Sample preparation effect on measured rheological properties of bitumen using Dynamic Shear Rheometer

Licentiate Thesis

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ISBN 978-91-8039-601-1 (print) / 978-91-8039-600-4 (pdf)
ISSN 1653-1930
CODEN:LUTVDG/(TVTT-3252) 1-112/2023
Printed by V-husets tryckeri LTH, Lund, Sweden
February 2023

Supported by:

LUND UNIVERSITY

SBUF
ID:13847

PEAB

Abstract

Through improved bitumen characterization, which will increase the road's durability, stability, and performance, pavement engineering can apply the sustainability notion that should be followed in all disciplines nowadays. The new techniques distinguish between the rheological properties of bituminous binders as a function of loading time and temperature to depict real-world behavior rather than only considering empirical properties. The complex shear modulus of bitumen, that can be measured using a dynamic shear rheometer (DSR), describes the stiffness and degree of viscous and elastic behavior at a given frequency and temperature.

Even though EN 14770:2012 primarily describes the procedure of specimen preparation and conditioning, various factors related to DSR testing setup may result in nonconformity. Therefore, an investigation was conducted to review several areas of test method practices and the underlying impact on results based on data from round-robin tests on bitumen types 50/70 in 2017 and 20/30 in 2018, as well as polymer-modified bitumen 45/80-55 in 2019 and 2020. The variability in testing conditions and how the testing conditions may affect the measures, as well as the precision of the test technique, are statistically examined. After that, a two-level, three-factor experimental design was used to investigate the effects of the oven setting temperature, bonding temperature to the rheometer, and trimming on $|G^*|$ and δ. This involved estimating the main and interaction effects of the factors from replicated runs as well as calculating the standard errors of the effects.

The results based on RR-test data demonstrate that ignoring the extreme values due to the use of inappropriate plate geometry or the small number of laboratories involved improves the accuracy range of phase angle to a repeatability limit of $r = 1\text{-}2°$ and reproducibility limit of $R = 3\text{-}6°$ for tested bitumen. While for $|G^*|$, the coefficient of variation under repeatability improves to a range of 2-8%, and the coefficient of variation under reproducibility varies between 7 and 18%. None of the studied bitumen were significantly affected by waiting times of longer than 72 h. Furthermore, no significant variations were found between the most often used brands of equipment and sample manufacturing procedures. With results usually falling between the upper and lower reported values, 15 minutes appears to be a suitable equilibrium duration. According to the correlation test based on RR- data, the bonding temperature, and sample production temperature exhibited a significant link in more test combinations than in other sample preparation phases. Finally, in the second part of the study, the investigation of the effect of these factors together with trimming on results showed a significant difference in trimmed and untrimmed samples when PP08 is applied. The bonding and oven heating temperatures take on varying degrees of significance depending on the materials and tested temperatures.

Acknowledgments

The work presented in this thesis was carried out at the Department of Technology and Society at Lund University. The author is thankful to those who have contributed with financing, research data, guidance, and moral support.

This study was financed by the Development Fund of the Swedish Construction Industry (SBUF) and co-financed by Lund University and Peab Asfalt AB. All support is gratefully acknowledged. Further, Study I relied on data from round-robin tests that were announced by Bureau de Normalisation du Pétrole.

I would like to express my appreciation to my supervisor Andras Varhelyi for his guidance and wise counsel. A special thanks to my co-supervisor Anders Gudmarsson for his specialist knowledge and expertise, and to Michael Langfjell at Peab Asfalt AB, for his indefatigable helpfulness in providing technical and practical support.

Other contributors I would like to thank are Sven Agardh at Lund Institute of Technology (LTH), Jiqing Zhu at Swedish National Road and Transport Research Institute (VTI) for their helpful conversations, and Lars Jansson at Peab Asfalt AB for his project management.

Finally, I want to thank the moral support I received from Ebbe Parhamifar and other members of the group Transport & Roads. The encouragement given to me by my family has also been invaluable.

List of Abbreviations

AASHTO	American Association of State Highway and Transportation Officials
ASTM	American Society for Testing and Materials
BT	Bonding temperature
DSR	Dynamic Shear Rheometer
EN	European Standard
HT	Heating Temperature
LVE	Linear Viscoelastic
PMB	Polymer modified bitumen
PG	Performance Grade
SAS	Strain amplitude sweep
T-f-sweep	Temperature-Frequency-Sweeps
VG	Viscosity Grade

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SAMMANFATTNING

Bakgrund

Polymermodifierade bitumen (PMB) i asfaltmassor bidrar till att förbättra viktiga beläggningsegenskaper som motstånd mot utmattningssprickor, deformationsmotstånd och reducerad temperaturkänslighet. PMB kan därför användas för att öka beläggningars livslängd eller för att reducera materialåtgången genom att möjliggöra tunnare beläggningstjocklek.

De empiriska testmetoderna penetration (EN 1426:2007) och mjukpunkt (EN 1427:2007), som används för att indikera styvhet och viskositet, kan särskilja penetrationsklassificerade omodifierade bitumen. Dessa testmetoder bidrar dock inte till en pålitlig karakterisering när temperatur och frekvensberoendet skiljer sig åt mellan olika produkter. Dessutom kan de empiriska metoderna endast appliceras så länge som tidigare kända förhållanden fortfarande gäller. Med tillåtandet av tyngre fordon i Sverige och med klimat i förändring ökar risken för att det empiriska systemet inte längre karakteriserar relevanta bitumenegenskaper. Med nya modifierade bitumen uppstår ett ökat behov av förbättrade bitumenspecifikationer som tar hänsyn till bindemedlets temperatur och frekvensberoende materialegenskaper.

Bitumen är viskoelastiskt, vilket betyder att materialegenskaperna beror på temperatur och belastningsfrekvens. Enligt SBUF raport (ID:12926), styvheten för traditionella omodifierade penetrationsklassificerade bitumen (t.ex. 50/70, 70/100 samt 160/220) uppvisar liknande temperatur och frekvensberoende. Temperatur och frekvensberoendet av PMB (t.ex. 45/80-65, 25/55-80 B och 25/55-80) varierar dock för varje unik produkt. Med hjälp av reologisk provning genom en dynamisk skjuvreometer (DSR) kan bitumens materialegenskaper bestämmas över ett intervall av temperaturer och frekvenser. Genom detta finns en potential att utveckla en bitumenklassificeringsmetod som kan urskilja relevanta materialegenskaper för effektivare val av bitumen. DSR metoder används redan idag i den amerikanska PG (Performance Grade) klassificeringen. PG systemet utvecklades mellan 1987 och 1993 i USA med syfte att ersätta bristerna i den empiriska penetrationsklassificeringen. Systemet utvecklades dock mot omodifierade bitumen och uppvisar därför brister i klassificeringen av modifierade bitumen. Detta visar sig bland annat genom beläggningar med dåliga deformationsegenskaper på svenska flygplatser där PMB kravställs enligt PG klassificeringen. På senare år har PG systemet kompletterats med provmetoder som ska kunna tydliggöra förbättrade egenskaper av PMB. Dessa nya DSR metoder är under process för att implementeras även i de europeiska specifikationerna och i exempelvis Norge finns det möjlighet

att ställa tilläggskrav på DSR provning. DSR möjliggör även en potential till provning av reologiska egenskaper av bitumen blandat med filler (asfaltbruk), vilket kan bidra till bättre proportionering av asfaltmassor. I dagens läge finns det dock ett utvecklingsbehov för att kunna standardisera metoder för DSR provning.

En viktig utmaning med DSR provning, av framför allt PMB, är att reproducerbarheten inte har visat sig vara tillräckligt noggrann. Detta beror i stor utsträckning på att nuvarande metodstandarder möjliggör ett stort utrymme för individuella tolkningar av provmetodiken och provberedningen. Då PMB är mycket känsligt för hanteringen av provet är det mycket viktigt att uppvärmning, provtagning och placering av provet i mätutrustningen utförs systematiskt och enligt bestämda rutiner.

Dagens avsaknad av ett vedertaget nationellt harmoniserat förfarande riskerar att försvåra tillämpningen av DSR provning. Eftersom DSR metoder med största sannolikhet kommer att införas i Trafikverkets kravspecifikationer i takt med det europeiska standardiseringsarbetet, så finns det ett stort behov av att utreda och utveckla metodiken för att säkerställa tillförlitliga resultat. DSR mätningar utförs idag inte rutinmässigt hos svenska entreprenörer vilket gör att det finns tydliga behov av utbildande nationella riktlinjer som bidrar till noggrann provning.

Internationella ringanalyser (Round-Robin tester) på DSR metoder utgör en grund för att identifiera potentiella metoder och utvecklingsmöjligheter. Tidigare arbeten vid Lunds Tekniska Högskola om implementering av DSR mätningar tillför ytterligare baskunskap och identifierade utvecklingsbehov. Med utgångspunkt ifrån pågående standardiseringsarbete i Europa kändes ett behov av ett sådant doktorandprojekt som kan öka vägbyggnadsbranschen förståelse för moderna mätmetoder av bitumen och därmed bidra till en samsyn och lyckad implementering.

Syfte och omfattning

Denna licentiatavhandling är en del av forskarutbildningsprojektet "Utveckling och implementering av reologisk provning av bitumen". Syftet med detta projekt är att utvärdera och vidareutveckla provmetodiker för DSR mätningar som kan förbättra utrustningens reproducerbarhet. Detta är nödvändigt för att kravställa bitumen utifrån DSR provning. Eftersom den nuvarande europeiska standarden (EN 14770:12) tillåter bred tolkning, används olika testförhållanden och provberedningsmetoder över hela landet, vilket gör det utmanande att uppnå harmoni och uppnå överensstämmelse i resultaten. Inför framtida implementering av DSR mätningar som ett krav i vägteknik i Sverige är det viktig att utveckla nuvarande metoder med avseende på framför allt provberedning och precision. Målet är att bestämma hur olika provberedningsfaser, såsom ugnsinställning, provtillverkningsmetoder, provlagringstid, provbindningstemperatur och trimningstillstånd, påverkar reologiskt resultat av DSR:en. Arbetet förväntas leda fram till rekommendationer för nationella riktlinjer som kan utgöra grund för utbildning och implementering av nya mätmetoder för bitumen i Sverige. Vidare i fortsättningen syftar projektet till att utvärdera nya prestandaparametrar för förbättrad bitumenklassificering som baseras på DSR mätningar.

Metod och Genomförande

Arbetet med att utvärdera provberedning och testutföranden har bedrivits via ett doktorandprojekt vid Lunds Tekniska Högskola i samarbete med Peab som har kompetens inom ämnesområdet och finansieras av Svenska Byggbranschens Utvecklingsfond (SBUF). Projektet inleddes med en litteraturstudie om provmetoder och viskoelastiska egenskaper av bitumen och om DSR provning med mål att få god kännedom om befintliga internationella metoder. Utifrån detta en inventering av behovet av nödvändig utrustning samt val av provberedningsmetoder och bitumentyper som skulle studeras utfördes. Därefter, en utvärdering av viktiga parametrar så som val av instrumentinställningar för olika reologiska provberedningar och dess betydelse för variation i resultat gjordes. Införskaffning av provmaterial för fortsatt arbete och beredning av provmaterial för testserier har även gjordes.

I *studie I* undersöktes resultaten av fyra interlaboratoriska round-robin (RR)-tester utförda av olika laboratorier inom Europa mellan 2017 och 2020. RR-testerna utfördes på typ 20/30 och 50/70 ren bitumen, samt polymermodifierad bitumen (45/80-55). Testerna utfördes efter kortvarig åldring i en rullande tunnfilmsugn (RTFOT) enligt EN 12607-1 (2014) 2017-2019 och på en original PMB utan åldring, d.v.s. ingen RTFOT eller PAV 2020 (45/80-55). Effekten av följande provberednings- och testförhållanden på de reologiska parametrarna som erhållits av DSR statistisk är: ugnsinställning för provtillverkning, provtillverkningsmetod, tid som förflutit mellan tillverkningen av provet och start av testning (lagringstid), temperaturer för provbindning (montering), diameter och gapstorlek på DSR-plattan.

I *studie II* utvärderades fyra olika typer av omodifierad bitumen, 50/70, 70/100 och två 160/220 av två olika ursprung, i ett laboratorieexperiment. De tre följande utvalda provberednings- och konditioneringsfaser då EN14770 används studerades för att fastställa effekterna på reologiska parametrer, skjuvmodulen (G*) och fasvinkel (δ). Ugnstemperaturinställningen, bindningstemperaturen och trimförhållandet i två utökade nivåer med en Anton Paar DSR, MCR302-modell studerats.

Resultat

Studie I: Dataanalys från internationella ringanalyser på ren och modifierad bitumen

Denna studie presenterar resultaten av fyra internationella round-robin tester utförda av olika laboratorier i Europa mellan 2017 och 2020. RR-testerna utfördes på typ 20/30 och 50/70 ren bitumen och 45/80-55 polymer- modifierad bitumen. För de olika laboratorierna varierade de fastställda diskreta testtemperaturerna från 10 till 65°C vid en frekvens på 1,59 Hz (10 rad/s).Variabilitet i provutförande och provberedning undersöktes genom att dela upp laboratorierna i två grupper (Gr. A och Gr. B) baserat på z-score och repeterbarheten av deras rapporterade resultat för dynamisk skjuvmodul (|G*|) och fasvinkel (δ) vid olika temperaturer och plattdimensioner vid användning av en dynamisk skjuvningsreometer (DSR), enligt den europeiska standarden EN14770:2012. Beredningen av bitumen för ursprungliga och korttidsåldrade tillstånd, förvaringstid och vila efter tillverkning av provkroppar, temperatur vid anläggning av provkroppar, temperaturstabiliseringstid innan data kunde samlas in vid varje ny testtemperatur (temperaturjämviktsvaraktighet), testning inom det viskoelastiska linjära området och den utrustning som användes undersöktes. Grupperingen gjordes baserat på de beräknade statistiska måtten av rapporterade resultat av deltagande laboratorier i varje round-robin-test. Den statistiska bearbetningen av data och statistiska kriterier för prestanda och z-score gjordes enligt standarder (ISO:5725 och ISO:13528). Laboratorier i Gr. A har alla de olika testkombinationsresultaten inom 1,5 standardavvikelse från det medelvärdet av RR, provutföranderepeterbarhet). Medan laboratorier i Gr. B har lägre repeterbarhet och faller utanför 1,5 SD från omgångens medelvärde för sina rapporterade resultat. För att kunna avgöra om det finns någon mätbar skillnad mellan metoderna som användes, analyserades statistisk hur provutförande och provberedning kan ha påverkat de reologiska resultaten (G* och δ) utan gruppering. Dessutom diskuteras repeterbarhets- och reproducerbarhetsanalysen av rapporterade resultat.

Resultaten visar att repeterbarhetsvärdena för G* ligger mellan 2 och 12 %, medan reproducerbarhetsvärdena varierar mellan 7 % och 20 %. Men när man eliminerar PP25-data vid låga test temperaturer på 10 till 20 grader och PP08-data vid höga temperaturer på 25 till 35 grader, förbättras precisionen avsevärt. Det oåldrade 45/80-55 (RR-2020)-resultatet har den högsta precisionen, följt av det korttidsåldrade 45/80-55 (RR-2019) vid högre testtemperaturer och 20/30 (RR-2018) vid lägre provningstemperaturer. För δ är variationskoefficienten under reproducerbarhetsförhållanden mycket bättre än kriterierna på 5 % som anges i EN14770: 2012 för alla testkombinationer, förutom vid lägre temperaturer som 10 °C och 15 °C testade med PP25. Om man ignorerar extremvärdena, uppnår oåldrad

45/80-55 högsta precision med en repeterbarhetsgräns på r =1° och reproducerbarhetsgräns på R=3°, följt av korttidsåldern 45/80-55 med en repeterbarhetsgräns på r =1° och reproducerbarhetsgräns på R=6° och 20/30 med en repeterbarhetsgräns på r =2° och reproducerbarhetsgräns på R=6°.

Laboratorier från två olika grupper valde olika metoder, vilket visar värdet av tidigare expertis med relaterat material och DSR-testning i termer av testresultat. Till exempel är det känt från EN14770:12 att ett tillverkat prov kan placeras i ett kylskåp i maximalt 30 minuter före avformningen och bindning av provmaterialet till DSR-anordningen, men endast 19%-30% av Gr. A föredrar att göra det, medan 33%-67% av Gr. B väljer det här alternativet. Detta kan bero på att olika grupper av operatörer har olika förkunskaper om materialets hårdhet. Dessutom visar resultaten att Gr. A med bättre precision för sina rapporterade resultat överensstämmer inte med den rekommenderade provlagringstiden.

Undersökning av tre kategorier av utrustningsmärke och provtillverkningsmetod visade att G* var högre när Anton Paar och en droppe av bitumen hällt på ett ark användes, medan Malvern och silikonformar gav ett högre δ värde. Men inga statistiskt signifikanta skillnader i ett vanligt testtillstånd upptäcktes mellan de två mest använda utrustningsmärkena och provtillverkningsmetoderna. Dessutom verkar δ vara mer mottagligt för tillverkningsmetoder än G*. Det verkar också som att temperaturen vid anläggning av provkroppar, har en liten men signifikant inverkan på fler testbetingelser än andra steg när man jämför variationen av erhållna reologiska parametrar. Dessutom bör uppvärmningstemperaturen som används för att tillverka prover övervägas ytterligare eftersom den har visat sig ha ett statistiskt signifikant samband med de inhämtade resultaten. Icke desto mindre krävs en distinkt experimentdesign och känslighetsanalys av resultaten för att mer exakt kunna fastställa i vilken grad varje fas av provberedningen och konditioneringen påverkar resultatet.

Studie II: Laboratorie experiment på bitumen av olika ursprung

I studie II utfördes en serie Temperatur-frekvenssvep på fyra standard penetrationsbitumen av graden 50/70, 70/100 och två160/220 från olika leverantörer för att bedöma ugnstemperaturinställningen (HT), temperaturen för placering av provet på DSR (BT) och trimning läge i två utökade nivåer med en Anton Paar DSR, MCR302-modell. För varje material två serier av provkroppar användes, en som ger en perfekt cylinder på en PP25 och PP08 vid 1,05 och 2,1mm gap respektivt en som inte behöver trimmas (exakt mängd kan räknas ut med hjälp av bitumendensitet och volymen mellan plattorna) och en som överskottar och måste trimmas bort för att få samma geometri som den andra provkroppen före provkörningen. Det skulle kunna anses att både ger samma resultat (utan signifikant skillnad) om provet i det första fallet är väll centraliserat och det andra trimmat noggrant, med syftet att belysa detta speciellt när det gäller modiferad bitumen som kan vara svårt att trimma. Enligt EN

14770:12 ska bitumen värmas upp till (85±5) °C över bitumensmjukpunkt eller till maximalt 180 °C. Två olika temperaturer användes till tillverkning av provkropparna. Den ena tillverkades vid 80°C över bitumensmjukpunkten och den andra vid 100°C över bitumensmjukpunkten. Den tredje faktorn som varierades i provutförandet var temperaturen på plattorna där bitumen anläggs på för att kunna mjuka upp och fästas inför teststart med eller utan trimning. Silikonformar från Anton Paar användes för alla provkropparna.

Temperatur-frekvenssvepkörningar mellan 0°C till 80°C delades i tre provkroppar med 10 °C intervaller för varje provkropp för att undvika termalhistorik på provkropparna. Varje temperatursteg kördes frekvenserna 0,1rad/sec upp till 10 rad/sec eller 100 rad/sec uppdelat i 10 steg med logaritmisk följd. Amplitudsvep gjordes på alla bitumenprovkroppar och temperaturintervall så att det höll sig inom linjärviskoelastiska området under Temperatur-frekvenssvep testen. Töjningar av 0,1 % respektive 0,5% valdes vid användning av PP08 och PP25. Den dynamiska skjuvmodulen (|G*|) och fasvinkeln (δ), som utgör basen för analysen, avlästes vid varje periodiskt steg i testprocessen. För att utvärdera huvud- och interaktionseffekterna av kombination av de tre variablerna, samt uppskatta standardavvikelsen på insamlade G* och δ från randomiserade testkörningar utfördes statistisk analys. Även för att kunna avgöra om det fanns någon mätbar skillnad mellan metoderna användes statistisk analys.

Resultaten visar att den komplexa modulen är mer känslig för förändringarna än fasvinkeln. Det observeras nästan samma mönster i både lägre och högre testade frekvenser. För 160/220_I Limningstemperaturen är den mest uppenbara faktorn som påverkar G*, och δ följt av ugnsvärmetemperatur som påverkar G* negativt och δ positivt. Variationen i ugnsvärmetemperatur påverkade starkt resultaten av 160/220_I i motsats till alla andra studerade material. Masterkurvan för detta bitumen avslöjade en svag överlappning vid förskjuten temperatur, vilket kan vara ett tecken på att det innehöll en annan naturlig kemisk komponent. Trimning av provet som testats vid lägre temperaturer på 0 C till 30 C för allt testat material tenderar att avsevärt öka δ och dekreterar komplexmodulen, vilket kan indikera högre känslighet hos mindre parallellplåtsdiameter (PP08) för trimning. BT hade en negativ inverkan på G* vid nästan alla temperaturer som utvärderades för 50/70, 70/100 och 160/220 I, vilket överensstämmer med tidigare studier på två typer av korttidsåldrat bitumen, 20/30 och 45/55-80. Emellertid uppvisar icke åldrade 45/55-80 i den tidigare studien och 160/220 II ett jämförbart positivt samband med G*. I två nivå samspel, i fallet med G*, har Trim:BT den starkaste effekten på alla material och temperaturer förutom 160/220 vid lägre temperaturer där Trim-HT påverkar mer. För δ var den mest effektiva tvåvägsinteraktionen Trim-BT för 70/100 och 160/220_II. Men för 160/220_I är Trim:HT och för 50/70 BT:HT den minst viktiga faktorn för δ. Totalt sett var HT enbart en obetydlig effekt på resultaten för alla material utom för 160220_I, men det blev viktigt i samverkan med andra faktorer. The δ kan tillskrivas det faktum att intervallet för ugnsvärmetemperatur (SP+80 och SP+100°C) i denna studie är mycket litet. Studien visade att G* och δ har påverkats av studerade faktorer, minst för 70/100, vilket följs av 50/70 och 160/220.

Slutsatser och rekommendation

Fynden visade att de undersökta parametrarna, beroende på testinställningarna, hade olika effekter på G* och δ. På grund av risken för stor påverkan på resultatet bör limningstemperaturen inte ställas in slarvigt. Med undantag för ett prov som betedde sig mer som modifierat bitumen och var känsligare för uppvärmningstemperaturen för provtillverkning, hade de övre och nedre inställda gränserna för ugnens temperatur i studie II ingen märkbar inverkan på ren bitumen. Det måste dock undersökas om den valda övre gränsen kan användas för modifierad bitumen också utan att ha en signifikant effekt, vilket indirekt kunde observeras från studie I. Slutligen avslöjade trimning av proverna som undersöktes i studie II nästan inga signifikanta förändringar när PP25 användes jämfört med när PP08 användes.

För fortsatt forskning rekommenderas att bedöma effekterna av olika faktorer på modifierat bitumen med ett bredare intervall på både höga och låga nivåer av de faktorer som används i den presenterade studien för att fånga gränsen som ledde till signifikanta skillnader.

1 Introduction

The road's performance, stability, and longevity are ultimately determined by the qualities of the binder, which represent considerable value in the form of limited resources. The Strategic Highway Research Program (SHRP) that was carried out in the U.S.A. between 1988 and 1993 (Anderson et al., 1994; Petersen et al., 1994) led to increasing interest in performance-based methods for characterizing bituminous binders and numerous new purchase specifications and standards. The new methods distinguish the rheological properties of bituminous binders as a function of loading time and temperature to simulate actual behaviour in practice rather than empirical properties such as softening point temperature and penetration value, among others. The complex shear modulus (G*) is one of the most used rheological properties for performing bitumen analysis and modelling, which can be defined as $G^* = |G^*|\, e^{i\delta}$. The dynamic shear modulus |G*| describes the material's stiffness, and phase angle δ, describes the extent of viscous and elastic behavior of the material at a given frequency and temperature, and i is the imaginary unit ($i^2 = -1$). In accordance with numerous standards, a dynamic shear rheometer (DSR) can be used to estimate the complex shear modulus (AASHTO-T312, 2010; ASTM-D7175, 2015; EN14770, 2012).

However, there is concern about ensuring conformity between laboratories on sample preparation and or testing methods which may lead to discrepancies in results. The procedure for preparing and conditioning the specimens is primarily specified in the method EN 14770:12, which is utilized in Sweden and other European countries. It appears there is so much leeway in the standard that operators have too much latitude to influence the test results when running the tests, which is implying the need to clarify factors related to the set-up of the laboratory testing that could possibly affect the final outcomes.

Blending pure bitumen with additives which is essential to achieve desired bitumen qualities and improve performance in a variety of traffic and temperature conditions makes the DSR testing of modified bitumen more sensitive to variations in the test set-up than the testing of neat bitumen (Airey, 2003; Soenen et al., 2008; Zhu et al., 2017). Various studies have shown how variables such as the limit of linear viscoelasticity (Airey et al., 2002) , the equipment sensitivity to measure torque (Divya & Krishnan, 2019), and the sample manufacturing method (Airey et al., 2017) affect the consistency of results. The dependence of bitumen rheological properties on the impact of various plate geometries and gap sizes were also investigated in a different study with inconsistent result due to variation of susceptibility to changes for different studied material (Carswell et al., 1998; Liu et al., 2020; Singh et al., 2016) . While other research has emphasized the importance of applying the same thermal treatment to achieve acceptable precision on the complex modulus, phase angle, and SHRP parameter (G*/sin δ) (Eckmann et al., 2012; Mouillet et al., 2004; Soenen et al., 2005). Numerous investigations emphasized the overfilling or underfilling of the gap as well as concerns with reproducibility since the sample diameter is not accurately captured by the DSR in the low-temperature test (Alisov, 2017; Laukkanen, 2017).

Since the present European Standard (EN 14770:12) allows for broad interpretation, various test circumstances and sample preparation methods are used by different laboratories, making it challenging to achieve harmony and obtain conformity in results. This is also of interest to the company PEAB, who noticed the discrepancy in results during smaller scale interlaboratory testing in which they participated. This led to a collaboration for a doctoral project between PEAB which has expertise in the subject, and the Faculty of Technology (LTH) at Lund University of which has been active with rheological studies on bitumen and its additives since 2011 in form of master thesis projects.

This licentiate work was carried out as part of an ongoing doctoral project with the subject, "Development and implementation of rheological testing of bitumen". The aim of the Ph.D. project is to evaluate and further develop methods and techniques for the rheological testing of bitumen.

2 Aim and Scope

This licentiate thesis aims to assess current methods for testing bitumen with a DSR and to further develop a practicable approach that can provide DSR testing with an acceptable degree of precision. This project will assist in achieving SBUF's aims and guidelines by creating better conditions for entrepreneurs and the industry to assure sample preparation conformance rather than going their own way and interpreting the EN standard differently. The findings will, ideally, be used in course activities in schools and industry to facilitate communication amongst parties involved.

The first goal of this work is to review the existing sample preparation and conditioning methods for testing the rheological properties of bitumen. The precision of the DSR testing will also be evaluated. The second goal is to determine how various conditions and sample preparation phases, such as oven setting, sample manufacturing methods, sample storage duration, specimen bonding (installation) temperature, and trimming state, affect complex modulus and phase angle, as measured by a DSR.

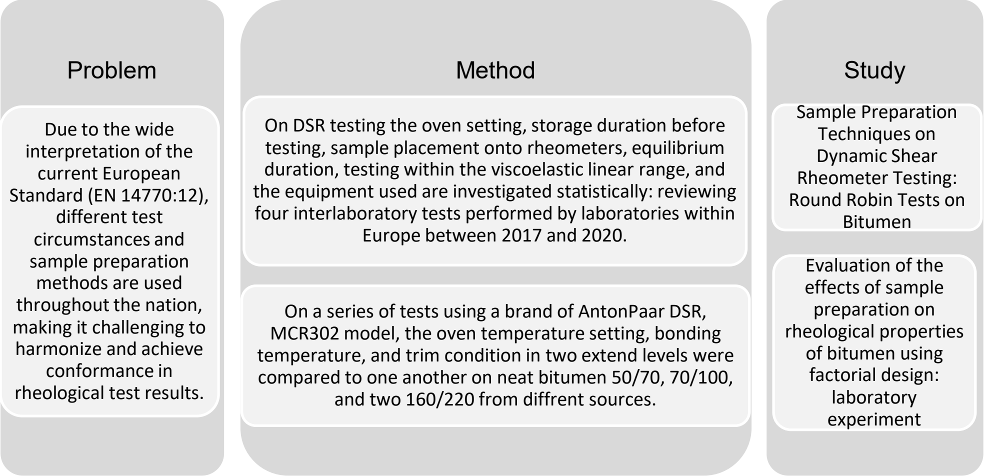
In this study, data from four interlaboratory round-robin (RR) tests performed by laboratories within Europe between 2017 and 2020 were investigated, which were sponsored by Eurobitumen Franc, Routes de France and Cerema Ouest. The RR tests were carried out on type 20/30 and 50/70 neat bitumen, as well as 45/80-55 (polymer-modified bitumen). Additionally, four different types of unmodified bitumen, 50/70, 70/100, and 160/220, were evaluated in a laboratory experiment at Lund university to determine the impacts of three selected sample preparation and conditioning phases when EN14770 is utilized.

3 Method

Figure 3.1 illustrates a schematic work frame description in this thesis. The project began with a review of the literature on bitumen rheological testing methods with a DSR to highlight the importance of following the same sample preparation and conditioning procedures and illustrates their potential effect on the results. Database searches were undertaken to gain a thorough perspective of sample preparation for the DSR testing. To find relevant literature, Web of Science, and Scopus were used. In order to accommodate more contemporary references, the database search procedure was updated in November 2022. Since some broad terms, such as "dynamic shear rheometer," might be relevant to numerous fields, not just bitumen, the first search result numbers were more than 2000 papers. The following terms—bitumen/asphalt, complex modulus/complex shear modulus, sample preparation/specimen preparation, pouring temperature, gap size, and bonding temperature—were combined and added to reduce the number of hits. Later, the screening method was used to further filter the results. Based on this technique and the elimination of duplicate records, the number of relevant papers was decreased and made more pertinent; a substantial proportion of studies that did not explore the impact of sample preparation procedures were excluded. Additionally, citation searching was applied to the retrieved papers to identify further studies from Google Scholar.

The objectives were reached by first studying the data from the different steps of sample preparation methods used on four interlaboratory round-robin tests performed by laboratories within Europe. The tests were carried out on the penetration-graded bitumen 50/70 in 2017 and 20/30 in 2018, as per EN 12591(2009), and the polymer-modified bitumen 45/80-55 in 2019 and 2020, as per EN 14023 (2010). The tests were conducted after short-term ageing in a rolling thin film oven (RTFOT) according to EN 12607-1 (2014) in 2017-2019 and on an original PMB without ageing i.e., no RTFOT nor PAV in 2020 (45/80-55). The precision analysis is expressed using the repeatability standard deviation (Sr), reproducibility standard deviation (SR), repeatability coefficient of variation (CV-r), and reproducibility coefficient of variation (CV-R). The R program was used to compare the means and variances of the results for all test combinations and materials, except for bitumen 50/70 due to the small number of participants, to explore the underlying impacts on G* and δ caused by variations in participant practices.

In study II, a series of Frequency-Temperature sweep tests were performed on unmodified bitumen 50/70, 70/100, and 160/220 from two different suppliers to assess the oven temperature setting, bonding temperature, and trim condition in two extend levels using an Anton Paar DSR, MCR302 model. The DSR testing technique is then detailed, as well as the selection of instrument parameters for various rheological sample preparations applying RheoCompass software.



Problem

Due to the wide interpretation of the current European Standard (EN 14770:12), different test circumstances and sample preparation methods are used throughout the nation, making it challenging to harmonize and achieve conformance in rheological test results.

Method

On DSR testing the oven setting, storage duration before testing, sample placement onto rheometers, equilibrium duration, testing within the viscoelastic linear range, and the equipment used are investigated statistically: reviewing four interlaboratory tests performed by laboratories within Europe between 2017 and 2020.

On a series of tests using a brand of AntonPaar DSR, MCR302 model, the oven temperature setting, bonding temperature, and trim condition in two extend levels were compared to one another on neat bitumen 50/70, 70/100, and two 160/220 from diffrent sources.

Study

Sample Preparation Techniques on Dynamic Shear Rheometer Testing: Round Robin Tests on Bitumen

Evaluation of the effects of sample preparation on rheological properties of bitumen using factorial design: laboratory experiment

Figure 3. 1. Schematic structure of this thesis project.

4 Literature Review

This literature review primarily focuses on the influence of various stages of sample conditioning and preparation on the rheological properties of bitumen using a Dynamic Shear Rheometer with parallel plates. EN 14770:2012 and ASTM D7175:15 are standards from European and American institutes, respectively, that are related to measurements with DSR.

The first three sections are focused on bitumen in terms of definition and measurement methods. Furthermore, the most often used traditional equipment for measuring bitumen rheology, the dynamic shear rheometer (DSR) with parallel plate, is briefly discussed. An overview of linear viscoelastic concept is also provided to help understand the viscoelastic behaviour of bitumen.

The last five sections of the literature review before the summary discuss the effect of the following sample preparation and test circumstances phases on the rheological parameters obtained by DSR:

- manufacturing of sample,
- oven setting for sample manufacturing,
- time elapsed between the manufacture of the sample and the start of testing (storage time),
- temperature for sample bonding (mounting),
- diameter and gap size of the DSR plate.

4.1 Bitumen

Bitumen (Asphalt) is one of the oldest building and road engineering materials. It was widely used in all early civilizations, including Babylonia, Mesopotamia, Sumeria, Chaldea, Mohenjo-daro and Harappa, Phoenicia, China, and later, Greece and Rome (Abraham, 1918; Boulangé et al., 2013; Murali Krishnan and & Rajagopal, 2003).

Unlike other building materials, its use has increased steadily. About 100 Mt/year of the bitumen produced globally is used by the pavement industry to create asphalt mixes. According to the European specification EN12597 (2014), bitumen is simply defined as a “virtually not volatile, adhesive and waterproofing material derived from crude petroleum, or present in natural asphalt, which is completely or nearly completely soluble in toluene, and very viscous or nearly solid at ambient temperatures”.

Bitumen is mostly constituted of hydrocarbon molecules with small amounts of heteroatoms, which can significantly affect bitumen characteristics. Heavy metals, in addition to heteroatoms, may exist in very low contents. Bitumen is a complicated material whose properties change depending on the crude oil source, production method, additives, and other chemical-physical treatments (Halstead, 1984; Zakar, 1971). The complicated chemical composition of bitumen makes precise identification and classification of all these forms difficult. The separation of bitumen elements based on polarity and solubility into a saturated, aromatic, resin, and asphaltene fraction (SARA) is a widely established analytical method (ASTM-D4124, 2018; Corbett, 1969; Lesueur, 2009). The ratio of resins to asphaltenes determines whether the bitumen is soluble (SOL) or gelatinous (GEL) (Figure 4.1) (Whiteoak et al., 1990). Aromatics, which comprise of the bitumen's lowest molecular weight naphthenic aromatic compounds, represent the majority of the dispersion medium for the bitumen's body asphaltenes and impact its adhesion and ductile qualities, while saturates and resins influence its viscosity and flow (Halstead, 1984).

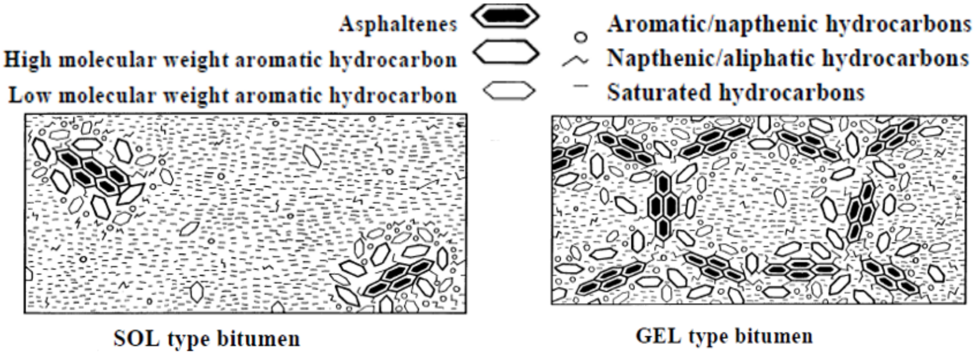


Figure 4. 1. Schematic representation of SOL and GEL type bitumen (Whiteoak et al., 1990).

4.2 DSR

For the quality assessment of manufactured bitumen, various standard tests have been devised. Individual countries, such as the United Kingdom, the United States, and Germany, have standardized these test techniques. However, bitumen's viscoelastic behavior is too complex to be represented by only traditional consistency metrics like penetration and softening point. The necessity for complete bitumen characterization has long been recognized. When compared to other viscoelastic materials used in construction, bitumen is a complicated material, and understanding its rheological properties is a challenging process. Rheology is the study of material flow and deformation characteristics. In other words, bitumen rheology is the study of the relationship between stress and strain in bitumen. When a viscoelastic substance, such as bitumen, is stressed, some of the distortion is recovered, but some remains. These new test systems produce new parameters, such as complex shear modulus (G*) and phase angle (δ), that better characterize the rheological behavior of bitumen. The phase angle is the angle at which the harmonic oscillating strain response lags behind the sinusoidal stress. The phase angle value is dictated by the material's elastic and viscous response. It is 0° for elastic materials and 90° for viscous materials. As a result, the phase angle of bitumen as a viscoelastic material ranges from 0 to 90°.

Dynamic shear rheometers are used to study the dynamic characterization of the rheological behaviour of bitumen. In this test, bitumen is sandwiched between two parallel plates, which can be rotated at different frequencies (Goodrich, 1991). During DSR testing, the responding strain/stress is measured by applying a torque to a disc-shaped bituminous sample in response to the applied stress/strain. In the strain-controlled mode, a sinusoidal strain with constant amplitude is applied to the sample, whereas in the stress-controlled mode, a sinusoidal stress with constant amplitude is delivered to the sample. Figure 4.2 depicts oscillatory shear readings using a DSR test. The collected data at various frequencies and temperatures, which may contain one or more parameters can be illustrate by different plots, such as stiffness modulus or phase angle versus temperature or frequency.

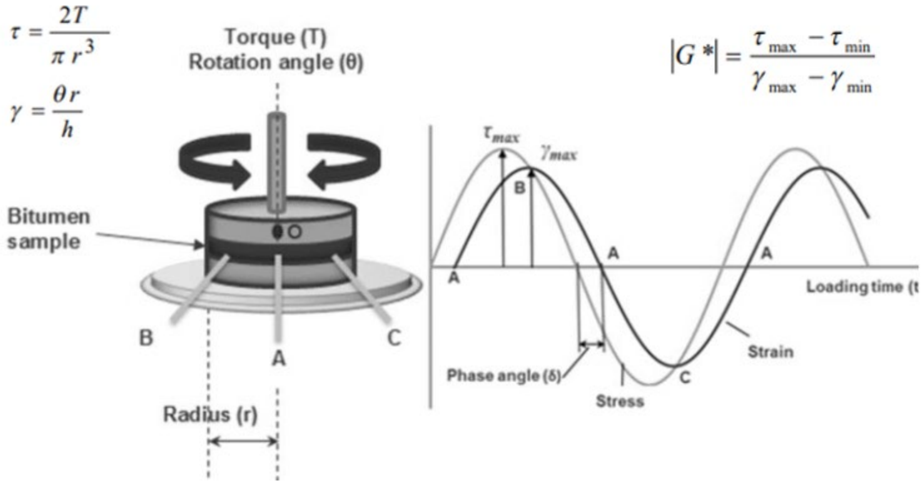


Figure 4. 2. Dynamic shear measurements using parallel plat geometry (Mukandila et al., 2015).

Since, the DSR only takes two measurements, namely torque and angular rotation, all the results (e.g., strain, stress, and G*) are calculated from these measurements. The following equations are used for calculating strain and stress in this instrument (Airey & Brown, 1998; Stroup-Gardiner & Newcomb, 1995): where h is the gap between plates (mm), r is the plate radius (mm), T is the applied torque (N-m), and θ is the rotation angle (radians). The absolute complex modulus (G*) was measured based on shear strain (γ) and shear stress (τ).

$\gamma = \theta r / h$

$\tau = 2T / \pi r^3$

$| G^*(\omega) | = \tau \max / \gamma \max$

4.3 Testing within the viscoelastic linear range

Linear elasticity is the fundamental basic material model, where stress is proportional to strain (Hooke's law). However, when bitumen is dynamically tested as a viscoelastic material, two behaviour domains emerge: the linear domain and the non-linear domain (Boussad et al., 1996). Non-linearity responses of bitumen generally become minimal at strain amplitudes less than a particular limit (LVE limit), and the material behaviour can then be effectively represented by a linear viscoelastic approximation. Most bitumen behaviour is non-linear under most conditions, and determining this linear limit is an important element of the rheological research of these materials.

Rheological properties can be obtained from the conventional frequency sweep at different temperatures according to EN 14770 or as suggested in several studies, with temperature ramping at a fixed frequency, which takes less time (Porot et al., 2020). Since time-temperature dependency is only valid in the linear region of behaviour, determining the linear limit is required in order to use the time-temperature superposition concept to interpret the results. The linear region can be found from a strain amplitude sweep test, in which the applied strain is increased during the test and then the linear region can be found by plotting complex modulus versus shear strain.

According to SHRP, the linear region can be defined as the point beyond which the measured value of complex modulus decreases to 95% of its maximum value. While according To EN 14770 to remain in the linear range, the value of G' and G'' must not differ by more than 5 % of the initial value over the stress or strain range chosen. The initial value can be taken as the intercept of a regression line fitted to the first measured values. It has been observed that testing within the strain range of 0.5% and 10% lies within the linear range for most of the binders, while for PMBs the range is much less (EN14770, 2012). Nevertheless, according to Rahimzadeh (2002), The LVE stress and strain limits were higher than the SHRP recommended stress and strain levels. There was no significant reduction of the linearity range for the modified bitumen, while the strain and stress LVE limits for the studied multigrade 35/50 bitumen were somewhat lower than those discovered for the other four studied bitumen (50 pen unmodified, EVA modified, and two SBS modified). Additionally observed, all the binders demonstrated dominant elastic behaviour in the first region, high frequency (low temperatures), with slight variances in the viscoelastic characteristics of the bitumen. At intermediate frequencies (temperature), the SBS PMBs began to exhibit viscoelastic variations from the other binders. This was especially apparent in the phase angle master curves, where the SBS polymers began to dramatically increase the elasticity of the modified binders. Except for the SBS PMBs, all bitumen began to lose elasticity and approach a viscous state at very low frequencies (high temperatures).

4.4 Sample Manufacturing

According to EN 14770 (2012), sample can be manufactured in 3 different ways, as follows: Pour into moulds or sheets, directly onto plates, and apply vials. Using a vial is not recommended for polymer modified binders. However, a previous study shows that repeatability of the hot pour onto plate method is slightly higher than the repeatability of using a mould (silicon), and weighing methods, which involves pouring a pre- weighted amount of hot bitumen directly onto a plate without trimming before testing. The G* obtained by silicon mould is slightly higher, which follows with direct hot pour method and weighing method (Airey et al., 2017) .

4.5 Pre-heating time and temperature for manufacturing sample

Previous studies, shows that for the SBS modified binder, the pouring or homogenisation temperature of the binder affects rheological tests more than storage time. The result of a test conducted at temperatures between 120°C and 200°C on two elastomer modified samples (50/70 + 3.5% SBS and 70/100 + 5% SBS), shows that the G* increases by pouring temperature, and for lower pouring temperatures viscosity η^* stabilises as frequency decreases from 10Hz toward 0.001Hz when tested at 50°C (Soenen et al., 2005) . Also, another study shows the influence of pouring temperature and oven setting duration for sample manufacturing prior to testing on rheological properties of bitumen. It investigates a pure performance graded bitumen PG 64-22 and two PMBs PG 70-22 and PG 76-22 in original, and after short- and long-time ageing processes at temperatures of 143 °C and 185 °C for a duration of 1/2, 2 and 4 h. result indicate an increase in the G* by increase in pouring temperature for all ageing condition for almost all the tested material. Phase angle showed more dependency on heating duration than temperature compared to the G*. The study recommends 2 hours of oven heating at 143 °C for sample manufacturing, taking into account the sample's short-term ageing index and absolute decline in complex viscosity (Dessouky et al., 2011). Furthermore, Büchner et al. (2020) reveals that most of the 20 European road asphalt laboratories that took part in an interlaboratory study heated pure bitumen, 50/70 (penetration at 25°C: 64 1/10 mm; ring and ball softening point: 51,3°C), and polymer-modified bitumen 25/55-55 (penetration at 25°C: 46 1/10 mm; ring and ball softening point: 58,0°C) between 90°C and 180°C for the manufacturing of samples for DSR testing. A heating temperature of about 150°C was used by the majority of participants. However, only a small number of participants chose to vary heating temperature based on the type of binder, choosing a slightly lower temperature for the neat bitumen than polymer modified. In a different study, an oven heating temperature of approximately the softening point temperature plus 100°C is applied to all of the distinct types of studied bitumen (unmodified, elastomer, and plastomer modified)(Airey et al., 2017).

Current bitumen sample preparation in EN12594 (2014) states that heating unaged bitumen to 100 °C above softening point temperatures should be avoided; this is declared as a temperature of (85±5) °C above softening point but not higher than 180 °C in EN14770 (2012). However, for modified bitumen obtained under an ageing test, heating within 180-200 °C may be needed, but it should not be heated above 200 °C (EN14023, 2010). According to EN 14770, the duration of melting, homogenising, and moulding should not exceed 135 min, but reheating time differs between a maximum of 30, 60, or 120 min depend on the mass of sub-samples, for instance, in case there is RTFO prior to manufacturing the sample.

4.6 Waiting period from manufacturing sample to test start (storage time)

Previous studies reveal that the waiting time at room temperature between the bitumen sample casting and the start of the test procedure does not have a significant impact on the test result for pure bitumen and styrene–butadiene-styrene (SBS) modified binder (Soenen et al., 2006). However, for waxy bitumen and special polymer modified binder different interpretations can be represented, which demonstrates the importance of reaching bitumen's full equilibrium when considering slow isothermal crystallisation during storage for polymer modified binders. For instance, for the plastomer modified bitumen, ethylene-vinyl acetate (EVA), about 6° increase is observed for the phase angle (δ) as frequency decreases from 10Hz toward 0.1Hz over a maximum of 24 h time delay. However, no effect was observed for modulus (Eckmann et al., 2012) . Another study shows that the complex viscosity ($\eta^*= G^* (\omega)/ \omega$) changes more than a decade at 0.001 Hz for the EVA modified binder (B70/100+5 % EVA). This influence is also observed in the phase angle, which decreases at 0.001 Hz over the waiting time drastically (Soenen et al., 2006).

4.7 Sample bonding (mounting) temperature

When placing a test sample onto the DSR a high temperature of the parallel plates is, to some extent, essential for make secure bonding between binder and plate, especially true for viscous binders like polymer modified binders. However, an upper limit needs to be chosen carefully to avoid ageing the sample before testing. A study shows that, for the tested plastomer modified binder, the bonding at either high (80 °C) or low (30 °C) temperatures does not have a significant impact on the measured stiffness modulus |G*|. However, lower phase angles have been observed for the sample that was bonded at a higher temperature. It was believed that there may have been other factors at play in addition to the higher bonding temperature, such as the initial reheating and the extended interval between bonding and testing for the specimen mounted at 80 ° C. (Eckmann et al., 2012). Airey et al. (2017) theorizes that the slightly higher G* for the moulded sample method compared to the hot pour method may be brought on by differing temperature applied when samples mounted in the DSR. Bitumen is viscous and above its softening point in the hot pure method, but elastic and at room temperature in the silicone mould method, which can expose the sample to internal stress when it is pressed between the plates before testing. However, it appeared that the tested neat bitumen with a lower softening point compared to the SBS modified bitumen had a smaller effect which can be due to its more viscous behaviour at room temperature when placed in the DSR. Specimen mounting temperatures applied by participants of an interlaboratory test for asphalt binders 50/70 and 25/55–55 ranged from 10°C to 90°C with a slightly higher temperature for the PMB when T-f-sweep test were performed at temperature range between 0 to -30 °C (Büchner et al., 2020). The softening point of neat and modified bitumen for tested material were 58.0°C and 51.3°C respectively, according to the ring and ball method.

According to the standard EN 14770:12, the temperature of both the upper and lower plates of the rheometer need to be set to a maximum of the binder's softening point, namely plus (20 ± 5) °C, or at (90 ± 5) °C, whichever is the lower, for at least 30 min to facilitate acceptable bonding of the test sample to the plates. Also, it is understood that a manufactured sample can be placed in a refrigerator for a maximum of 30 min at about 5°C prior to the de-moulding and bonding of the sample material to the DSR device.

4.8 Plate diameter and gap size

According to a study finding (Scholz & Brown, 1996), using the DSR is a useful way to learn more about how bitumen behaves in thin films that are subjected to shear stresses. However, the bitumen film thickness coating aggregates in actual asphalt mixtures can range from a few microns to a few millimetres (Mack, 1957; Wu et al., 2021; Wu et al., 2020).

According to EN 14770:12, the operational ranges are as follows: at low temperature, the 8 mm plate geometry and 2 mm rheometer gap size are preferred (suitable for stiffnesses |G*| in the range 100 kPa to 10 MPa), while at intermediate to high temperature, the 25 mm plate geometry and 1 mm rheometer gap size are preferred (suitable for stiffnesses in the range 1 kPa to 100 kPa). According to Airey et al. (2017) utilizing a 25 mm plate for complex modulus larger than 100 kPa overestimates elastic response and underestimates bitumen stiffness.

In a different study, the effects of plate geometry and gap size were explored in relation to two viscosity graded neat bitumen, VG10 and VG30, as well as two modified bitumen (VG10 + SBS and VG10 + EVA). Gap sizes of 1, 2, and 3 mm for plates with a 25 mm diameter and 1, 2 mm for plates with an 8 mm diameter were investigated. At the higher test temperatures (40, 60, and 70 °C), a higher gap resulted in a lower complex modulus and phase angle, but the opposite was true at the lower test temperatures (10, 20, and 30 °C). According to the study, 8mm diameter plates generate higher values for phase angles at higher temperatures (Singh et al., 2016).

Liu et al. (2020) have also investigated how the size of the gap between the plates affects the results. The outcome shows that for the tested neat bitumen (penetration at 25°C: 70.6 1/10 mm; ring and ball softening point: 47.6°C), the complex modulus tends to decrease when the gap size reduces from 1mm to 10 µm. The modified bitumen (PG-76 + 4% SBS) showed the same trend, but with a different sensitivity. It exhibits more viscous behaviour when the gap size is reduced from 250 to 50 m, in contrast to neat bitumen, which exhibits more elastic behaviour.

Effect of different gap size on two neat performance graded bitumen (PG58-28) from different sources, two elastomers modified (SBS), four plastomer modified bitumen, and two oxidized bitumen were all evaluated by (Zhai et al., 2000). It was discovered that the G* falls constantly when the gap size decreases from 1mm to 10 µm. The bitumen modified with elastomers was the most susceptible to changes in gap size, whilst the oxidized bitumen showed the least sensitivity to gap changes. However, for the majority of bitumen, the phase angle remains constant as the gap size varies.

Another study shows that, based on the fitted master curves of the G* and δ at a reference temperature using the 2S2P1D (two springs, two parabolic elements, and one dashpot) model, similar rheological properties can be obtained with both the 1.75 mm and 3 mm gap size with 4 mm geometry for five distinct types of bitumen (50/70, 70/100, SBS modified, and short-term and long-term aged 70/100). However, the sample preparation procedure with the 3 mm gap appears to generate more consistent samples than the 1.75 mm sample preparation, leading to higher repeatability (Wang et al., 2019).

4.9. Summary of the literature review

Different phases of preparation and conditioning of sample on DSR testing were investigated in different studies. Frequency sweep tests were performed in different range of temperatures and strain levels to measure the complex modulus and phase angle to be able to describe the rheological behaviour of bitumen under different conditions.

Testing withing the LVE regime of the material is essential to model the bitumen rheological behaviour. The LVE limit for all bituminous increases with test temperature, though with varying degrees of temperature dependence. Using an inappropriate plate diameter can also over or underestimate the elastic and stiffness response of the bitumen. Thus, to prevent measuring under non-linearity conditions and over or underestimating the rheological parameters, it is required to determine the LVE range and take the complex modulus value into account when selecting plate geometry (100 kPa – 10 MPa for PP08 and 1 kPa – 100 kPa limit for PP25) for all planed test temperatures. Conclusions relating the effect of gap size on bitumen rheological properties were conflicting among studies, which could be attributed to material and test condition dependency. According to the findings of the studies, modified bitumen is more susceptible to gap change than unmodified and aged bitumen. However, the complex modulus appears to be more sensitive to gap size variation than the phase angle. Complex modules were shown to increase as gap size increased, except for the tested material at higher temperatures. Therefore, it is crucial to maintain the standard recommended gap size of 2 mm with PP08 and 1 mm with PP25 to avoid gap size dependency of outcomes. Sample manufacturing method can be generally divided in three ways. The results showed that the mould and weighing mould methods produced slightly higher and lower G* values than the hot pour method, and that the repeatability of each method varied only slightly. The effect of pouring temperature and oven setting period on sample fabrication demonstrates that higher temperatures can result in a rise in G*. However, interlaboratory research indicate that operators use a diverse range of temperatures, highlighting the need for deeper research with different types of materials in the subject. In contrast to plastomer modified binder, waiting time between bitumen sample casting and the start of the test method does not have a major impact on the test outcome for neat and SBS modified bitumen. However, viscoelasticity appears to be more susceptible to storage time than stiffness. In the case of bonding temperature, the DSR plates temperature when mounting the samples on appears to have no major impact on stiffness modulus G*, but rather on phase angle. A slightly higher G* was observed for the moulded specimen than with the hot pure method, which was mounted onto DSR at a lower temperature than the hot pure approach. A general conclusion, on the other hand, may necessitate additional testing.

5 Study I: Review of variations in test performance and its effect on results: interlaboratory round-robin (RR) tests

5.1 Aim

The practices, techniques, and interpretations utilized by several laboratories in a series of interlaboratory round-robin tests are examined in this research. The aim is to identify the sample preparation phases that are crucial to achieving consistent results and to underline the importance of employing the same sample preparation and test conditioning approaches because of the possible impact on the outcome. The goal of this study is to determine how various sample preparation factors, such as equipment brand, sample manufacturing method, oven setting, storage time of the manufactured sample, bonding temperature, and equilibrium duration, affect the outcomes. Furthermore, the precision of the EN14770:2012 method is evaluated utilizing repeatability and reproducibility assessments of reported G* and δ values.

5.2 Material and Method

In this study, the practices, techniques, and interpretations of EN14770:2012 that have been used by participants in four round-robin tests are investigated. The number of participating laboratories were 13 in 2017, 37 in 2018, and 46 in 2019 and 2020. The tests were carried out on the penetration graded bituminous binders 50/70 in 2017 and 20/30 in 2018, as per EN 12591 (2009), and the polymer modified bitumen PMB 45/80-55 in 2019 and 2020, as per EN 14023 (2010). The tests were conducted after short term ageing in a rolling thin film oven (RTFOT) according to EN 12607-1 (2014) in 2017-2019 and on an original binder without ageing i.e., no RTFOT nor PAV in 2020. Table 5.1 shows the material properties and testing plan.

Table 5. 1. G* and δ testing plan at different temperatures with corresponding parallel plate dimensions.

Material type Volume (L)	PEN (0.1mm) EN 1426	SP (°C) EN 1427	Test Temperatures (°C) and parallel plate dimensions (PPmm)
50/70 1	58	54	15, 20 and 25°C (PP08) 40, 45 and 50°C (PP25)
20/30 1 or 0.5	31	60.4	35, 30, 25 and 20°C (PP08) 20, 25, 30,35, 55, 60 and 65°C (PP25)
45/80-55 0.5 or 0.25	51	58.4	20, 15 and 10°C (PP08) 10, 15, 20, 40, 45 and 50°C (PP25)
45/80-55 0.25	49	61	20, 15 and 10°C (PP08) 40, 50 and 60°C (PP25)

The variation on the preparation of the bituminous binders for original and short-term aged condition, sample manufacture method, storage time of sample before testing, bonding temperature of sample onto rheometer, equilibrium duration on different testing conditions, testing within the viscoelastic linear range, and the brand of equipment used are investigated for two group of participants (Gr. A and Gr. B). The statistical processing of data and statistical criterion of the performance are done according to ISO standards (ISO 5725). The overall trueness of the reported results by participant labs were measured by applying statistical tests to the relative deviation from the theoretical value of all samples analysed (z-score) according to ISO 13528 (2015) by round robin organizer per year . Grouping of the laboratories is done based on the repeatability and closeness of their reported results in relation to the mean of the corresponding test. Laboratories that obtain a |Z-score| ≤ 1.5 and a within-laboratory repeatability (SD) less than or equal to the repeatability of the round (Sr) for their reported results are classified as group A (Gr. A). The remaining laboratories are named as group B (Gr. B). The repeatability standard deviation (Sr), reproducibility standard deviation (SR), repeatability coefficient of variation (CV-r), and reproducibility coefficient of variation (CV-R) are used to express the precision analysis. According to ISO 5725-6 (2001), the difference between results obtained in the same or alternate laboratory is significant, if it is greater than the repeatability limit ($r = 1.96 \times \sqrt{2}$ Sr) or the reproducibility limit ($R = 1.96 \times \sqrt{2}$ SR), respectively.

Underlying effects on G* and δ due to variation in practices used by all participant without grouping also investigated. The means and variances of G* and δ are compared using R (version 4.1.0) (R CoreTeam, 2013) to conduct each statistical test on all test combinations except for bitumen 50/70 due to limited number of participants (13). In order to determine which following statistical test, Welch's

ANOVA or Kruskal-Wallis (H-test), should be used, the assumption of normality is first verified in accordance with Zimmerman's (2011), guideline. When results indicate that specific experimental conditions have a statistically significant mean difference, or that there is a particular aspect between the group means, multiple comparisons post hoc tests, such as Pairwise tests, and Dunn's test are used to pinpoint the precise groups with the significant differences between them. For all performed statistical tests, a level of significance of 0.05 is used which indicates, a 5% risk of concluding that a difference exists when there is no actual difference. A Pearson correlation measure performed for all test conditions to quantify the direction and strength of the relationship (Correlation Coefficient: r) in case of numeric variables, which followed by a statistical significance test to determine whether the correlation between variables is significant.

5.3 Results and Discussion

Repeatability and reproducibility

The reproducibility of 10% for the complex shear modulus and 5% for the phase angle are the recommended values in EN 14770:2012 based on the Rilem TC 180-PEB work (Sybilski et al., 2004). The repeatability for G* and δ was determined to be 5% and 1% for paving grade bitumen and 8% and 2% for polymer modified bitumen from a study conducted by Eckmann et al. (2008). The coefficient of variation under repeatability conditions (COV_r) and the coefficient of variation under reproducibility conditions (COV_R) for each of the testing combinations and sample material are calculated at a frequency of 1.59 Hz (10 rad/s) (Table 5.2).

Additionally, the |G*| and δ obtained from 8- and 25-mm parallel plates at the overlap temperatures were examined. The values of |G*| and δ deviate from the mean of |G*| and δ at the overlap temperatures by less than 6 % and 0.6°, respectively, which is significantly better than the recommended values of 15% for |G*| and 3° for δ.

It is found that the repeatability values are within a range of 2 - 11.8% for |G*| and 0. 2 – 3.4% for δ. Reproducibility values vary between 7% and 19.6% for |G*|. However, when eliminating the PP25 data at low test temperatures of 10 to 20 °C and the PP08 data at high temperatures of 25 to 35 °C, the precision improves significantly. Nevertheless, the result for unaged 45/80-55 obtains the best precision, which is followed by RTFO conditioned 45/80-55 at higher testing temperatures and 20/30 at lower testing temperatures. For the phase angle, the coefficient of variation under reproducibility conditions meets the criteria given in EN14770: 2012 for all the results, except at temperatures as low as 10 °C and 15 °C for RTFO conditioned 45/80-55 and 20 °C for 20/30 in PP25 geometry. For the phase angle a reverse relationship can be noticed between the coefficient of variation and temperature, as mentioned by Büchner J. et al. (2020) for smaller parallel plate geometry. On the contrary, another interlaboratory study with a smaller number of participants using the same type of bitumen, but with different Pen and SP values, shows a wider spread of data around the mean for phase angles than the complex shear modulus (Błażejowski et al., 2016).

Table 5. 2. Reproducibility coefficient of variation (COVR) and repeatability coefficient of variation (COVr) for |G*| and δ for all combination. The bold font indicates that an 8-mm parallel plate is used.

2017	COV$_r$	COV$_R$	2018	COV$_r$	COV$_R$	2019	COV$_r$	COV$_R$	2020	COV$_r$	COV
Complex shear modulus (**PP08**/ PP25)											
			35°C	**9.0**	**19.4**	**25°C**	**5.5**	**12.8**			
25°C	**7.7**	**15.6**	**30°C**	**7.5**	**14.7**	**20°C**	**5.7**	**17.9**	**20°C**	**4.4**	**11.9**
20°C	**7.7**	**15.7**	**25°C**	**7.2**	**15.9**	**15°C**	**7.3**	**14.7**	**15°C**	**3.5**	**15.3**
15°C	**7.6**	**15.8**	**20°C**	**4.7**	**10.6**	**10°C**	**6.3**	**14.9**	**10°C**	**4.0**	**12.9**
40°C	7.2	16.2	55°C	6.9	12.9	40°C	4.7	9.5	40°C	3.7	10.9
45°C	5.4	11.4	60°C	6.4	12.3	45°C	4.5	8.8	50°C	2.1	7.5
50°C	5.7	11.4	65°C	6.7	13.2	50°C	4.5	9.4	60°C	2.0	7.0
			25°C	3.1	11.5	10°C	11.8	19.6	40°C	3.5	10.7
			30°C	4.0	9.9	15°C	4.1	13.6	50°C	3.0	8.0
			35°C	5.2	10.2	20°C	2.9	13.1	60°C	2.2	7.1
Phase angle (**PP08**/ PP25)											
			35°C	**1.2**	**2.3**	**25°C**	**0.7**	**1.3**			
15°C	**1.0**	**2.0**	**30°C**	**1.0**	**2.2**	**20°C**	**0.9**	**2.2**	**20°C**	**0.7**	**1.7**
20°C	**1.1**	**2.1**	**25°C**	**1.1**	**2.1**	**15°C**	**0.7**	**2.3**	**15°C**	**0.6**	**1.9**
25°C	**0.6**	**3.5**	**20°C**	**1.4**	**2.7**	**10°C**	**0.9**	**2.9**	**10°C**	**0.7**	**2.6**
40°C	0.3	1.8	55°C	0.8	1.7	40°C	0.5	1.5	40°C	0.3	0.5
45°C	0.2	1.3	60°C	0.9	1.7	45°C	0.3	0.9	50°C	0.2	0.4
50°C	0.2	0.5	65°C	0.9	1.8	50°C	0.4	0.7	60°C	0.2	0.6
			20°C	1.0	8.2						
			25°C	1.0	5.0	10°C	3.4	20.2	40°C	0.3	0.5
			30°C	1.2	3.1	15°C	2.1	8.7	50°C	0.2	0.4
			35°C	0.9	2.3	20°C	0.7	4.7	60°C	0.2	0.6

To prevent temperature reliance, for the δ, the repeatability limit (r) and reproducibility limit (R) related to the arithmetic mean value are expressed in absolute value as shown in Figure 5.1, which will remain constant regardless of temperature. Ignoring the extreme values due to selection of unsuitable plate geometry or low number of involved laboratories, unaged 45/80-55 achieves the highest precision with a limit of r=1° and R=3°. There is an overall improvement in precision over the years, which may be attributed to round-robin instruction experience and the removal of the influence of various individual practices on RTFOT technique performed by each participating laboratory from the most recent year, where tests were performed on unaged bitumen.

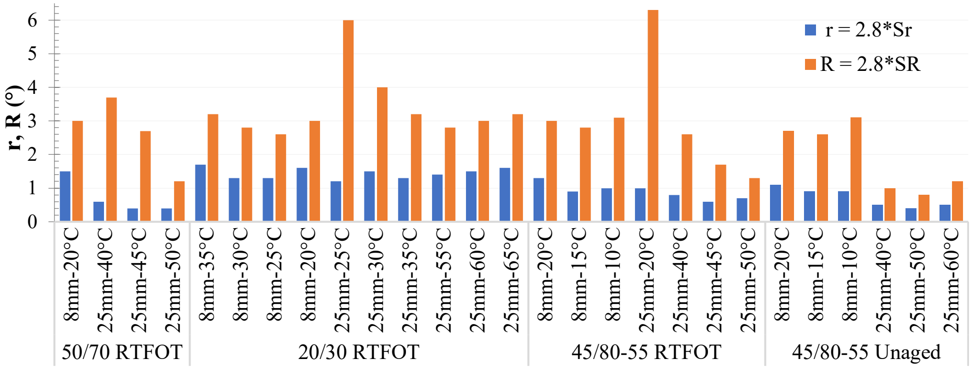
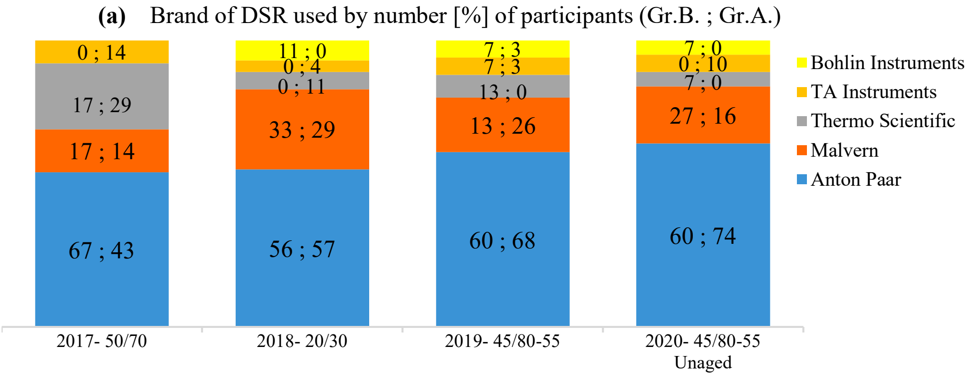


Figure 5. 1. Standard deviation under repeatability conditions (S_r) and standard deviation under reproducibility conditions (S_R) for δ.

Equipment

Figure 5.2 shows the percentage of different equipment from different manufacturers used by all the participant laboratories. Most laboratories used various models of Anton Paar manufactured rheometers (MCR 101, 102, 301, 302, 501, 502, 702, smart pave 102/301, and EC-Twist 502). Malvern Panalytical rheometers represented six different models (Kinexus DSR, DSR+, Pro, Pro+, Ultra+, and KNX). The Discovery HR 2 and AR1000 models, and the Haake Mars 2 and 3 models were produced by TA Instruments and Thermo Scientific, respectively. The least amount of used DSR devices were produced by Bohlin, representing one model (DSR2). Results indicate that laboratories with better precision (Gr. A.) used Anton Paar devices more than Gr.B. for all the round-robin tests, except in 2017.



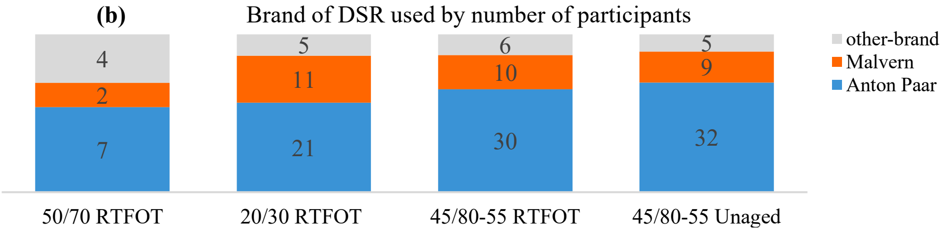


Figure 5. 2. Brand of DSR applied by laboratories.

Figure 5.3 shows the average G* and δ by equipment brand. The term 'other-brand' refers to the combination of the three least used brands (TA Instruments, Thermo Scientific, and Bohlin). A black outline denotes test conditions that were shown to have statistically significant differences. For the G*, there are statistically significant differences between Malvern brand and other-brand for the RTFOT 20/30 at test condition PP25- 55 °C (F (2) = 3.42, p-value = 0.049). In terms of δ, Malvern brand was significantly different from other-brand for the RTFOT 20/30 at PP08- 20 °C (H(2)= 9.52, p-value = 0.009), for the RTFOT 45/80-55 at PP08- 10 °C (F(2)= 5.835, p-value = 0.009), and for the Unaged 45/80-55 at PP08- 15 °C (F(2)= 13.17, p-value = 0.001), at PP08- 10 °C (F(2)= 7.95, p-value = 0.019), and at PP25- 60 °C (F(2)= 12.93, p-value = 0.002).

When comparing the Malvern and Anton Paar brands, the Malvern resulted in a higher G* when applying PP025 and lower G* with PP08. However, the differences are only significant in the two test conditions listed here: for Unaged 45/80-55 at PP08- 20 °C (H (2) = 6.20, p-value = 0.045), and at PP08- 15 °C (H (2) = 7.10, p-value = 0.029). In terms of the δ , Malvern brand resulted in a higher value than Anton Paar for all test conditions except testing at 40 °C and 50 °C with PP25 for Unaged 45/80-55. The differences, however, are not significant.

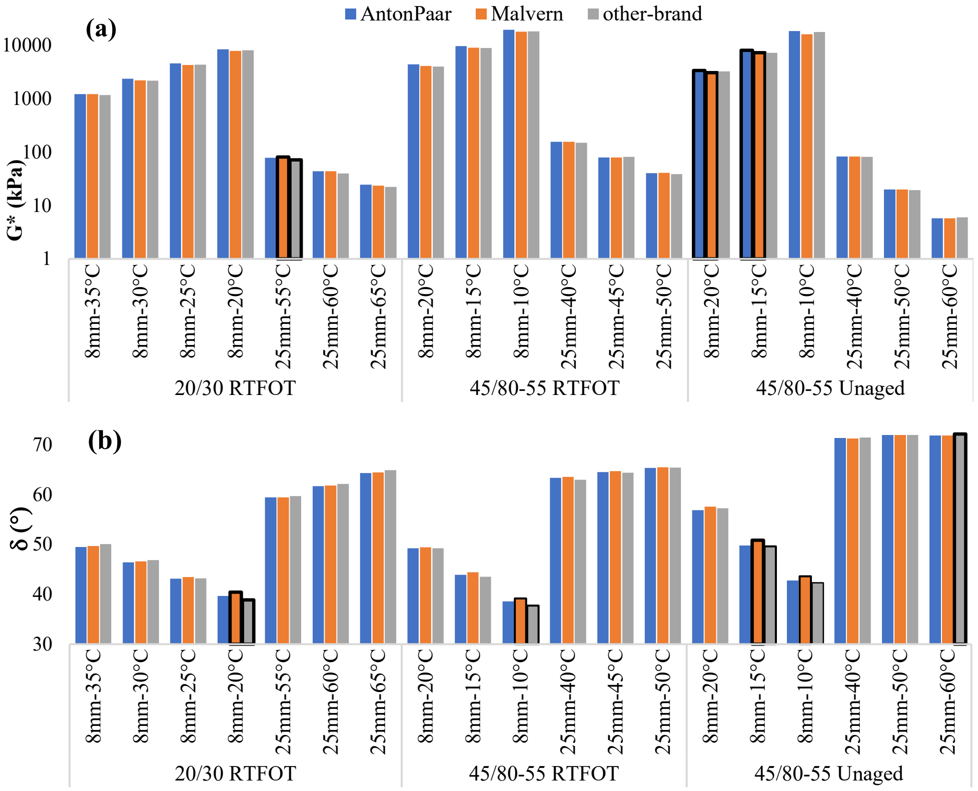
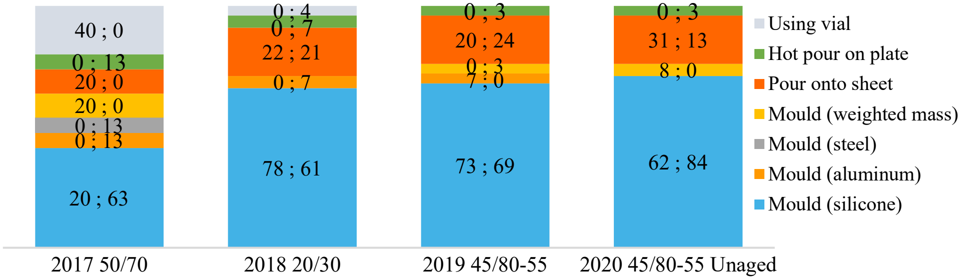


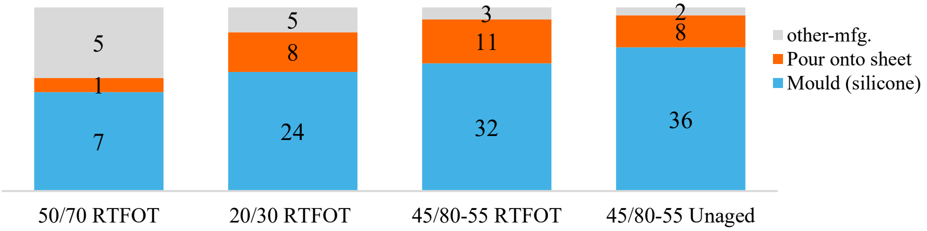
Figure 5. 3. The significant difference per brand is represented by a black outline for each test combination; (a) for the average G*of measurements, and (b) for the average δ of measurements.

Sample manufacturing

Figure 5.4 shows that the most frequently used method during the round robin test between 2017 and 2020 is the silicon mould. The trend for using the silicone mould is increasing, which can be the result of a larger amount of Anton Paar equipment among participant laboratories. Nevertheless, only a few laboratories mentioned their mould supplier. There is otherwise one laboratory in each year that used the hot pouring onto plate for the 25 mm plate dimension.



(a) Sample manufacturing method used by number [%] of participants (Gr.B. ; Gr.A.)



(b) Sample manufacturing method used by number of participants

Figure 5. 4. Different sample manufacturing methods (mould, sheet, direct onto plate, and vial) used by laboratories of Gr. B and A (%).

Statistical tests were used to examine the differences in the G* and δ based on the sample manufacturing method used by participants (Note that as mentioned before statistical test exclude 50/70 due to low number of participants). The term ‘other-mfg.’ refers to the combination of manufacturing methods other than sheet and silicone mould. The sheet manufacturing method resulted in a higher G*, and a lower δ on average than mould manufacturing method for all test conditions, but there were no significant differences between the two methods. There are, however, statistically significant differences ($p < 0.05$) between other-mfg. and sheet, as well as between other-mfg. and mould at few testing conditions as it follows: For G*, for the RTFOT 45/80-55 at 15 °C and 20 °C when P25 applied. For δ, for the RTFOT 20/30 at 35°C, 30°C and 25°C with PP08, for the RTFOT 45/80-55 at 10°C with PP08 and at 50°C with PP25, and for the unaged 45/80-55 at 60°C with PP25.

Pre-heating time and temperature for manufacturing sample

In this study only in the 2020 round test are DSR samples directly manufactured after heating, while 2017-2019’s round tests undergo short term ageing and samples manufacturing in the three following different ways:

Case1: without additional heating
Case2: cooling slightly and reheating to a defined temperature and duration
Case3: cooled and stored for later reheating to a defined temperature and duration.

Case 2 is the least preferred method have been applied by 14-21% of laboratories. Table 5.3 shows the oven heating duration and pouring temperature of sample materials for sample manufacturing directly without additional heating compared to the recommended limits of EN 14770. As expected, Gr. A are more likely than Gr. B to be in accordance with EN recommended limits for pure bitumen 50/70. On the contrary, for pure bitumen 20/30, Gr. A exceeds the recommended upper limit range of EN, while Gr. B follows it strictly. In case of PMB in both conditions, aged and unaged, almost the same range of temperature (155-185 °C) is used by more than 80% of both groups. The outcome presents a tighter range of temperatures within the range limits of EN, which leads to satisfactory test results. Figure 6 illustrates that the heating duration preference noticeably differs between Gr. B and A for all materials, except for the unaged 45/80-55. An oven sitting duration between 60 to 90 min for pure bitumen, and no longer than 60 min for PMB regardless of aging condition, are chosen by the majority in Gr. A. Results also indicate, in this case for Gr. A, that heating temperature decreases as oven sitting duration increases.

Table 5. 3. Pre-heating temperature (°C) and duration (min) range used by laboratories of Gr. A and B compared to limits according to EN 14770 and EN12594 in case 1.

Material -Yr.	50/70- 2017	20/30- 2018	45/80-55 -2019	45/80-55-2020
Range according to EN14770 or **EN12594**	SP + (85±5) =144-139 **max: 54 + 100 = 154**	SP + (85±5) =150-140 **max: 60 + 100 = 160**	**Max 200** (12594:7.4. b)	**Max 180-200** (12594:7.1.1)
Percentage of each group in accordance with EN recommended range				
Gr. B	17%	100%	100%	100%
Gr. A	100%	93%	100%	100%
The heating temperature range (°C) used by at least 80% of each group				
Gr. B	135-163 (100%)	145-160 (88%)	160-180 (87%)	155-180 (80%)
Gr. A	140-150 (100%)	150-165 (88%)	160-185 (82%)	155-180 (83%)
Percentage of each group that used heating duration range as follows: <=60; 60-90; >90-120 (min)				
Gr. B	17%; 83%; 0%	12%; 44%; 44%	40%; 53%; 7%	92%; 8%; 0%
Gr. A	50%; 50%: 0%	44%; 49%; 7%	70%; 20%; 10%	84%; 16%; 0%

Figure 5.5 shows the reheating temperature and duration for sample preparation in case 2. The average heating temperature used in this case is less than two other cases. Nevertheless, this approach, as the least favourable method, has results obtained based on only two measures or missing data noted as not applicable (n.a).

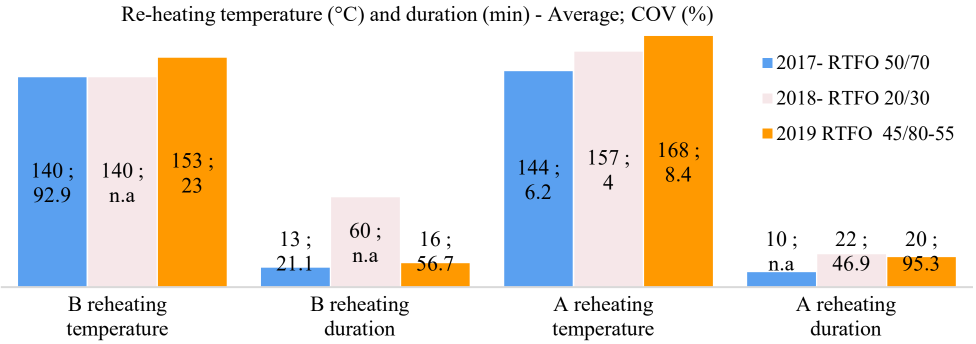


Figure 5. 5. Re-heating temperature and duration for sample manufacturing after RTFO (case 2).

Figure 5.6 shows that the time delay (storage duration) from RTFO residue cools completely and reheated for the sample manufacturing applied in case 3. The storage and reheating duration results are widely distributed, which makes drawing a general conclusion difficult. Differing from case 1, heating temperature increases for sample materials in this case, and the reheating duration and storage duration also increases for Gr. A. For all the cases and sample materials, Gr. A. prefers a slightly higher heating temperature than Gr. B. on average. The COV for time duration is much higher than the heating temperature, however, and Gr. A. has a lower variation in its data set than Gr. B.

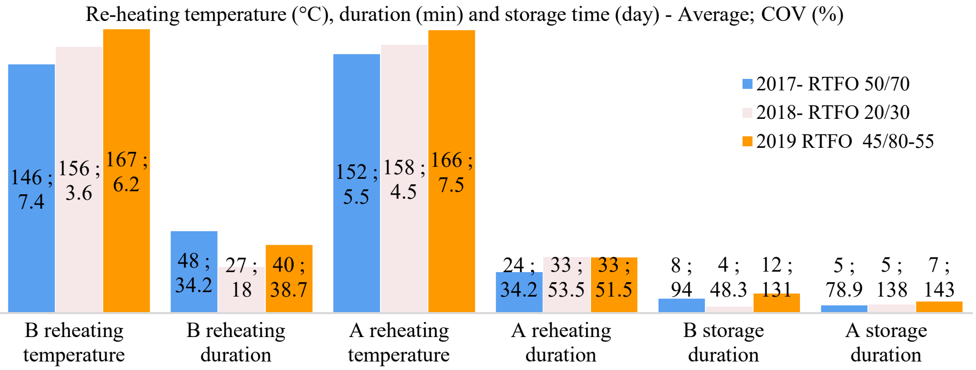


Figure 5. 6. Re-heating temperature, duration, and storage time for sample manufacturing after RTFOT (case 3).

Table 5.4 shows the direction of the correlation coefficient r (+, −) between the G* and δ with the heating temperature and heating duration without grouping the participants. In general, a negative sign signifies a reduction in G* or δ when heating temperature or duration increases. These factors appear to be positively correlated with each other, meaning that if one increases, the other one also tends to increase. For RTFOT 20/30, the G* shows a statistically significant moderately negative relationship with heating temperature, which is shown in bold in Table 5.5. While the correlation for δ is positive. However, for RTFOT 45 /55-80, G* and δ were found to be insignificantly negatively correlated with heating temperature for sample manufacturing. The unaged 45/55-80, on the other hand has a different tendency to aged ones at higher test temperatures (PP25), which may be attributed to its stronger relationship (greater r) to heating duration rather than temperature in comparison to other materials.

Table 5. 4. The sign of the correlation coefficient r (+, −) between G* and δ, with the heating temperature and duration at different test conditions.

20/30 RTFOT		45/55−80 RTFOT		45/55−80 Unaged		20/30 RTFOT		45/55−80 RTFOT		45/55−80 Unaged	
PP08	PP25	PP08	PP25	PP08	PP25	PP08	PP25	PP08	PP25	PP08	PP25
Direction of relationship between G* and Heating Temp.						Direction of relationship between δ and Heating Temp.					
−	**−**	−	−	−	+	+	+	−	−	−	+
Direction of relationship between G* and Heating Dur.						Direction of relationship between δ and Heating Dur.					
−	−	−	−	−	**+**	+	+	+	+	+	−

Table 5. 5. Correlation coefficients (r) and p-values obtained for G* and δ, with heating temperature used by labs at different test conditions.

20/30 RTFOT	G* vs. Heating Temp.							δ vs. Heating Temp.						
Test COND	PP08				PP25			PP08				PP25		
	35°C	30°C	25°C	20°C	55°C	60°C	65°C	35°C	30°C	25°C	20°C	55°C	60°C	65°C
r	**-0.38**	**-0.39**	**-0.36**	**-0.32**	**-0.40**	**-0.31**	**-0.24**	**0.27**	**0.32**	0.24	0.26	0.19	0.17	0.17
p-value	0.002	0.002	0.004	0.03	0.001	0.01	0.04	0.03	0.01	0.05	0.06	0.11	0.16	0.16

When manufacturing samples of pure bitumen 20/30, approximately 25% of laboratories exceed the temperature of 160 °C. However, for the PMB in both aged and unaged conditions, approximately 30% of laboratories use temperatures lower than 160 °C, yielding more accurate results in terms of coefficient of variation and standard deviation for G* and δ, respectively, when compared to laboratories using temperatures higher than 160 °C. These comparisons show that the upper limit for pure bitumen (145 - 160 °C) is likewise appropriate for the PMB. It should be noted that the softening points of all materials are near to one another.

Tables 5.6, 5.7, and 5.8 display the G* and δ values for lower and higher temperature ranges together with the related heating durations. The results are in accordance with the results from correlation test carried out on applied heating temperature shown in Table 5.4. As contrary to aged bitumen, for unaged 45/55-80 the heating duration decreases from 58 min to 50 min when heating temperature increases from lower than 160 °C to higher than 160°C. Nevertheless, the differences are insignificant for all test conditions and materials, expect for the δ at PP25- 60 °C for unaged 45/55-80 shown on bold.

Table 5. 6. The average G* and δ by sample manufacturing in an upper and lower temperature and its duration for 20/30 RTFOT.

Heating Temp. - Dur.	<160°C - 70 min		>160°C - 79 min		<160°C - 70 min		>160°C - 79 min	
20/30 RTFOT	G*(kPa)	CV%	G*(kPa)	CV%	δ (°)	SD	δ (°)	SD
8mm- 35 °C	1230.5	17	1081.0	15	49.5	1.0	50.2	1.3
8mm- 30 °C	2355.7	15	2102.3	9	46.4	1.0	47.1	0.8
8mm- 25 °C	4511.8	15	4133.2	9	43.1	0.9	43.6	0.8
8mm- 20 °C	8374.3	10	7839.5	9	39.8	1.1	40.2	0.7
25mm- 55 °C	80.0	12	71.7	10	59.3	1.0	60.0	0.7
25mm- 60 °C	44.1	12	40.2	10	61.6	1.1	62.3	0.7
25mm- 65 °C	24.3	14	22.4	10	64.2	1.2	64.9	0.7

Table 5. 7. The average G* and δ by sample manufacturing in an upper and lower temperature and its duration for 45/80-55 RTFOT.

Heating Temp. - Dur.	<160 °C - 62 min		>160 °C - 67 min		<160 °C - 62 min		>160 °C - 67 min	
45/80-55 RTFOT	G*(kPa)	CV%	G*(kPa)	CV%	δ (°)	SD	δ (°)	SD
8mm- 20 °C	4443.8	14	4189.3	19	49.3	0.9	49.2	1.1
8mm- 15 °C	9590.0	12	9324.7	16	44.2	1.2	43.9	0.9
8mm- 10 °C	19999.7	9	18663.4	16	38.8	1.2	38.5	1.1
25mm- 40 °C	155.5	6	154.9	11	63.6	0.5	63.2	1.0
25mm- 45 °C	79.2	6	79.4	10	64.7	0.4	64.5	0.7
25mm- 50 °C	40.1	7	40.1	10	65.4	0.3	65.3	0.5

Table 5. 8. The average G* and δ by sample manufacturing in an upper and lower temperature and its duration for 45/80-55 Unaged.

Heating Temp. - Dur.	<160 °C - 58 min		>160 °C - 50 min		<160 °C - 58 min		>160 °C - 50 min	
45/80-55 Unaged	G*(kPa)	CV%	G*(kPa)	CV%	δ (°)	SD	δ (°)	SD
8mm- 20 °C	3376.9	9	3271.9	13	57.0	0.8	57.1	1.0
8mm- 15 °C	7912.2	14	7770.6	16	50.0	0.9	50.0	0.9
8mm- 10 °C	18251.9	10	17643.2	14	42.9	1.0	42.8	1.1
25mm- 40 °C	81.4	9	83.7	11	71.3	0.4	71.3	0.3
25mm- 50 °C	19.9	8	20.1	8	71.9	0.2	72.0	0.3
25mm- 60 °C	5.8	7	5.8	7	**71.7**	0.4	**71.9**	0.4

Waiting time between manufacturing of sample and test start (storage time)

Figure 5.7 shows the average storage time for laboratories in Gr. B, Gr. A, and Gr. A after excluding the extreme values (the maximum and minimum). Results are very widely distributed, especially for the first two years' round test and for pure bitumen compared to modified bitumen. In all the rounds, Gr. A has a slightly shorter waiting time on average after removing the extreme values. A relatively higher storage time is obtained for 20/30 compared to other materials, which is due to two laboratories having a time delay as high as 7 and 14 days. However, both mentioned laboratories belong to Gr. A., and have used similar sample manufacturing method (mould), but with two different brands of equipment and a heating temperature as high as 160 °C and 150 °C, as well as an oven setting duration of up to 70 min and 60 min, both of which are close to the average of the corresponding round at 157 °C and 69 min (for the oven setting).

According to EN14770 (2012) , the maximum delay recommended is 72 h regardless of the type of bitumen, with a minimum storage duration of 2 and 12 h for pure and polymer-modified bitumen, respectively. Nevertheless, Table 5.9 reveals that Gr. A. prefers to wait less than 2h for pure bitumen and at least 12h for PMB. Furthermore, three days of waiting time is exceeded by Gr. A more often than Gr. B in all cases, except for aged 45/80-55. For Gr. B., 24 h is the most preferred time delay, which follows Gr. A with a lower rate for all types of material.

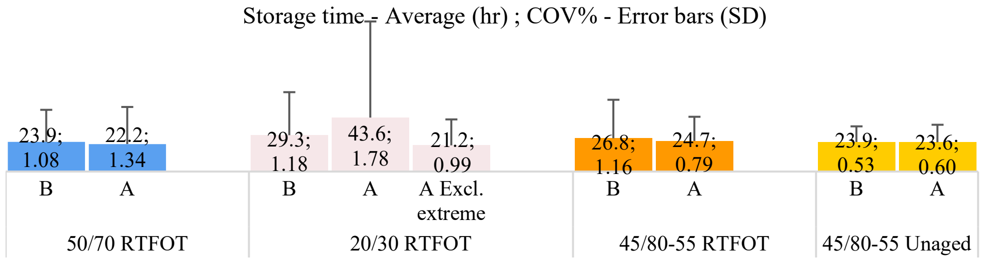


Figure 5. 7. Storage time (waiting time) between the bitumen sample casting and the start of the test procedure used by laboratories of Gr.A. and Gr.B. The error bars show the standard deviation.

Table 5. 9. Storage time (h) used by numbers [%]of Gr. A. and Gr. B.

Stored (h)	50/70 B	50/70 A	20/30 B	20/30 A	45/80-55 B	45/80-55 A	45/80-55 B	45/80-55 A
<2	17	20	0	9				
≤12					48	22	17	13
24	33	20	50	26	24	28	37	24
> 72	0	10	8	11	10	2	0	0
Other	50	50	42	54	18	48	46	63

The statistical tests were conducted to examine the differences on results by the three categories of waiting time applied by laboratories as follows:

short: <=2 h in case of unmodified bitumen and <=12 h for PMB
medium: 2< h <72,
long: >=72 h

In most test conditions, long waiting time resulted in a higher G*, and a lower δ when compared to short waiting time. Also, as waiting time passes, the ratio of standard deviation to mean for G* and standard deviation for δ drops (reduced variability). However, there are statistically significant differences only for 45/80-55 in unaged condition as follows: for the G* at test condition PP08- 20 °C (H (2) = 6.18, p-value = 0.046) between long waiting time and medium, and at 50 °C (H (2) = 6.97, p-value = 0.031) and 60 °C (H (2) = 7.68, p-value = 0.021) with PP25, between the long and short waiting time. In terms of the δ, significant difference was found only at PP25- 65 °C (H (2) = 7.91, p-value = 0.019) (Figure 5.8).

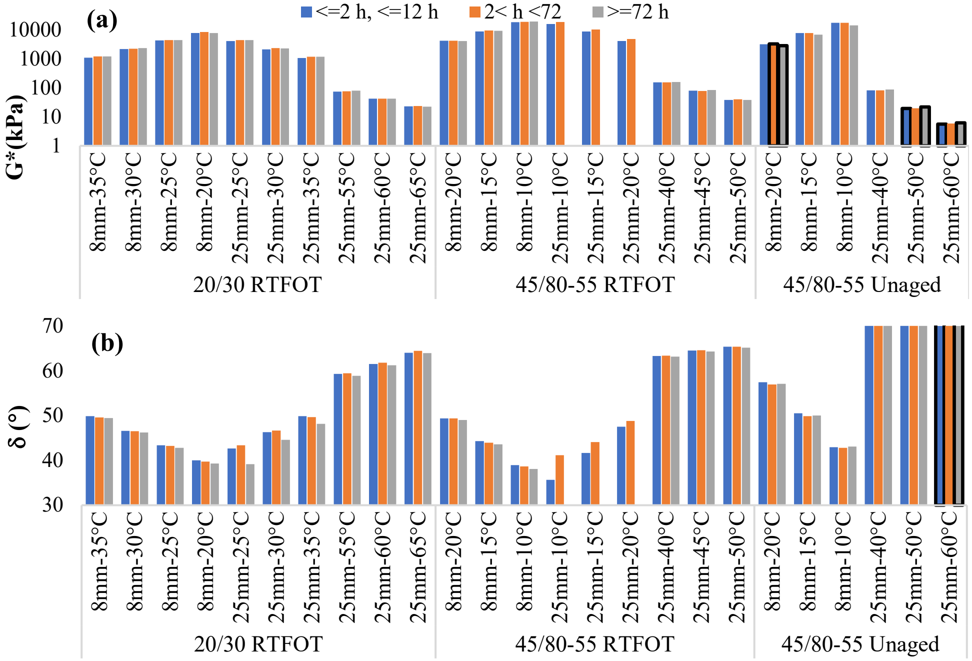


Figure 5. 8. The significant difference by waiting time is represented by a black outline for each test combination; (a) for the average G* of measurements, and (b) for the average δ of measurements.

Specimen bonding (mounting) temperatures

However, the result illustrated in Table 5.10 shows that only 19%-30% of Gr. A prefer to place the sample in a refrigerator, while 33%-67% of Gr. B opt for this, which can be due to the former knowledge of different groups of operators concerning the hardness of material. For pure bitumen the chosen temperature range for bonding bitumen are in accordance with the recommended upper limit, while PMBs exceed the limit. In cases that involve bitumen 20/30 the reported result of one participant is eliminated from the data set, when using 162°C as bonding temperature. This may be a misunderstanding, and this might be a result of the temperature used for binders poured directly onto the plate.

Table 5.10. Practice and bonding temperature onto rheometer used by the laboratories of Gr. A and B.

Material	50/70	20/30	45/80-55	45/80-55
Soft. Point (°C)	54	60.4	58.4	61
Bonding Temp. upper limit according to EN14770	SP+(20±5) = 79-69	SP+(20±5) = 85.4-75.4	SP+(20±5) = 83.4-73.4	SP+(20±5) = 86-76
The temperature range (°C) used by each group (most frequent used Temp.)				
Gr. B	40-74 (40, 60 &75)	50-90 (55&60)	25-90 (50,55,60,65&75)	25-90 (60 &70)
Gr. A	30-80 (40&60)	30-90 (50&80)	45-90 (60&80)	45-90 (60&80)
Percentage of each group placed the sample in a refrigerator before de-moulding				
Gr. B	67%	33%	47%	33%
Gr. A	29%	30%	27%	19%

Figure 5.9 demonstrates that the bonding temperature is slightly higher for the modified binder in general and for Gr. A compared with Gr. B. The duration of the bonding sample is wildly varied for all sample materials; however, it is not exceeded by 30 min except for one laboratory for bitumen 20/30, which is delayed by 45 min. An average of around 15.2 min for pure bitumen and around 12.6 min for modified bitumen are preferred by Gr. A.

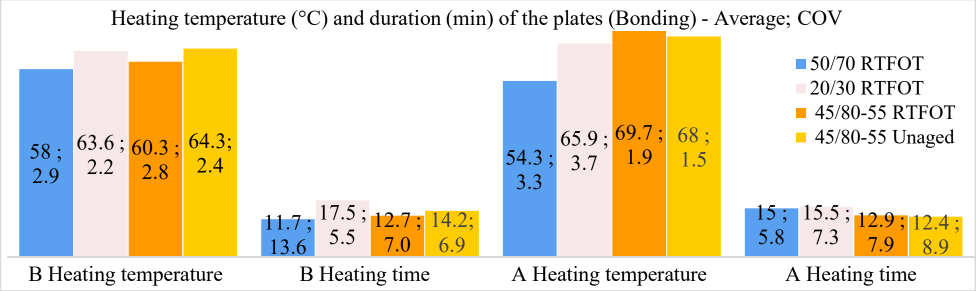


Figure 5. 9. Heating Temperature and duration of the rheometer plates including the sample of material prior to testing.

Without grouping the participants, a slightly higher bonding temperature are chosen for the modified bitumen than pure bitumen, regardless of their softening point. The duration of the bonding sample is wildly varied for all sample materials (1 - 30 min); only one laboratory for bitumen 20/30, is delayed by 45 min. An average duration of around 16 min for pure bitumen and around 13 min for modified bitumen are preferred by labs. The Pearson's correlation test shows a positive relationship between the G* and bonding duration for 20/30, and a negative relationship for 45/55-80 regardless of ageing condition. However, this is only significant at 40 °C and PP25 for RTFO aged 45/55-80 (r = − 0.24, p-value = 0.037).

Based on result of the Pearson's correlation test, there is a negative relationship between the bonding temperature used by participants and the G* for RTFO aged bitumen. Table 5.11 shows these significant values in bold. However, for unaged bitumen, the bonding temperature positively correlated with the G* across test conditions, although this impact peaked statistically at PP25- 40 °C (r = 0.2, p-value = 0.02). This effect might be brought on by ageing of fresh bitumen at higher bonding temperatures. The result of correlation test between the δ and bonding temperature indicates a positive relationship at test temperature between 40 to 65 °C for all the materials. However, the relationship between these variables is negative, for 45/55-80 at test temperature between 10 to 20 °C. For the δ similarly to G*, for most test conditions, the larger coefficient of correlation is indicated with bonding temperature rather than bonding duration.

Table 5. 11. Correlation coefficients (r) and p-values obtained between G* and the bonding temperatures used by labs at different test conditions.

relationship between G* and Bonding Temp.													
Material	20/30 RTFOT							45/80-55 RTFOT					
Test COND.	PP08				PP25			PP08			PP25		
	35 °C	30 °C	25 °C	20 °C	55 °C	60 °C	65 °C	20 °C	15 °C	10 °C	40 °C	45 °C	50 °C
r	**− 0.3**	**− 0.4**	− 0.2	− 0.03	**− 0.2**	− 0.2	− 0.2	− 0.1	**− 0.2**	**− 0.2**	**− 0.3**	−0.04	−0.02
p-value	0.02	0.003	0.23	0.85	0.06	0.10	0.21	0.28	0.04	0.04	0.02	0.72	0.84

Additionally, a statistical test was conducted to examine the differences on G* by the three bonding temperature spans used by participant laboratories. The temperature spans are chosen based on the softening point (SP) of the materials as follows: lower than SP: 25 - 55 °C, around SP: 55 - 75 °C, and higher than SP: 80 - 90 °C. For both RTFO aged bitumen, the G* value is higher for a bonding temperature lower than the softening point (25 - 55 °C), compared to the higher softening point (80 - 90 °C) for almost all test conditions (Figure 5.10). Significant differences (p-value < 0.05) were found among the half of the test conditions, indicating the need of maintaining unambiguous upper and maybe lower bonding temperature limits.

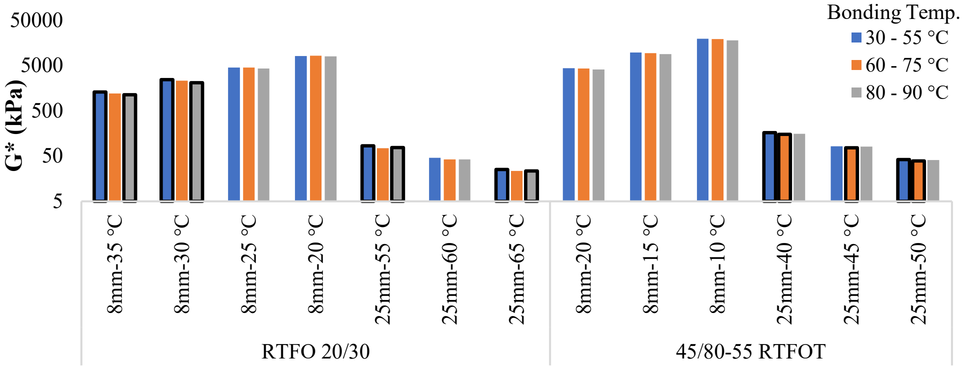


Figure 5. 10. The significant difference by the three bonding temperature spans is represented by a black outline for each test combination for the average G* of measurements.

Testing within the viscoelastic linear range

With respect to current activity, for Gr A, only one laboratory in 2017 and between two and three laboratories in 2019-2020 round test reported that they have not studied the viscoelastic linear range of the sample material. They may have chosen the suitable shear strains or stresses based on their experiences with the material and the device used. Stress-controlled mode was used by five laboratories in 2018 and two laboratories in 2017 and 2020, while most of the laboratories applied strain-controlled mode. Figure 5.11 illustrates strain values chosen for the tests by laboratories in Gr. A at different testing conditions. Some of the participants varied strain amplitude as a function of temperature, while others applied a strain as a function of plate dimension.

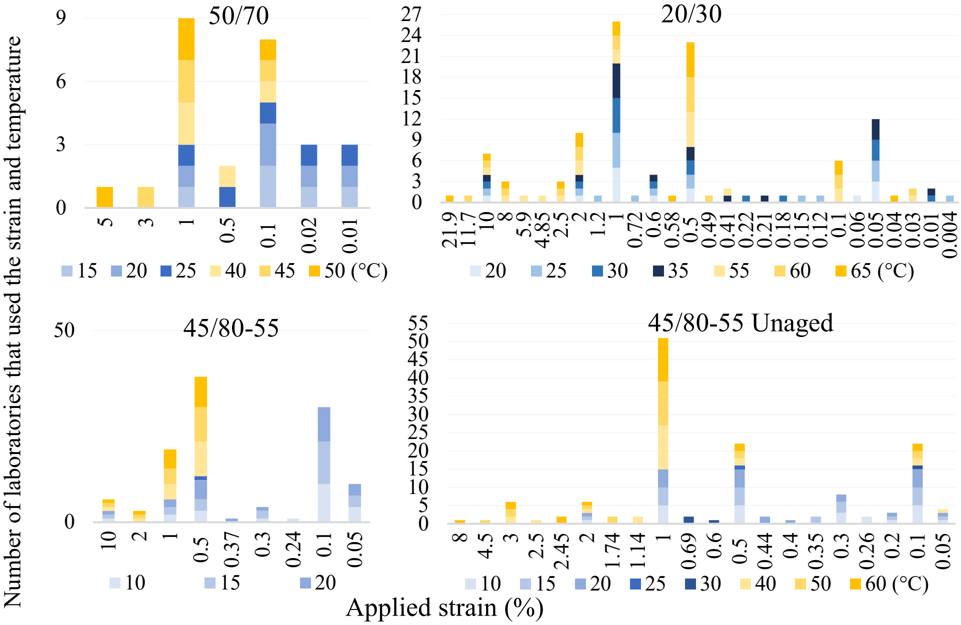


Figure 5. 11. Strain value (%) applied by a different number of laboratories.

Duration of Equilibrium time

Determining time for the temperature of the bitumen to reach thermal and mechanical equilibrium is outlined in EN 14770; however, there is no data if participants have followed the procedure. The findings show that, 15 min is the most frequently used equilibrium duration regardless of testing temperature, plate geometry, material type, and ageing condition. Also, a closer look into data reveals that laboratories with similar model and brand of DSR have chosen different durations of equilibrium time. Only three-five laboratory measures carried out increasing and decreasing testing temperatures with PP08 and PP25 geometry, respectively. Nevertheless, for the last year-round tests, all the measures with PP08 were done with a decreasing trend on temperatures, where an increasing trend was used when applying the PP25.

The correlation test for all the tested bitumen reveals a negative relationship between G* and equilibrium duration at test temperatures ranging from 40 to 65 ° C. Only at 50 °C the link was significant, but marginal ($r = 0.18$, p-value = 0.020). At majority of the test conditions, however, δ increases as the equilibrium duration grows.

A statistical test was also done to examine the differences in G* driven by the three durations of equilibrium timespans chosen by participant laboratories as follows: 5 - 15 min is utilized by 6 - 11% of labs, 15 min is used by 67 - 75% of labs, and 15-30 min is used by 14 - 27 % of labs. Result in Table 5.12 shows that, at test temperatures ranging from 40 to 65 °C and PP25, the average G* is lower for equilibrium duration of 15 to 30 min compared to other timespans for all the studied bitumen. However, significant variations were detected only for unaged 45/80-55 at 40 °C (H (2) = 17.64, p-value < 0.001). Nevertheless, when PP08 is used, the converse effect is observed for practically all measured temperatures ranging from 10 to 35 °C (Table 5.13); a lower average G* for applied equilibrium duration of 5 - 15 min, with no significant effect.

Table 5. 12. Average G* by the equilibrium duration timespan and temperature using a PP25.

Eq. Dur. (min)	Avg. G* (kPa) at different test combinations using a PP25								
	20/30 RTFOT			45/80-55 RTFOT			45/80-55 Unaged		
	55°C	60°C	65°C	40°C	45°C	50°C	**40°C**	50°C	60°C
5-15	82.3	45.0	24.9	161.1	82.5	40.4	93.5	20.3	6.0
15	79.4	43.7	24.0	156.5	79.3	40.4	83.7	20.1	5.8
15-30	73.1	40.9	23.0	147.5	77.4	38.6	**78.3**	19.5	5.7

Table 5. 13. Average G* by the equilibrium duration timespan and temperature using a PP08.

Eq. Dur. (min)	Avg. G* (kPa) at different test combinations using a PP08									
	20/30 RTFOT				45/80-55 RTFOT			45/80-55 Unaged		
	35°C	30°C	25°C	20°C	20°C	15°C	10°C	20°C	15°C	10°C
5-15	1146.6	2217.3	4107.9	8586.8	4145.7	9386.6	18248.8	3117.8	7421.4	16519.0
15	1222.6	2297.2	4446.3	8295.5	4344.3	9425.6	19196.3	3304.1	7769.2	17814.5
15-30	1151.2	2291.4	4492.8	7877.2	4061.3	9617.0	19302.1	3353.9	8073.6	18345.1

5.4 Conclusions of study I

This study presents different practices and their impacts on lab results in four interlaboratory tests on RTFO-aged neat bitumen 50/70, 20/30, and PMB 45/80-55 in RTFO-aged and unaged conditions using a DSR at frequency of 1.59 Hz and temperatures ranging from 10 to 65 °C. The practices used by labs differed noticeably, emphasizing the importance of having a well-defined protocol for interpreting EN14770:2012 for determining G* and δ.

It is found that the coefficient of variation under repeatability are within a range of 2-12% for |G*| and 0. 2-3.4% for δ. Coefficient of variation under reproducibility vary between 7% and 20% for |G*| and 0.4-20.2 for δ, which is not fulfilling the EN14770:12 recommended reproducibility values of 10% for the complex shear modulus and 5% for δ. However, Ignoring the extreme values attributed to the selection of unsuitable plate geometry or the small number of laboratories engaged improves the precision range of the phase angle to a repeatability limit of r =1-2° and reproducibility limit of R=3-6° for tested bitumen. Additionally, the |G*| and δ obtained from 8- and 25-mm parallel plates at all the overlap temperatures deviate from the mean of |G*| and δ by less than 6 % and 0.6°, respectively, which are significantly better than the recommended values of 15% for |G*| and 3° for δ.

Remarkable differences were identified in terms of the specimen preparation and conditioning of the sample when screening practices used by two groups of participants which were grouped based on their z-score and repeatability on reported results. A closer examination of the methods utilized by groups reveals that the group with greater precision has not strictly followed all the preparation processes according to the existing norm, such as waiting time. Also, better repeatability than reproducibility means that discrepancies in results between laboratories may be due to differences in testing methodology rather than operator skill. An examination of three categories of equipment brand and sample manufacturing methods used by participants found that, the G* was higher when brand of Anton Paar and sample manufactured by dropping bitumen on a sheet were used, while Malvern and moulding method had a higher δ. However, no significant differences were discovered across the two most used brands of equipment and sample manufacturing methods. Furthermore, the δ value appears to be more sensitive to manufacturing method than the G*. The long waiting time before testing the manufactured sample had a higher G* and a lower δ value in comparison to the short waiting time. However, none of the studied bitumen would be significantly affected by waiting times of less than 2 hours or longer than 72 h. A statistically significant variations between different equilibrium timespan (5-15 min and 15-30 min) were detected only for unaged 45/80-55 at 40 °C. On the other hand, in most test conditions, the value of δ increases along with the equilibrium time. 15 minutes appears to be a suitable equilibrium duration, with results frequently falling between the upper and lower reported values. Finally, the bonding temperature and the sample production temperature showed a significant association in more test combinations than other sample preparation steps did, according to the correlation test. However, a sensitivity analysis, which examines each stage as the only variable in each experiment while holding all other conditions constant, is required to ascertain how much each phase of the sample preparation and conditioning influences the outcome.

6 Study II: Effects of various selected test techniques on Dynamic Shear Rheometer test results: laboratory experiments on unmodified Bitumen

6.1 Aim

The aim is to assess current methods for testing bitumen with a DSR in accordance with EN:14770. The goal of this study is to determine whether and under what conditions the heating temperature for sample manufacturing, bonding temperature, and radial trimming of the sample on DSR testing have a significant impact on the results when the testing set-up variables are varied. Additionally, the interaction effects of these testing conditions, as well as simple instructions on the test procedure and sample and Rheometer preparation employing parallel plate geometry, will be described.

6.2 Material and Method

Table 6.1 lists the Material type used in this research. The needle penetration test (PEN) according to EN 1426, the softening point (SP) test according to EN 1427, and the density of the bitumen which is used for calculating the amount of bitumen needed for sample preparation are also provided.

Table 6. 1. Type of bitumen used in Study II.

Sample ID	PEN (0.1mm) EN 1426	SP (°C) EN 1427	Density kg/m^3
50/70	61	48.4	1030
70/100	77	46.0	1022
160/220_I	160	41.2	1000
160/220_II	161	39.5	1013

A 2^3 factorial designed experiment conducted to evaluate the effects of selected factors on G* and δ at frequencies of 0.1rad/s and 10 rad/s at different temperatures. These variables are two quantitative factors – heating temperature (HT) for manufacturing the samples and the bonding temperature (BT) when placing samples onto DSR –and a qualitative factor – trim state. Table 6.2 and Figure 6.1 illustrate the 8 combinations of three factors and two levels. The numbers in parenthesis (-1 or +1) refer to coded values for the factors under study at their low (-1) and high (+1) levels, respectively. The data values measured for G* and δ are averaged over two genuine replicated runs. Randomization of run order for all 16 runs is done to ensures that variation between runs made at the same experimental conditions. All the other remaining phases of preparation and conditioning of the sample were constant and fixed according to EN-14770:2012.

After processing of responses G* and δ data, the significant parameters were determined manually also with a statistical analyse tool, R (ANOVA test), after the main and interactions effects and their standard errors were calculated as described in result section.

The main effect is defined as the difference between the average outcomes of the factor at two defined levels. The interaction is the effect between two factors (HT:BT) which can be defined as the mean difference between the effects of heating temperature and bonding temperature at both levels. There is also an interaction effect between three factors, which is also defined as the mean difference between the effects of three factors at same levels.

Table 6. 2. Combination of a two-level three factor factorial design with two replicates.

Standard Run Order	Randomized Run Order	Trimming state Trim	Bonding Temp. °C BT	Heating Temp. °C HT
1	6 & 13	Yes (-1)	SP (-1)	SP+80 (-1)
2	2 & 4	No (+1)	SP (-1)	SP+80 (-1)
3	1 & 16	Yes (-1)	SP+25 (+1)	SP+80 (-1)
4	5 & 10	No (+1)	SP+25 (+1)	SP+80 (-1)
5	8 & 12	Yes (-1)	SP (-1)	SP+100 (+1)
6	9 & 14	No (+1)	SP (-1)	SP+100 (+1)
7	3 & 11	Yes (-1)	SP+25 (+1)	SP+100 (+1)
8	7 & 15	No (+1)	SP+25 (+1)	SP+100 (+1)

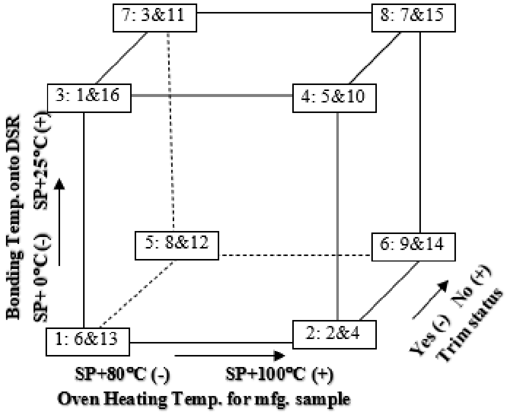


Figure 6. 1. Three parameters for sample preparation and conditioning in DSR testing are shown in a two-level factorial design matrix (2^3) with their high (+) and low (-) values.

6.3 Test procedure and preparation of sample and Rheometer

Testing plan: Strain and frequency sweep tests

An Anton Paar MCR302 dynamic shear rheometer with RheoCompass software was used for measuring complex shear modulus (G*) and phase angle (δ) in an oscillatory-type testing mode using 8 mm and 25 mm parallel plate testing geometries. The storage modulus (G′) and the loss modulus (G″) can be represented as |G∗|cos(δ) and |G∗|sin(δ), respectively. As per current standard (EN 14770), the LVE region is considered as the range of strain up to which the value of G′ and G″ differ by less 5% of the initial value over the chosen shear strain range. The strain amplitude sweep (SAS) test was performed at all the tested temperatures and materials. However, in this study the strain amplitude limits for 8 mm and 25 mm geometries were selected at range of 0.1% (0.001 mm/mm) and 0.5%, respectively, where the shear modulus is relatively independent of shear stress.

The Temperature-Frequency-Sweeps (T-f-sweep) tests were performed for each of the 8 conditions set (runs) and material. Each T-F-sweep involved 3 samples. For instance, one in the temperature range of 30°C to 0°C using a parallel plate with a diameter of 8mm (PP08) and gap height of 2mm, and the two others, in the range of 30°C to 50°C and 60°C to 80 °C using a parallel plate with a diameter of 25mm (PP25) and a 1mm gap height. The same operator performed 48 T-f-sweeps per material in total, with two repeats, for each tested bitumen over the 8 runs. A frequency range of 0.1 to 10 Hz was used to collect 10 points from the logarithmic ramp pattern for each constant temperature. Note that, in this investigation, only data at the high and lower end of the frequency ranges were employed. The tests were performed at intervals of 10°C by allowing the material to remain at the test temperature for 15 min within the tolerance of ±0.2°C. Consequently, T-f-sweep per sample took about 2.5 hours.

Sample preparation

In this study, samples were manufactured after heating the material at the planned temperatures (SP+80°C and SP+100°C) for a max of 20 min or 40 min depending on the size of the container 100 or 200 gr (Figure 6.2). The heated bitumen was poured into silicone molds after stirring and homogenization. The bitumen moulds were stored at room temperature without being exposed to light, and before testing the disc shape samples were removed from the mould after 5 min refrigeration and stored on silicone sheets for 10 min (one with pink color in figure below) at 5C temperature . Only 3 samples were manufactured at a time in order to maintain a constant storage time before testing the samples between 1 to 10 hours. Samples were then tested one after another; the first specimen stored just 1h, second specimen stored for about 4 h, and the third one for maximum of 10 h at room temperature. Note that same pattern used at all the runs. Zero gap were carried out after waiting 10 min at the mid-point of the expected testing temperature range, for instance at 40 °C if the plan was to carry out a T-F-sweep at 30, 40, and 50 °C. At the planned bonding temperature (SP+0°C and SP+25°C), in the case of trimming method, sample was loaded onto the lower plate of the DSR, and the upper plate was then gradually lowered to a gap of 1,05 mm and 2,1mm for PP25 and PP08 respectively, and after 60 sec the excess material was removed from the radial surface of the samples with a heated spatula. Trimming was done in several steps, wiping away the bitumen residue each time, and not trimming with a too warm and smoked spatula. At least the test gap was set to 1mm and 2mm. Meanwhile, in without the trimming method, the upper plate was removed from the rheometer (this is possible for Anton Paar instruments), and with help of a form centralized the weighed sample into upper plate . Then gradually lower the upper plate toward lower plate to reach a bulge around the periphery of the plates at the final gap of 1mm and 2mm for PP25 and PP08 respectively. To eliminate trimming, the exact amount of sample material is estimated based on the volume of the sample and the density of the materials. The amount of bitumen used in samples with trimming was somewhat higher than in samples without trimming. Normal force-controlled mode was used for gap compensation, where compensate specimen shrinking at low temperatures by adjusting the gap to keep the normal force zero. The normal force is set to zero N. However, because we only work with a narrow temperature range, this may not be critical. It would be more relevant if the temperature range were wider.

Figure 6. 2. Moulds, silicon sheets, and centralizer forms were employed in the manufacturing and installation of the specimen into the DSR

6.3 Results and Discussion

The main and interaction effect of the selected factors on G* and δ and relevant standard errors were calculated at frequencies of 0.1 rad/s and 10 rad/s (1.59 Hz), and at temperature range between 0 °C to 80 °C for 50/70 and 70/100, and between 0 °C to 70 °C for the softer bitumen 160/220. Averaging individual measures of effects and differences between two averages is done to calculate the main effects, two-factor interactions, and three factor interaction as following (Box et al., 1978). The standard test order used to illustrate:

$$\mathrm{Trim}(G^{*}) = ((G^{*}8 - G^{*}7) + (G^{*}4 - G^{*}3) + (G^{*}6 - G^{*}5) + (G^{*}2 - G^{*}1))/4$$

$$\mathrm{BT}\,(G^{*}) = ((G^{*}3 - G^{*}1) + (G^{*}4 - G^{*}2) + (G^{*}7 - G^{*}5) + (G^{*}8 - G^{*}6))/4$$

$$\mathrm{HT}\,(G^{*}) = ((G^{*}8 - G^{*}4) + (G^{*}7 - G^{*}3) + (G^{*}6 - G^{*}2) + (G^{*}5 - G^{*}1))/4$$

$$\mathrm{Trim.HT}\,(G^{*}) = (G^{*}1 + G^{*}3 + G^{*}6 + G^{*}8)/4 - (G^{*}2 + G^{*}4 + G^{*}5 + G^{*}7)/4$$

$$\mathrm{Trim.BT}\,(G^{*}) = (G^{*}8 + G^{*}5 + G^{*}4 + G^{*}1)/4 - (G^{*}7 + G^{*}6 + G^{*}3 + G^{*}2)/4$$

$$\mathrm{BT.HT}(G^{*}) = (G^{*}8 + G^{*}7 + G^{*}2 + G^{*}1)/4 - (G^{*}3 + G^{*}4 + G^{*}5 + G^{*}6)/4$$

$$\mathrm{Trim.BT.HT}\,(G^{*}) = (G^{*}8 + G^{*}5 + G^{*}3 + G^{*}2)/4 - (G^{*}7 + G^{*}6 + G^{*}4 + G^{*}1)/4$$

For brevity calculation of effects, and estimation of standard error shown only for G* at angular frequency of 0.1 rad/s and temperature of 30 °C when used a PP08 for 50/70. Average response values are used to calculate the effects. Randomized run order for all 16 runs is indicated in Table 6.3 by superscripts.

Table 6. 3. Estimating the variance at each set of conditions for G* at frequency of 0.1 and temperature of 30° C for 50/70.

Result from 8 runs	Average of duplicate (kPa)	Estimated Variance: (diff. of duplicate)^2/2
G^{*6} & G^{*13}	$G^*1 = (G^{*6} + G^{*13})/2 = 14\ 270$	$(G^{*6} - G^{*13})^2/2 = (2\ 997)^2/2$
G^{*2} & G^{*4}	$G^*2 = (G^{*2} + G^{*4})/2 = 15\ 524$	$(G^{*2} - G^{*4})^2/2 = (1\ 554)^2/2$
G^{*1} & G^{*16}	$G^*3 = (G^{*1} + G^{*16})/2 = 13\ 081$	$(G^{*1} - G^{*16})^2/2 = (634)^2/2$
G^{*5} & G^{*10}	$G^*4 = (G^{*5} + G^{*10})/2 = 14\ 487$	$(G^{*5} - G^{*10})^2/2 = (1\ 274)^2/2$
G^{*8} & G^{*12}	$G^*5 = (G^{*8} + G^{*12})/2 = 14\ 131$	$(G^{*8} - G^{*12})^2/2 = (812)^2/2$
G^{*9} & G^{*14}	$G^*6 = (G^{*9} + G^{*14})/2 = 17\ 739$	$(G^{*9} - G^{*14})^2/2 = (129)^2/2$
G^{*3} & G^{*11}	$G^*7 = (G^{*3} + G^{*11})/2 = 13\ 688$	$(G^{*3} - G^{*11})^2/2 = (518)^2/2$
G^{*7} & G^{*15}	$G^*8 = (G^{*7} + G^{*15})/2 = 16\ 402$	$(G^{*7} - G^{*15})^2/2 = (316)^2/2$
Average of the Estimated Variance of 8 observation:		904 132.6

Standard error of an effect (main or interaction), SE is square root of the average of estimated variance of 8 observations for an effect, which itself is difference between average of two levels according to equation below. A significant value of t distribution with 8 degrees of freedom (DF) at the 5% level ($\alpha=0.05$) is 2.3; thus the 95% confident interval for the estimated effects are given by Pr ($|t_{0.05,8}|>2.3$). The effects which are almost certainly real and not by chance are shown in bold type (Table 6.4). The estimated effects and its standard errors for all tested material are shown in Table 6.6 to Table 6.9. The variation in the experimental errors rises with the test temperature rather than having a constant variance for G*. In order to compare various materials and temperatures, it is also necessary to calculate the coefficient of variation (CV), which is equal to the standard deviation (SD) divided by the mean (Appendix 1).

$$\text{SE (effect)} = \sqrt{\text{Var(effect)}} = \sqrt{\left(\frac{1}{8}+\frac{1}{8}\right)\cdot\left(\sum_{n=1}^{n=8}\frac{diff_n^2}{2}\right)/\,\boldsymbol{n})} = \sqrt{\left(\frac{1}{4}\right).(904\ 132)} = 475.4$$

Table 6. 4. Average Effect with Standard Error on G* (ω=0.1 rad/s) at 30°C.

Factors	Average Effect ± Standard Error
Main Effects	
Trim	**2245 ± 475.4**
BT	– 1002 ± 475.4
HT	**1150 ± 475.4**
Two-factor interactions	
Trim:HT	916 ± 475.4
Trim:BT	– 186 ± 475.4
BT:HT	112 ± 475.4
Three-factor interactions	
Trim:BT:HT	– 262 ± 475.4

The two-level interaction is shown in Figure 6.3. According to the plots, there is no evidence of interactions, particularly for BT:HT and Trim: BT. The influence of the main factors cannot be interpreted separately if there is a significant interaction between them, even if the main factors involved are not significant. Interaction effects are plotted for all material and temperatures (Appendix 2).

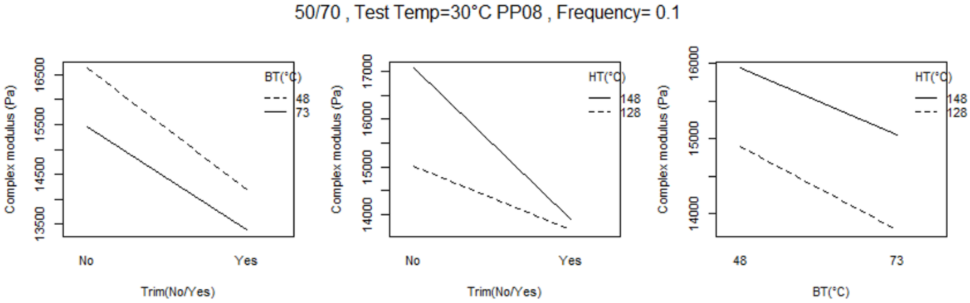


Figure 6. 3. Two-level interaction diagrams.

Table 6.5 shows the F-ratios and the P-values from ANOVA. The statistically significant effects of the factors were examined using P-values, which is in accordance with result in Table 6.4. It was found that the main effects, Trim and BT, statistically significant because the P-values from the ANOVA test were less than the confidence limit (α = 0.05) at which the null hypothesis could be rejected.

Table 6. 5. Analyse of variances, ANOVA for the G* (ω=0.1 rad/s) at 30°C_PP25.

Factor	DF	Sum of Square	Mean Square	F value	*P*-value (Prob > F)
Trim	1	20 169 081	20 169 081	22.31	**0.00**
BT	1	4 014 012	4 014 012	4.44	0.07
HT	1	5 285 401	5 285 401	5.85	**0.04**
Trim:HT	1	3 354 392	3 354 392	3.71	0.09
Trim:BT	1	137 641	137 641	0.15	0.71
BT:HT	1	49 729	49 729	0.06	0.82
Trim:BT:HT	1	274 052	274 052	0.30	0.60
Pure Error	8	7 233 061	904 133		
Total	15	40 517 369			

Figure 6.4 displays a normal probability of effect estimates and a significance level of α = 0.05 on the response G*. The graph is used to show the impact of sample preparation techniques, showing whether or not it is statistically significant. All calculated effects are plotted against a straight line which represents the normal

distribution line. The outliers, indicated by squares, are thought to be the factors that have the most governing parameters on estimating the result (G*). On this plot, the effects that are not statistically significant are typically distributed as a straight line with mean zero and variance σ^2, whereas the significant main effects and interactions have nonzero means and do not lie along the straight line (Daniel, 1959). The results show that the Trim and HT stay away from the normal distribution line. Thus, the effects are considered statistically significant at the 5% significance level. Factors Trim and HT have a stronger effect on the G* since they both lie farther from the straight line. The effects of factors Trim.HT, and BT are slightly not significant since they are closer to the straight line. By contrast, factors BT.HT, Trim.BT, and Trim.BT.HT are not significant.

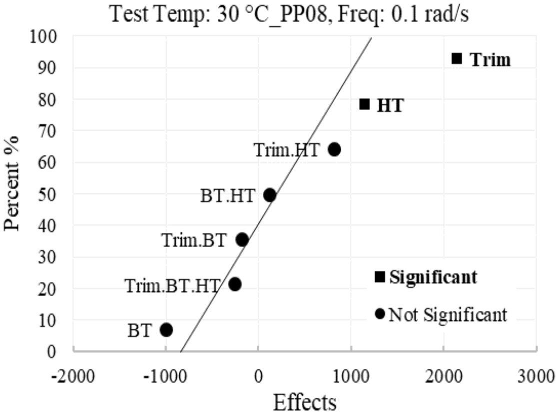


Figure 6. 4. Normal probability of effect estimates at significance level of α=0.05.

From result shown in Table 6.6 to Table 6.9 can be observe that, the complex modulus is more sensitive to the variation on studied factors than the phase angle. However, results do not show major difference when comparing the lower and higher tested frequencies; the almost same pattern is observed in term of significance of effects. The bonding temperature is the most evident factor affecting the G*, and the δ followed by oven heating temperature contrariwise in case of the 160/220_I, however since the two bonding temperatures level are not very high, higher G* and lower δ cannot be due to ageing rather than better adhesion between the bitumen and plates in temperature slightly above softening point. The variation in oven heating temperature strongly affected the results of the 160/220_I contrary to all other studied materials. Although not shown in this study, the master curve of this bitumen showed a weak overlapping on shifted temperature, which may be a hint that it had a different natural chemical component. Comparing the bounding temperature effect at frequency of 10 rad/s and the first tested temperatures of the 3 samples used for the T-f-sweep tests (30 °C with PP08 and PP25, and 60 °C) indicate that, bonding temperature effect is not significant at the higher test temperature (60 °C) for both G* and δ. Trimming the sample tested at lower temperatures of 0 °C to 30 °C for all tested material tends to significantly increase the δ and decrees the complex modulus, which may indicate higher sensitivity of smaller parallel plate (PP08) to trimming. Higher G* in case of without trimming can be due to more amount of excess material left on periphery of the plates compare to trimmed ones. The BT has a negative impact on G* at medium test temperatures for 70/100 and almost on all the evaluated temperatures for 50/70 and 160/220_II.

In 2-way interaction, in the case of G*, Trim:BT has the strongest effect on all material and temperatures except for 160/220 at lower temperatures where Trim-HT affects more. For δ, the most effective two-way interaction was Trim:BT for 70/100 and 160/220_II. For 160/220_I the Trim:HT, and for 50/70 the BT:HT is the least important factor for δ. However, it may be more relevant to study interaction effect of Trim:BT at only first tested temperatures after loading the plates with bitumen. Except for 160220_I, HT do not generally have a statistically significant impact on the results; but, when it interacted with other parameters, it has a substantial impact. This may be attributed to the fact that the range of oven heating temperature (SP+80 and SP+100°C) in this study is very small. According to the investigation, the studied parameters have an impact on G* and δ to varying degrees. The factors that have the greatest impact on are 160/220, followed by 50/70 and 70/100.

Table 6. 6. Effect estimates and Standard Errors (SE), for 50/70.

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=0.1 rad/s)								
0	**1 100 063**	**-619 763**	233 588	108 688	-282 663	246 713	223 563	215 687
10	**187 925**	**-93 575**	51 175	33 225	-34 175	36 975	28 625	37 414
20	**22 566**	-9 819	8 499	7 209	-2 389	3 014	1 044	4 602.0
30 PP08	**2 246**	-1 002	**1 150**	916	-186	112	-262	475.4
30 PP25	-487	426	677	469	728	-35	-167	343.1
40	-44	**157**	68	37	72	5	-17	44.40
50	-3.1	**19.5**	10.7	5.1	6.8	1.3	-0.4	6.44
60	-0.8	-1.8	**3.6**	0.6	1.7	-0.4	-1.1	0.86
70	-0.26	0.28	**0.65**	-0.08	0.15	-0.01	-0.36	0.23
80	-0.13	0.12	0.15	-0.06	-0.03	0.08	-0.11	0.09
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=10 rad/s)								
0	**5 930 000**	**-4 064 750**	1 211 250	601 000	-1 963 000	1 267 250	1 153 000	1 070 849
10	**1 606 000**	**-949 750**	379 000	190 750	-434 500	346 000	291 750	298 394
20	**335 575**	**-167 525**	101 300	66 075	-66 050	59 525	38 800	60 246.2
30 PP08	**52 660**	**-29 903**	**23 180**	15 890	-10 268	5 588	-2 063	9 512.4
30 PP25	-11 005	14 870	16 963	12 930	14 993	-1 090	-553	7 875.6
40	-1 207	**5 074**	3 363	2 442	2 422	-87	-278	1 683.05
50	-230.8	**868.5**	627.0	395.3	379.8	18.0	-16.3	354.13
60	-57.9	-102.5	**268.0**	68.4	140.2	-46.4	-78.5	63.12
70	-21.7	16.9	**51.8**	4.8	23.3	-10.7	-26.3	15.67
80	**-7.9**	6.3	**10.4**	-1.1	6.1	-1.9	**-8.5**	3.16
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
δ (ω=0.1 rad/s)								
0	**-0.46**	-0.03	-0.14	-0.08	-0.11	-0.11	-0.07	0.19
10	-0.42	0.06	-0.30	-0.31	-0.16	-0.02	0.03	0.23
20	0.05	-0.07	-0.31	**-0.52**	-0.20	0.16	0.11	0.21
30 PP08	-0.11	-0.11	-0.50	**-0.52**	-0.27	0.21	0.32	0.23
30 PP25	0.24	0.05	-0.19	-0.01	-0.29	0.05	0.25	0.16
40	0.08	**-0.68**	-0.06	0.10	-0.13	-0.16	0.09	0.10
50	-0.05	**-0.42**	0.02	0.13	-0.02	-0.13	0.03	0.07
60	-0.06	0.10	0.00	0.20	0.18	-0.20	-0.08	0.09
70	0.32	-0.25	-0.21	0.39	0.52	-0.63	0.16	0.33
80	0.49	-0.39	-0.33	1.30	1.96	-1.47	0.37	0.96
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
δ (ω=10 rad/s)								
0	**-0.29**	-0.06	-0.08	0.01	-0.09	-0.07	-0.04	0.11
10	**-0.39**	-0.09	-0.16	-0.10	-0.13	-0.06	-0.02	0.17
20	-0.43	-0.04	-0.28	-0.29	-0.20	0.03	0.14	0.21
30 PP08	-0.35	0.10	-0.38	-0.36	-0.18	0.08	0.30	0.21
30 PP25	0.10	0.23	-0.14	-0.04	-0.31	0.02	0.26	0.15
40	0.00	-0.20	-0.06	0.03	-0.20	-0.02	0.15	0.10
50	-0.01	**-0.26**	-0.03	0.05	-0.07	-0.03	0.05	0.07
60	-0.03	**0.16**	-0.09	0.03	-0.02	0.02	0.01	0.04
70	**0.10**	-0.04	**-0.13**	0.10	-0.01	**-0.09**	0.02	0.04
80	0.04	-0.07	-0.01	0.00	-0.13	-0.03	0.09	0.19

Table 6. 7. Effect estimates and Standard Errors (SE), for 70/100.

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=0.1 rad/s)								
0	254 463	108 363	-323 888	-172 538	424 763	111 763	82 463	234 343
10	28 270	8 815	-27 803	-14 250	38 418	10 385	3 933	21 525
20	2 815	622	-2 541	-1 186	3 400	1 025	225	2 091.1
30 PP08	295	41	-278	-124	351	120	20	238.1
30 PP25	53	-153	117	98	102	26	-2	67.6
40	10	-13	17	13	12	-1	-4	8.80
50	1.4	-0.9	2.3	1.6	1.6	0.4	-0.2	1.45
60	**0.7**	0.0	-0.1	0.2	0.4	0.3	0.0	0.26
70	**0.10**	0.05	-0.04	0.05	**0.15**	0.06	-0.03	0.04
80	0.03	0.00	**-0.04**	**0.04**	0.03	0.02	0.03	0.01

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=10 rad/s)								
0	1 951 125	1 029 375	-3 840 625	-2 202 125	4 072 375	1 236 625	1 372 625	2 573 870
10	431 950	213 300	-703 300	-394 050	826 550	257 300	217 050	480 270
20	73 925	30 400	-104 725	-55 050	127 575	41 175	26 800	72 653.4
30 PP08	10 021	3 524	-16 514	-8 701	18 906	6 951	3 764	11 671.0
30 PP25	1 084	-5 411	6 664	5 909	4 724	719	-246	3 014.2
40	473	-813	1 243	965	690	-118	-299	544.06
50	88.8	-77.3	209.8	151.7	119.4	20.4	-24.9	110.10
60	**63.7**	-8.5	-5.4	19.4	34.3	35.9	-3.1	25.03
70	**9.7**	3.7	-0.3	3.0	**16.9**	7.3	-5.1	3.55
80	1.8	-0.5	-0.7	1.8	**3.9**	3.2	0.1	1.41

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
δ (ω=0.1 rad/s)								
0	**-0.50**	-0.07	0.07	-0.03	-0.37	-0.03	0.15	0.19
10	**-0.59**	0.01	0.08	-0.03	-0.25	-0.01	0.20	0.24
20	-0.54	0.11	0.06	-0.08	-0.13	0.00	0.21	0.27
30 PP08	-0.54	0.15	0.05	-0.10	-0.02	0.06	0.29	0.26
30 PP25	-0.14	**0.24**	0.01	0.03	-0.07	-0.06	-0.02	0.07
40	-0.06	0.06	0.00	0.01	-0.05	-0.02	0.00	0.04
50	**-0.03**	0.00	0.02	0.02	0.00	-0.02	0.00	0.01
60	0.08	-0.07	0.10	-0.12	0.11	0.09	-0.13	0.12
70	0.25	-0.11	0.24	-0.10	0.23	0.09	-0.17	0.24
80	0.10	-0.20	0.29	-0.66	0.12	0.06	-0.59	0.48

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
δ (ω=10 rad/s)								
0	**-0.24**	-0.07	0.04	-0.01	**-0.27**	-0.03	0.05	0.10
10	-0.35	-0.06	0.06	-0.03	-0.32	-0.06	0.10	0.15
20	**-0.45**	0.00	0.07	-0.02	-0.28	-0.05	0.15	0.18
30 PP08	-0.43	0.09	0.07	-0.03	-0.16	-0.06	0.17	0.20
30 PP25	-0.19	**0.29**	-0.01	0.00	-0.11	-0.06	0.01	0.09
40	-0.14	0.17	-0.01	0.00	-0.12	-0.05	-0.01	0.08
50	-0.09	0.05	0.00	-0.01	-0.08	-0.03	-0.01	0.06
60	-0.03	-0.01	0.03	-0.01	**-0.07**	**-0.06**	0.02	0.02
70	-0.02	0.02	**0.20**	0.00	-0.02	0.07	0.02	0.07
80	-0.08	-0.26	0.19	-0.12	-0.21	0.11	0.26	0.24

Table 6. 8. Effect estimates and Standard Errors (SE), for 160/220_I.

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=0.1 rad/s)								
0	**434 950**	**611 475**	**-520 175**	-70 500	-68 350	185 675	238 350	117 939
10	49 393	**156 418**	**-112 123**	-9 945	-19 575	21 340	33 663	25 366
20	2 427	**23 623**	**-17 452**	-2 616	-4 748	240	2 730	4 333.2
30 PP08	127	**1 565**	**-1 262**	-620	-432	52	-60	377.4
30 PP25	73	**1 163**	**-862**	**-722**	**-684**	**586**	229	165.6
40	8	**77**	**-47**	**-72**	**-65**	**77**	**21**	8.12
50	0.8	2.0	1.2	**-6.4**	**-6.3**	**9.3**	2.4	1.32
60	**1.0**	-0.9	**1.3**	0.2	-0.3	-0.3	-0.4	0.42
70	0.18	-0.15	**0.30**	0.09	-0.09	-0.09	-0.10	0.09
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=10 rad/s)								
0	**2 844**	1 165 500	**-1 876 250**	-714 750	-239 000	781 500	1 165 000	667 751
10	**595 613**	**660 813**	**-614 088**	-138 813	-93 563	205 488	281 213	157 821
20	**88 005**	**173 628**	**-140 773**	-38 613	-31 968	32 705	37 210	33 575.5
30 PP08	13 052	**21 899**	**-20 642**	-12 311	-6 478	4 829	1 531	5 903.3
30 PP25	1 534	**18 738**	**-13 366**	**-10 741**	**-8 013**	**9 789**	3 838	2 743.5
40	292	**1 896**	**-1 270**	**-1 958**	**-1 267**	**2 052**	334	365.34
50	40.8	18.4	31.8	**-237.2**	-137.0	**290.0**	9.3	69.49
60	53.2	-64.5	**80.5**	12.4	-24.2	-20.1	-33.6	31.57
70	10.2	-11.1	**21.7**	6.9	-9.3	-6.0	-11.2	8.43
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
δ (ω=0.1 rad/s)								
0	0.00	**-5.03**	**2.95**	-0.31	0.56	-1.49	-0.24	0.80
10	0.82	**-6.51**	**4.04**	-0.16	0.70	-1.58	0.03	1.12
20	3.03	**-6.74**	**4.85**	0.38	0.31	-1.01	-0.05	1.45
30 PP08	**6.91**	**-3.72**	2.99	2.32	-0.88	0.08	-0.12	1.60
30 PP25	0.34	**-6.53**	**4.94**	**2.89**	**3.17**	**-3.10**	**-1.93**	0.69
40	0.68	**-4.58**	**2.96**	**2.59**	**3.71**	**-3.12**	**-2.31**	0.65
50	-0.55	-1.68	-1.33	2.70	**3.05**	**-5.66**	-2.46	1.28
60	**-1.23**	**0.87**	**-0.93**	0.06	-0.51	0.14	-0.21	0.23
70	**-1.46**	-0.82	-0.80	-0.51	-0.65	-0.34	-0.28	0.60
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
δ (ω=10 rad/s)								
0	-0.34	**-2.98**	**1.62**	-0.25	0.33	-0.88	-0.42	0.49
10	0.00	**-4.27**	**2.26**	-0.19	0.54	-1.05	-0.13	0.63
20	0.43	**-4.66**	**2.64**	0.17	0.79	-0.94	0.29	0.75
30 PP08	0.42	**-3.29**	**1.75**	0.67	0.89	-0.34	0.62	0.64
30 PP25	0.11	**-3.18**	**2.42**	**1.49**	**1.89**	**-1.48**	-0.87	0.48
40	-0.06	**-1.28**	**0.75**	**0.97**	**1.13**	**-1.12**	**-0.47**	0.16
50	-0.09	**-0.38**	-0.03	0.25	**0.51**	**-0.54**	**-0.31**	0.12
60	-0.25	0.05	-0.13	-0.08	-0.04	0.04	0.03	0.11
70	-0.29	0.10	-0.27	-0.08	-0.11	0.28	**-0.43**	0.18

Table 6. 9. Effect estimates and Standard Errors (SE), for 160/220_II.

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
$G^*(\omega=0.1$ rad/s$)$								
0	**271 050**	**-63 150**	**70 500**	**82 825**	**67 425**	32 625	15 250	24 875
10	**23 889**	-4 585	4 911	5 904	**6 775**	1 564	-379	2 614
20	**2 332**	-433	445	524	**724**	85	-122	293.7
30 PP08	**263**	-56	54	59	79	10	-19	37.2
30 PP25	7	**-58**	-15	34	29	9	-38	18.5
40	1	**-7**	-2	5	3	2	**-6**	2.06
50	0.2	**-0.8**	-0.1	0.7	0.4	0.5	**-1.0**	0.33
60	-0.1	0.0	0.0	0.0	0.1	0.1	-0.2	0.07
70	0.0	0.01	-0.01	0.01	**0.06**	0.02	-0.02	0.02
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
$G^*(\omega=10$ rad/s$)$								
0	**4 456**	**-1 363 000**	**1 601 000**	**1 881 000**	679 000	864 250	642 750	380 875
10	**665 213**	**-185 663**	**207 188**	**249 913**	**147 463**	110 788	63 163	59 801
20	**93 995**	**-24 098**	**25 710**	**30 670**	**26 478**	12 663	5 243	9 181.5
30 PP08	**14 279**	**-3 981**	**3 800**	**4 468**	**4 339**	1 500	344	1 647.0
30 PP25	239	**-3 099**	-890	1 841	1 694	435	-1 775	1 169.0
40	62	**-576**	-152	360	268	100	**-421**	166.07
50	18.4	**-79.9**	-17.8	**68.9**	43.8	31.5	**-76.5**	29.15
60	0.4	-2.3	-3.1	1.6	14.5	5.6	-8.3	6.44
70	-0.8	1.4	-0.8	-0.2	**4.9**	2.9	-1.0	1.71
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
δ $(\omega=0.1$ rad/s$)$								
0	**-1.26**	-0.02	0.13	0.15	**-0.60**	0.24	0.38	0.24
10	**-1.42**	-0.04	0.12	0.15	-0.57	0.36	0.50	0.31
20	**-1.23**	-0.03	0.05	0.14	-0.42	0.33	0.51	0.30
30 PP08	**-0.86**	-0.27	-0.06	0.14	-0.16	0.32	0.47	0.30
30 PP25	-0.03	**0.16**	-0.01	**-0.06**	-0.03	-0.06	**0.18**	0.02
40	0.01	0.00	-0.03	0.04	**0.07**	**-0.09**	**0.10**	0.02
50	-0.02	0.03	-0.01	0.12	0.17	-0.14	0.04	0.08
60	0.02	-0.17	-0.23	0.14	0.14	-0.27	0.11	0.13
70	-0.18	0.17	-0.25	-0.03	0.26	-0.22	0.12	0.17
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
δ $(\omega=10$ rad/s$)$								
0	**-0.50**	0.02	0.06	0.07	**-0.42**	0.06	0.14	0.09
10	**-0.77**	0.01	0.09	0.12	**-0.52**	0.13	0.26	0.15
20	**-0.88**	0.01	0.09	0.12	**-0.49**	0.19	0.32	0.19
30 PP08	**-0.87**	0.09	0.03	0.03	-0.41	0.18	0.31	0.22
30 PP25	-0.09	**0.33**	0.05	**-0.18**	**-0.11**	-0.05	**0.25**	0.04
40	-0.05	**0.14**	0.01	**-0.10**	**-0.08**	-0.05	**0.18**	0.04
50	-0.03	0.01	-0.01	-0.03	-0.05	-0.02	**0.12**	0.03
60	-0.06	0.03	0.07	0.09	-0.08	-0.08	-0.09	0.07
70	0.02	-0.14	-0.29	-0.01	0.13	-0.38	0.00	0.21

6.4 Conclusions of study II

In this study, a 2^3 experimental design matrix is carried out to evaluate the effect of pre-heating temperature for manufacturing specimen (HT), bonding temperature onto rheometer (BT), and trimming state on rheological properties of bitumen when applying Dynamic shear rheometer (DSR). Verifications of the observed effects on complex shear modulus |G*| and phase angle (δ) were performed with two replication at various temperatures between 0 °C and 80 °C at two frequencies of 0.1 and 10 rad/s (1.59Hz). The tests were executed for four neat bitumen of type 50/70, 70/100, and 160/220 from two sources according to the European standard method EN14770 (2012). It is observed that the complex modulus is more sensitive to the changes than the phase angle. However, the almost same pattern in both lower and higher tested frequencies is observed. The bonding temperature is the most obvious factor affecting the G*, and the δ followed by oven heating temperature contrariwise for the 160/220_I. The variation in oven heating temperature strongly affected the results of the 160/220_I contrary to all other studied materials. Although not shown in this study, the master curve of this bitumen showed a weak overlapping on shifted temperature, which may be a hint that it had a different natural chemical component. Trimming the sample tested at lower temperatures of 0 °C to 30 °C for all tested material tends to significantly increase the δ and decrees the complex modulus, which may indicate higher sensitivity of smaller parallel plate (PP08) to trimming. The BT had a negative impact on G* at almost all of the temperatures evaluated for 50/70 and 160/220_II.

In 2-way interaction, in the case of G*, Trim:BT has the strongest effect on all material and temperatures except for 160/220 at lower temperatures where Trim-HT affects more. For δ, the most effective two-way interaction was Trim-BT for 70/100 and 160/220_II. However, for 160/220_I the Trim:HT, and for 50/70 the BT:HT is the least important factor for δ. Except for 160220_I, HT do not generally have a statistically significant impact on the results; but, when it interacted with other parameters, it had a substantial impact. This may be attributed to the fact that the range of oven heating temperature (SP+80 and SP+100°C) in this study is very small. The study showed that G* and δ have affected by studied factors, least for 70/100, which is followed by 50/70 and 160/220.

7 Discussion and Conclusions

In this section, the main conclusions of this work, carried out in form of study I and II, as well as recommendations for further studies, are made.

The precision analysis was carried out under the first part of study, on data from round-robin tests at frequency of 10 rad/s at temperatures ranging from 10 °C to 65 °C on short term aged bitumen type of 50/70, 20/3, and 45/80-55 between 2017 to 2019, and in 2020 on an original bitumen 45/80-55 without ageing. The practices used by labs differed noticeably, emphasizing the importance of having a well-defined protocol for interpreting EN14770:2012 for determining G* and δ.The coefficient of variation under repeatability are almost twice better than reproducibility values for all the tested materials. However, precision values differ across the material type and temperature. After ignoring the extreme values due to the use of inappropriate plate geometry or the small number of laboratories involved the accuracy range for |G*| improves to a range of COV_r = of 2-8%, and COV_R = 7 and 18%, while for δ, a repeatability limit of r =1-2° and reproducibility limit of R=3-6° are observed. The lower producibility compared to repeatability could be due to laboratory differences. However, a groupwise analysis of three categories of equipment brand (Anton Paar, Malvern, and other) and sample manufacturing method (mould, sheet, and other) revealed that no significant variations were found between the two most often used brands of equipment and sample manufacturing procedures were used by laboratories. Furthermore, the statistical analyses were performed to assess the differences in outcomes by waiting time of samples before start testing categories (short: <=2h (in case of neat bitumen) and <=12h (in case of PMB), medium: 2< h <72, and long: >=72h). The long waiting time had a higher G* and a lower δ value than the short waiting time. However, this was significant in only few test conditions. It observed that all the bitumen studied, namely the neat and PMB, are appropriate for having a waiting period of less than 2 h and an upper limit greater than 72h without a significant impact on outcomes. In comparison to PP25 at temperatures ranging from 40 to 65 °C, PP08 at temperatures ranging from 10 to 35 °C demonstrated a lower average G* for applied equilibrium durations of 5 to 15 minutes compared to 15 to 30 minutes. However, statistically significant variations between different timespans were detected only for unaged 45/80-55 at 40 °C. On the other hand, in most test conditions, the value of δ increases along with the equilibrium time. 15 minutes appears to be an appropriate equilibrium duration, with results typically falling between the upper and lower recorded values. Finally, the correlation test based on RR- data, the bonding temperature, and sample production temperature exhibited a significant link in more test combinations than in other sample preparation phases. Therefore, in the second part of the study, the investigation on effect of these factors together with trimming carried out at frequencies of 0.1 rad/s and 10 rad/s, and at temperature range between 0 °C to 80 °C for 50/70 and 70/100, and between 0 °C to 70 °C for the softer bitumen 160/220 from two resources. The results showed a significant difference in trimmed and untrimmed samples when PP08 is applied. The bonding and oven heating temperatures take on varying degrees of significance depending on the materials and tested temperatures. In contrast to all other investigated materials, the variation in oven heating temperature had a significant impact on the results of the 160/220_I, which could indicate that it may has a different natural chemical composition. The G* and δ tends to change in the opposite direction with sample manufacturing temperature, for all tested material except for short-term aged 50/70 in study I. However, drawing a general conclusion and comparison on oven heating between two study was difficult due to only one unaged bitumen (45/55-80) in study I. At the same test condition, in terms of frequency and the temperature tested in relation to the bitumen softening point, comparisons indicate similar results for the 45/55-80 and 160/220_I; the G* drops with oven heating temperature while δ increases. The bonding temperature had a negative impact on G* at practically all the evaluated temperatures for 50/70, and 160/220_II, which is consistent with earlier studies on two types of short-term aged bitumen, 20/30 and 45/55-80. However, unaged 45/55-80 in the prior study and 160/220_II in second one exhibits a comparable positive relationship between G* and bonding temperature.

Future research is advised to assess the effects of various factors on modified bitumen with a wider range at both high and low levels of the factors used in the presented study to capture the limit which leads to significant differences.

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APPENDIX 1. The coefficient of variation of the effect estimates and SE.

Table 1. The coefficient of variation (CV) of the effect estimates and standard errors (SE) for 50/70.

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=0.1 rad/s)								
0	**0.14**	**-0.08**	0.03	0.01	-0.04	0.03	0.03	0.03
10	**0.16**	**-0.08**	0.04	0.03	-0.03	0.03	0.02	0.03
20	**0.16**	-0.07	0.06	0.05	-0.02	0.02	0.01	0.03
30 PP08	**0.15**	-0.07	**0.08**	0.06	-0.01	0.01	-0.02	0.03
30 PP25	-0.04	0.04	0.06	0.04	0.06	0.00	-0.01	0.03
40	-0.03	**0.09**	0.04	0.02	0.04	0.00	-0.01	0.03
50	-0.01	**0.07**	0.04	0.02	0.03	0.00	0.00	0.02
60	-0.02	-0.04	**0.08**	0.01	0.04	-0.01	-0.02	0.02
70	-0.02	0.02	**0.06**	-0.01	0.01	0.00	-0.03	0.02
80	-0.04	0.03	0.04	-0.02	-0.01	0.02	-0.03	0.03
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=10 rad/s)								
0	**0.13**	**-0.09**	0.03	0.01	-0.04	0.03	0.02	0.02
10	**0.13**	**-0.08**	0.03	0.02	-0.04	0.03	0.02	0.02
20	**0.14**	**-0.07**	0.04	0.03	-0.03	0.02	0.02	0.02
30 PP08	**0.13**	**-0.07**	**0.06**	0.04	-0.03	0.01	-0.01	0.02
30 PP25	-0.03	0.04	0.05	0.04	0.04	0.00	0.00	0.02
40	-0.02	**0.07**	0.05	0.03	0.03	0.00	0.00	0.02
50	-0.01	**0.06**	0.04	0.03	0.02	0.00	0.00	0.02
60	-0.02	-0.03	**0.07**	0.02	0.04	-0.01	-0.02	0.02
70	-0.02	0.02	**0.05**	0.00	0.02	-0.01	-0.03	0.02
80	**-0.02**	0.02	**0.03**	0.00	0.02	-0.01	**-0.03**	0.01

Table 2. The coefficient of variation (CV) of effect estimates and standard errors (SE) for 70/100.

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G* (ω=0.1 rad/s)								
0	0.05	0.02	-0.06	-0.03	0.08	0.02	0.01	0.04
10	0.06	0.02	-0.06	-0.03	0.08	0.02	0.01	0.05
20	0.07	0.02	-0.06	-0.03	0.08	0.03	0.01	0.05
30 PP08	0.07	0.01	-0.06	-0.03	0.08	0.03	0.00	0.06
30 PP25	0.01	-0.04	0.03	0.02	0.03	0.01	0.00	0.02
40	0.02	-0.02	0.03	0.02	0.02	0.00	-0.01	0.02
50	0.01	-0.01	0.02	0.02	0.02	0.00	0.00	0.02
60	**0.03**	0.00	-0.01	0.01	0.02	0.02	0.00	0.01
70	**0.02**	0.01	-0.01	0.01	**0.03**	0.01	-0.01	0.01
80	0.01	0.00	**-0.02**	**0.02**	0.02	0.01	0.01	0.01
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G* (ω=10 rad/s)								
0	0.03	0.02	-0.06	-0.03	0.06	0.02	0.02	0.04
10	0.04	0.02	-0.06	-0.03	0.07	0.02	0.02	0.04
20	0.04	0.02	-0.06	-0.03	0.07	0.02	0.02	0.04
30 PP08	0.04	0.01	-0.07	-0.04	0.08	0.03	0.02	0.05
30 PP25	0.00	-0.02	0.03	0.02	0.02	0.00	0.00	0.01
40	0.01	-0.02	0.03	0.02	0.02	0.00	-0.01	0.01
50	0.01	-0.01	0.03	0.02	0.01	0.00	0.00	0.01
60	**0.03**	0.00	0.00	0.01	0.02	0.02	0.00	0.01
70	**0.02**	0.01	0.00	0.01	**0.03**	0.01	-0.01	0.01
80	0.01	0.00	0.00	0.01	**0.02**	0.02	0.00	0.01

Table 3. The coefficient of variation (CV) of effect estimates and standard errors (SE) for 160/220_I.

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=0.1 rad/s)								
0	**0.19**	**0.26**	**-0.22**	-0.03	-0.03	0.08	0.10	0.05
10	0.14	**0.44**	**-0.31**	-0.03	-0.05	0.06	0.09	0.07
20	0.06	**0.55**	**-0.41**	-0.06	-0.11	0.01	0.06	0.10
30 PP08	0.04	**0.44**	**-0.35**	-0.17	-0.12	0.01	-0.02	0.11
30 PP25	0.03	**0.42**	**-0.31**	**-0.26**	**-0.25**	**0.21**	0.08	0.06
40	0.02	**0.23**	**-0.14**	**-0.22**	**-0.19**	**0.23**	**0.06**	0.02
50	0.02	0.04	0.03	**-0.14**	**-0.13**	**0.20**	0.05	0.03
60	**0.12**	-0.12	**0.16**	0.03	-0.03	-0.04	-0.05	0.05
70	0.08	-0.07	**0.13**	0.04	-0.04	-0.04	-0.04	0.04
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G*(ω=10 rad/s)								
0	**0.18**	0.07	**-0.12**	-0.05	-0.02	0.05	0.07	0.04
10	**0.17**	**0.19**	**-0.17**	-0.04	-0.03	0.06	0.08	0.04
20	**0.14**	**0.27**	**-0.22**	-0.06	-0.05	0.05	0.06	0.05
30 PP08	0.14	**0.24**	**-0.22**	-0.13	-0.07	0.05	0.02	0.06
30 PP25	0.02	**0.25**	**-0.18**	**-0.14**	**-0.11**	**0.13**	0.05	0.04
40	0.02	**0.13**	**-0.09**	**-0.13**	**-0.09**	**0.14**	0.02	0.02
50	0.01	0.01	0.01	**-0.08**	-0.05	**0.10**	0.00	0.02
60	0.08	-0.10	**0.12**	0.02	-0.04	-0.03	-0.05	0.05
70	0.05	-0.05	**0.10**	0.03	-0.04	-0.03	-0.05	0.04

Table 4. The coefficient of variation (CV) of effect estimates and standard errors (SE) for 160/220_II.

Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G* (ω=0.1 rad/s)								
0	**0.19**	**-0.04**	**0.05**	**0.06**	**0.05**	0.02	0.01	0.02
10	**0.22**	-0.04	0.05	0.05	**0.06**	0.01	0.00	0.02
20	**0.23**	-0.04	0.04	0.05	**0.07**	0.01	-0.01	0.03
30 PP08	**0.22**	-0.05	0.05	0.05	0.07	0.01	-0.02	0.03
30 PP25	0.01	**-0.06**	-0.01	0.03	0.03	0.01	-0.04	0.02
40	0.01	**-0.05**	-0.01	0.03	0.02	0.01	**-0.04**	0.01
50	0.01	**-0.02**	0.00	0.02	0.01	0.01	**-0.03**	0.01
60	-0.01	0.00	0.00	0.00	0.01	0.01	-0.02	0.01
70	0.00	0.00	0.00	0.00	**0.02**	0.01	-0.01	0.01
Temp. (°C)	Trim	BT	HT	Trim:HT	Trim:BT	BT:HT	Trim:BT:HT	SE (effect)
G* (ω=10 rad/s)								
0	**0.15**	**-0.05**	**0.05**	**0.06**	0.02	0.03	0.02	0.01
10	**0.16**	**-0.05**	**0.05**	**0.06**	**0.04**	0.03	0.02	0.01
20	**0.18**	**-0.05**	**0.05**	**0.06**	**0.05**	0.02	0.01	0.02
30 PP08	**0.18**	**-0.05**	**0.05**	**0.06**	**0.05**	0.02	0.00	0.02
30 PP25	0.00	**-0.04**	-0.01	0.03	0.02	0.01	-0.02	0.02
40	0.00	**-0.04**	-0.01	0.03	0.02	0.01	**-0.03**	0.01
50	0.01	**-0.03**	-0.01	**0.02**	0.01	0.01	**-0.03**	0.01
60	0.00	0.00	0.00	0.00	0.02	0.01	-0.01	0.01
70	0.00	0.01	0.00	0.00	**0.02**	0.01	0.00	0.01

APPENDIX 2. Interaction effect between two variables.

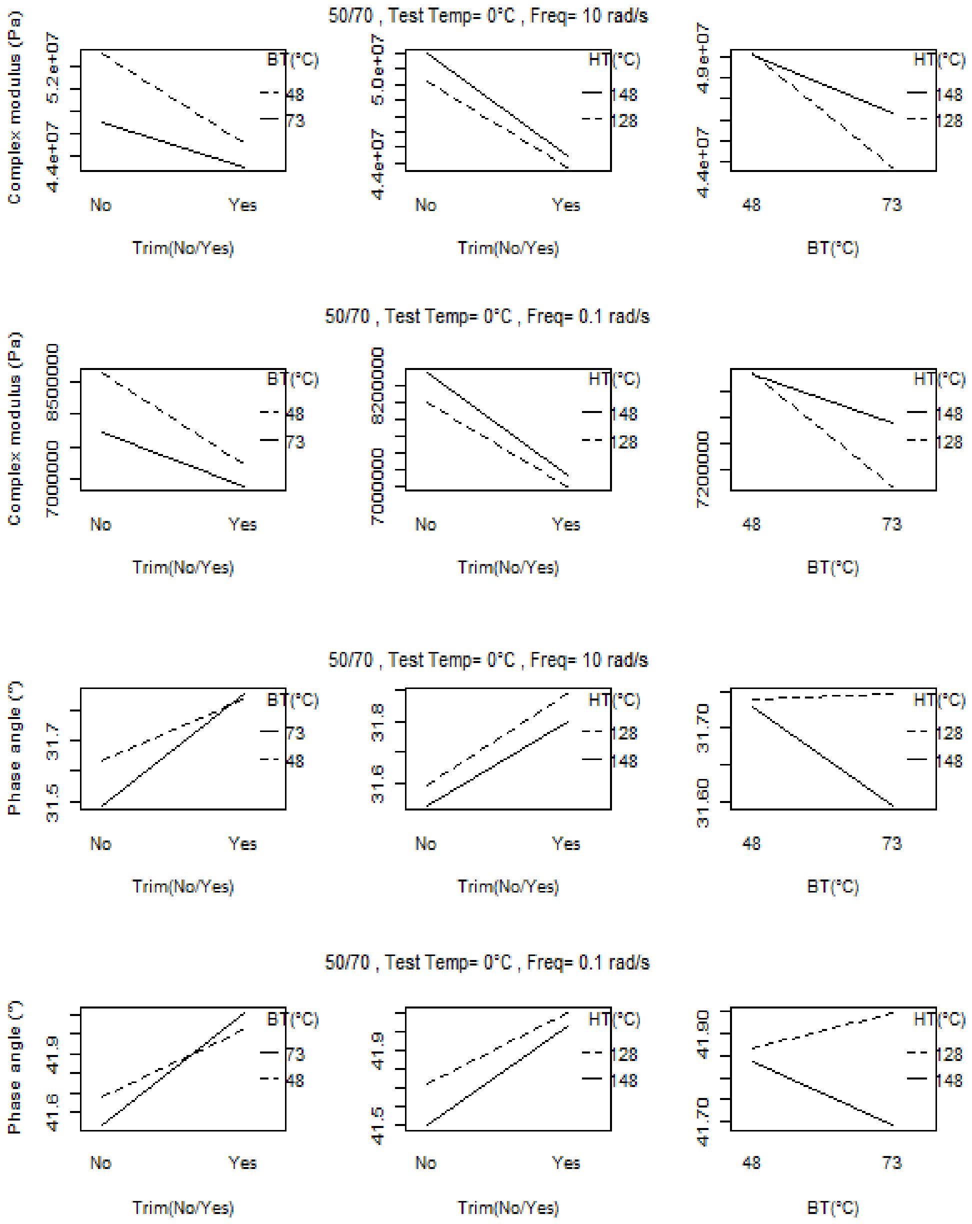


Figure 1. Interaction effect between two variables at different frequencies.

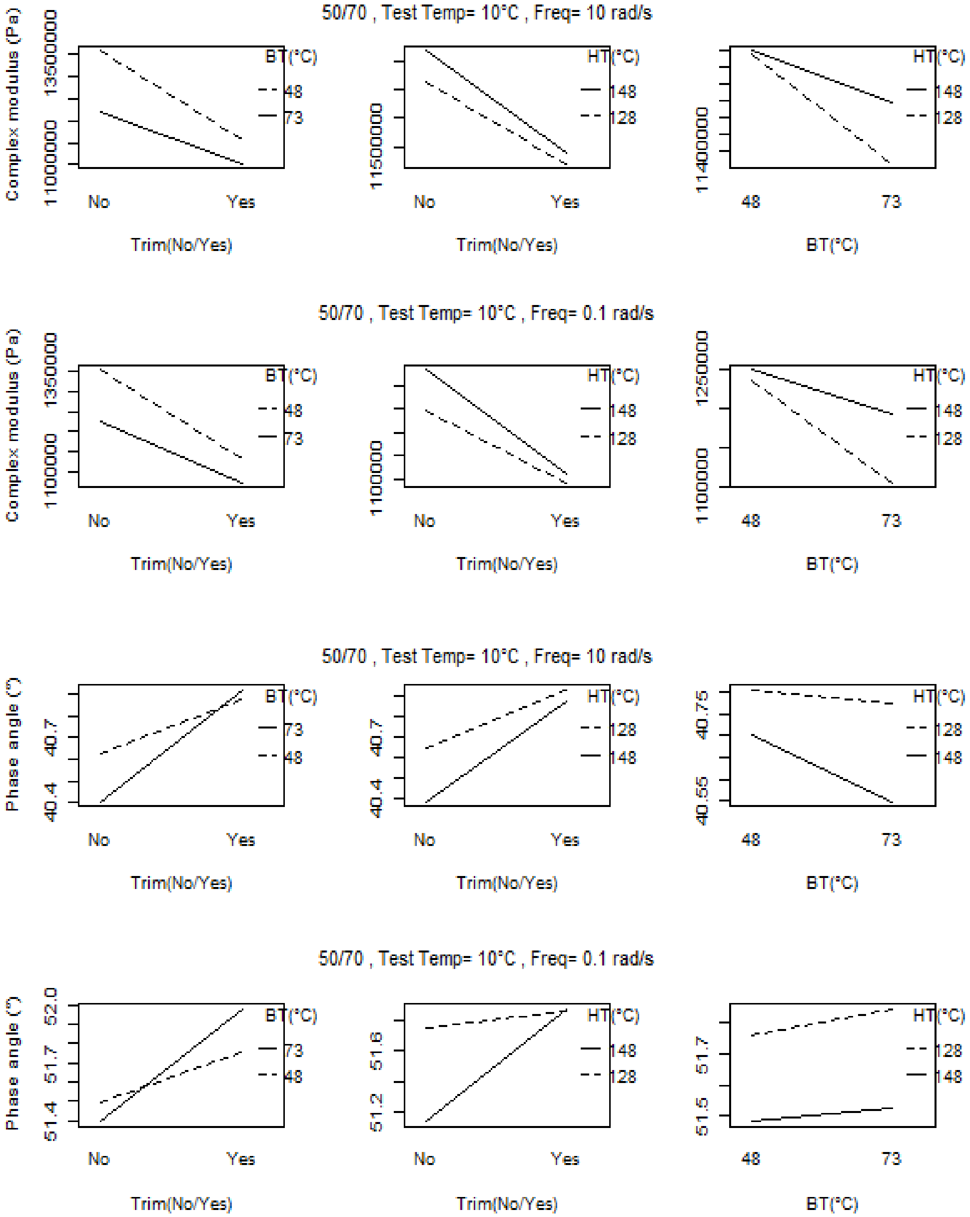


Figure 2. Interaction effect between two variables at different frequencies.

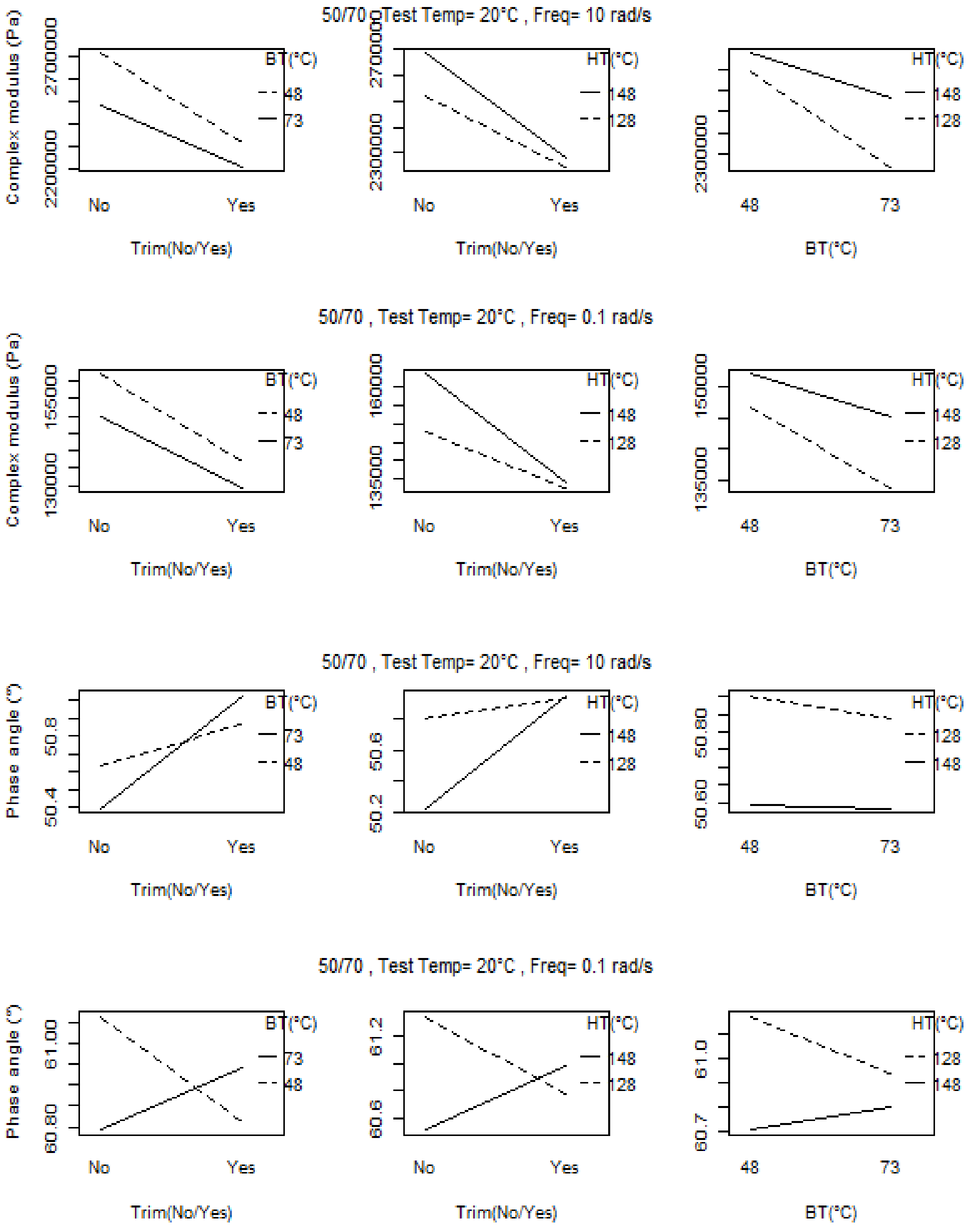


Figure 3. Interaction effect between two variables at different frequencies.

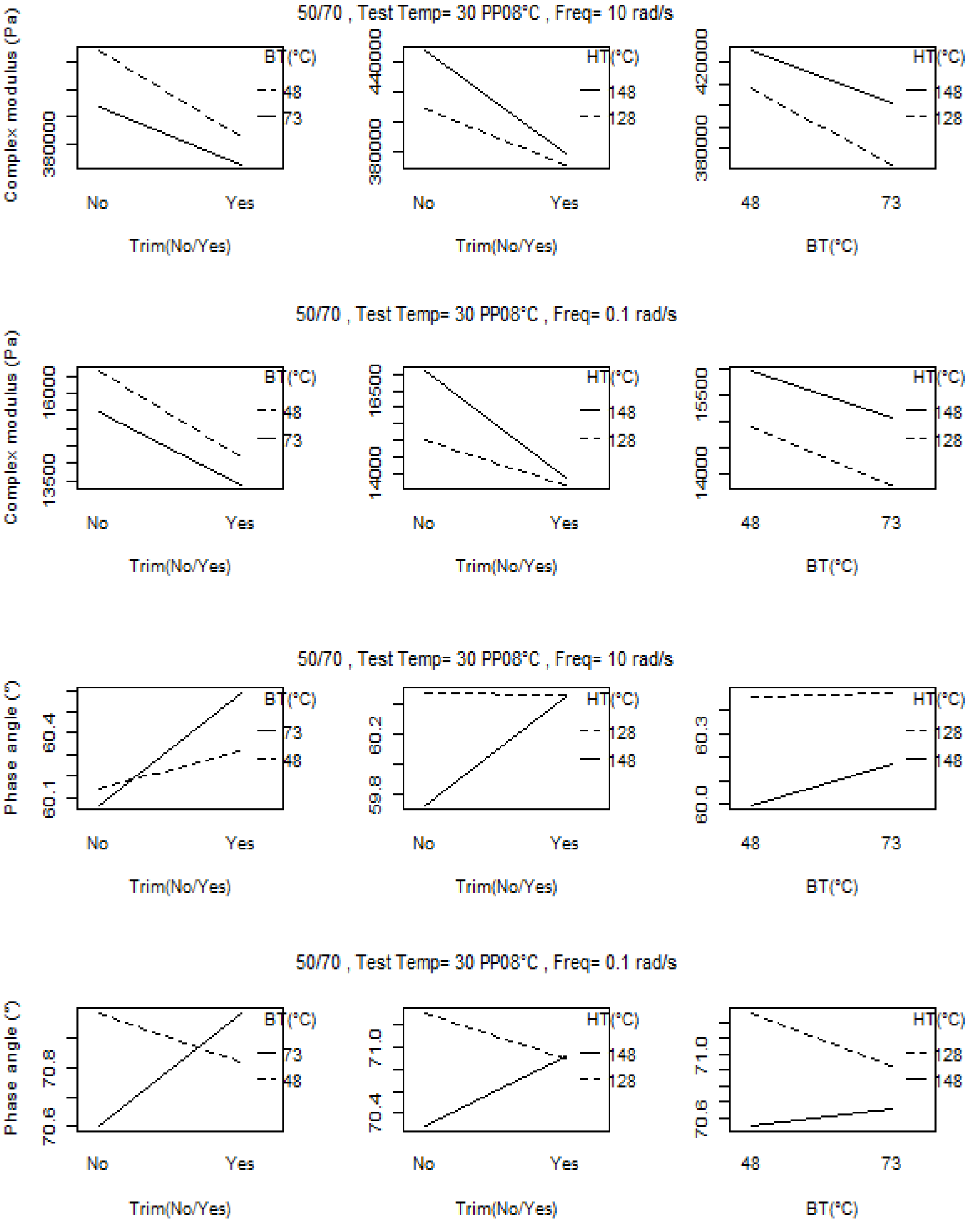


Figure 4. Interaction effect between two variables at different frequencies.

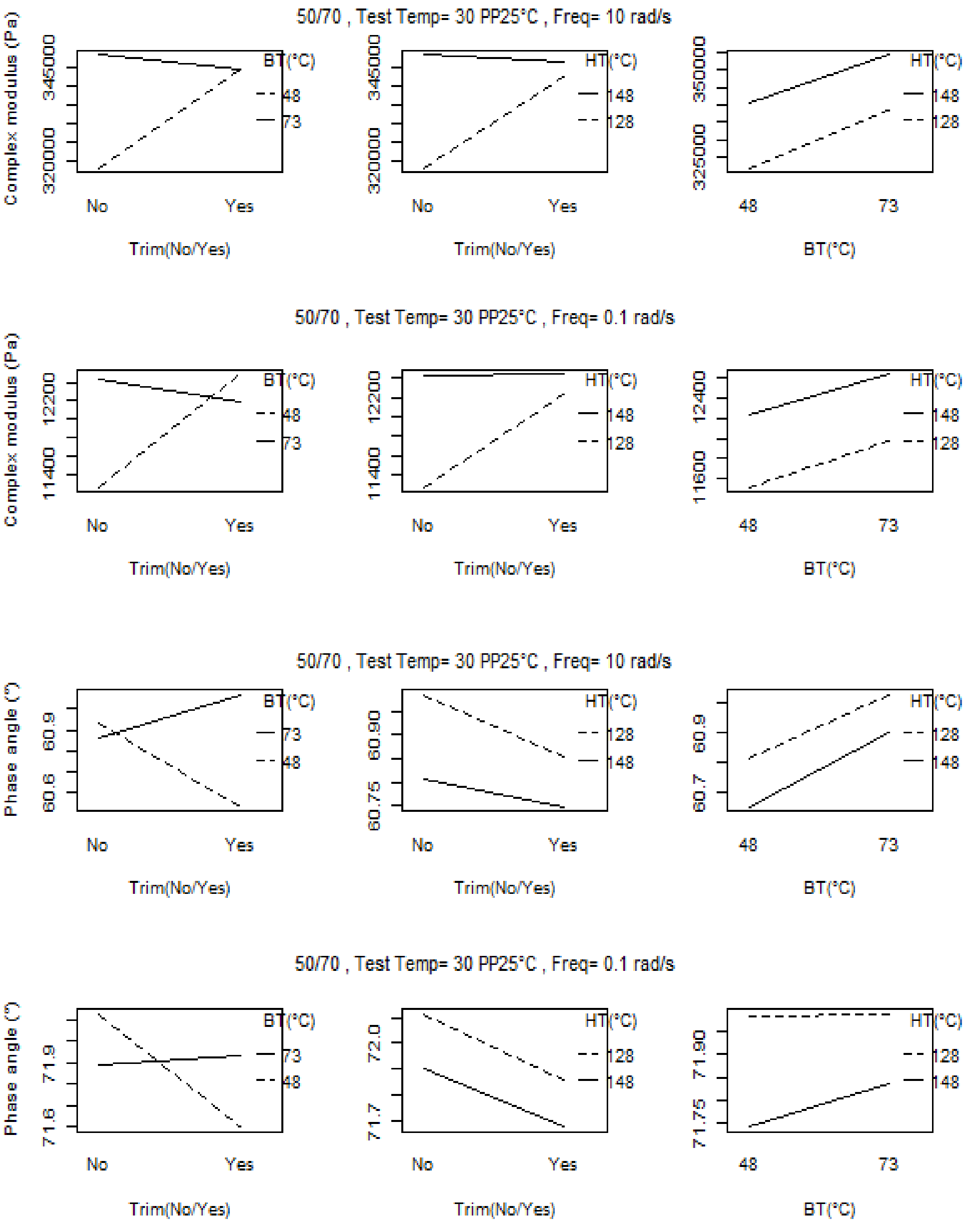


Figure 5. Interaction effect between two variables at different frequencies.

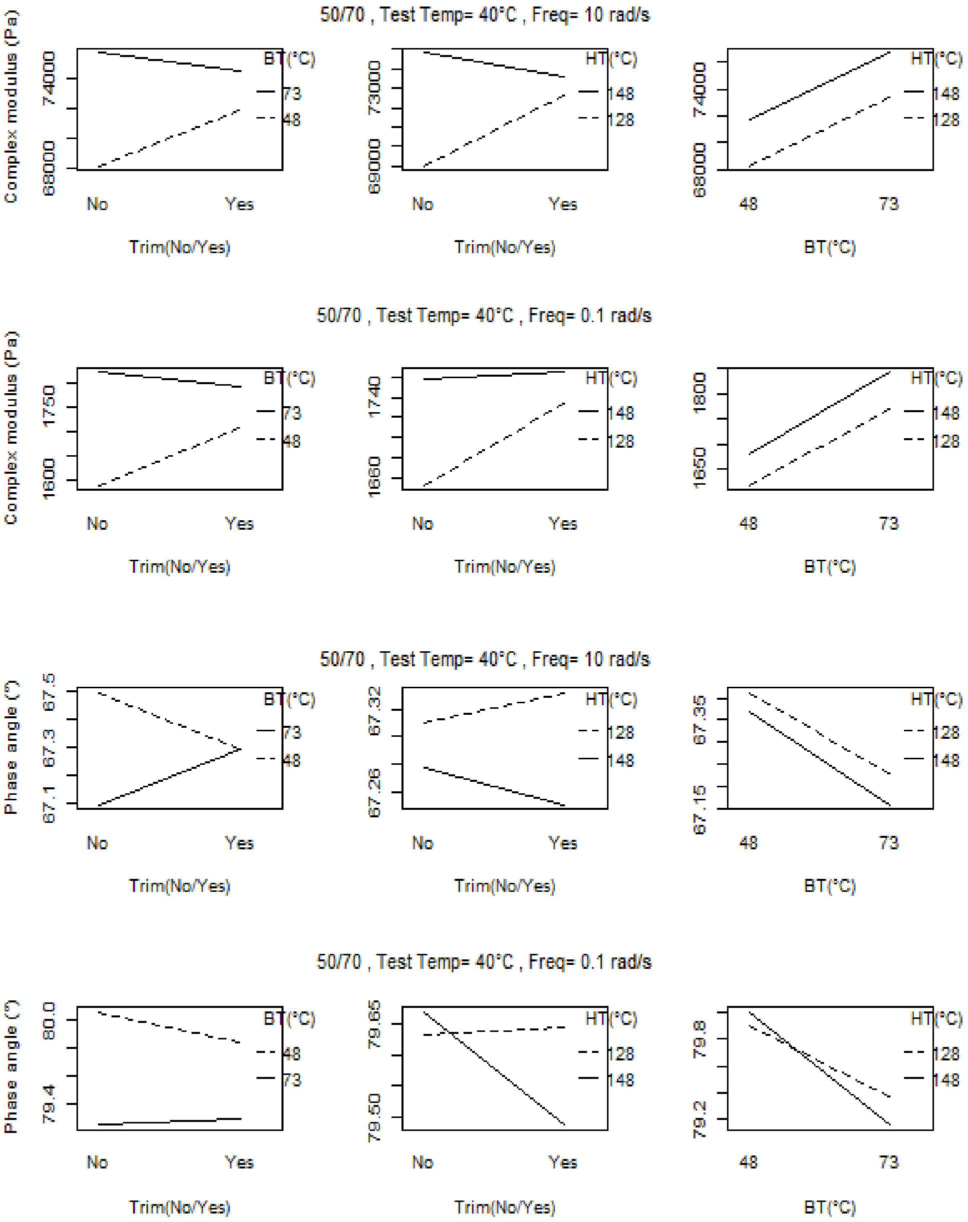


Figure 6. Interaction effect between two variables at different frequencies.

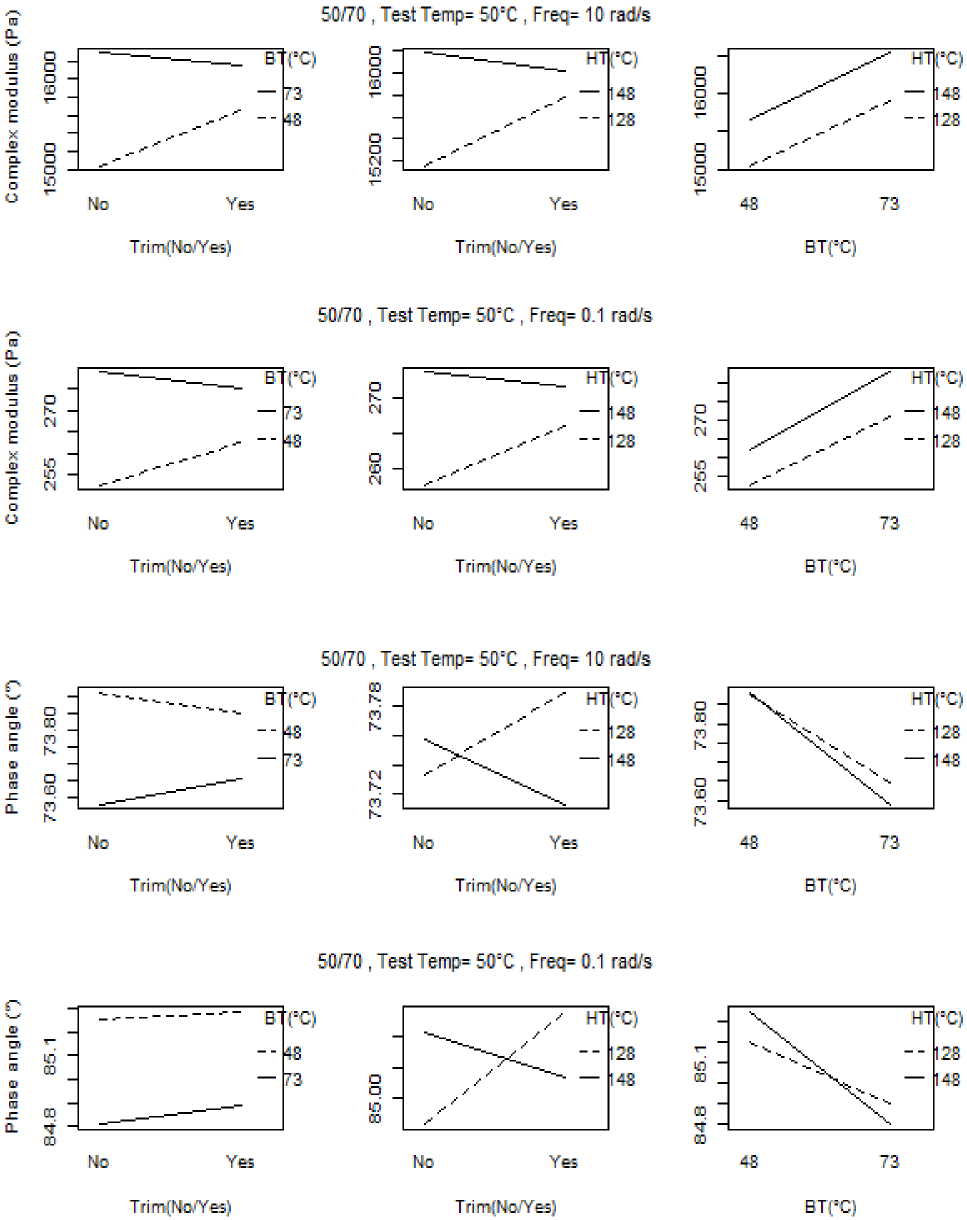


Figure 7. Interaction effect between two variables at different frequencies.

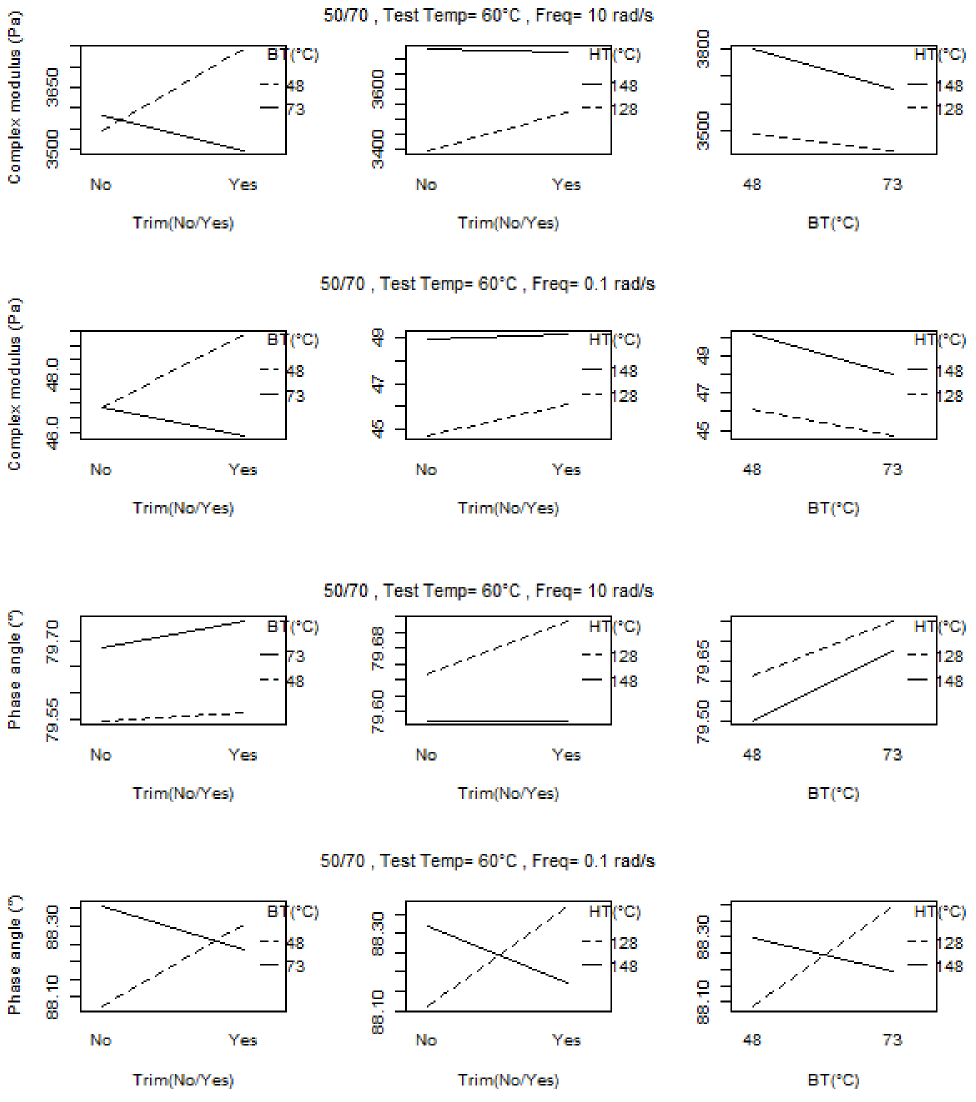


Figure 8. Interaction effect between two variables at different frequencies.

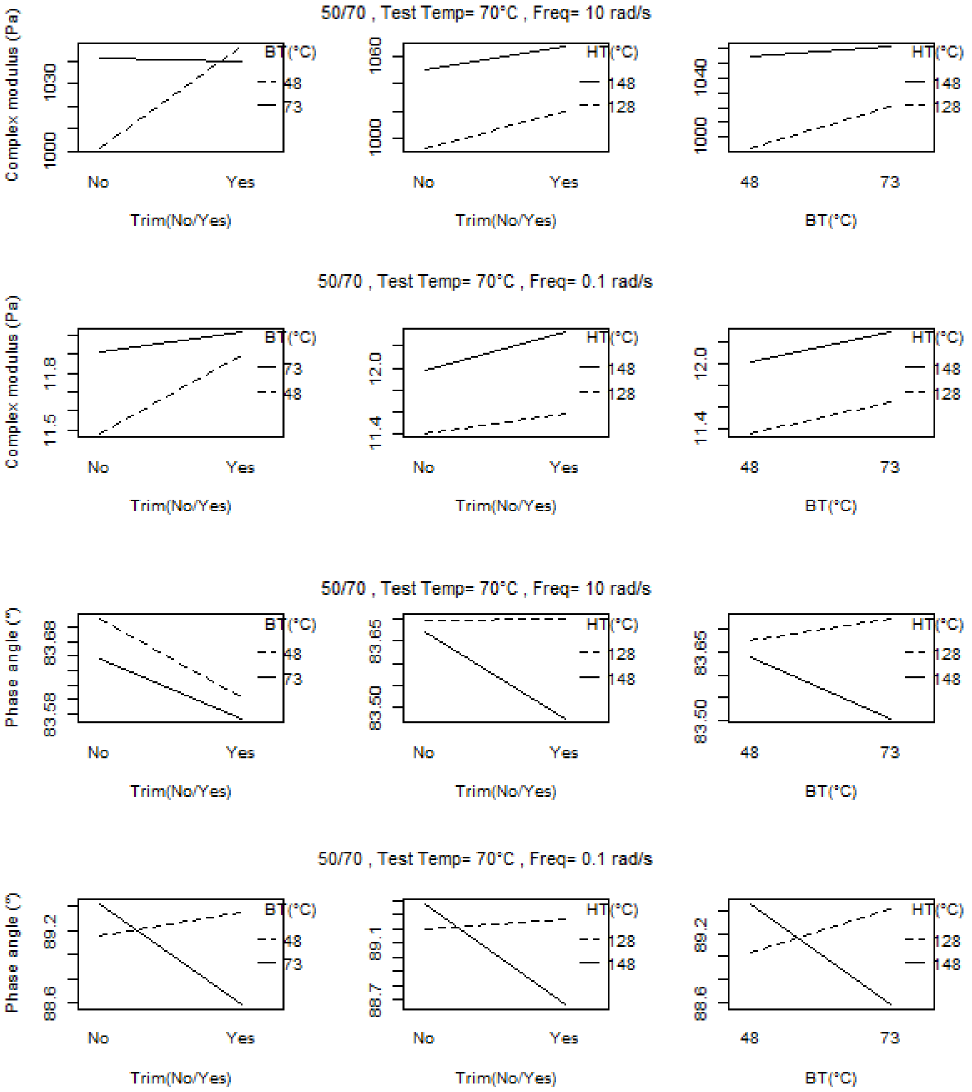


Figure 9. Interaction effect between two variables at different frequencies.

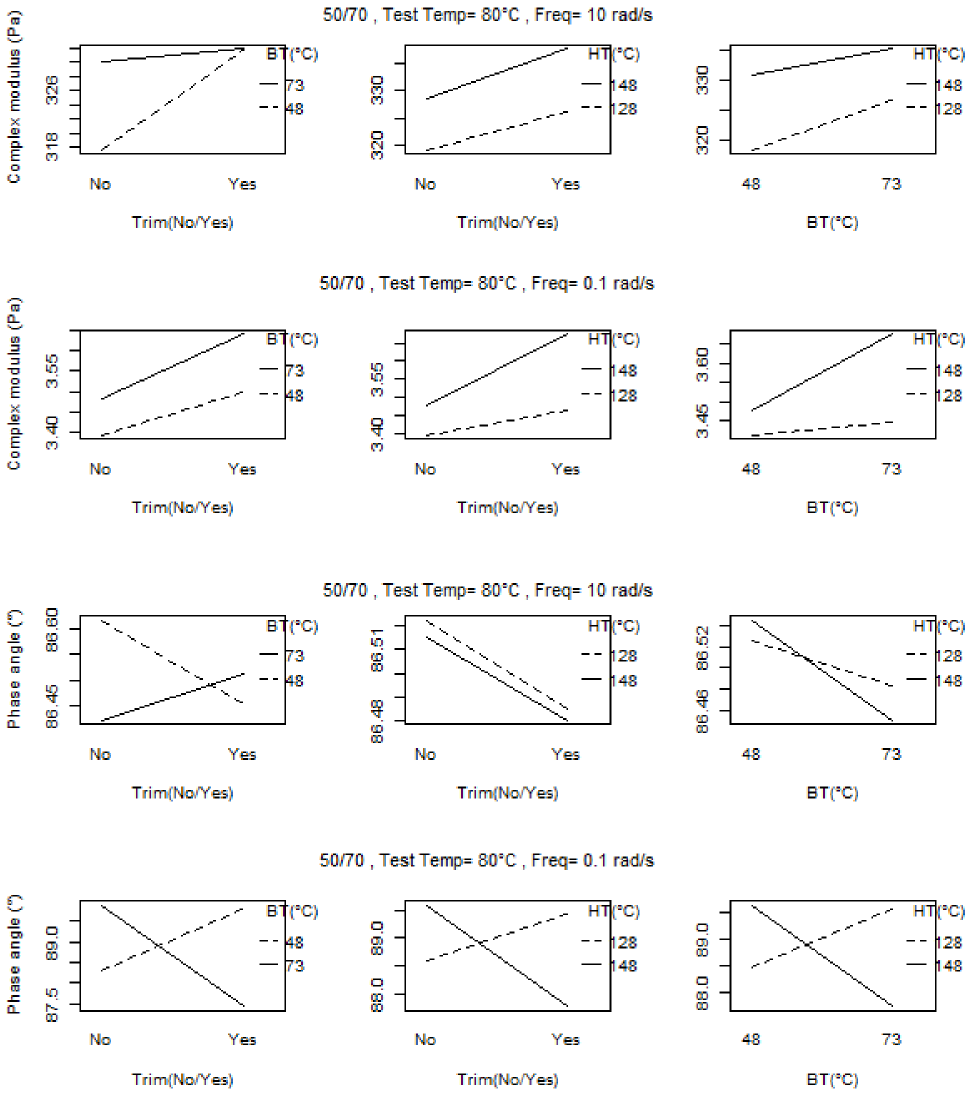


Figure 10. Interaction effect between two variables at different frequencies.

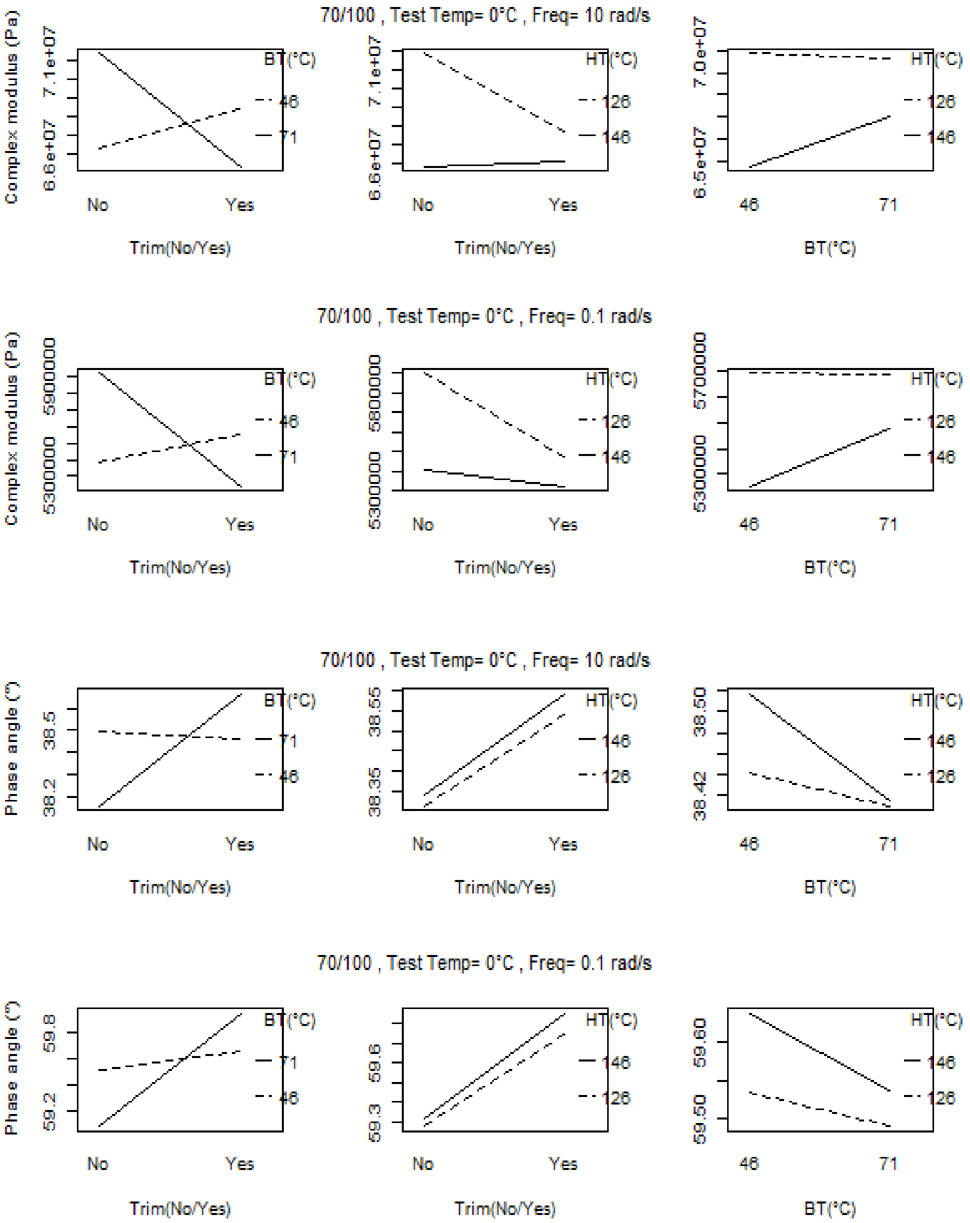


Figure 11. Interaction effect between two variables at different frequencies.

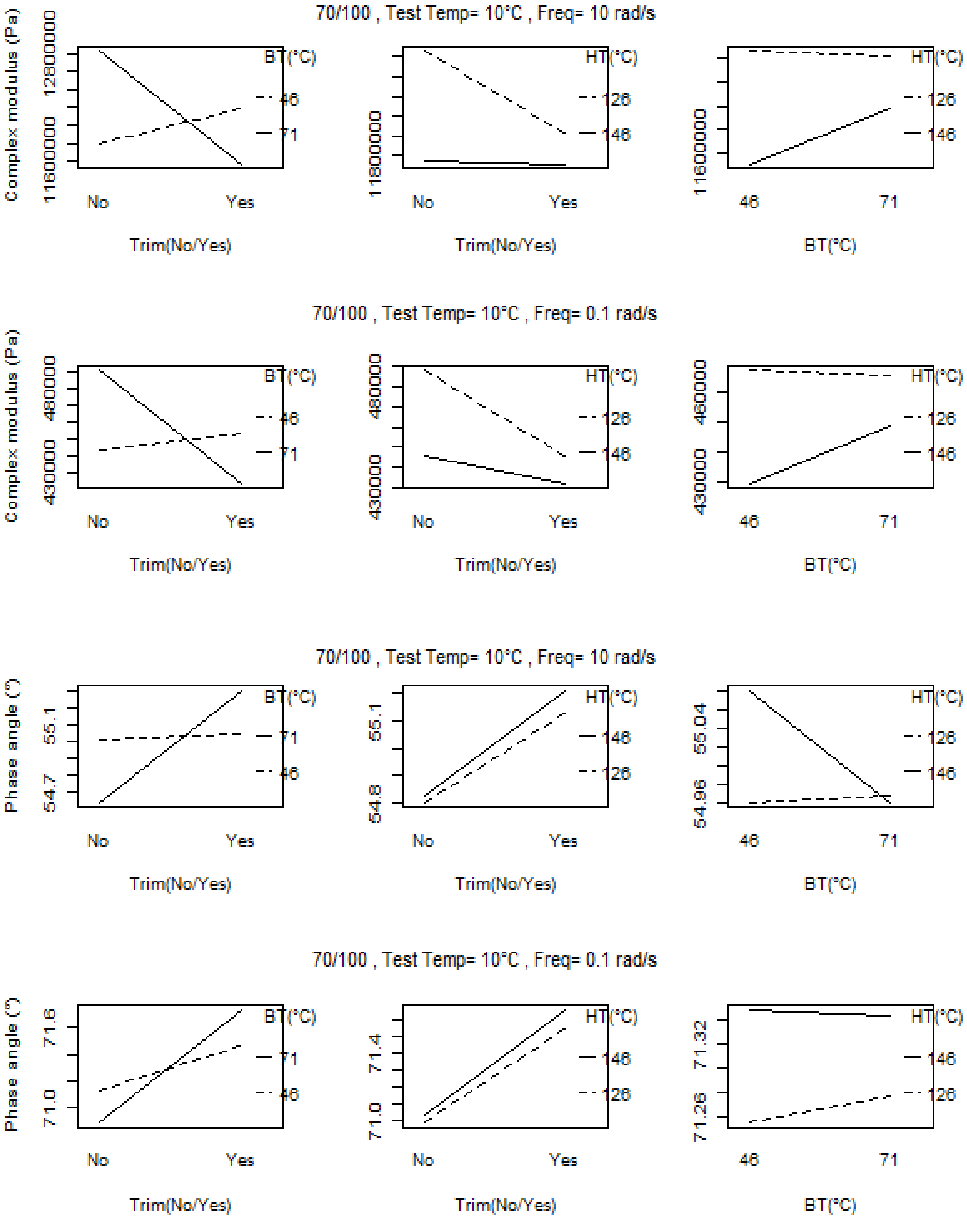


Figure 12. Interaction effect between two variables at different frequencies.

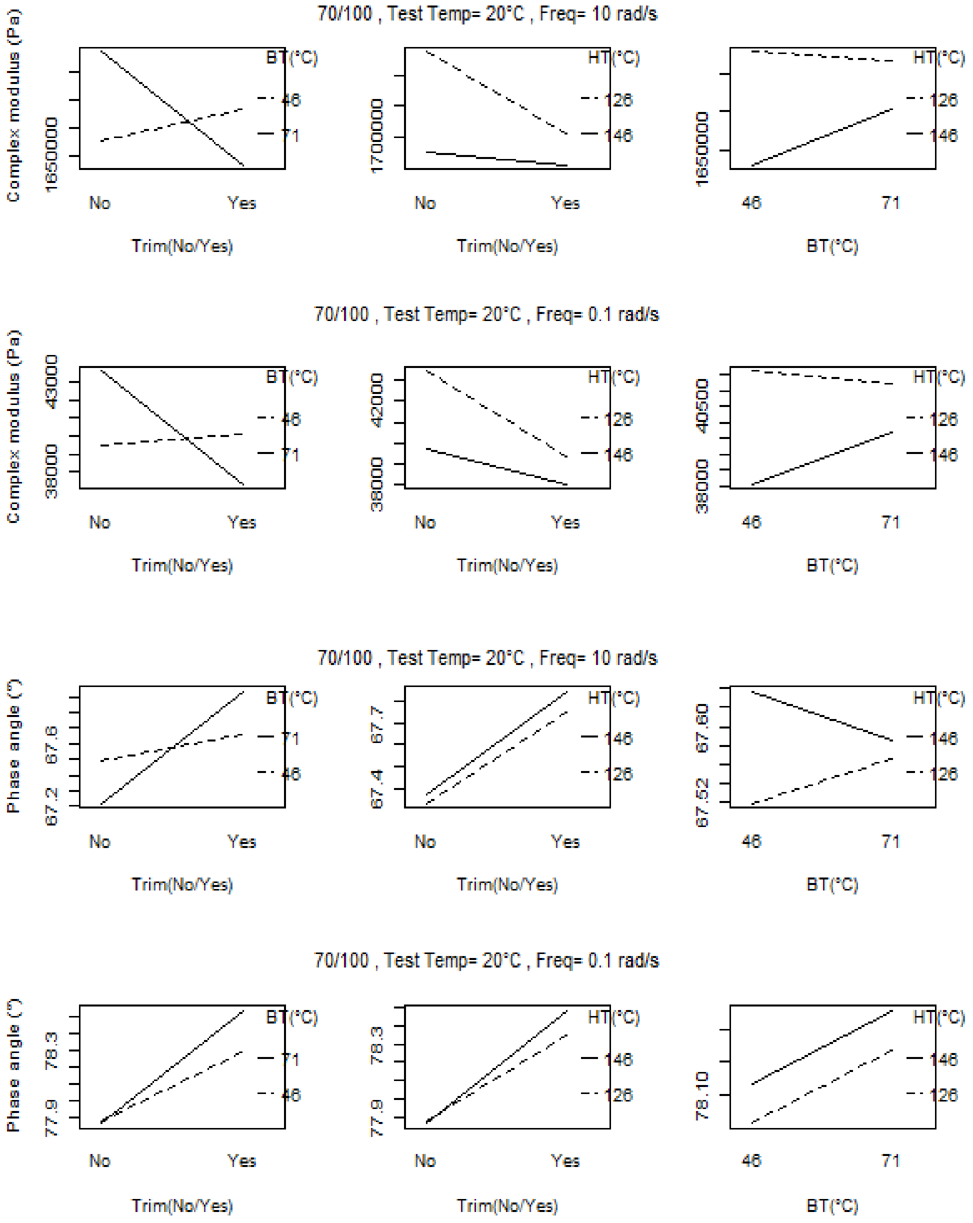
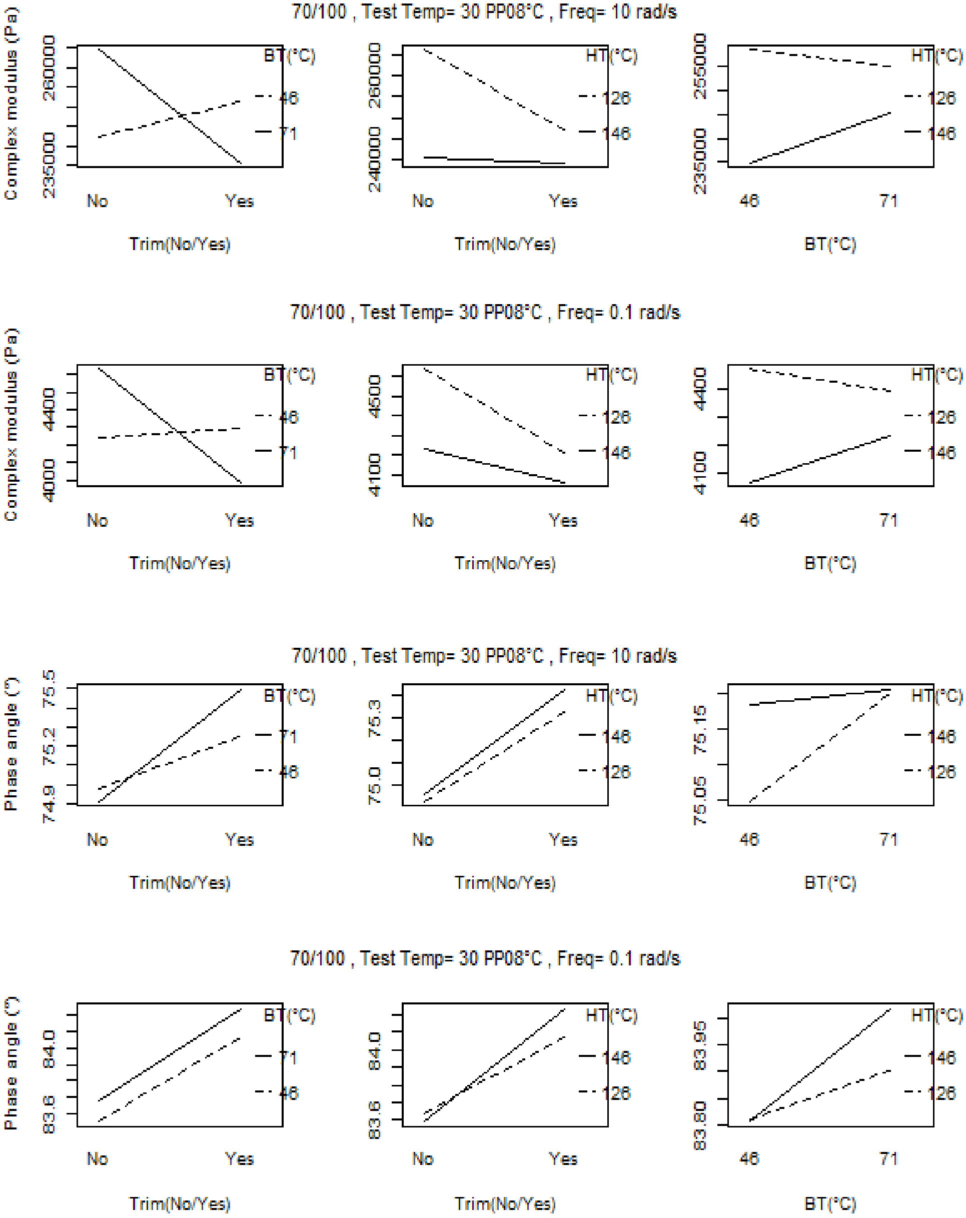


Figure 13. Interaction effect between two variables at different frequencies.


70/100 , Test Temp= 30 PP08°C , Freq= 10 rad/s

70/100 , Test Temp= 30 PP08°C , Freq= 0.1 rad/s

70/100 , Test Temp= 30 PP08°C , Freq= 10 rad/s

70/100 , Test Temp= 30 PP08°C , Freq= 0.1 rad/s

Figure 14. Interaction effect between two variables at different frequencies.

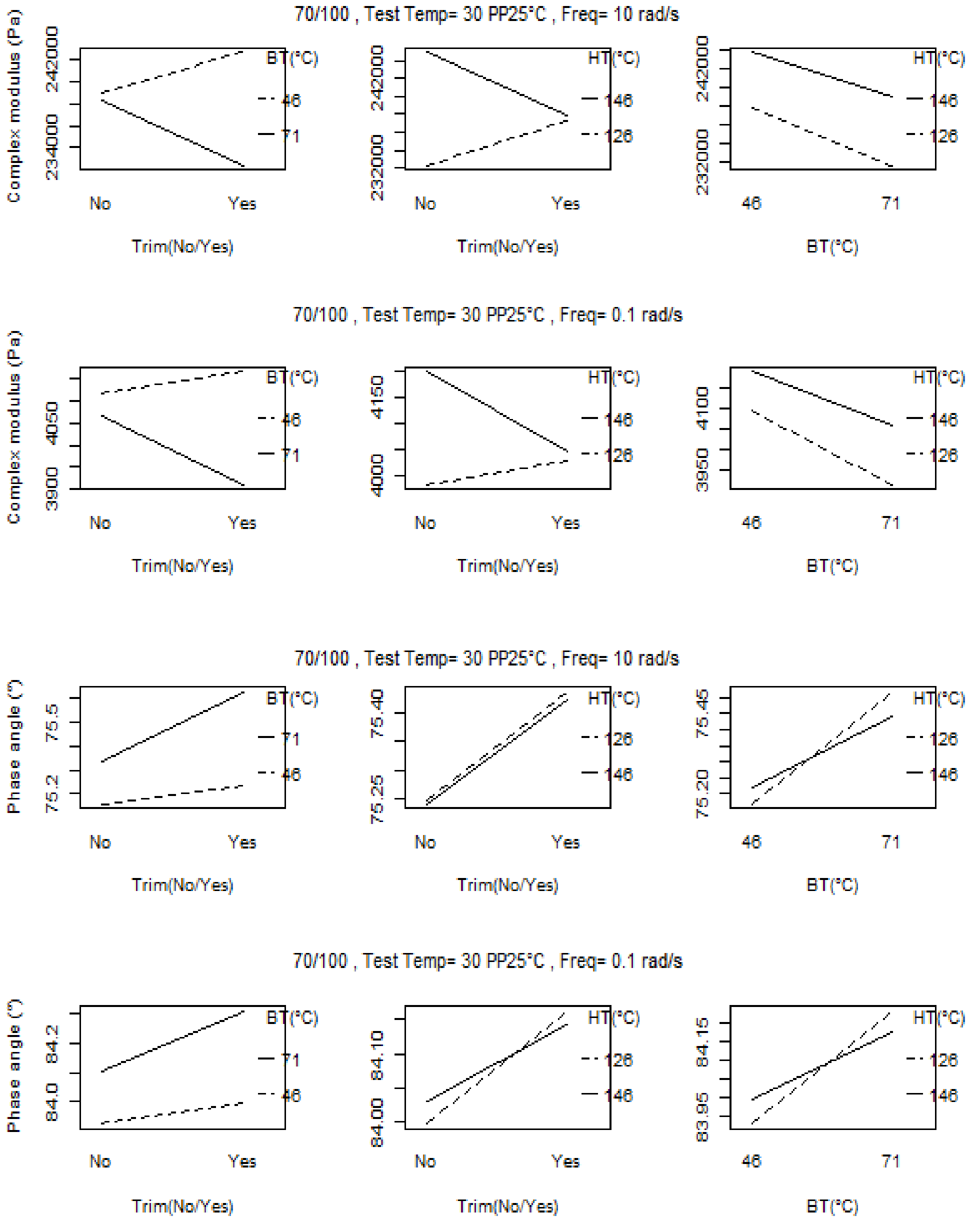


Figure 15. Interaction effect between two variables at different frequencies.

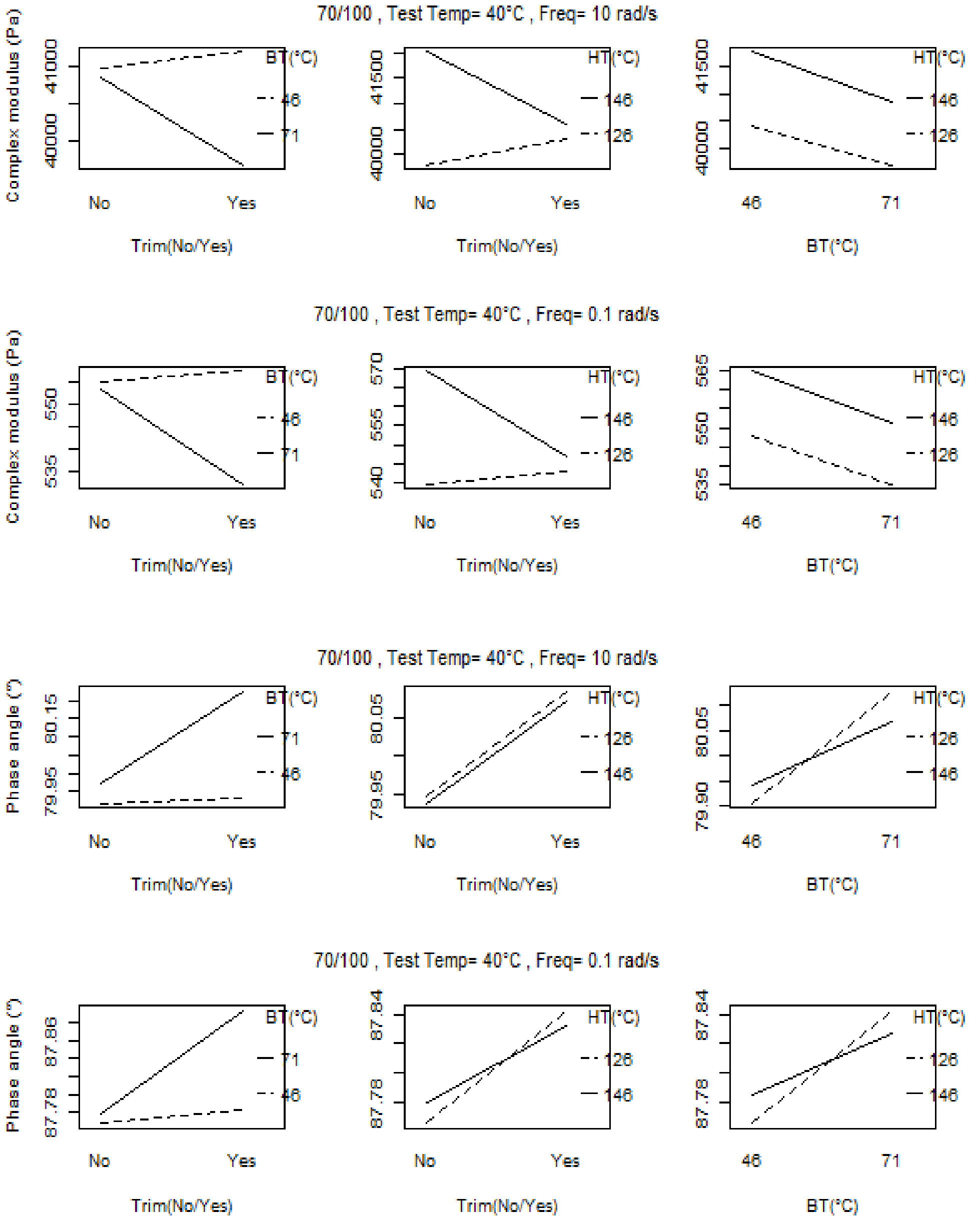


Figure 16. Interaction effect between two variables at different frequencies.

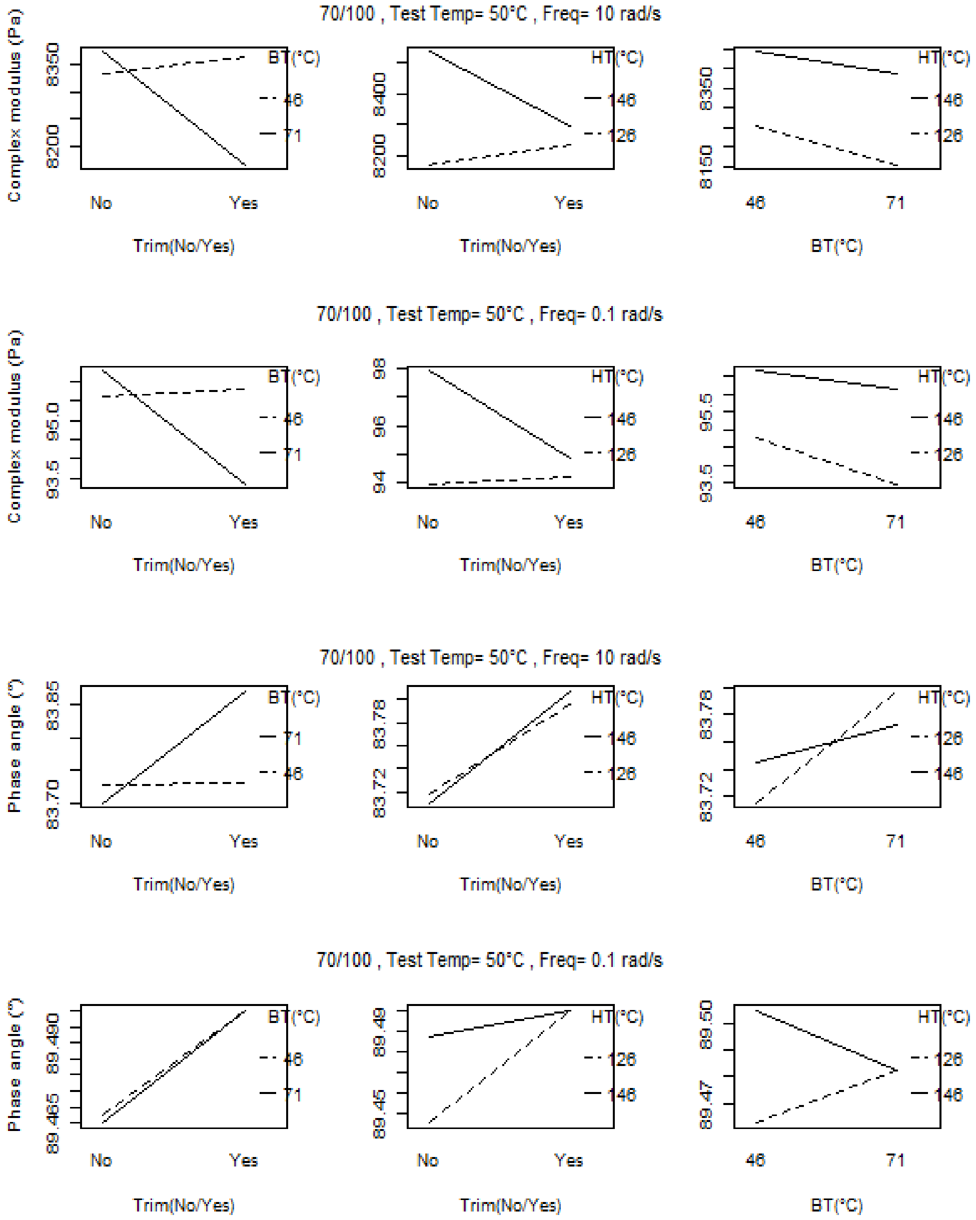


Figure 17. Interaction effect between two variables at different frequencies.

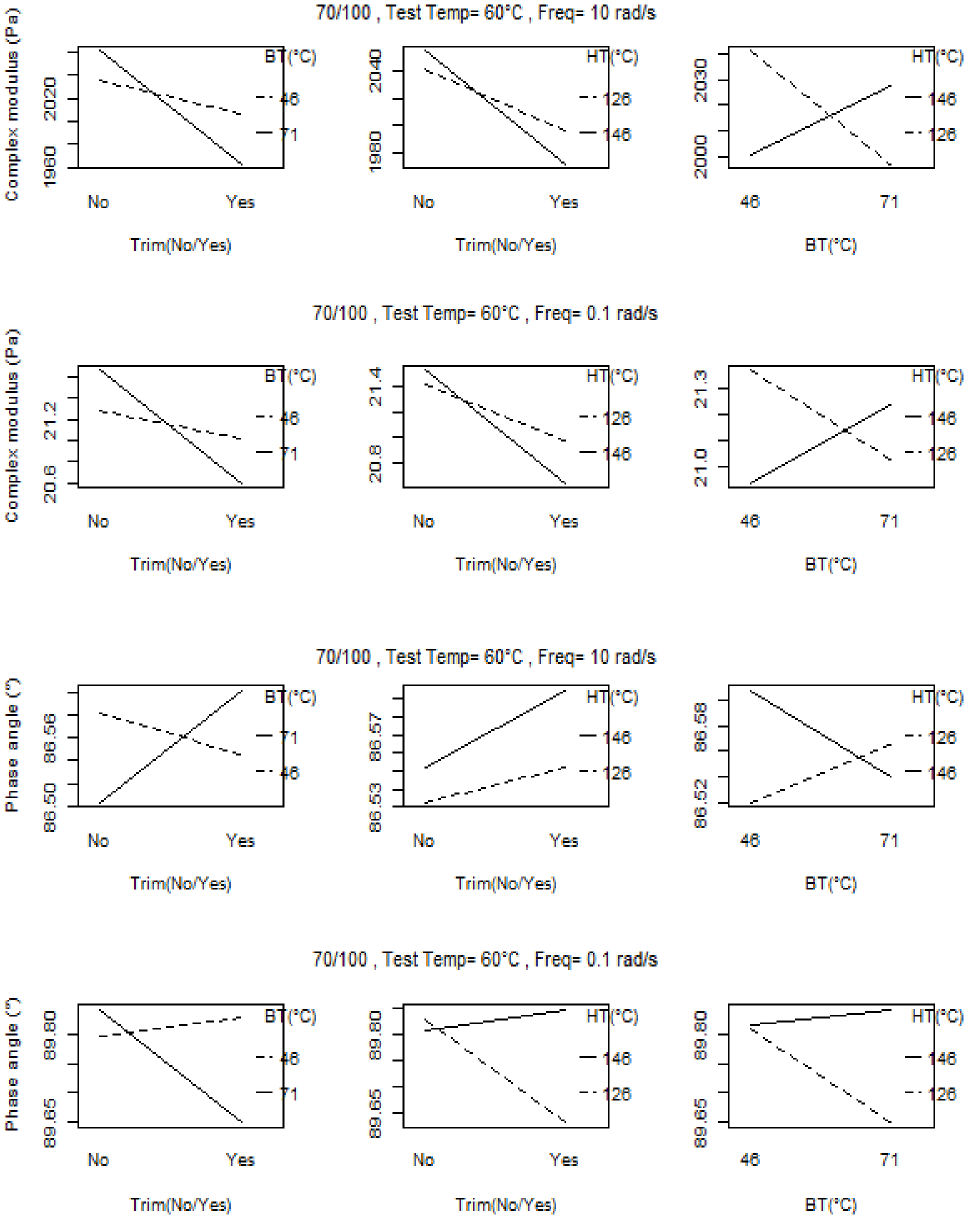


Figure 18. Interaction effect between two variables at different frequencies.

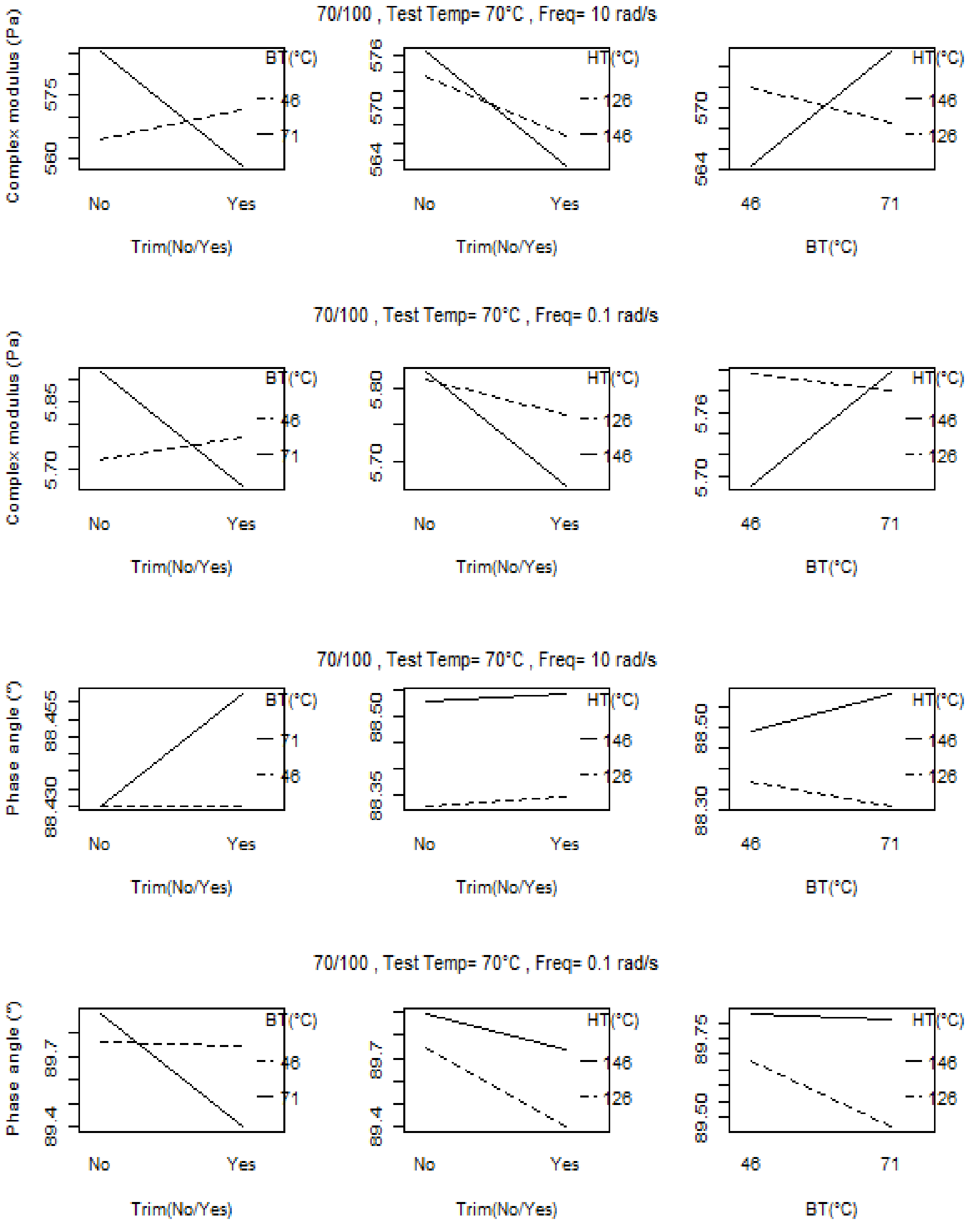


Figure 19. Interaction effect between two variables at different frequencies.

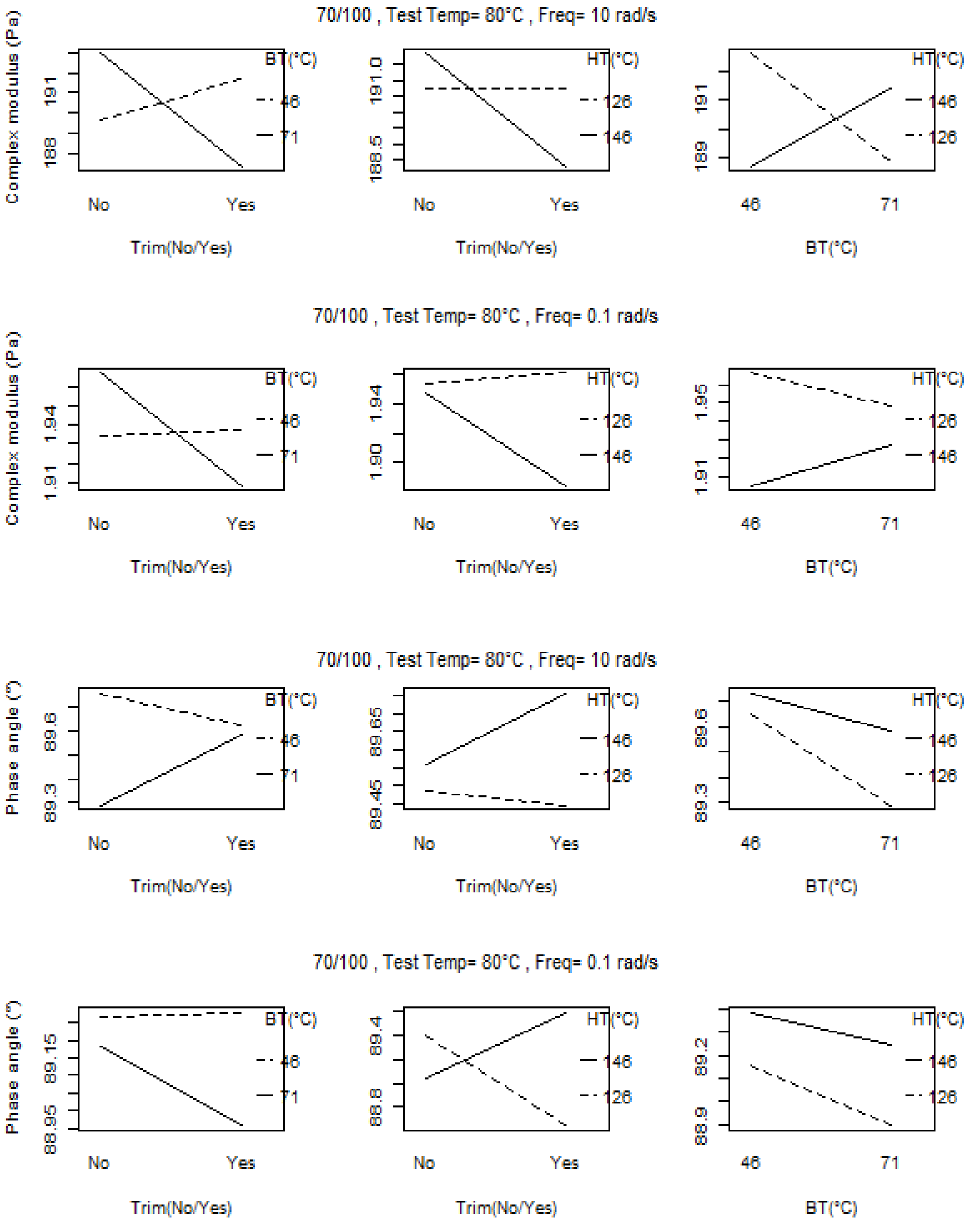


Figure 20. Interaction effect between two variables at different frequencies.

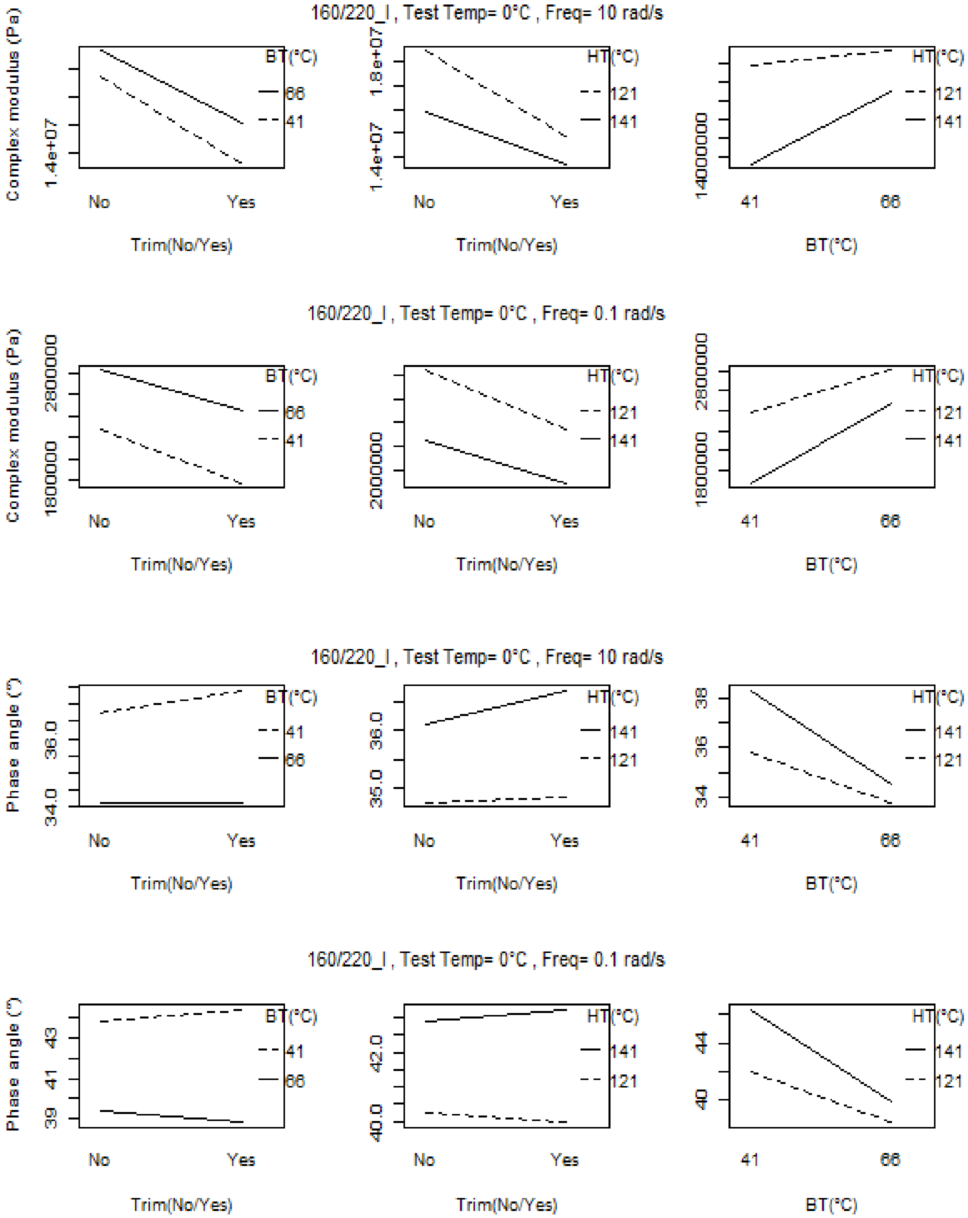


Figure 21. Interaction effect between two variables at different frequencies.

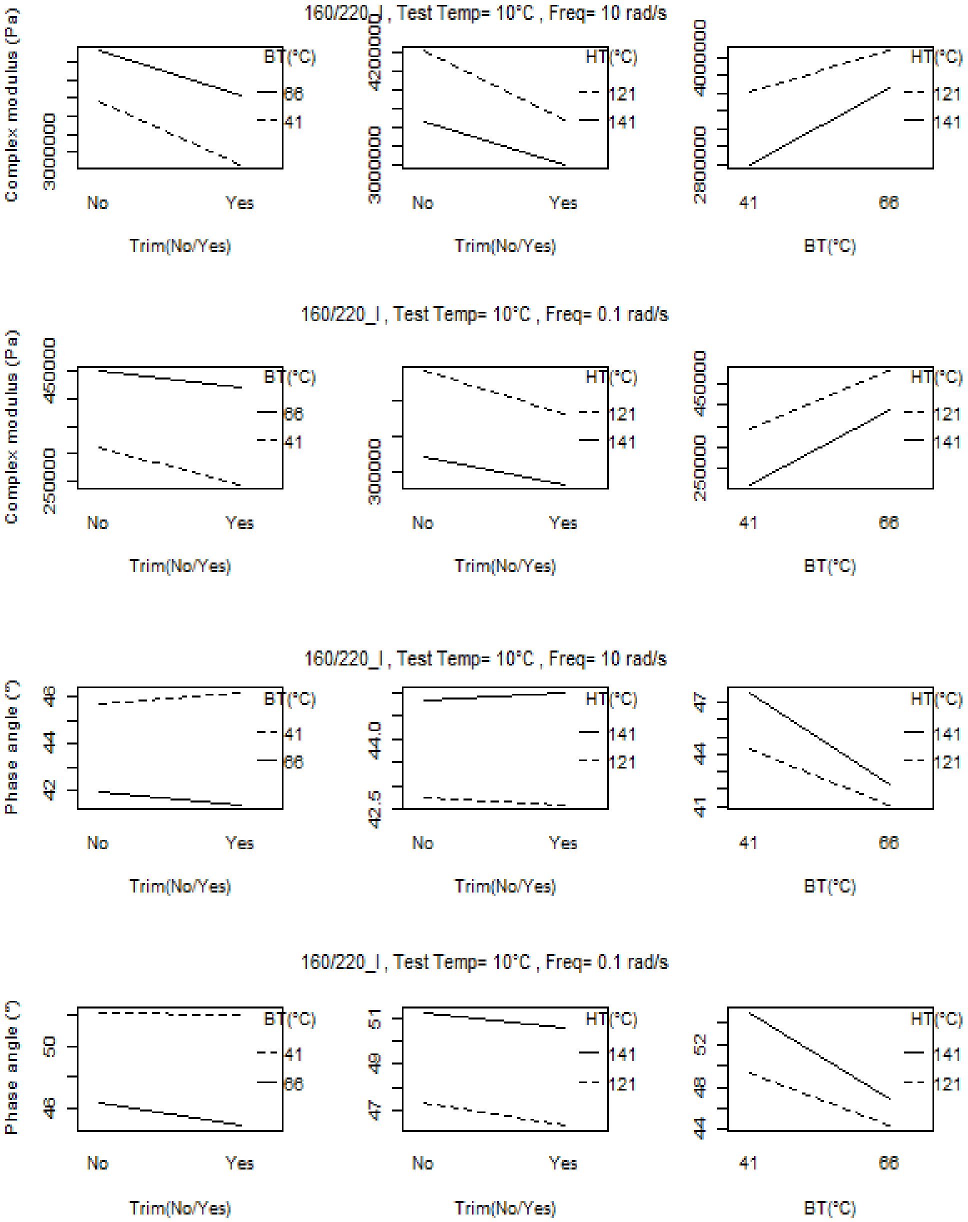


Figure 22. Interaction effect between two variables at different frequencies.

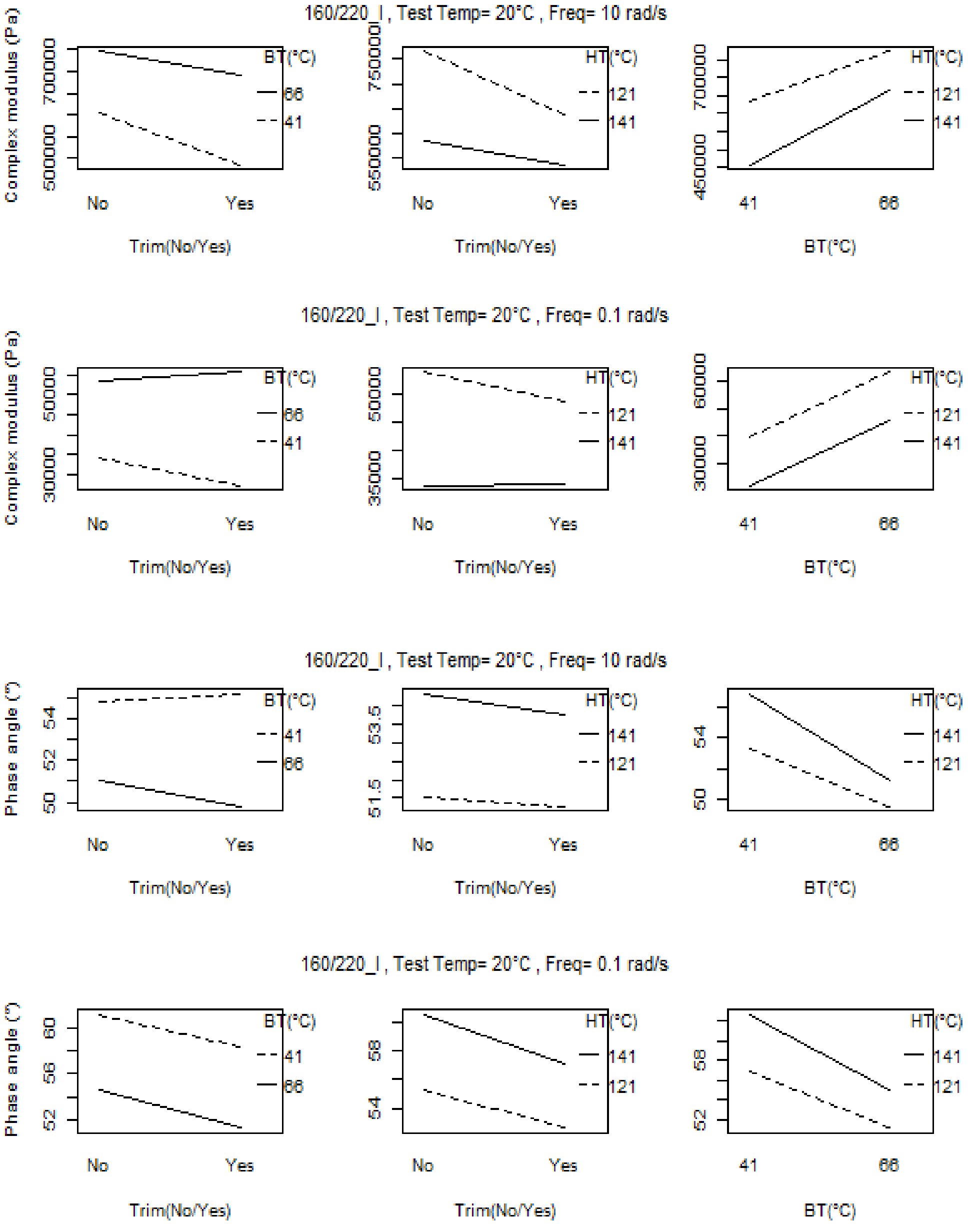


Figure 23. Interaction effect between two variables at different frequencies.

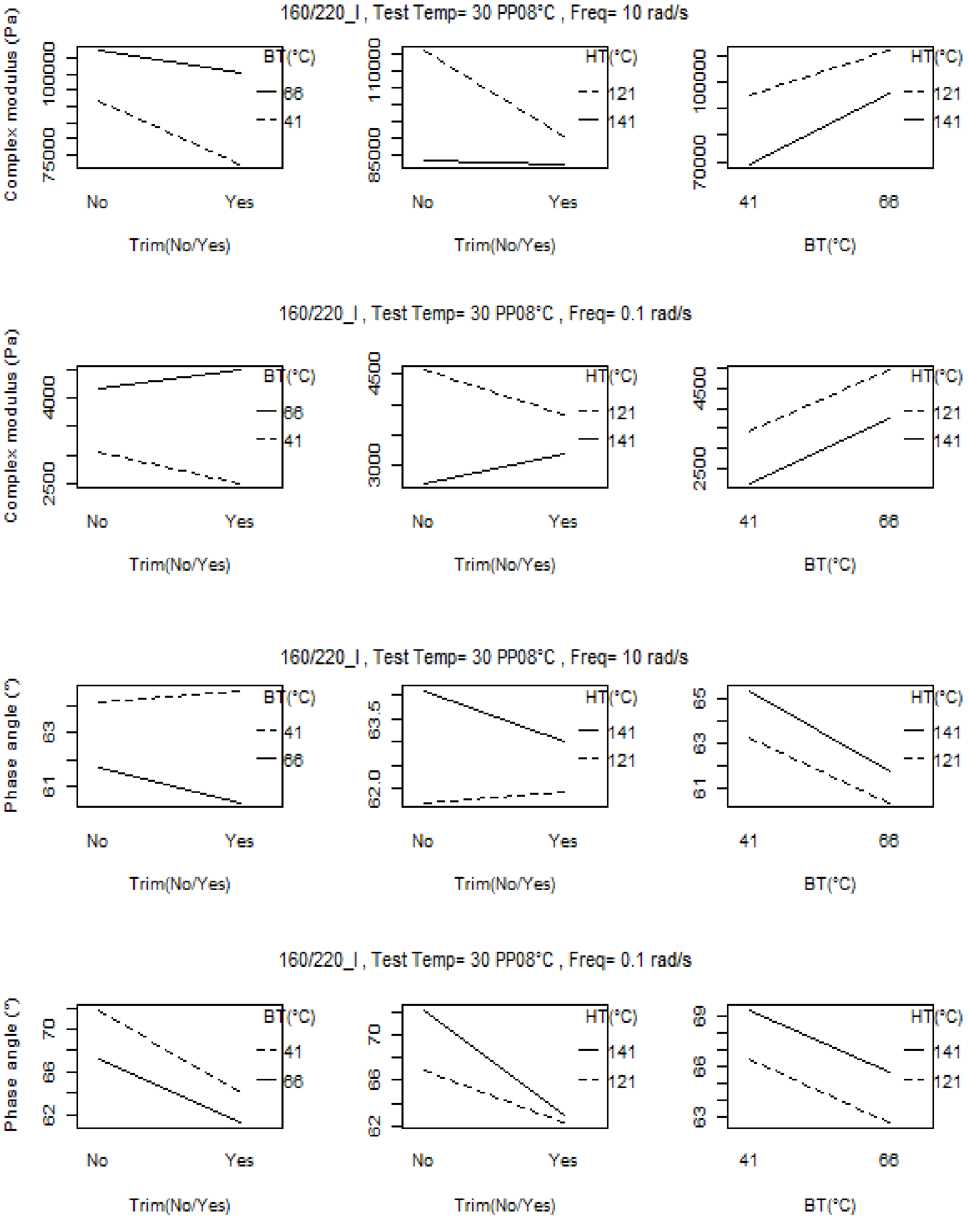


Figure 24. Interaction effect between two variables at different frequencies.

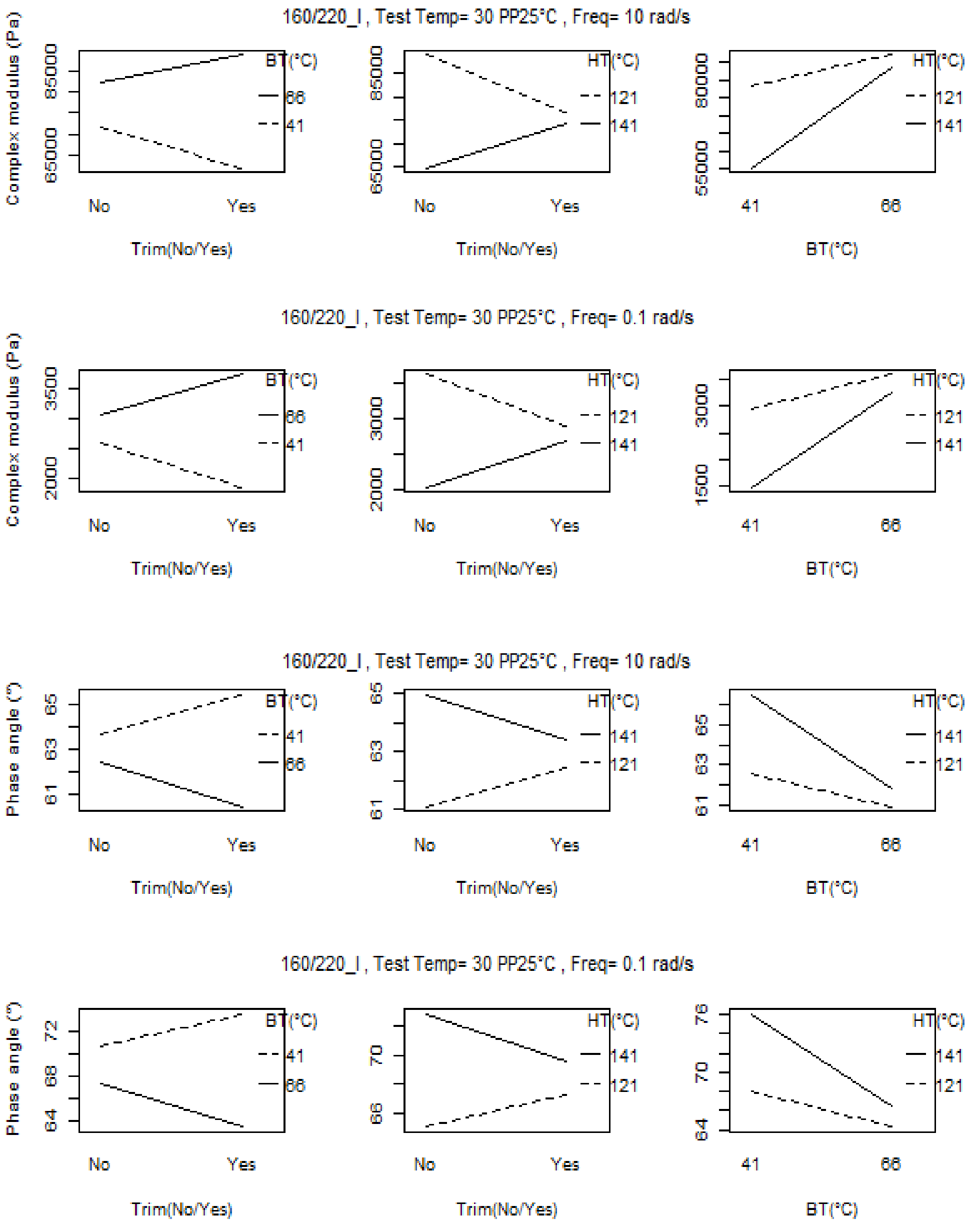


Figure 25. Interaction effect between two variables at different frequencies.

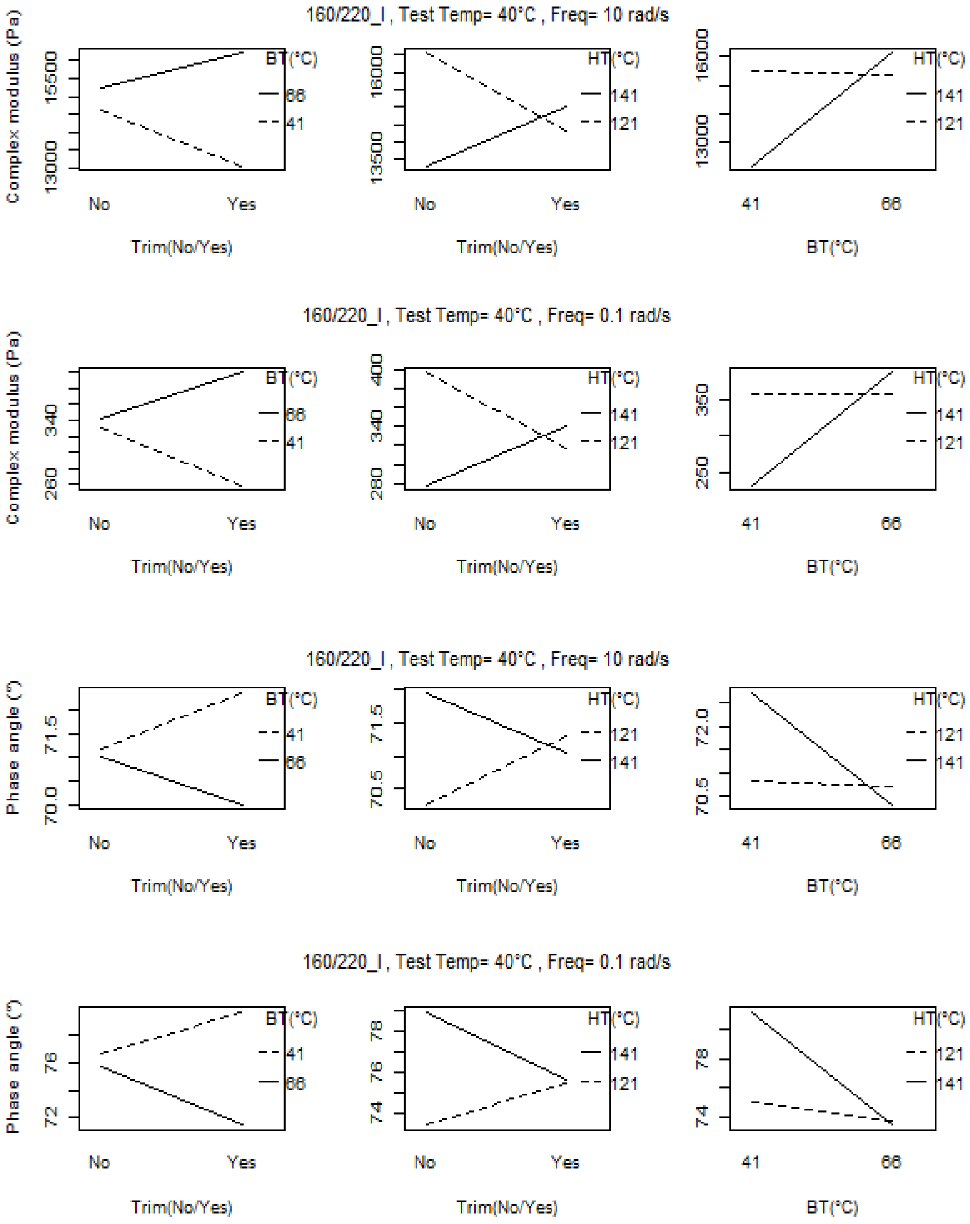


Figure 26. Interaction effect between two variables at different frequencies.

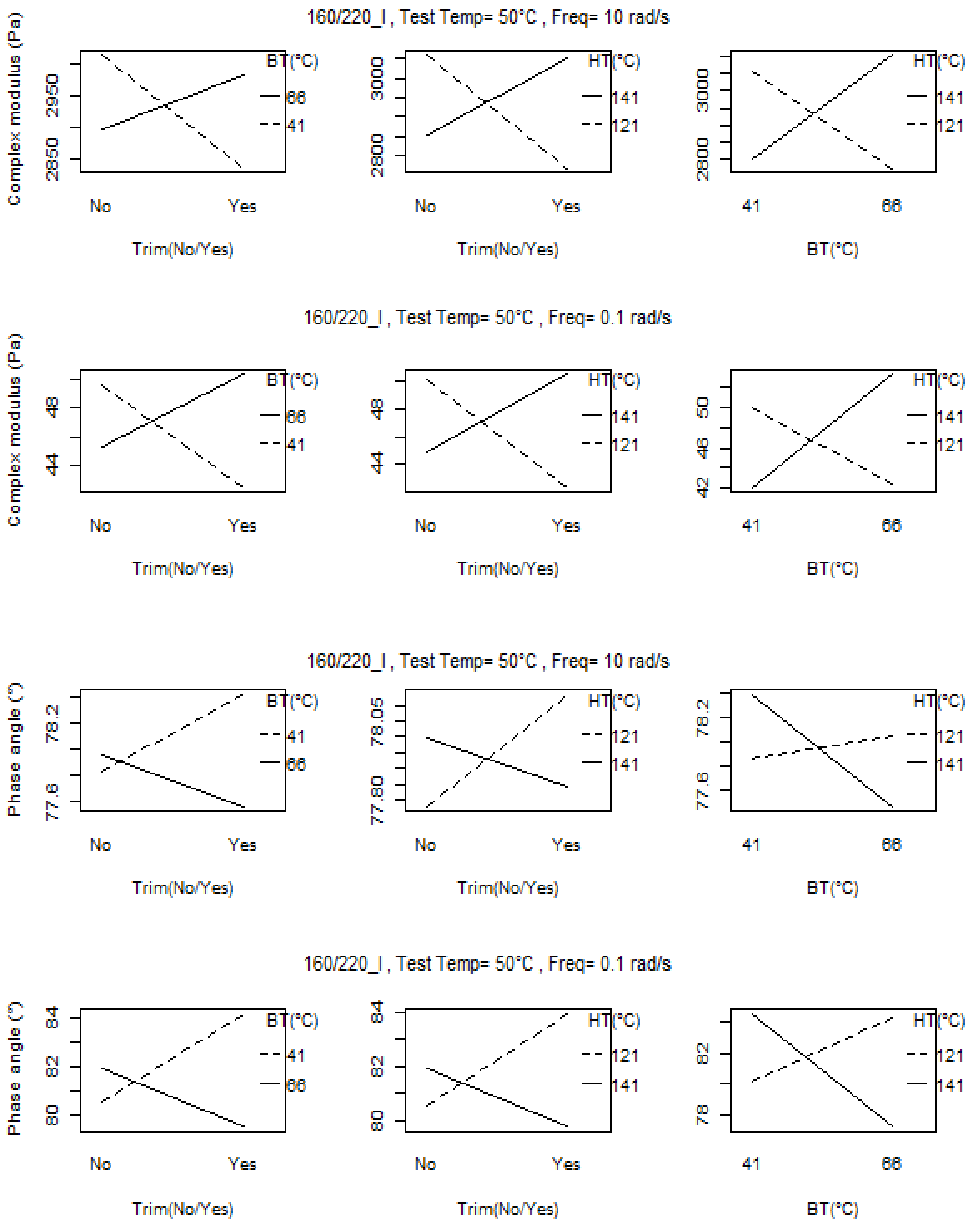


Figure 27. Interaction effect between two variables at different frequencies.

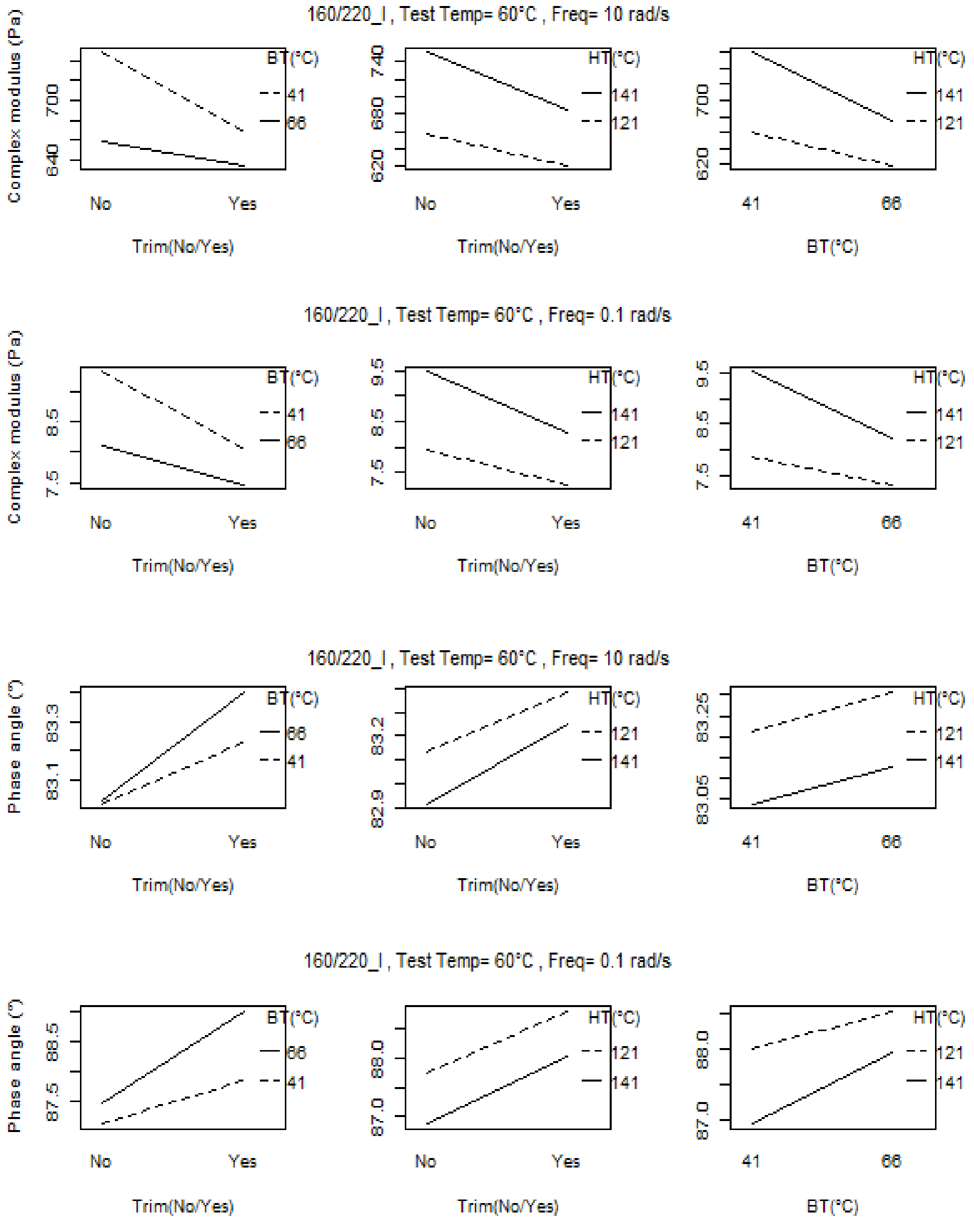


Figure 28. Interaction effect between two variables at different frequencies.

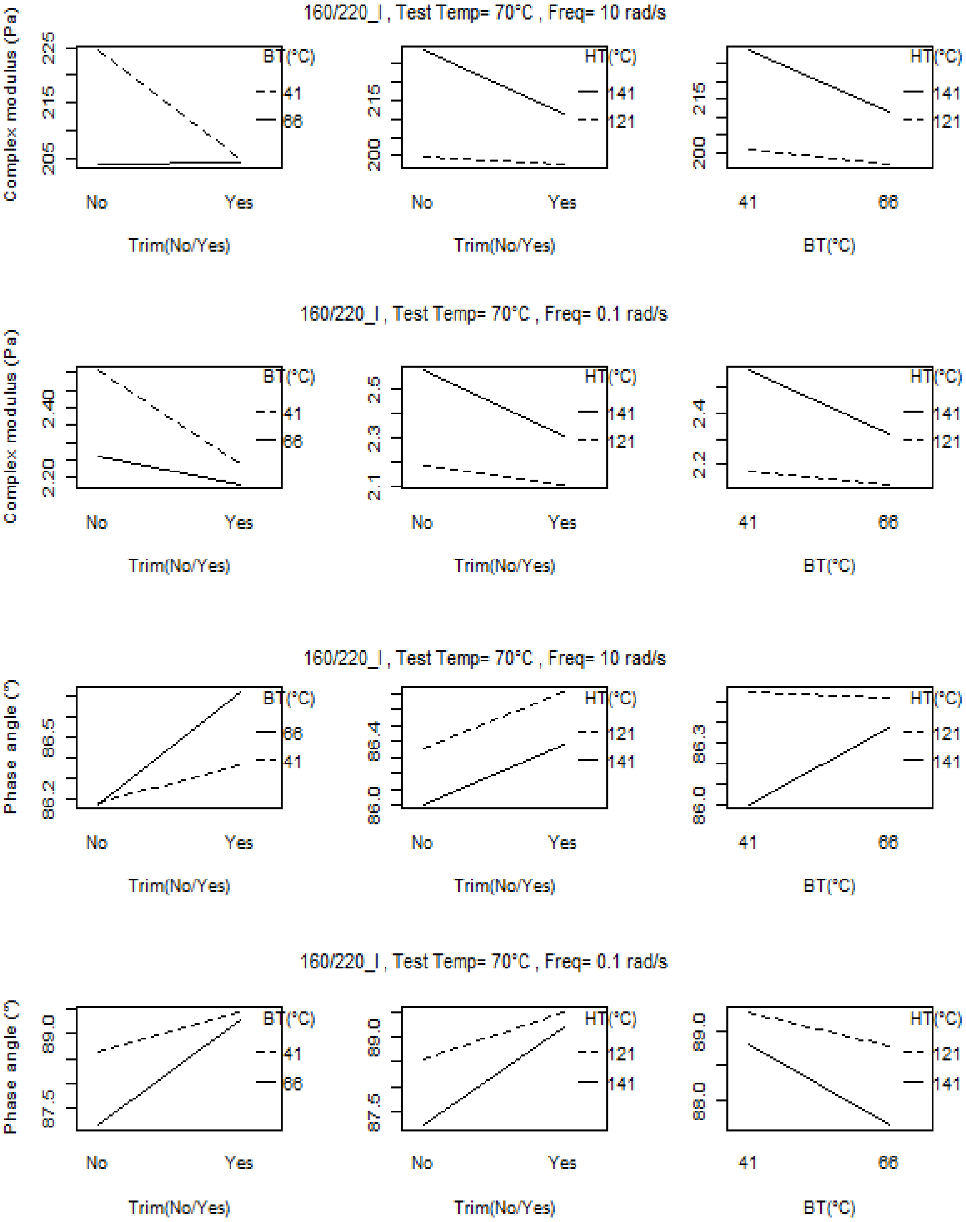


Figure 29. Interaction effect between two variables at different frequencies.

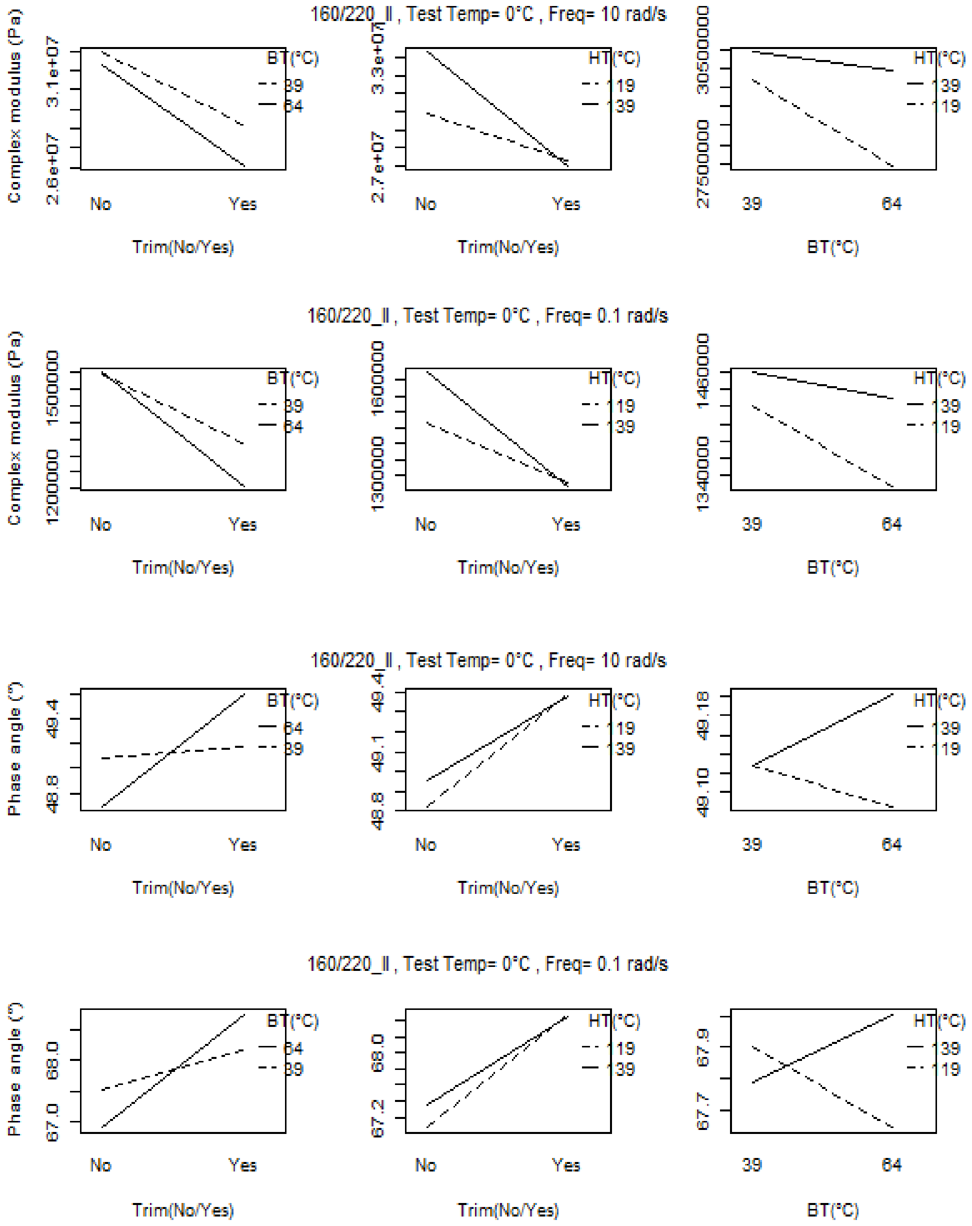


Figure 30. Interaction effect between two variables at different frequencies.

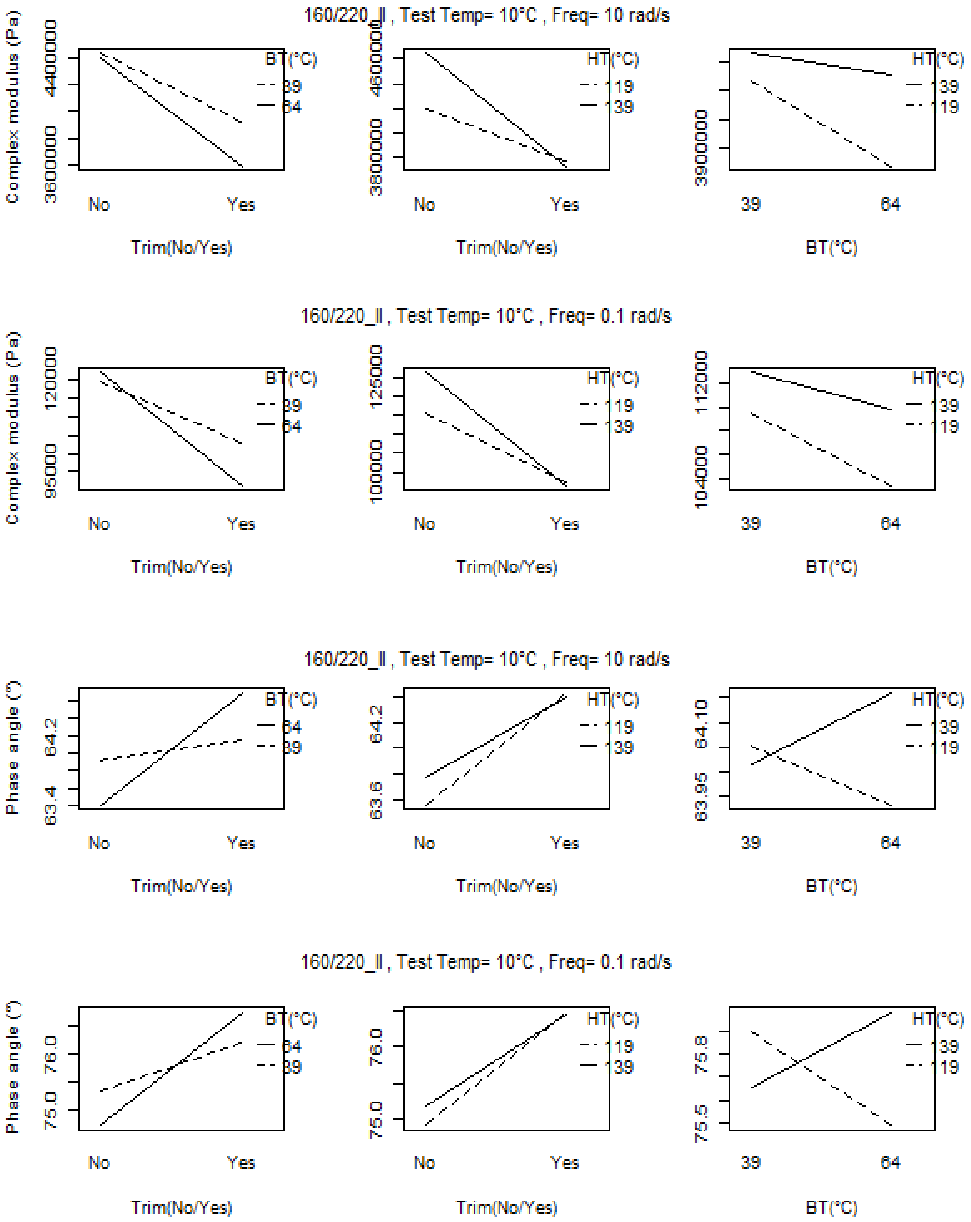


Figure 31. Interaction effect between two variables at different frequencies.

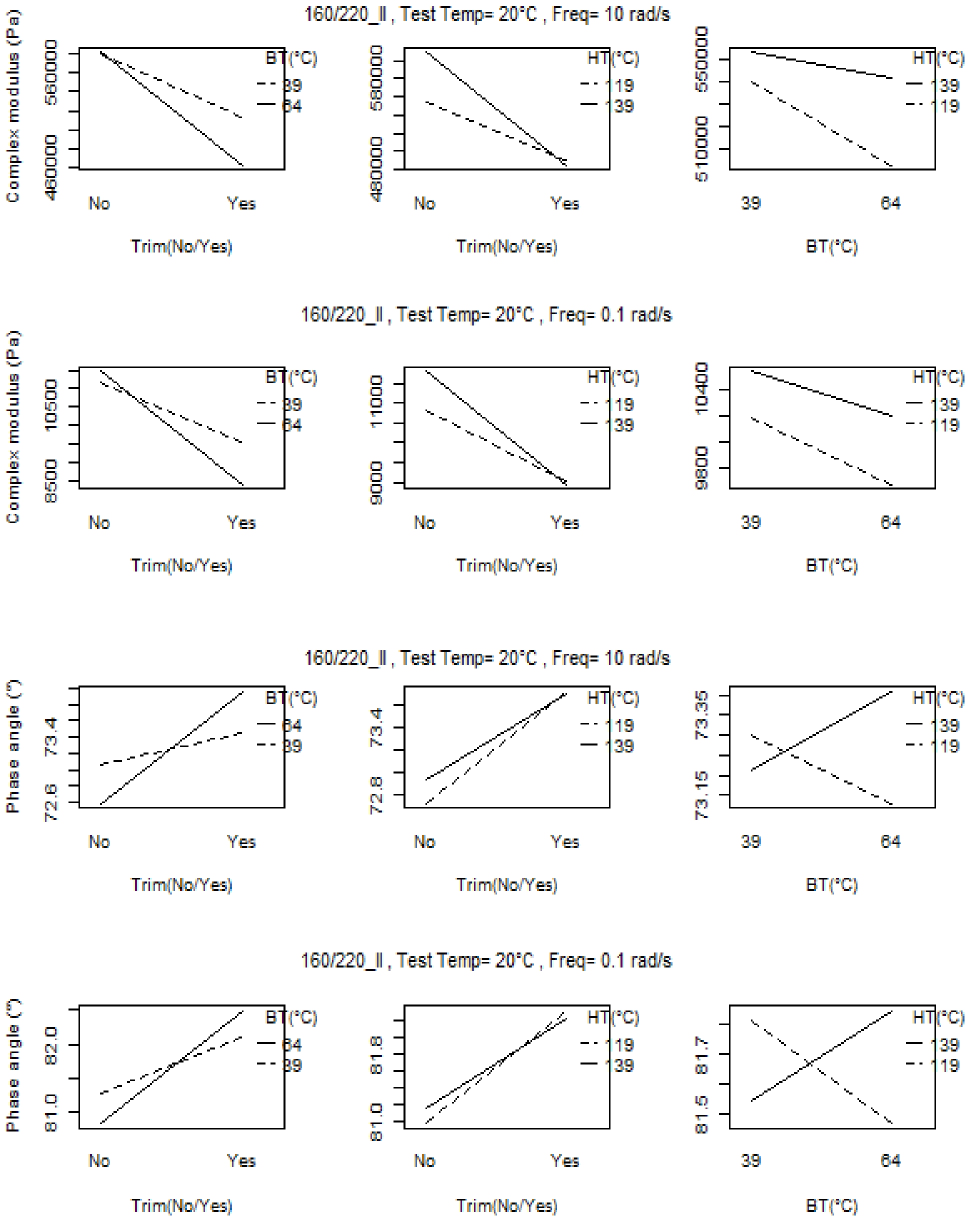


Figure 32. Interaction effect between two variables at different frequencies.

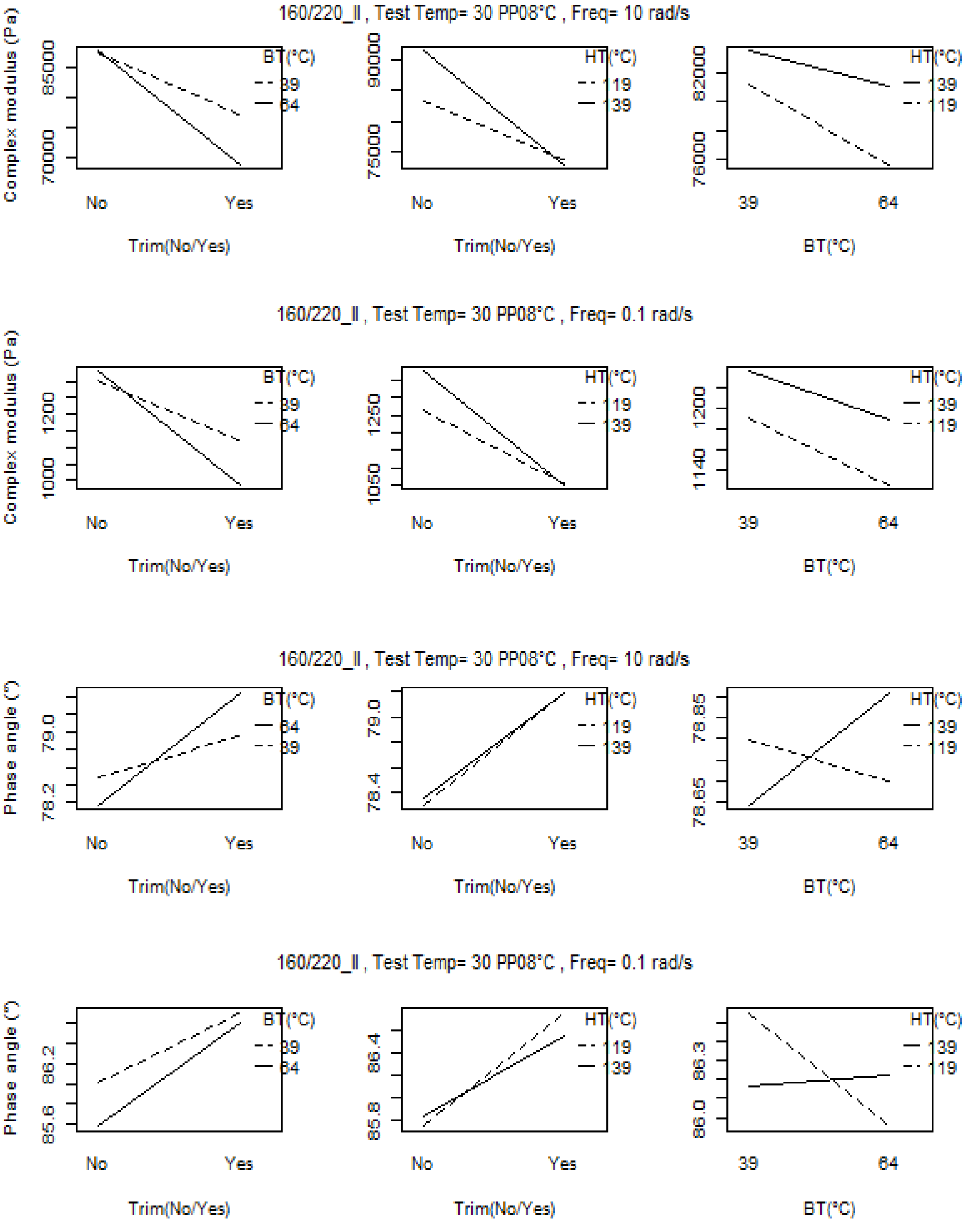


Figure 33. Interaction effect between two variables at different frequencies.

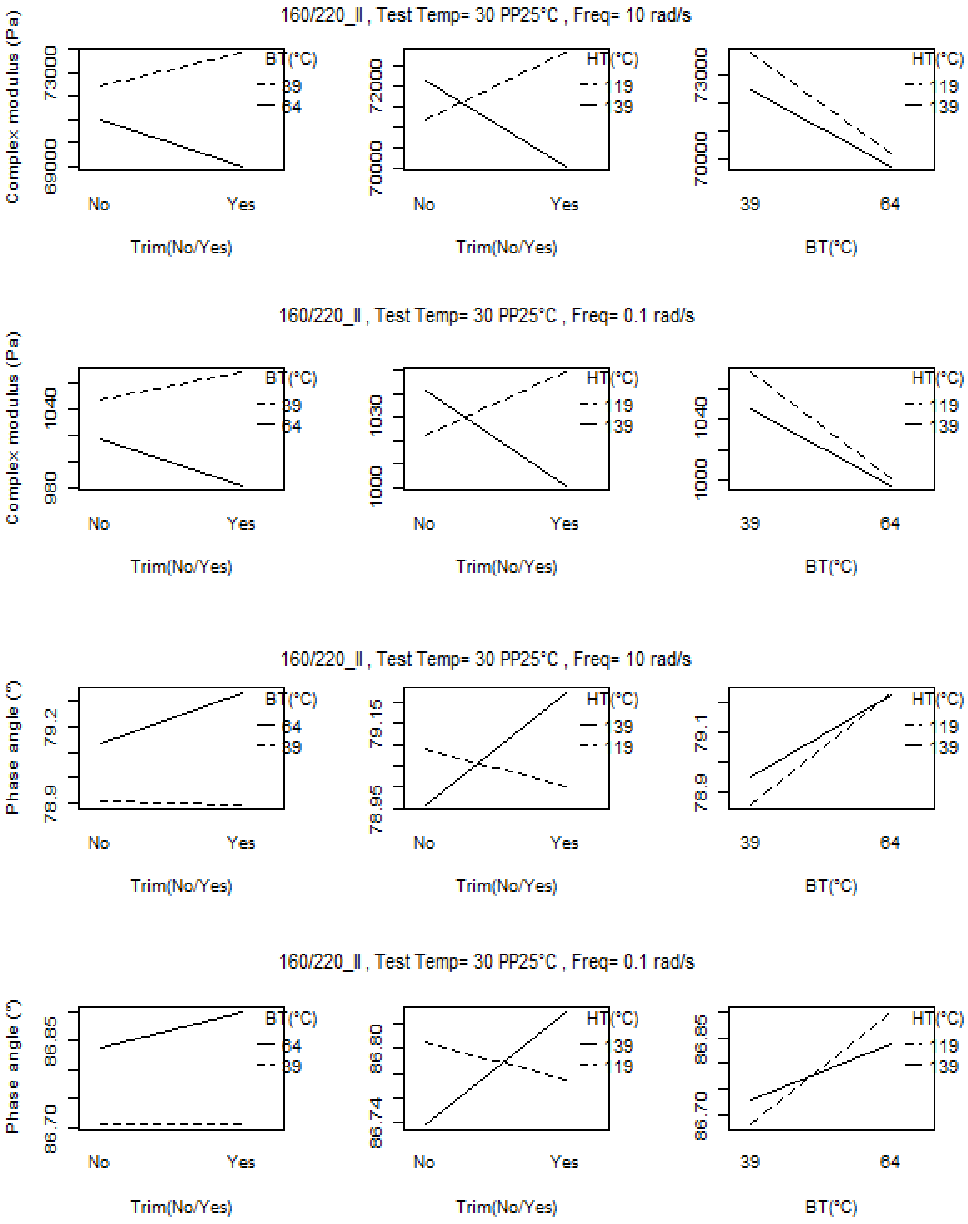


Figure 34. Interaction effect between two variables at different frequencies.

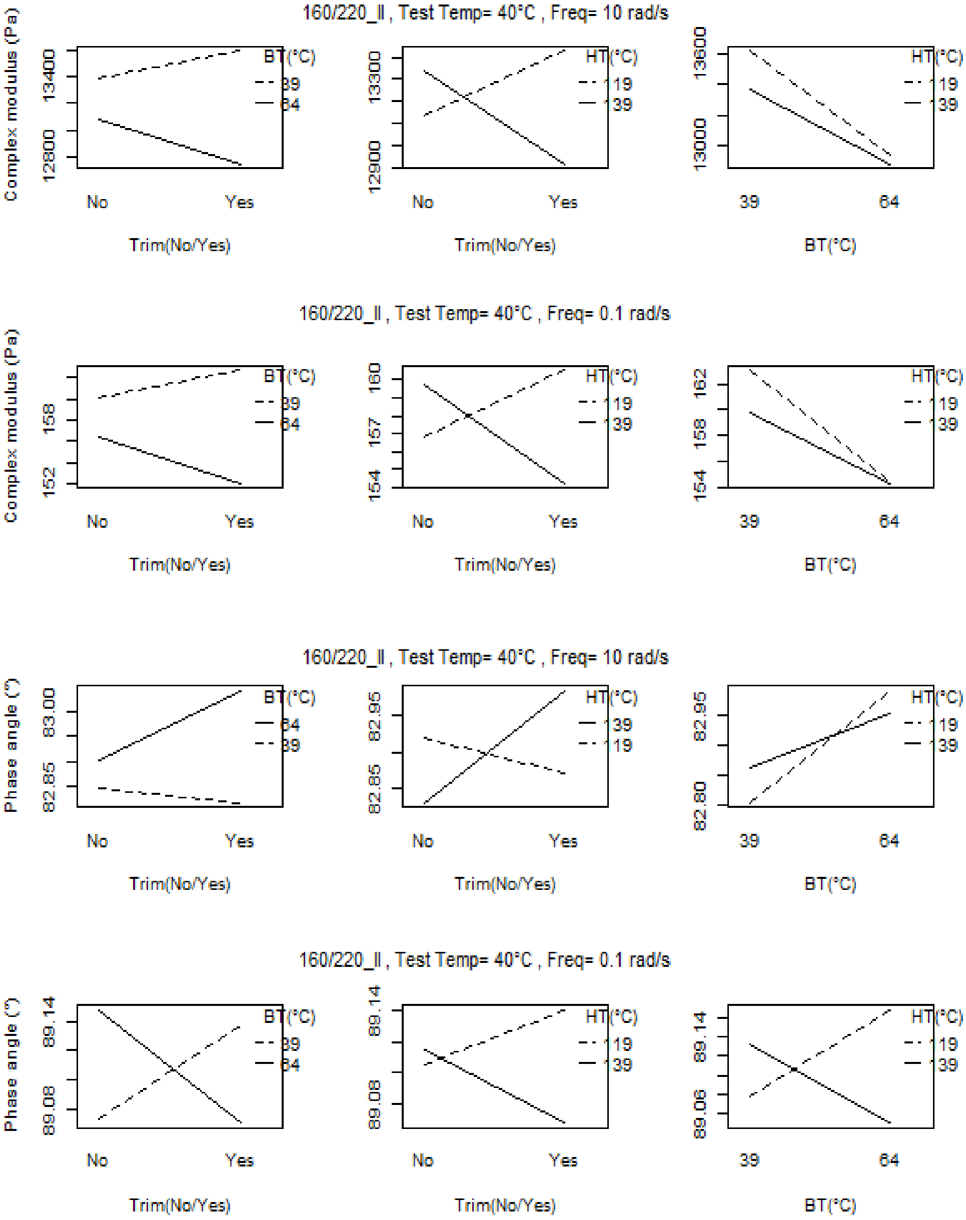


Figure 35. Interaction effect between two variables at different frequencies.

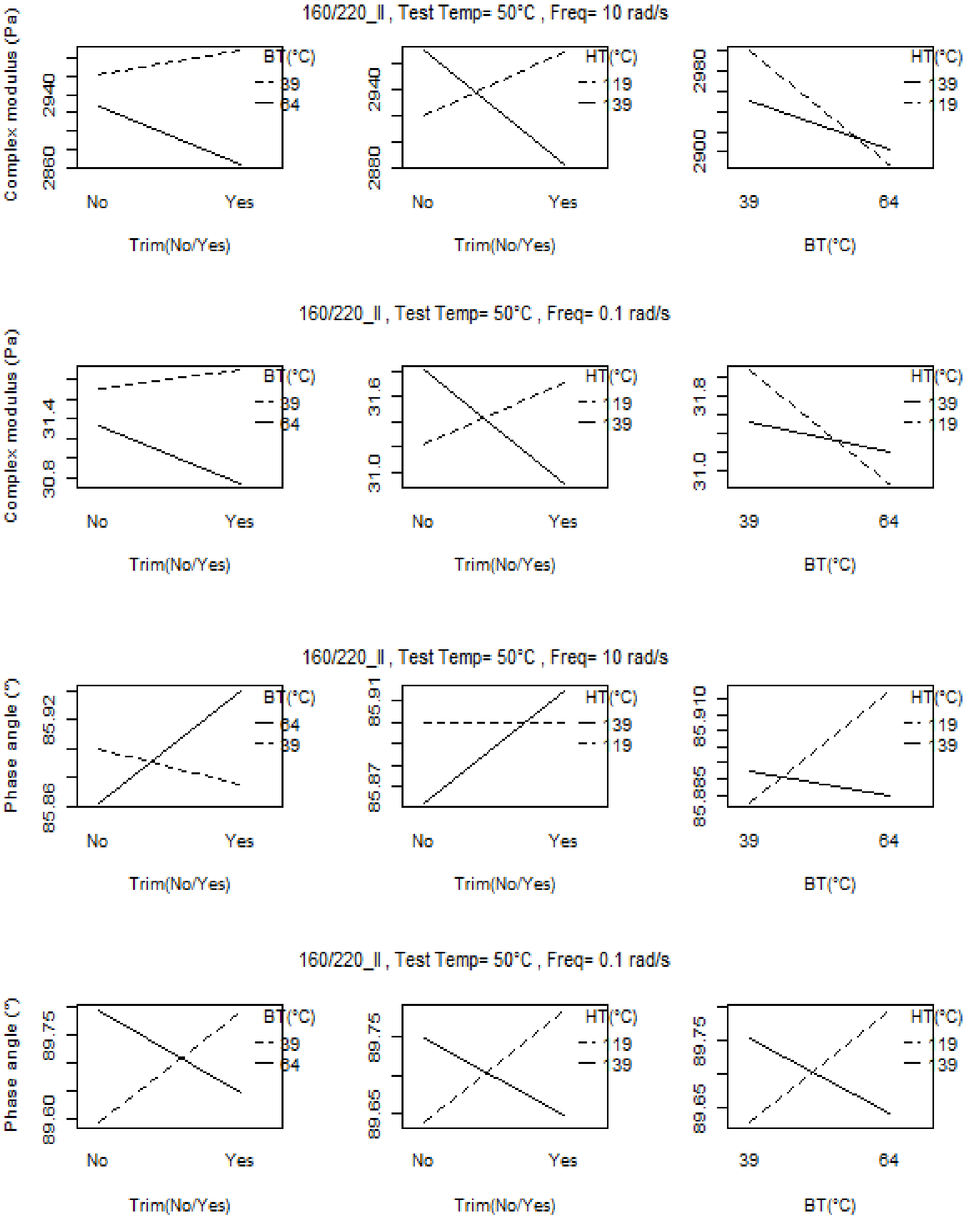


Figure 36. Interaction effect between two variables at different frequencies.

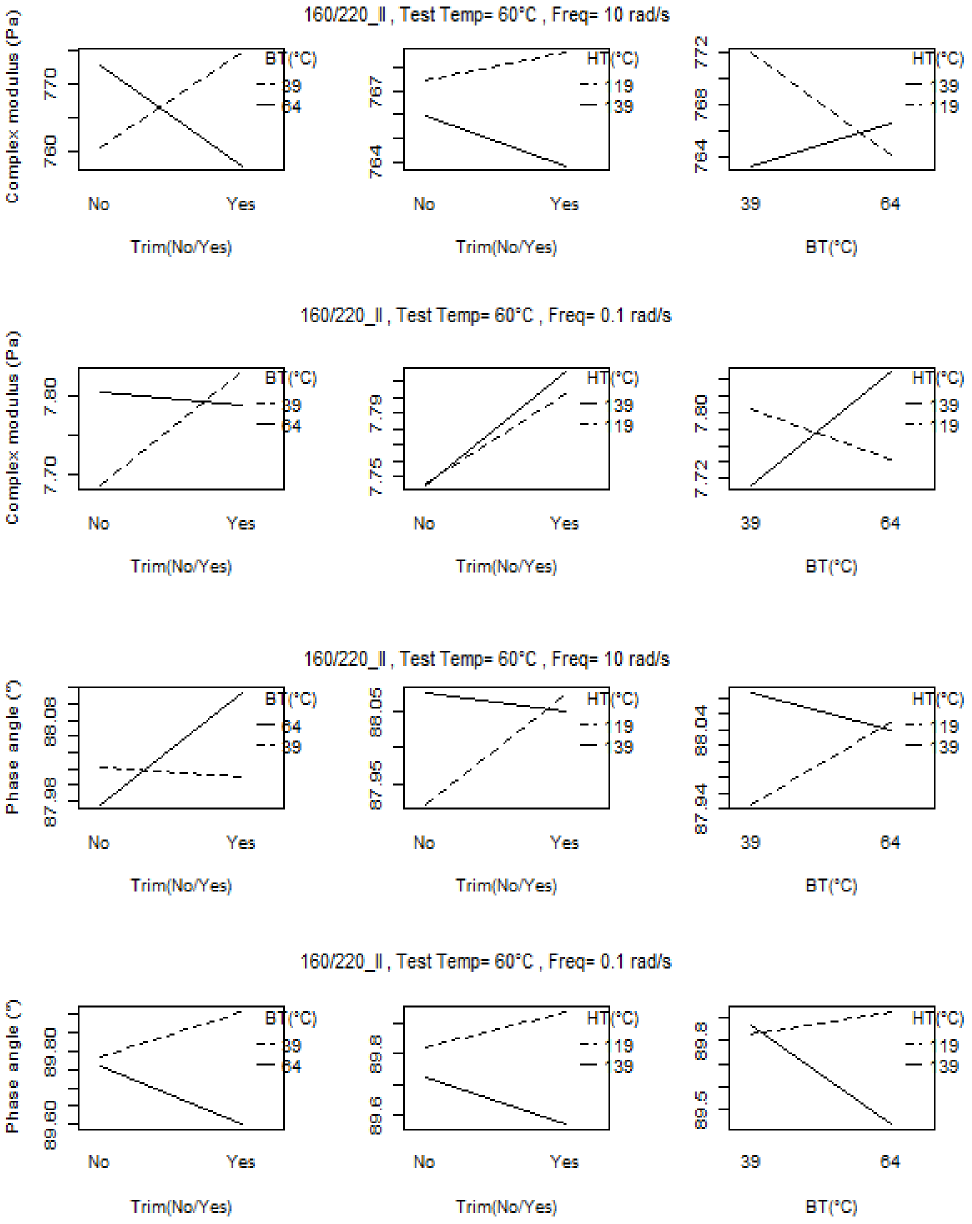


Figure 37. Interaction effect between two variables at different frequencies.

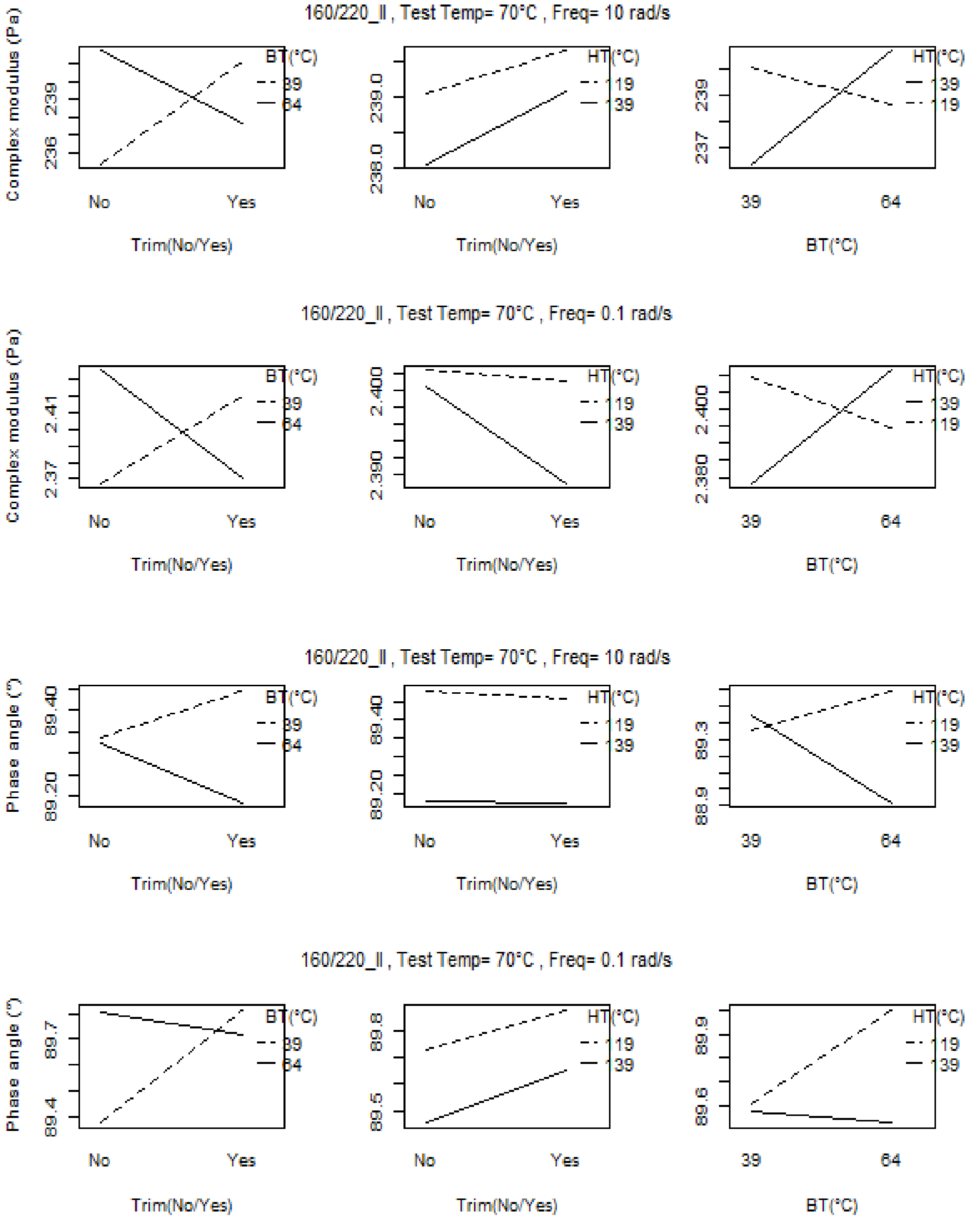


Figure 38. Interaction effect between two variables at different frequencies.