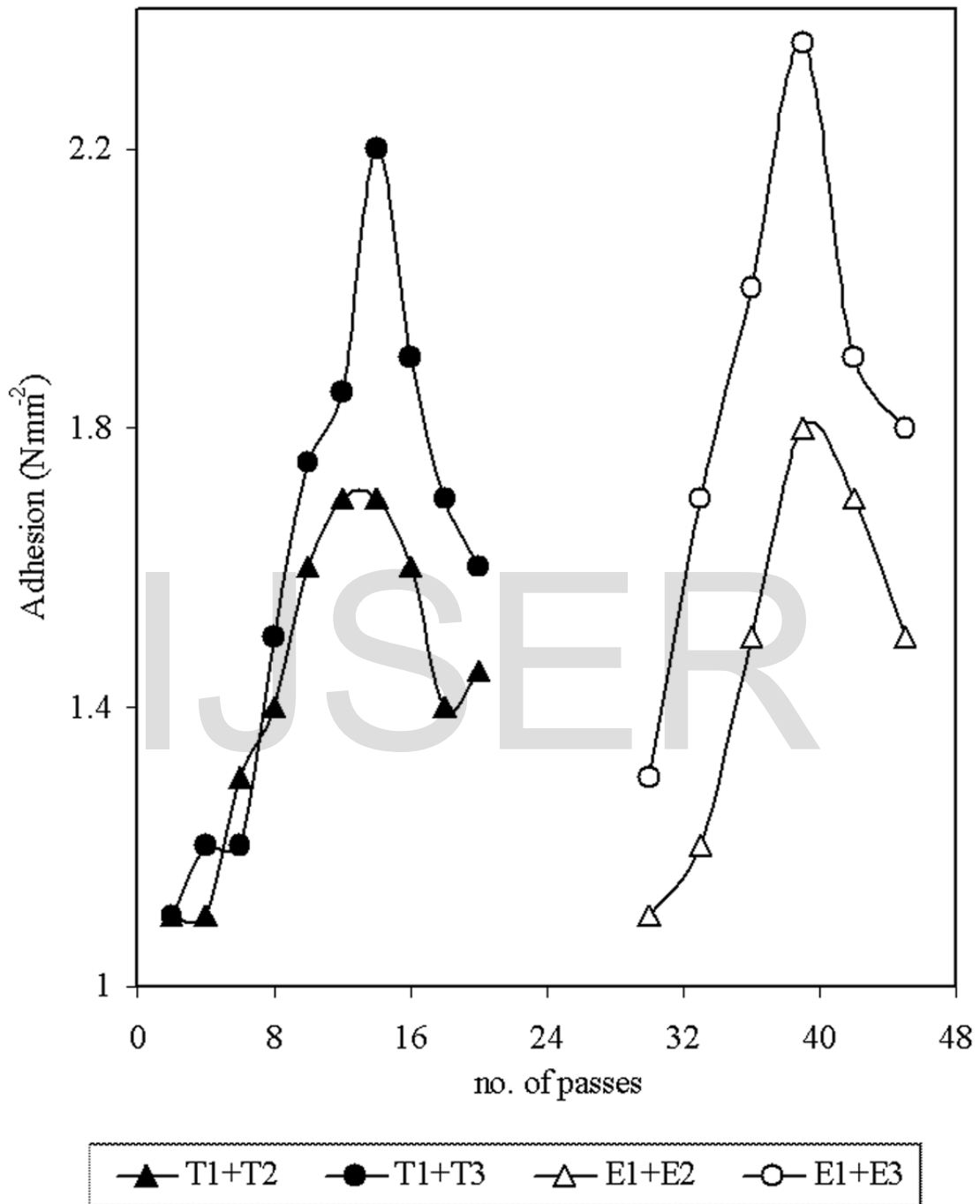


Fig. 10

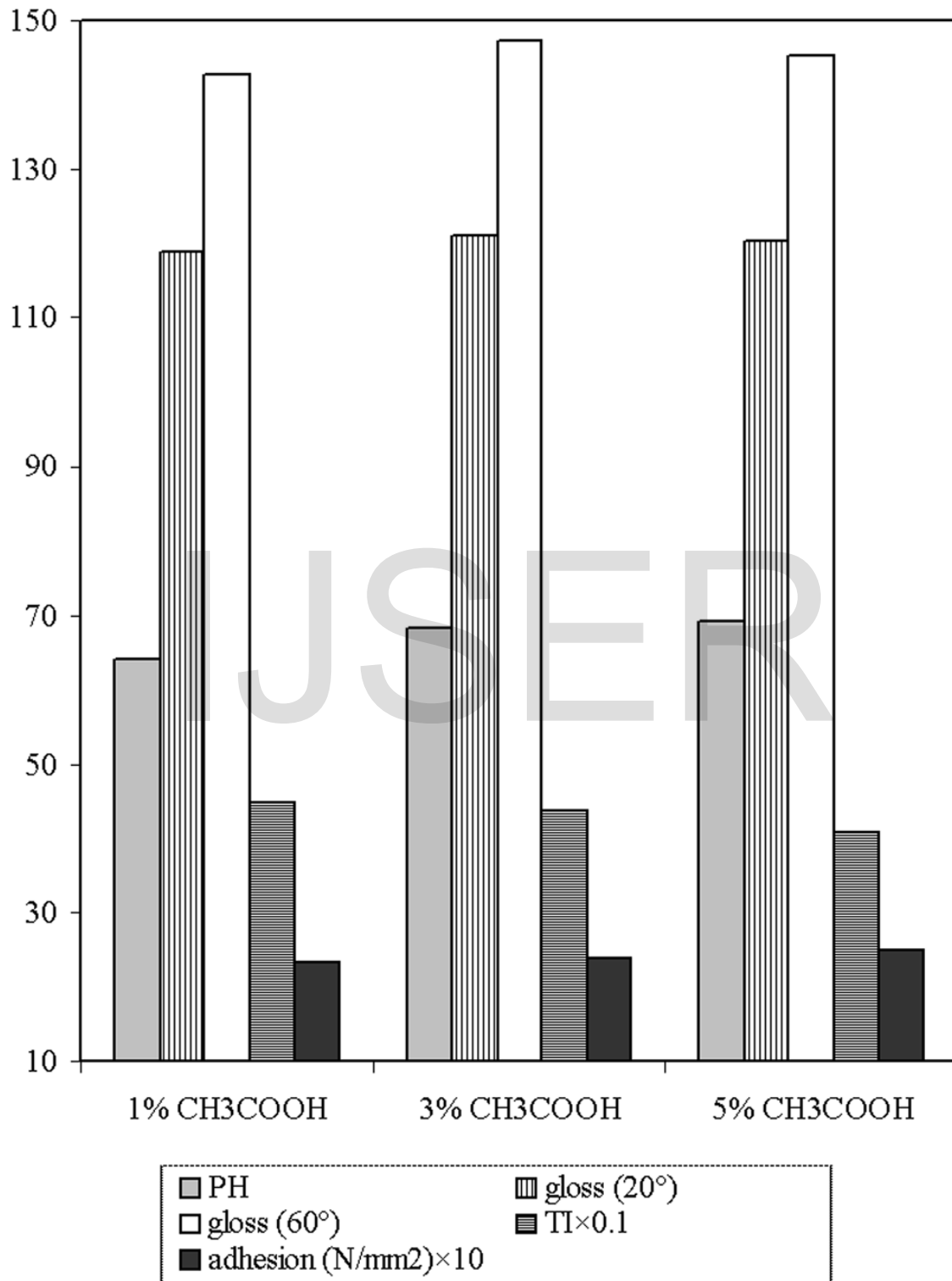


Fig. 11

