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MISTING IN THE TRANSFER NIP OF A FILM COATER

GABRIEL ASCANIO

DÉPARTEMENT DE GÉNIE CHIMIQUE

ÉCOLE POLYTECHNIQUE DE MONTRÉAL

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ÉCOLE POLYTECHNIQUE DE MONTRÉAL

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MISTING IN THE TRANSFER NIP OF A FILM COATER

présentée par: ASCANIO Gabriel

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M. LEGROS Robert, Ph.D., président

M. TANGUY Philippe A., Ph.D., et directeur de recherché

M. CARREAU Pierre J., Ph.D., et co-directeur de recherché

M. VLACHOPOULOS John, Ph.D., membre externe

M. JOLICOEUR Mario, Ph.D., membre

M. FRAPPIER Clément, représentant du Doyen

To my wife Patricia

To my son Fernando

To my mother Guadalupe

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RÉSUMÉ

Le couchage du papier consiste à déposer une mince couche de pigment sur une trame de papier afin d'améliorer ses propriétés optiques et d'impression. De nombreux procédés sont disponibles. Cependant, de nos jours des efforts sont faits pour développer de nouvelles techniques qui permettent d'atteindre des vitesses élevées tout en réduisant les problèmes de machinabilité, qui entraînent des arrêts du procédé. Les machines les plus utilisées pour le couchage du papier à haute vitesse sont actuellement les coucheuses à lame et les presses de couchage par film (PCF). Cependant, la littérature montre que tous les nouveaux investissements, particulièrement en Europe, portent sur les PCF. Les raisons du succès croissant des PCF sont bien connues. Les PCF nécessitent un investissement relativement bas pour transformer un système simple de couchage par rouleaux en un nouveau système de couchage par dosage de film. Elles présentent également une bonne machinabilité et permettent de développer de nouveaux produits tel que le papier journal couché. Le couchage par film est une méthode plus profitable et plus flexible comparativement aux coucheuses à lame pour les raisons suivantes : (i) la possibilité d'utiliser un papier moins dense avec un contenu élevé de pâte mécanique ou avec des fibres recyclées; (ii) l'emploi de sauces de couchages avec une teneur en matières solides supérieure à 60%; (iii) la possibilité de couchage à haute vitesse avec une contrainte plus faible appliqué sur le papier et (iv) une couche plus épaisse. En ce qui concerne le produit fini, les PCF offrent une plus grande flexibilité en traitement de surface, et une opacité et brillance plus élevées.

Cependant, en raison de la présence de la séparation du film à la sortie de l'entrefer d'application, le lustre, le lissé et l'uniformité de la surface sont inférieurs à ceux obtenus avec les coucheuses à lame. Cette séparation du film promeut la formation de "peau d'orange" et quand le couchage a lieu à haute vitesse un phénomène indésirable "le brouillard" (connu en anglais comme "misting") apparaît particulièrement pour les

grammages élevés. Il y a plusieurs variables à considérer afin de réduire les effets de la séparation du film. Dans la plupart des cas, la configuration des coucheuses et le type de papier sont fixés et peuvent être difficilement modifiés. En pratique, les variables des formulations de sauces de couchage sont les seuls paramètres qui peuvent être manipulés pour améliorer la machinabilité et la qualité du couchage par film.

L'objectif de cette recherche est de comprendre le comportement de l'écoulement des fluides dans l'entrefer d'application d'une PCF, plus particulièrement la séparation du fluide et le phénomène de "misting", un des défauts associés à la machinabilité. Pour ce faire, une coucheuse de transfert par film a été mise au point afin de mener la caractérisation hydrodynamique des suspensions de sauces de couchage. Cette coucheuse consiste fondamentalement en un rouleau déformable (rouleau de transfert), un rouleau rigide (rouleau de support) et un système d'alimentation comprenant une lame de dosage qui contrôle l'épaisseur de la couche du fluide à transférer. De plus, la vitesse des rouleaux, la force entre les rouleaux et le débit peuvent être facilement ajustés.

La distribution de la pression des fluides s'écoulant dans l'entrefer a été déterminée à l'aide d'un mini-capteur de pression dynamique installé sur le rouleau de support. Ce capteur est relié à une carte d'acquisition de données en utilisant une bague collectrice, de sorte que les données de pression sont enregistrées en continu. Plusieurs profils de pression sont obtenus à chaque tour de rouleau.

L'écoulement à la sortie de l'entrefer a été étudié à l'aide d'un système de visualisation à haute vitesse. Ce système comprend une camera vidéo à haute vitesse CMOS ayant une résolution de 1024 x 1024 pixels carrés et un système d'illumination basé sur un laser à vapeur de cuivre. La vitesse maximale d'enregistrement est 462 images/s et un total de 512 images peuvent être stockées à la pleine résolution.

Les propriétés rhéologiques en cisaillement des fluides étudiés ont été déterminées avec un rhéomètre AT-2000 de la compagnie TA Instruments en utilisant une configuration Couette avec un cylindre extérieur de 30 millimètres et un cylindre intérieur de 28 millimètres. Les propriétés élongationnelles des suspensions ont été évaluées à l'aide d'un rhéomètre à orifice basé sur la méthode de perte de charge. Dans cette méthode, la viscosité élongationnelle est une fonction du débit et de la perte de pression du fluide lors de son passage par un petit orifice circulaire.

Cette recherche a montré que le "misting" peut être produit par deux mécanismes différents : la séparation du fluide à la sortie de l'entrefer et un effet d'atomisation dû à l'entraînement d'air dans l'entrefer. Ce dernier effet n'est obtenu que dans le cas d'une couverture de rouleau en élastomère très mou pour des rouleaux tournant à haute vitesse et avec une charge entre les rouleaux faible. L'entraînement d'air peut être évité si la charge appliquée est augmentée; cependant, l'écoulement devient périodique. Il a été montré que le rapport de Trouton pour une sauce de couchage excède considérablement la valeur théorique qui devrait être obtenue pour des fluides newtoniens. Il a également été prouvé que les sauces de couchage ayant une viscosité élongationnelle plus élevée et un écrouissage plus prononcé produisent moins de "misting". Cependant cette viscosité doit être maintenue au niveau le plus bas possible afin d'éviter la formation de peau d'orange.

ABSTRACT

Paper coating consists of depositing a thin film onto the paper in order to improve its printing and optical properties. Although many processes are available, nowadays many efforts are being made in developing new techniques that allow operating at high speeds reducing runnability problems, and therefore shut downs.

The most common machines for paper coating at high speed are currently blade coaters and Metering Film Coaters (MFC). Up to date, 80% of worldwide production of coated paper is carried out with blade coaters. However, reports show that all the new investments, especially in Europe are for MFC machines. The reasons for the growing success of the metered size press in these areas are well recognized. MFC requires minimal rebuild and a relatively low investment. An additional key feature is its good runnability as well as the possibility to have brand new product concepts such as coated newsprint. MFC is a more profitable and flexible method compared to blade coaters taking into account the following reasons: (i) the possibility of using a less dense paper with a high mechanical pulp content or with recycled fibers; (ii) a solid content higher than 60%; (iii) high speed with a low stress applied to the web and, (iv) larger coating thickness. In regard to the finished product, a higher covering, a higher opacity and a higher brightness are obtained when using a MFC.

However, due to the presence of film split at the exit of the transfer nip, the gloss, smoothness and uniformity of the coating surface are lower than those obtained with blade coaters for high quality paper and higher coat weights. Because of the film split, there is a tendency toward patterning of the coating or "orange peel" and when coating is applied at high speed the undesirable phenomenon of misting appears especially at high coat weights. There are several variables to consider in reducing the effects of the film splitting. In most cases the coating equipment and base paper are fixed and can be

modified with great difficulty, therefore this leads to consider the adjustment of the formulation variables as an easier way to improve the runnability and quality of the film coating.

The present work is dedicated to the development of the understanding of the behavior of the fluid flow in the transfer nip focusing on the film splitting process as well as one major runnability issue related to it, misting. For that purpose, a laboratory film coater was developed for the hydrodynamic characterization of Newtonian fluids and suspensions. It consists basically of a deformable roll (transfer roll), a rigid roll (backing roll) and a delivery system including a doctor blade to control the amount of fluid to be transferred. The roll speed, the applied load between the rolls and the flow rate can be easily varied with the film coater.

The pressure distribution of the fluids into the nip was determined by means of a miniaturized dynamic pressure transducer mounted on the backing roll. This transducer is connected to a data acquisition card via a slip ring, so that the pressure data are continuously recorded. Several pressure profiles are obtained for every roll revolution.

The flow downstream the nip was visualized by means of a high-speed video system, which consists of a high-speed CMOS camera having a full resolution of 1024 x 1024 square pixels and a diode copper vapor laser as illumination system. The maximum record speed is 462 frames/s and a total of 512 images can be stored at full resolution.

The shear rheological properties of the tested fluids was determined with the AR-2000 rheometer from TA Instruments in a Couette configuration of 30 mm and 28 mm for the inside and outside diameter, respectively. The extensional properties of the suspensions were evaluated by means of an orifice flowmeter based on the pressure entry technique, in which the extensional viscosity is a function of the flow rate and the pressure drop data of the tested fluid passing through a small orifice.

This investigation has shown that misting can be generated by two different mechanisms: film splitting and a spray effect due to air entrainment into the nip. Spray was found to occur with a soft deformable gap at high speed and a low load is applied between the rolls. The air entrainment can be avoided if the applied load is increased; however, the flow can become periodic.

From a rheological standpoint, both the suspensions and coating colors investigated had shown little elasticity at high strain. Results from the extensional rheology tests showed that the Trouton ratio for coating colors can exceed considerably the theoretical value expected for Newtonian fluids. It has been demonstrated that misting is reduced when coating colors exhibiting a pronounced strain hardening are used. However, the extensional viscosity must be kept as low as possible in order to avoid orange peel formation.

CONDENSÉ EN FRANÇAIS

De nos jours, l'industrie du couchage joue un grand rôle économique. Le papier, les adhésifs, les bandes magnétiques, les peintures et les pellicules photographiques sont quelques exemples de produits issus de l'industrie du couchage.

Dans l'industrie audio et vidéo par exemple, les bandes magnétiques sont fabriquées à partir de particules magnétiques dispersées dans un agent liant qui est couché sur un support en polyester. Dans l'industrie informatique, le couchage est l'une des technologies les plus importantes; la majeure partie des informations traitées par les ordinateurs est stockée sur des supports magnétiques couchés, tels que les disques durs et les disquettes. L'enduction des adhésifs relève également de l'industrie du couchage. Pensons simplement au marché dynamique des autocollants et des articles décoratifs. Dans l'industrie de la photocopie, le couchage du tambour de sélénium est une étape clé dans le processus de copie. Celui-ci est couché avec l'encre du toner qui est ensuite transféré au papier. L'industrie du graphisme repose également sur le couchage; le papier utilisé pour les magazines est couché, ce qui permet de lui donner le lustre et la capacité d'impression requis et d'augmenter sa résistance. D'un point de vue économique, la fabrication des papiers d'impression et d'édition est la branche de l'industrie du couchage la plus importante.

La vitesse du procédé de couchage s'est considérablement accrue ces dernières années, ce qui a nécessité le développement de nouvelles techniques de couchage. Actuellement, deux technologies de couchage sont utilisées pour le couchage du papier à haute vitesse : la coucheuse à lame et la presse de couchage par film dosé (PCF). La technique de couchage par lame consiste à appliquer un excès de fluide sur une trame de papier, puis à racle l'excédent avec une lame. La vitesse du couchage par lame peut atteindre 2500

m/min; cependant, à haute vitesse, en raison de la force centrifuge du rouleau, la trame du papier est soumise à une tension élevée et la fréquence des casses tend à augmenter.

Dans le procédé de couchage par film dosé, le fluide est pré-dosé avant d'être transféré à la trame de papier en mouvement, de sorte que la pression hydrodynamique et le mouillage du papier sont limités. Le procédé est peu sensible à la précision du couchage; il permet l'application des couches minces sur des trames de papier se déplaçant à haute vitesse.

La vitesse caractéristique typique d'une PCF est d'environ 1000 m/min pour des faibles poids de couche ($10\text{--}12\text{ g/m}^2$ par côté) sur des papiers à base de pâte chimique et 1400 m/min pour des poids de couche moyens ($7\text{--}10\text{ g/m}^2$ par côté) sur des papiers à base de pâte mécanique.

La PCF comprend un rouleau de transfert et un rouleau de support. Ces rouleaux tournent à la même vitesse en mode contra-rotatif. La trame de papier se déplace dans l'entrefer d'application entre les deux rouleaux où elle entre en contact avec la couche qui a été précédemment appliquée sur le rouleau de transfert. La quantité de sauce de couchage sur les rouleaux de transfert est contrôlée par une barre de dosage tournant dans le même sens que le rouleau de transfert, mais à une vitesse plus faible. Dans les coucheuses de laboratoire, des rouleaux mous ou rigides peuvent être employés. Cependant, dans le cas des équipements industriels, une couche d'élastomère déformable recouvre souvent un des deux rouleaux.

La technologie de couchage par film présente beaucoup d'avantages dont la possibilité de travailler avec une vitesse de couchage élevée et d'utiliser des fluides fortement pigmentés. Cependant, comme elle est basée sur l'utilisation de rouleaux, des instabilités de type rides peuvent apparaître à la surface du film. Par ailleurs, à mesure que la vitesse des rouleaux augmente, le procédé est exposé à deux problèmes de machinabilité

trouvant leur origine dans l'entrefer d'application : le brouillard et la formation de peau d'orange.

Le brouillard est défini comme l'éjection de gouttelettes lors la séparation du film à la sortie de l'entrefer d'application. L'apparition de ce brouillard dépend de nombreux facteurs, mais il apparaît habituellement pour des vitesses très élevées. Bien que le brouillard n'affecte pas la qualité du couchage, il réduit le poids de la couche et représente une source très importante de pollution de l'air pour la machinerie et l'usine; en conséquence, des lavages fréquents de l'équipement sont nécessaires.

Étant donné le peu d'information disponible sur les mécanismes de formation de brouillard, le premier objectif de cette thèse est de déterminer les facteurs contrôlant le mécanisme de séparation du fluide à la sortie de l'entrefer d'application. Le deuxième objectif vise à comprendre le rôle que la rhéologie de sauces de couchage joue sur le phénomène de brouillard.

Pour atteindre ces objectifs, des essais expérimentaux ont été réalisés avec une coucheuse à film de laboratoire conçue et construite spécifiquement aux fins de la présente thèse. Le système comprend trois parties : le rouleau de transfert, le rouleau support et le système d'alimentation. Le rouleau de transfert est un cylindre recouvert d'une couche d'élastomère déformable.

Le rouleau de support est un cylindre rigide ayant une surface lisse chromée. Les deux rouleaux tournent à contre-sens et sont supportés à l'aide de roulements auto-alignés. Ils sont actionnés par un moteur électrique ayant chacun une puissance de 746 W. La vitesse des moteurs est soigneusement contrôlée par un variateur électronique de fréquence régulé en mode feedback par un encodeur de vitesse. Le contrôle de vitesse permet de fonctionner entre 0-1600 m/min et également de changer les pentes de décélération et d'accélération afin d'éviter une surcharge des moteurs en cas de fonctionnement à vitesse

élevée. Chaque rouleau est équilibré dynamiquement dans deux plans en trois points différents jusqu'à 6250 t/min. Une lame de raclage est utilisée sur chaque rouleau pour enlever l'excès du fluide. La charge appliquée entre les rouleaux peut être modifiée en utilisant un cylindre pneumatique attaché à une structure rigide. C'est cette structure qui supporte le rouleau de transfert.

La distribution de pression du fluide dans l'entrefer d'application est mesurée à l'aide d'un capteur dynamique de pression de 2,5 millimètres de diamètre installé à la surface du rouleau de support. La membrane de ce capteur a été soigneusement usinée et polie au même rayon du rouleau de support afin d'éviter les effets d'intrusion. Pendant une révolution du rouleau de support, divers profils sont obtenus, dont la moitié correspond à la pression dans l'entrefer d'application et l'autre à la pression de la lame de raclage sur le rouleau. Le capteur est capable de mesurer la pression entre 0-690 kPa avec une résolution de 0,069 kPa à une fréquence de 250 kHz.

Les données de la distribution de pression dans l'entrefer sont saisies par un ordinateur au moyen d'une bague collectrice de quatre circuits reliés à une carte d'acquisition de données. Les parties internes de la bague collectrice sont faites de métaux précieux qui réduisent au minimum le bruit et servent aux signaux d'instrumentation à bas niveau. La bague est installée à l'extrémité de l'axe du rouleau de transfert; les fils du signal venant du capteur de pression sont introduits dans l'axe du rouleau et un adaptateur est utilisé pour attacher le rotor de l'anneau glissant à l'axe de roulement.

Un système pressurisé est utilisé pour le stockage et l'alimentation du fluide vers le rouleau de transfert. Ce système permet d'assurer un débit de sauce constant pendant les expériences. Une vanne de dosage à boule permet de réguler le débit à la valeur requise et une chambre d'alimentation distribue uniformément le fluide le long de la surface à couler. L'épaisseur du fluide sur le rouleau de transfert est contrôlée à l'aide d'une lame de raclage placée à la sortie de la chambre d'alimentation.

Dans un premier temps, on a employé des solutions aqueuses de polyéthylène glycol à deux concentrations (20% et 30% massique), qui sont des fluides newtoniens inélastiques. Ces formulations ont été choisies pour leur stabilité rhéologique. La viscosité des fluides a été mesurée avec un rhéomètre AR-2000 en configuration Couette. La pression de vapeur de ces fluides a été calculée à l'aide de la fraction molaire de chaque concentration.

Un système vidéo à haute vitesse a été installé pour visualiser l'écoulement en aval de l'entrefer. Un laser de vapeur de cuivre sert de source de lumière pulsée (fréquence et durée des pulses variables). Les images sont obtenues avec une caméra vidéo CMOS de résolution 1024 x 1024 pixels carrés permettant d'obtenir jusqu'à 462 images/s à cette résolution. Un total de 150 images est obtenu pour chaque expérience à une vitesse d'enregistrement de 231 images/s. L'obturateur est réglé à 2,2 ms et un facteur d'agrandissement de 2 permet d'observer l'écoulement à la sortie de l'entrefer.

Contrairement à ce que mentionne la littérature scientifique sur l'enduction des adhésifs, aucun phénomène de cloison ("septum") n'a été observé. On a par contre noté la présence de filaments. Par ailleurs, on a trouvé que la dépression à la sortie de l'entrefer n'est pas suffisamment intense pour provoquer un phénomène de cavitation. Quand on applique une faible force entre les rouleaux, le brouillard peut être généré par deux mécanismes différents : la séparation du fluide et l'effet de pulvérisation dû à l'entraînement d'air dans l'entrefer d'application. L'entraînement d'air pourrait être évité en augmentant la force entre les rouleaux; mais on a noté que l'écoulement devenait périodique à mesure que la vitesse des rouleaux augmentait, ce qui pourrait mener à un couchage non uniforme. L'utilisation d'un élastomère plus rigide combiné à des forces entre les rouleaux plus élevées pourrait permettre d'éviter ces phénomènes.

Dans un deuxième temps, on a utilisé des suspensions et des sauces couchage. Quatre suspensions ont été préparées avec de l'argile delaminé dispersé dans du polyéthylène glycol et de l'eau. Deux de ces suspensions ont été préparées avec un épaississant, la carboxyméthyle cellulose (CMC). Les suspensions finales ont une teneur en matière solide totale de 52% et 62%. Par ailleurs, quatre sauces de couchage ont été formulées avec la même argile delaminé. Toutes les sauces ont été préparées avec de la CMC et un latex de styrène-butadiène. Le pH est ajusté entre 8,0 et 8,5.

Les filaments formés avec les fluides newtoniens présentent un profil en forme de "C" à la sortie de l'entrefer. Pour les suspensions, on a obtenu des profils de filaments droits. Le mode de rupture de ces filaments est également très différent; ils peuvent se casser en deux ou davantage de morceaux selon les conditions opératoires et la teneur en matière solide. Un filament de fluide peu concentré a tendance à se casser en un plus grand nombre de morceaux que dans le cas des suspensions plus concentrées. Par ailleurs, on a observé que la couche d'élastomère qui recouvre le rouleau de transfert est grandement déformée si l'on emploie une faible force entre les rouleaux à une vitesse de rotation très élevée, ce qui fait qu'une petite quantité d'air peut pénétrer dans l'entrefer d'application. Dans ces conditions, le brouillard est généré simultanément par la séparation du fluide et par un effet de pulvérisation dû à l'entraînement d'air. En ce qui concerne les profils de pression, on a observé la même tendance que pour les fluides newtoniens, à savoir que plus la vitesse de rouleaux est importante, plus le pic de pression est élevé. Le même effet a été observé à la sortie de l'entrefer; cependant le niveau de dépression observé pour les suspensions et les sauces de couchage est moins élevé que pour les fluides newtoniens. Dans ce cas, les forces visqueuses pour les fluides pigmentés deviennent plus petites et l'air éprouve moins de difficulté à pénétrer dans l'entrefer d'application.

Le deuxième objectif de ce travail consiste à clarifier le rôle de la rhéologie dans la genèse du brouillard. Dans cette optique, une série des tests en cisaillement en régime permanent et oscillatoire ont été réalisés au moyen de la géométrie des cylindres

concentriques (écoulement Couette). Toutes les suspensions et les sauces de couchage ont présenté un comportement rhéofluidifiant à des vitesses de cisaillement moyennes. À des vitesses de cisaillement plus élevées, le comportement rhéologique est généralement rhéoépaississant et cet effet devient plus prononcé à mesure que le contenu de matière solide augmente. En ce qui concerne les tests oscillatoires, les suspensions et les sauces de couchage montrent un comportement viscoélastique à de très faibles déformations. Mais à des déformations très élevées, l'effet élastique devient moins important que l'effet visqueux. Dans tous les cas, pour réduire les effets élastiques, il est très recommandable de maintenir la teneur de liant aussi faible que possible lors de la préparation de la sauce de couchage.

Un rhéomètre à orifice a également été utilisé pour caractériser la viscosité élongationnelle des différentes formulations. Ce rhéomètre à orifice comprend deux cylindres en acier de 2 m de long chacun et d'un diamètre intérieur égal à 0,0762 m. Le liquide à tester est placé dans un cylindre. Les cylindres sont séparés par un disque en acier de 12,7 mm d'épaisseur possédant un petit orifice au centre. On dispose au laboratoire de plusieurs orifices de diamètres différents, tous composés d'une section convergente et divergente avec des angles de 45°. Lors d'un test, le fluide est pompé d'un cylindre à l'autre jusqu'à ce que le premier soit vide. Ce processus est ensuite inversé. La perte de pression est mesurée grâce à un capteur dynamique de pression différentielle localisée aux extrémités de l'orifice. Cette localisation permet d'accéder directement à la pression qui règne dans chaque cylindre. On suppose en fait que la perte de pression dans les réservoirs est négligeable. Trois différents capteurs sont disponibles pour mesurer la chute de pression et ils peuvent être choisis en fonction de la résolution et de la plage de pression (1% @ 0-70 kPa, 1% @ 0-345 kPa et 2% @ 0-1380 kPa). Le fluide est mis en mouvement à l'aide d'un système de pistons hydrauliques. Le circuit hydraulique qui contrôle le déplacement des pistons a été spécialement choisi pour assurer des débits très stables. La vitesse des pistons est contrôlée par des capteurs de déplacement (LVDT) localisés aux extrêmes de chaque cylindre.

La configuration de ce rhéomètre a deux caractéristiques intéressantes : premièrement, il est possible d'employer le même échantillon liquide pour une série de tests car le mouvement est réversible, et deuxièmement, on peut étudier la dégradation mécanique dans des conditions de taux de déformation élevé parce que ce même fluide est réutilisable indéfiniment. On peut faire fonctionner le rhéomètre à orifice manuellement ou automatiquement à partir d'un ordinateur. L'ordinateur permet d'enregistrer toute l'information sur la perte de pression et sur le débit du fluide à travers l'orifice.

Le rhéomètre a été initialement étalonné en traçant la courbe du nombre d'Euler en fonction du nombre de Reynolds pour des fluides newtoniens au moyen des données de la perte de pression et du débit du fluide à l'orifice. La courbe d'étalonnage est alors employée pour déterminer la viscosité élongationnelle des suspensions et des sauces de couchage.

La viscosité élongationnelle des suspensions et des sauces de couchage a été mesurée dans une plage de taux de cisaillement de 10^3 - $12,5 \times 10^3 \text{ s}^{-1}$. On a observé que les suspensions atteignent une viscosité de plateau pour des taux de déformation élevés. Par contre, les sauces de couchage présentent d'abord un comportement rhéofluidifiant en élongation puis deviennent écrouissants. Finalement, il est intéressant de comparer la viscosité élongationnelle à la viscosité de cisaillement au moyen du rapport de Trouton.

Un rapport de Trouton de 5 à 12 a été obtenu pour les suspensions. Les résultats avec les sauces de couchage sont très différents puisque des rapports de Trouton de 10 à 85 ont été calculés. On explique ces différences par les interactions entre les composants employés dans la préparation des sauces de couchage.

Bien qu'il soit pratiquement impossible d'obtenir un écoulement élongationnel sans cisaillement aux taux de déformation élevés, le concept de pression d'entrée utilisé dans le rhéomètre à orifice est considéré comme la méthode la plus simple. À partir des mesures de la perte de pression et du débit, on peut obtenir des données d'élongation pour une plage de taux de déformation et de viscosité élevée. Cependant, il est important de ne pas perdre de vue la structure complexe de l'écoulement d'entrée, particulièrement pour les suspensions concentrées et les fluides viscoélastiques. Pour cette raison, d'autres recherches seraient nécessaires pour évaluer les hypothèses dans l'analyse de l'écoulement à travers un petit orifice et ainsi démontrer l'intérêt de cette méthode simple.

En résumé, cette recherche permet de mieux comprendre l'hydrodynamique de l'écoulement dans l'entrefer d'application d'une PCF, les mécanismes de génération du brouillard et le rôle que la rhéologie joue dans le procédé. On a observé que la couche d'élastomère du rouleau de transfert peut être grandement déformée à des vitesses de rotation très élevées. De plus, si on utilise une faible force entre les rouleaux, une petite quantité d'air peut entrer dans l'entrefer d'application, ce qui est une des causes du développement du brouillard. En fait, deux mécanismes différents peuvent être envisagés pour expliquer la génération du brouillard : la séparation du fluide à la sortie de l'entrefer et l'effet d'atomisation dû à l'entraînement d'air dans l'entrefer. Par ailleurs, l'entraînement d'air peut être évité avec une plus grande force entre les rouleaux, mais dans ces conditions l'écoulement peut devenir périodique. Une façon de supprimer en même temps l'entraînement d'air et l'écoulement périodique serait de combiner des élastomères plus durs sur les deux rouleaux à des forces plus élevées entre eux. En termes rhéologiques, on observe que la région de viscosité linéaire diminue à mesure que le contenu de matière solide augmente, ce qui signifie que les effets élastiques peuvent être négligés en considérant les conditions opératoires utilisées ici. Les tests de rhéologie élongationnelle montrent que le rapport de Trouton obtenu pour les suspensions et les sauces de couchage est plus élevé qu'avec des fluides newtoniens. Finalement, on a vérifié que les sauces de couchage présentant une viscosité élongationnelle plus élevée

donnent une meilleure machinabilité. Cependant, la taille des gouttelettes éjectées à la sortie de l'entrefer est plus petite pour ce type de sauces de couchage, ce qui pourrait causer des problèmes de qualité de l'air dans l'usine. Par ailleurs, même si une meilleure performance de procédé de couchage peut être obtenue avec des formulations ayant un contenu en matière solide et une viscosité élongationnelle plus élevés, il y a risque de formation de peau d'orange. Un compromis est donc nécessaire.

Les résultats obtenus dans cette thèse ouvrent de nouvelles perspectives de recherche. Il serait intéressant d'étudier l'hydrodynamique dans l'entrefer d'application et le rôle que la rhéologie des fluides viscoélastiques joue dans le couchage. Un autre axe de recherche important serait l'étude numérique du procédé de transfert à haute vitesse. Il pourrait permettre d'associer les profils de pression et les profils de vitesse aux phénomènes produits à haute vitesse de procédé, tel que le brouillard. Cependant, la difficulté d'imposer des conditions frontières sur deux surfaces libres constitue un vrai défi.

INDEX

	Page
Resumé	vi
Abstract	ix
Condensé en français	xii
Index	xxii
List of tables	xxv
List of figures	xxiv
Nomenclature	xxxix
1. Introduction	1
1.1 Coating methods	1
1.2 Defects	3
1.3 Coating colors formulations	6
1.4 General objective	7
2. Literature survey	8
2.1 Forward roll coating	8
2.1.1 Rigid gaps	8
2.1.2 Deformable gaps	12
2.1.3 Mechanisms of film splitting	14
2.2 Misting	18
2.3 Rheology of suspensions and coating colors	27
2.3.1 Shear rheology	28

	xxiii
2.3.2 Extensional rheology	30
2.4 Specific objectives	31
3. Experimental set-up	32
3.1 Laboratory film coater	32
3.1.1 Pressure transducer	33
3.1.2 Slip ring	35
3.1.3 Fluid delivery system	36
3.2 Orifice flowmeter	37
4. Forward deformable roll coating at high speed with Newtonian fluids	39
4.1 Abstract	40
4.2 Introduction	40
4.3 Materials and methods	44
4.3.1 Apparatus	44
4.3.2 High-speed imaging	47
4.3.3 Test conditions	47
4.4 Results	48
4.4.1 Pressure profiles	48
4.4.2 Imaging analysis	52
4.5 Conclusions	60
5. Non-Newtonian effects on forward deformable roll coating at high speed	61
5.1 Abstract	62
5.2 Introduction	62
5.3 Materials and Methods	65
5.3.1 Film coater	65
5.3.2 High-speed video	67
5.3.3 Fluids	67
5.4 Rheology	72

5.4.1 Steady state shear	
72	
5.4.2 Oscillatory shear	74
5.4.3 Extensional rheology	77
5.5 Image analysis	82
5.6 Pressure profiles	85
5.7 Analysis	89
5.8 Concluding remarks	93
6. Orifice flowmeter for measuring extensional rheological properties	94
6.1 Abstract	95
6.2 Introduction	95
6.3 Description of the flowmeter	98
6.4 Operating principle	100
6.5 Materials and methods	105
6.5.1 Calibration	105
6.5.2 Fluids	106
6.6 Results and discussion	107
6.6.1 Calibration	107
6.6.2 Coating color characterization	108
6.7 Conclusions	114
7. Conclusions and recommendations	115
References	118

LIST OF TABLES**Chapter 4**

Table 4.1 Laboratory Coater Characteristics

Table 4.2 Test conditions

Chapter 5

Table 5.1 Composition and physical properties of suspensions

Table 5.2 Composition and physical properties of coating colors

Table 5.3 Power-law parameters from steady state shear and key
parameters from strain sweep tests.

Table 5.4 Pressure ratio of suspensions and coating colors.

Chapter 6

Table 6.1 Coating color composition

Table 6.2 Power law parameters for coating colors

LIST OF FIGURES

Chapter 1

- Figure 1.1 Metered Film Coater.
- Figure 1.2 (a) An example of a rib pattern (Réglat, 1997); (b) Photograph of the cascade instability on the metered film (top view, roll surface moves from bottom right to top left). The irregular cross-web waves are regularly spaced in the downweb direction (Kistler and Schweizer, 1997).
- Figure 1.3 Example of printed strips (Roper et al, 1999)
- Figure 1.4 Bubble with streak (Guttoff and Cohen, 1995).
- Figure 1.5 (a) Example of misting (Roper et al, 1997); (b) Coating color spitting at the metering rod nip outlet (Grön et al, 1998).

Chapter 2

- Figure 2.1 Effect of Ca on flow field: (a) 0.1; (b) 0.2; (c) 0.5 (Coyle et al, 1982).
- Figure 2.2 Flow structures in the bead of a meniscus roll coater for $R/H_0 = 100$ and $\lambda_2 = 0.075$: (a) $S = 1/2$, $Ca_2 = 2.25 \times 10^{-3}$; (b) $S = 1$, $Ca_2 = 1.80 \times 10^{-3}$; (c) $S = 2$, $Ca_2 = 7.0 \times 10^{-4}$ (Gaskell et al, 1995).
- Figure 2.3 A sequence of dye traces showing the path of the primary transfer jet through the bead for forward-mode meniscus coating ($R/H_0 = 68$, $S = 0.7$, $\lambda_1 = 0.076$, $Ca_2 = 8.0 \times 10^{-3}$): time $t =$ (a) 1 s, (b) 2 s, (c) 3 s from when the primary transfer jet first enters the bead. The broken vertical line indicates the minimum nip position (Gaskell et al, 1998).
- Figure 2.4 Laser illuminated particles for the case of forward-mode

meniscus roll coating: $R/H_0 = 100$, $S = 1.0$, $\lambda_i = 0.048$, $Ca_2 = 2.3 \times 10^{-3}$. The image, shown in negative form, is made up from eight frames, grabbed sequentially and added together electronically. The broken vertical line indicates the minimum nip position (Gaskell et al, 1998).

- Figure 2.5 Sketch of film-split meniscus. The curvature of the meniscus gradually changes from a constant curvature to a plane film (Carvalho and Scriven, 1997).
- Figure 2.6 (a) Flow between two counter-rotating rolls (Agassant et al, 1991); (b) Pressure distribution within nip and in region beyond nip exit (MacPhee, 1997a).
- Figure 2.7 Effect of transfer roll speed (a) and viscosity (b) on pressure profile (Réglat and Tanguy, 1997).
- Figure 2.8 Change in character of film splitting in the nip exit versus speed (MacPhee, 1997a).
- Figure 2.9 (a) Depiction of the smooth film splitting that occurs at very low roll surface speeds (MacPhee, 1997a); (b) stages in the formation of cavities and filaments during at high roll speed (MacPhee, 1997b).
- Figure 2.10 Effect of base paper on misting and orange peel (Roper et al, 1999).
- Figure 2.11 Video images of misting at 1200 and 1500 m/min on wood free base paper. Top: uncalendered; Bottom: calendered (Roper et al, 1997).
- Figure 2.12 Photomicrographs (10X) of droplets collected at 1200 m/min (left) and 1500 m/min (right) from uncalendered wood free base paper (Roper et al, 1997).
- Figure 2.13 Effect of solids on misting and orange peel (Roper et al, 1999).
- Figure 2.14 Effect of CMC level on misting and orange peel formation (Roper et al, 1999).
- Figure 2.15 Step shear experiment (Roper et al, 1999).

- Figure 2.16 Effect of leveling (Roper et al, 1999).
- Figure 2.17 Effect of high solids, thickener and latex (Roper et al, 1998).
- Figure 2.18 Effect of coat weight on misting (Salminen et al, 1996).
- Figure 2.19 Coating adhesion dependence on misting and machine speed (Grön et al, 1998).
- Figure 2.20 Misting amount as a function of transfer ratio at 1950 m/min (Grön et al, 1998).

Chapter 3

- Figure 3.1 General view of the film coater
- Figure 3.2 Pressure sensor: (a) General view; (b) Mounting.
- Figure 3.3 Slip ring mounting.
- Figure 3.4 Feeding chamber.

Chapter 4

- Figure 4.1 Metered film coater principle
- Figure 4.2 Forward deformable roll coating array with negative gap
- Figure 4.3 Experimental set-up
- Figure 4.4 Pressure profiles for PEG20 ($\mu = 0.13 \text{ Pa}\cdot\text{s}$), $U = 500, 800$ and 1100 m/min : (a) 2 kN/m ; (b) 4 kN/m
- Figure 4.5 Pressure profiles for PEG30; $\mu = 0.63 \text{ Pa}\cdot\text{s}$, $U = 500, 800$ and 1100 m/min : (a) 2 kN/m ; (b) 4 kN/m
- Figure 4.6 Images from the nip with PEG 20 ($\mu = 0.13 \text{ Pa}\cdot\text{s}$) at 2 kN/m : (a) 500 m/min ; (b) 800 m/min ; (c) 1100 m/min
- Figure 4.7 Images from the nip with PEG 30 ($\mu = 0.63 \text{ Pa}\cdot\text{s}$) at 2 kN/m : (a) 500 m/min ; (b) 800 m/min ; (c) 1100 m/min
- Figure 4.8 Images from the nip with PEG 20 ($\mu = 0.13 \text{ Pa}\cdot\text{s}$) at 4 kN/m : (a) 500 m/min ; (b) 800 m/min ; (c) 1100 m/min

- Figure 4.9 Images from the nip with PEG 30 ($\mu = 0.63 \text{ Pa}\cdot\text{s}$) at 4 kN/m:
(a) 500 m/min; (b) 800 m/min; (c) 1100 m/min
- Figure 4.10 Sequence of six images downstream the nip with PEG 30 ($\mu = 0.63 \text{ Pa}\cdot\text{s}$) at 4 kN/m and 800 m/min
- Figure 4.11 Negative region of pressure profile for PEG 30 at 4 kN/m

Chapter 5

- Figure 5.1 Laboratory film coater.
- Figure 5.2 Forward deformable roll coating arrangement with negative gap.
- Figure 5.3 Experimental set-up.
- Figure 5.4 Steady shear viscosity as function of shear rate for (a) suspensions and (b) coating colors.
- Figure 5.5 Dynamic moduli as functions of strain amplitude for (a) suspensions and (b) coating colors.
- Figure 5.6 Extensional viscosity as function of strain rate for (a) suspensions and (b) coating colors.
- Figure 5.7 Trouton ratio for (a) suspensions and (b) coating colors.
- Figure 5.8 Images downstream the nip with 10PEG52: (a) $Ca_E = 280$; (b) $Ca_E = 280$; (c) $Ca_E = 620$.
- Figure 5.9 Images downstream the nip with 10PEG62: (a) $Ca_E = 120$; (b) $Ca_E = 240$.
- Figure 5.10 Pressure profiles for suspensions at $W = 3 \text{ kN/m}$ and $U = 500$, 800 and 1100 m/min: (a) 10PEG52; (b) 10 PEG62; (c) 5PEG52-CMC; (d) 5 PEG62-CMC.
- Figure 5.11 Pressure profiles for suspensions at $W = 3 \text{ kN/m}$ and $U = 500$, 800 and 1100 m/min: (a) C52L8; (b) C62L8; (c) C52L12; (d) C62L12.
- Figure 5.12 Roll profiles versus X/R as a function of the elasticity number

(Es_E).

Figure 5.13 Experimental and predicted pressure profiles for $Es_E = 7.8 \times 10^{-6}$.

Chapter 6

Figure 6.1 Orifice plate

Figure 6.2 Experimental set-up: a) Hydraulic system; b) General configuration of the orifice flowmeter

Figure 6.3 Flow through a conical die.

Figure 6.4 Calibration curve for Newtonian fluids

Figure 6.5 Shear viscosity data for the coating colors

Figure 6.6 Extensional viscosity data for the coating colors

Figure 6.7 Orifice flow data for coating colors

Figure 6.8 Extensional viscosity/shear viscosity ratio as a function of effective strain rate

NOMENCLATURE

A	area, (m^2)
d	diameter, (mm)
e	thickness, (mm)
E	dissipated energy, (W)
E	Young modulus (N/m^2)
E_c	Cohesion energy, (mJ/m^3)
Eu	Euler number
G'	Storage modulus (Pa)
G''	Loss modulus (Pa)
h_n	Dimensionless nip gap
H_r	Nip gap (m)
l	extensional behavior index, (Pa.s^l)
m	shear consistency index, (Pa.s^m)
M_w	Molecular weight (g/mol)
n	shear behavior index
Ne	Modified elasticity number
p_e	Dimensionless pressure
P	pressure, (Pa)
P_v	Vapor pressure (kPa)
Q	Flow rate, (L/s)
q	Dimensionless flow rate
r	radius, (mm)
R	Roll radius (m)
Re	Reynolds number
t_c	Contact time (μs)
t	extensional consistency index
Tr	Trouton ratio
U	Roll speed (m/s)

v	velocity, (m/s)
W	Applied load (kN/m)
x	Dimensionless flow coordinate

Greek Symbols

δ	Nip length (μm)
γ_c	Critic strain
$\dot{\gamma}$	shear rate, (s^{-1})
$\bar{\dot{\gamma}}$	effective strain rate, (s^{-1})
Δ	Difference
$\dot{\epsilon}$	extensional rate, (s^{-1})
λ	Characteristic elastic time, (s)
η	shear viscosity, (Pa.s)
η_∞	High-shear rate viscosity, (Pa.s)
μ	Newtonian viscosity, (Pa.s)
ρ	Density, (kg/m^3)
ϕ	half-angle of convergence, (degrees)

Subscripts

<i>act</i>	Actual
<i>ave</i>	average
<i>corr</i>	corrected
<i>exp</i>	experimental
<i>E</i>	extensional
<i>en</i>	Entry
<i>K</i>	Kinetic

max maximum

S Shear

the theoretical

0 orifice

1 Pipe

CHAPTER 1

INTRODUCTION

1.1 Coating methods

In the 1850's, the first surface treatment of a web with clay coating was accomplished with the brush coater. The end product was a coated wallpaper that improved the brightness and printing quality patterns (Booth, 1970). Shortly after the introduction of the brush coater, its use expanded to label stock and other high grade painting papers. The development of coating machinery however started in this century with the replacement from hand painting shade cloth by moving brush. Brush coater machine speeds seldom exceeded 120 m/min. One of its main disadvantages is that brushes wear rapidly, thus requiring replacement or trimming to make them perform satisfactorily. Because of the speed limitation of the cumbersome operation of the brush coater and the fragility of the brushes, the air doctor coater eventually replaced it.

With the advent of the air doctor coater in the 1930's, the modern paper coating industry was born. It was the introduction of the air doctor coater under a patent held by the S.D. Warren Co. (Booth, 1970) that revolutionized paper coating and stimulated its growth in tonnage. Operating speeds increased from about 120 m/min to 300 m/min. Not only were speeds increased but also downtime was greatly reduced, particularly for wash-ups when compared to brush coatings. The new machines ran more reliably and improved quality.

In 1935 the roll coater was introduced. It has been used since many years for every conceivable type of coating and product. The roll coater was developed to deposit a coating or adhesive to the surface of paper to bond the fibers and is currently one of the most common coater machines.

In the 1950's appeared a new coating method, the blade coating. The method involves applying excess of a coating mix and then scraping the surplus away (similar to buttering bread or plastering a wall). Blade coating has been tested up to 2,500 m/min (Pauksta, 1998) but due to the centrifugal force, the web is subjected to high tension and the tendency of web breaks causing downtime increase.

In recent years the film coater has converted into a very useful coating equipment by using metering devices. The MFC principle is shown in figure 1. The equipment consists of a transfer roll and backing roll. These rolls rotate at the same speed in a counter-rotating mode. The paper web is fed to the transfer nip (application nip) between the two rolls and picks up the coat that has been previously applied to the transfer roll. The amount of coating color on the transfer roll is controlled by a metering rod or bar co-rotating with the transfer roll, but at a much lower speed.

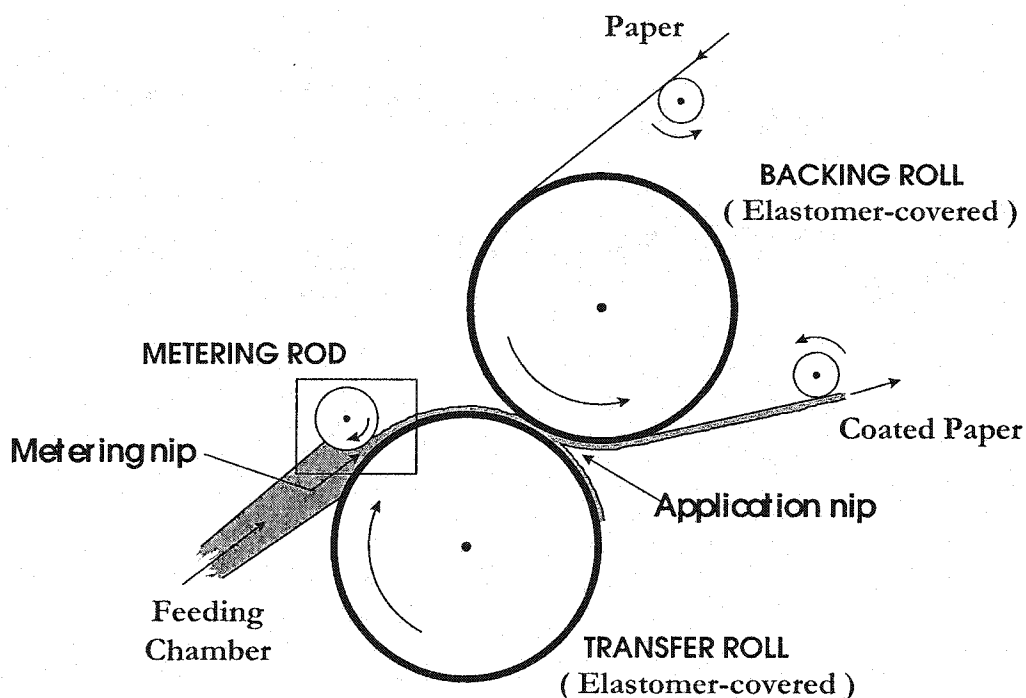


Figure 1.1 Metered Film Coater.

The MFC can be thought as a simple roll coater and in common with all roll coaters there is a tendency towards ribbing and patterning. As the web leaves the nip of the metered film coater after the application of the coating, the thin film splits, forming small coating filaments, which break and bounce back to the two diverging surfaces. These phenomena generate small droplets, which are thrown out of the nip exit and the coating color starts to "mist".

1.2 Defects

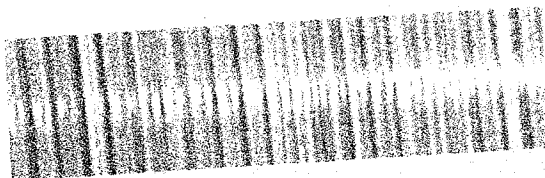
Coating process is subjected to many spurious phenomena that can lead to a wide variety of defects. A general classification of physical defects has been proposed (Guttoff and Cohen, 1995). The physical defects have been divided in two categories, namely:

a) Continuous defects

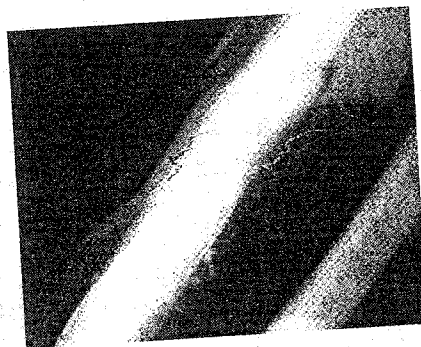
- Ribbing is the formation of lines running down the web and very uniformly spaced across the web (Figure 1.2a)
- Chatter is appearance of uniform cross-web or band of different coverage.
- Cascade is a periodic cross-web disturbance with a sawtooth pattern (Figure 1.2b).

b) Discrete defects

- Orange peel is a rough finish in the dried coating that is generally associated with surface-tension-driven flows (Figure 1.3).
- Mottle is an irregularly pattern defect that can be gross or quite subtle.
- Bubbles as a consequence of air entrapment in the coating (Figure 1.4).

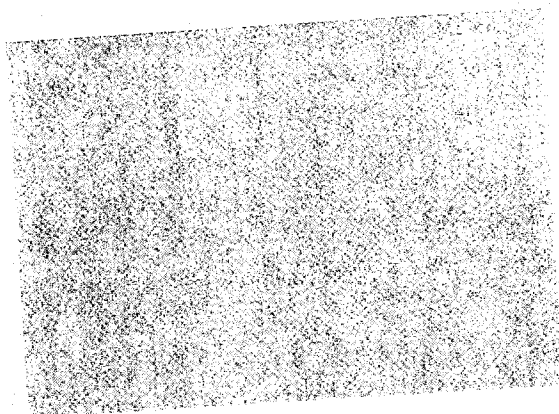


(a)

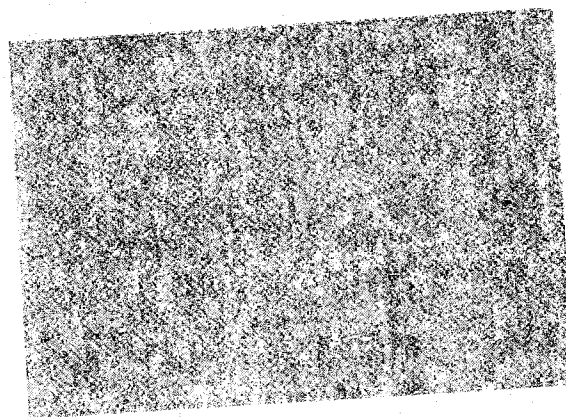


(b)

Figure 1.2 (a) An example of a rib pattern (Réglat and Tanguy, 1997); (b) Photograph of the cascade instability on the metered film (top view, roll surface moves from bottom right to top left). The irregular cross-web waves are regularly spaced in the downweb direction (Kistler and Schweizer, 1997).



Good orange peel



Bad orange peel

Figure 1.3 Example of printed strips (Roper et al, 1999).

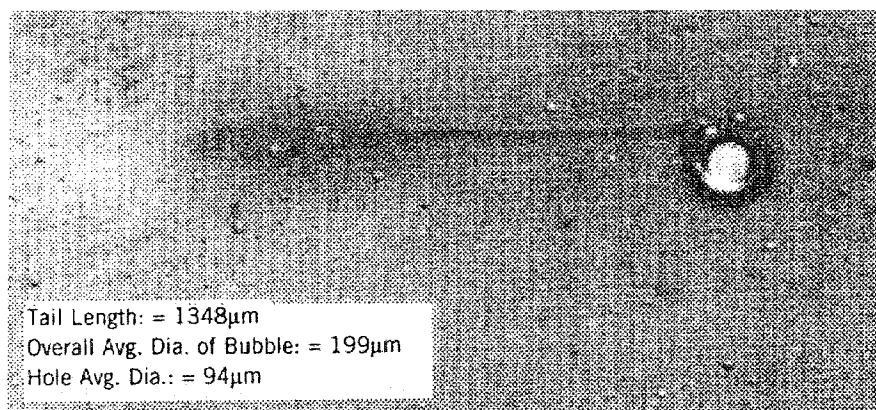
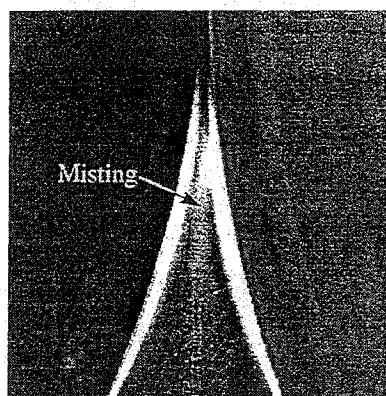
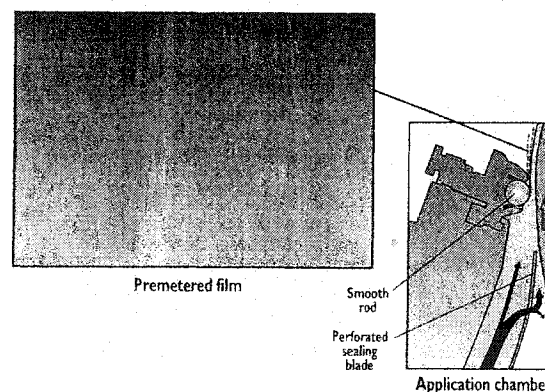


Figure 1.4 Bubble with streak (Gutoff and Cohen, 1995).

Two other phenomena, not considered as physical defects, generate runnability problems in MFC: misting and spitting. As defined before, misting is the ejection of small droplets in the film splitting at the exit of transfer nip and spitting is the ejection of large droplets from the metering nip. Misting should not affect the quality of the coating, although it might reduce the coverage. Figure 1.5 shows an example of misting and spitting in a MFC.



(a)



(b)

Figure 1.5 (a) Example of misting (Roper et al, 1997); (b) Coating color spitting at the metering rod nip outlet (Grön et al, 1998).

1.3 Coating colors formulations

The coating color is prepared to provide the desired mechanical, optical, and printability properties to the coated paper. A wide variety of coating color formulations can be applied to a paper web depending on the type of paper and the coating method among others. A typical formulation consists at least of three ingredients: pigments, water, and binder. Both kaolin clay in North America and calcium carbonate in Europe are currently the most common pigments and styrene/butadiene is the most used binder.

Because of the presence of suspensions and polymers coating colors behave as non-Newtonian fluids of very complex rheology. However, at industrial level there is the tendency to ignore their complexity and then they are usually treated as simple rheology fluids.

During metering film coating the coating color is submitted to very complex rheological behaviors such as thixotropy, shear-thinning, shear-thickening or dilatancy and viscoelasticity. Thus, the characterization of the rheological properties in the laboratory is necessary to mimic the flow process. Coating color is submitted to high shear rates and high extensional rates (of the order of 10^6 s^{-1}) in the metering nip and in the transfer nip, respectively. However, this shear or deformation rates are of extremely short duration (around 1 ms). After a rapid change in the shear rate, the coating needs some time to reach its steady-state, which sometimes is not possible specially when operating at high speed.

Finally, coating color is pressed onto the paper and water may migrate from the color to the paper yielding inhomogeneities in the fluid, which induces changes in the rheological properties. Therefore, dewatering properties play also an important role during coating process. Accordingly, a better understanding of the interaction of the coating color and paper is needed.

1.4 General objective

Metering film coating (MFC) has become a profitable and flexible technology for paper coating. Although the process is currently in operation the phenomena produced during the process, especially in the application nip have not been well understood. Nowadays, many efforts have been made for reducing the effects of those phenomena. However, results from these studies are generally lacking of scientific background.

The general objective of this work is to develop a state of knowledge on the film splitting process in the transfer nip for a better understanding of misting occurrence. The following points will be dealt:

- Factors that govern these phenomena
- the role that rheology plays in the process

More focused objectives will be defined after the literature analysis.

CHAPTER 2

LITERATURE SURVEY

2.1 Forward roll coating

2.1.1 Rigid gaps

Banks and Mill (1954) carried out one of the first studies of liquid flow between rotating rolls. They reported direct observations for cavitation in liquid films. Experiments were conducted starting from rest up to a certain speed. At moderate speeds spaced bubbles appear, but at higher speeds these bubbles pass over gradually to a continuous cavity and finally they break up due to a pressure drop of liquid.

The film-splitting profile plays an important role in determining the effects of phenomena generated during the process, especially in forward roll action. A numerical analysis from Coyle et al (1982) reveals a symmetric film splitting in which a “double-eddy” structure is generated. As Figure 2.1 shows, eddies shrink and tend to disappear when increasing the capillary number.

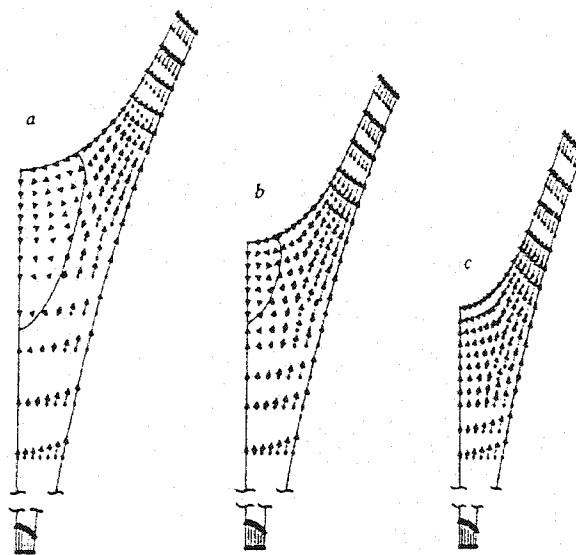


Figure 2.1 Effect of Ca on flow field: (a) 0.1; (b) 0.2; (c) 0.5 (Coyle et al, 1982).

A similar phenomenon was observed in dip coating by Tanguy et al (1984). From a numerical analysis of dip coating process, they noticed the displacement of the free surface between two consecutive iterations, which is clearly observed when the web is suddenly accelerated. The shape and position of the stagnation point of the free surface is highly dependent on the fluid properties as well as the speed ratio.

Gaskell et al (1995) developed three mathematical models for meniscus roll coating, with rigid rolls in which there is a two-dimensional and steady flow of a Newtonian fluid in the narrow nip between two counter-rotating rolls. These models are so-called zero flux, the small flux and the last one that incorporates the full effects of curved menisci and nonlinear free surface boundary conditions.

The finite element method reveals a "double-eddy" structure with eddy thickness dependent on the roll speeds ratio and a constant pressure gradient within the central core and two eddies, which was predicted by the zero flux model (see Figure 2.2). In the last figure R is the roll radius, H_0 is the nip half width, λ is a dimensionless flow rate, S is the roll speeds ratio and Ca is the capillary number. Subscript 2 is related to the lower roll.

These eddies were observed by Gaskell et al (1998), who developed an experimental technique for flow visualization by means of dye injection and particle imaging. To do that, some parameters were used to describe the coating regimes. The non-dimensional flow rate is given by

$$\lambda = \frac{H_i}{2H_0} \quad (2.2)$$

where H_i is the inlet film thickness and $2H_0$ is the minimum nip width.

The roll speed ratio (S) is defined by

$$S = \frac{U_1}{U_2} \quad (2.3)$$

where U_1 and U_2 are the peripheral speeds of upper and lower rolls, respectively. The effective roll radius $R = 2(R_1^{-1} + R_2^{-1})^{-1}$ and finally the capillary number Ca_2 , which was earlier defined.

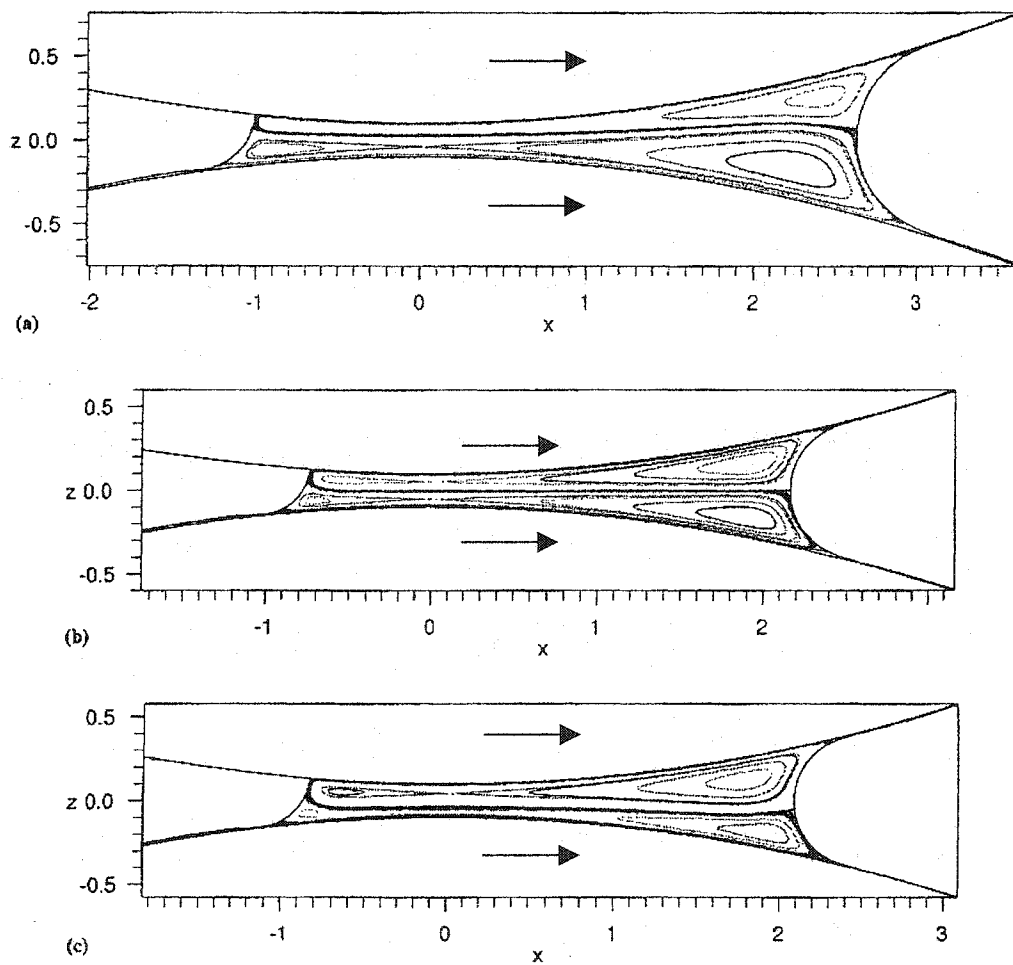


Figure 2.2 Flow structures in the bead of a meniscus roll coater for $R/H_0 = 100$ and $\lambda_2 = 0.075$: (a) $S = 1/2$, $Ca_2 = 2.25 \times 10^{-3}$; (b) $S = 1$, $Ca_2 = 1.80 \times 10^{-3}$; (c) $S = 2$, $Ca_2 = 7.0 \times 10^{-4}$ (Gaskell et al, 1995).

By injecting dyed liquid through a hypodermic needle into the upstream flow and viewing its passage through the nip, flow structure was highlighted. Figure 2.3 shows a sequence of dye traces. It can be observed that the incoming flux (lower left corner) travels along the bead to the downstream meniscus where it splits and a part continues with the lower roll and the remainder turns to flow in the reverse direction between two large counter-rotating eddies and jet turns once again at the upstream meniscus.

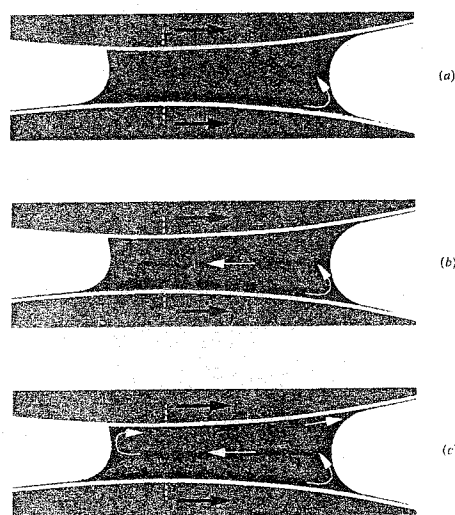


Figure 2.3 A sequence of dye traces showing the path of the primary transfer jet through the bead for forward-mode meniscus coating ($R/H_0 = 68$, $S = 0.7$, $\lambda_i = 0.076$, $Ca_2 = 8.0 \times 10^{-3}$): time $t =$ (a) 1 s, (b) 2 s, (c) 3 s from when the primary transfer jet first enters the bead. The broken vertical line indicates the minimum nip position (Gaskell et al, 1998).

In order to observe the flow details within the eddies 8 μm diameter hollow glass spheres were seeded throughout the liquid. A pulsed diode laser was used to illuminate a thin slice through the flow field. A particle image of the same flow field, reveals the existence of two sub-eddies with each of the large counter-rotating eddies (see figure 2.4).

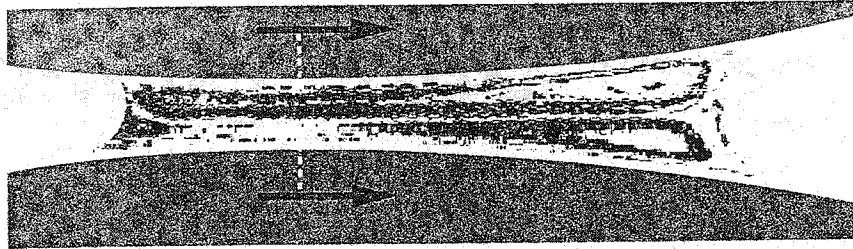


Figure 2.4 Laser illuminated particles for the case of forward-mode meniscus roll coating: $R/H_0 = 100$, $S = 1.0$, $\lambda_i = 0.048$, $Ca_2 = 2.3 \times 10^{-3}$. The image, shown in negative form, is made up from eight frames, grabbed sequentially and added together electronically. The broken vertical line indicates the minimum nip position (Gaskell et al, 1998).

2.1.2 Deformable gaps

Coyle (1984, 1988) carried out a dimensionless analysis on forward roll coating with a deformable gap. He studied the role of forces involved in the flow in the deformable nip, and he obtained a theoretical correlation for estimating the coating thickness. The validity of this correlation was experimentally confirmed by Cohu and Magnin (1997) in a study in which it was determined that the elastic modulus of rubber materials in cylinders may be time-dependent. Coyle (1984, 1988) proposed a one-dimensional elastohydrodynamic model suitable for small deformations for predicting the pressure profile between forward rolls.

Carvalho and Scriven (1994, 1997) and Carvalho (1996) analyzed the effect of a deformable gap on roll coating. They confirmed the results obtained by Coyle with a one-dimensional model to describe the roll deformation. They also proposed a Hookean two-

dimensional model for predicting the roll profile and the pressure distribution with both positive and negative gaps when the roll is highly deformed.

Carvalho and Scriven (1997) described the flow between a rigid and deformable counter-rotating roll. They determined that the curvature of the meniscus (R_m) gradually changes from a constant curvature to a plane film (Figure 2.5) and the meniscus position X_m is given by

$$H(X_m) = 2H_0 + \frac{X_m^2}{R} + D(X_m) = 1.644 \frac{Q}{2V} \frac{1+S}{1+S^{2/3}} (1+S^{2/3}) + R_m \left\{ \frac{1}{1+0.414(6CaS/(1+S))^{2/3}} + \frac{1}{1+0.414(6Ca/(1+S))^{2/3}} \right\} \quad (2.4)$$

where

$$R_m = \frac{1}{1.34Ca^{2/3}} \frac{Q}{2V} \frac{(S+1)}{1+S^{5/3}} \left(\frac{S+1}{2} \right)^{2/3} \quad (2.5)$$

and

$$S = \frac{V_A}{V_B} \quad (2.6)$$

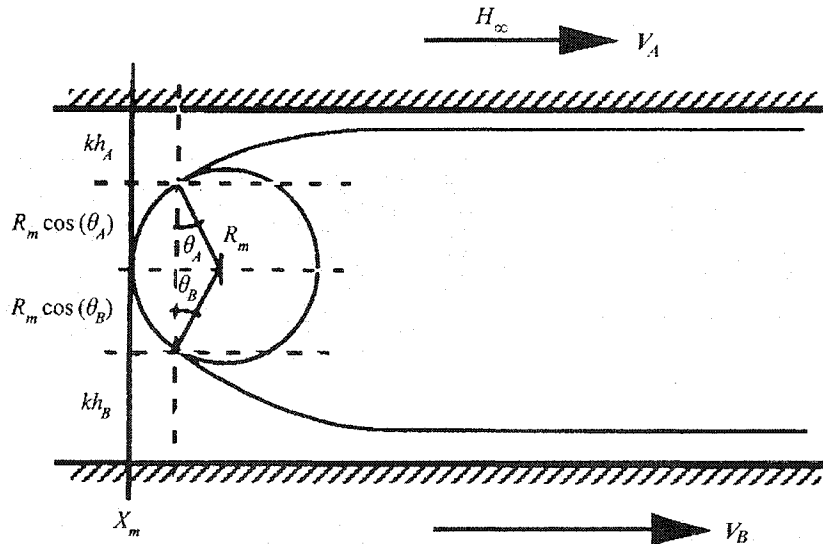


Figure 2.5 Sketch of film-split meniscus. The curvature of the meniscus gradually changes from a constant curvature to a plane film (Carvalho and Scriven, 1997).

2.1.3 Mechanism of film splitting

Film splitting is a very complex phenomenon dependent on many factors, such as roll speed, nip gap, coating color rheology and properties, web characteristics, etc. A full understanding of film splitting mechanism is the key to establishing strategies to reduce runnability issues such as misting and orange peel formation.

The film splitting action takes place at some small distance from the nip exit as a result of viscous pumping action whereby the fluid is drawn in separate directions by the diverging roll surface. During the process, the coating color is submitted to compressive and tensile forces equivalent to positive and negative pressures, respectively. The flow between two counter-rotating rolls and its corresponding pressure profile is illustrated in Figure 2.6.

The motion of the two rolls at the same angular speed U drags the fluid. The pressure profile is characterized by a pressure peak at point C, which is located slightly before the centerline at D. Upstream of point C, the average velocity of the coating color is then accelerated by the presence of the rolls and the pressure increases. Because of this pressure reaches certain value, coating color tends to return (Réglat and Tanguy, 1997). However, downstream of point C, the average velocity of the coating color is greater than that of the rolls; the coating color is then decelerated by the presence of the rolls and the pressure decreases. In this region (point D), the Poiseuille contribution is generated by the pressure field induced by the converging/diverging geometry. This leads to a parabolic velocity profile as shows Figure 2.6a, with a maximum velocity of about $1,5 U$ (Coyle, 1984). Finally, the contact is lost at point E where the average coating color velocity is again equal to that of the rolls.

It is clear that the pressure increases from zero at the entrance to a maximum at point C, then decreases to zero at the outlet, which define the nip width. However, under

certain conditions, a small negative pressure at the nip exit where the film splits may appear. The occurrence of this negative pressure zone result in vortices that grow and cavities start to form.

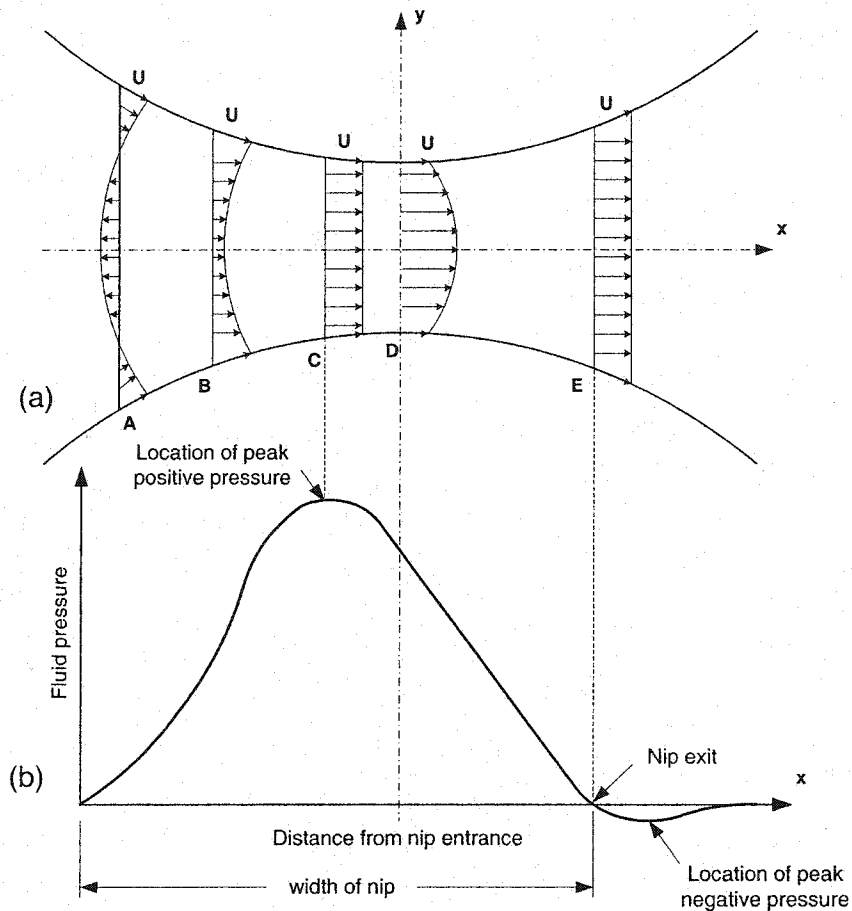


Figure 2.6 (a) Flow between two counter-rotating rolls;(b) Pressure distribution within nip and in region beyond nip exit (MacPhee, 1997a).

Réglat et Tanguy (1997) determined that both transfer roll speed and viscosity have an impact on the value of the minimum pressure. As Figure 2.7 shows, a minimum subatmospheric pressure appears.

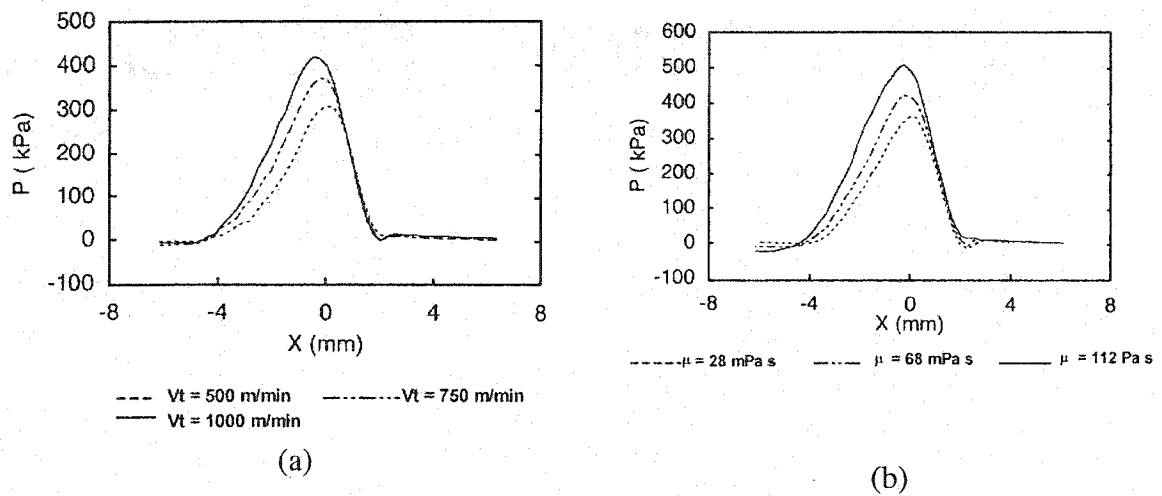


Figure 2.7 Effect of transfer roll speed (a) and viscosity (b) on pressure profile (Réglat and Tanguy, 1997).

MacPhee (1997) proposed that the character of film splitting at the nip exit undergoes three marked changes as printing press roll speed is increased (see Figure 2.8).

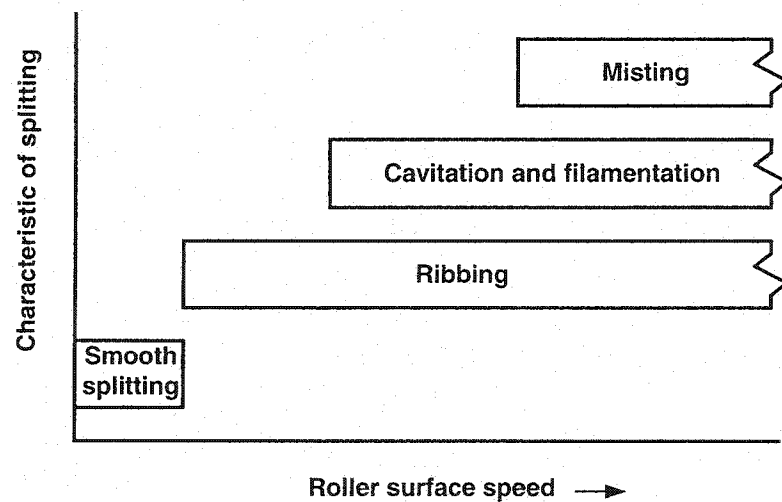


Figure 2.8 Change in character of film splitting in the nip exit versus speed (MacPhee, 1997a).

This is true if it only considers that speed is the main factor that controls the film splitting profile. However, it has demonstrated that film splitting depends on many factors such as the fluid rheology, nip gap, roll surface hardness, etc. When using a smooth flexible roll cover, ribbing can be reduced or delayed and then the character of film splitting could be very different from that shown in figure 2.8.

It is clear that at very low roll speed the film exiting the nip in a very smooth manner, as shown figure 2.9a. When the roll speed is further increased, cavitation and filamentation starts to appear (see figure 2.9b). These phenomena are produced due to negative pressure at the nip exit. When this pressure reaches some limiting level, closed cell cavities are then formed. The formation of these cavities depends basically on the fluid speed, fluid viscosity, and film thickness and roll diameter. As roll surfaces diverge, these cavities start to interconnect, forming filaments of liquid between the two surfaces and finally rupture of liquid occurs in the filaments and the ruptured filaments spring back to form a temporary orange peel surface (MacPhee, 1997a).

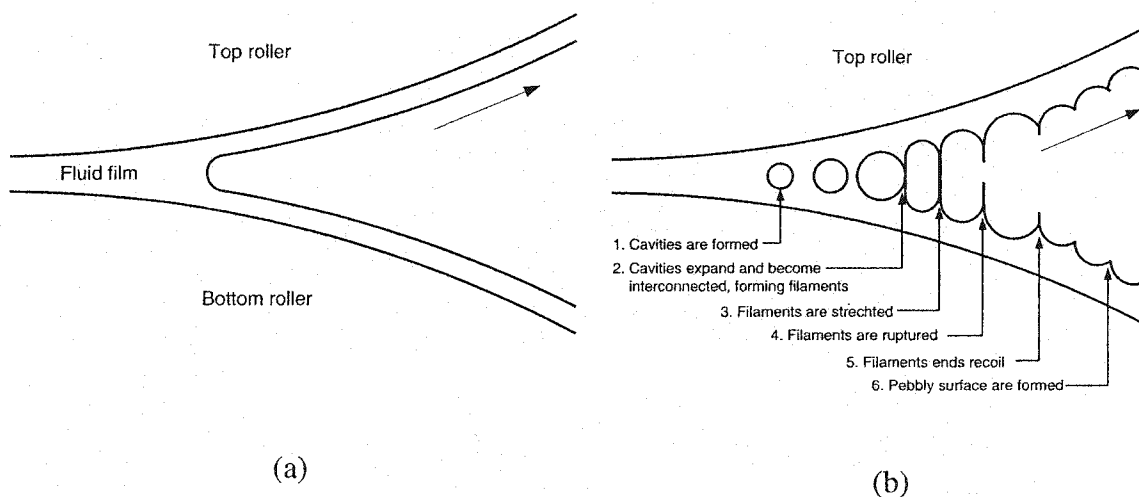


Figure 2.9 (a) Depiction of the smooth film splitting that occurs at very low roll surface speeds (MacPhee, 1997a); (b) stages in the formation of cavities and filaments during at high roll speed (MacPhee, 1997b).

2.2 Misting

Roper et al (1999) experimentally studied the effect of coating color, base paper and machine variables on orange peel pattern. To evaluate the degree of orange peel formation printed strips were used. When comparing the visual evaluation of printed strips to scanned samples, they found that orange peel is a two-dimensional defect that requires more sophisticated image analysis for a right correlation between the image parameters and the visual ranking.

The effect of base paper was also studied and it was found that a pre-calendered paper has a positive effect in reducing either misting level or orange peel formation (Figure 2.10).

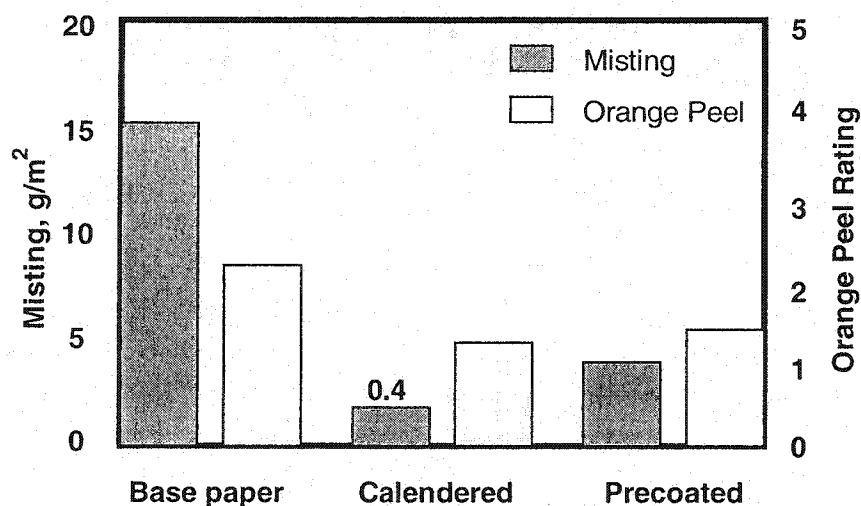


Figure 2.10 Effect of base paper on misting and orange peel (Roper et al, 1999).

A number of experiments were performed by Roper et al (1997) for assessing the degree of misting in a metered size press. For that purpose high-speed video and image processing techniques were used. The severity of misting was assessed by means of video images at 1200 and 1500 m/min using both calendered and uncalendered base paper (see figure 2.11).

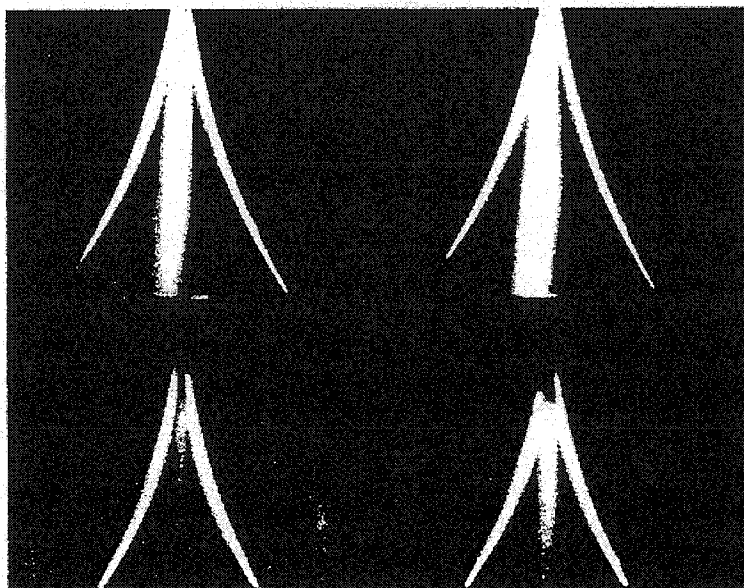


Figure 2.11 Video images of misting at 1200 and 1500 m/min on wood free base paper. Top: uncalendered; Bottom: calendered (Roper et al, 1997).

On the other hand, misting droplets were collected on black paper. Profilometry measurements on the drops were used for determining size droplets and the size distribution was calculated by using field separation techniques (image processing). Figure 2.12 shows a typical photomicrograph of misting droplets. Results from this study show that a smooth calendered paper showed very little misting compared to rough papers. This was true even though for this paper the rate of coating immobilization in the nip has been reduced.

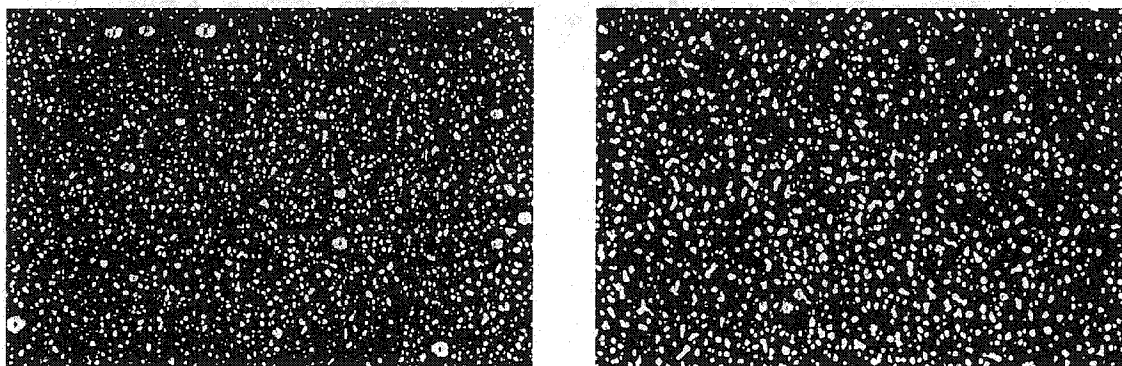


Figure 2.12. Photomicrographs (10X) of droplets collected at 1200 m/min (left) and 1500 m/min (right) from uncalendered wood free base paper (Roper et al, 1997).

As the solids content increases viscosity also increases up to a certain limit, called critical packing factor. Generally, the preference is for high-solids, more viscous-like coatings with low elasticity to avoid misting at the transfer nip (Triantafillopoulos and Smith, 1998). Figure 2.13 shows how orange peel is affected by the solids content. It can also be seen that misting is almost completely eliminated when the solids content is increased.

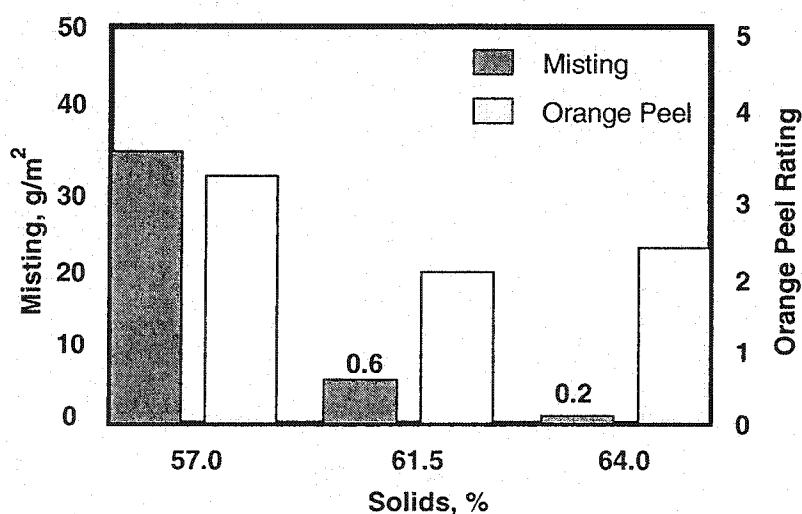


Figure 2.13. Effect of solids on misting and orange peel (Roper et al, 1999).

The thickener in the coating colors plays also a significant role. Figure 2.14 shows the effect of CMC level on orange peel formation. When the viscosity is increased, the rate of droplet ejection due to centrifugal forces will decrease because of the viscous drag and the greater cohesive energy of the coating, so that misting is reduced, but orange peel present a significant increase. By coating with higher solids content, both its runnability and the paper gloss are improved (Grön et al, 1997).

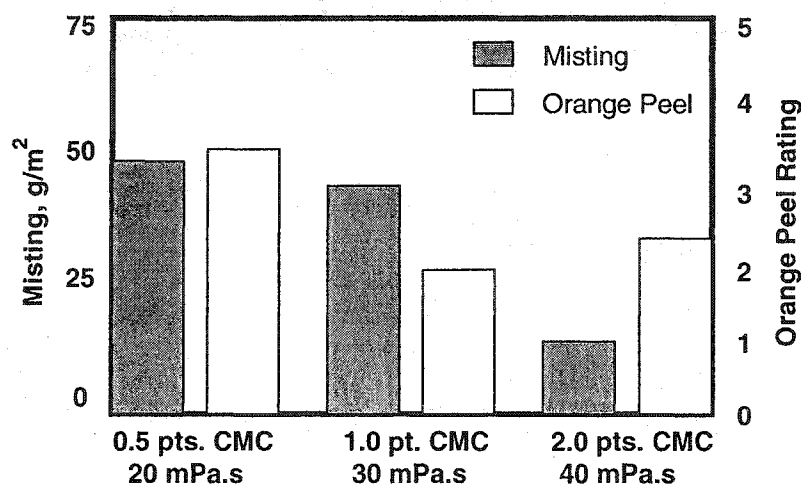


Figure 2.14. Effect of CMC level on misting and orange peel formation (Roper et al, 1999).

The choice of cobinder and thickener can play a vital role in the runnability and orange peel formation because they directly impact the water retention of the coating and consequently the misting. The effect of two different thickeners on misting amount at different speed was evaluated by Roper et al (1998). It was shown that CMC provides a lower high-shear viscosity and coating elasticity as well as a higher rate of immobilization than an alkali soluble thickener (AST). As a result, the misting amount was clearly reduced for high speeds.

Figure 2.15 shows the results of the rheological measurement of a step shear experiment to simulate the effect of sudden high shear on the coating followed by

recovery. Coating colors are first sheared at 1 s^{-1} during 5 seconds followed by a shear rate of 5000 s^{-1} for 1 second and finally they return to the lower shear rate. In this experiments two different thickener (high-leveling and low-leveling) were used. As show in this figure, the low-leveling coating recovers its viscosity much faster and almost return to the original viscosity.

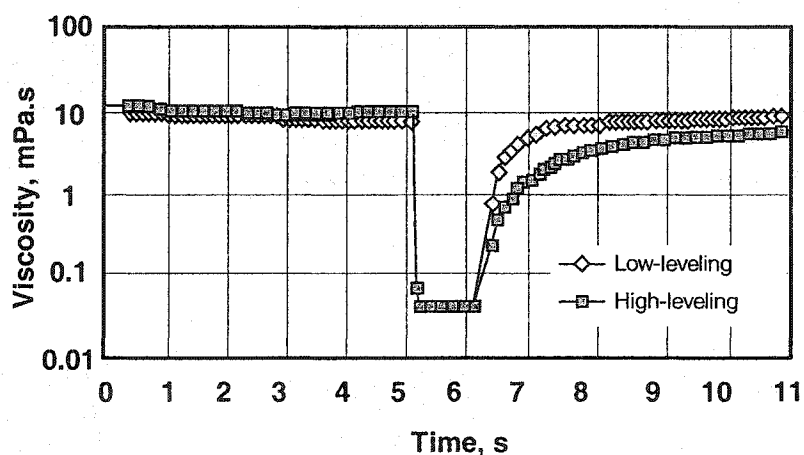


Figure 2.15 Step shear experiment (Roper et al, 1999)

Results of experiments in which two different thickeners were used show that a high-leveling thickener gives a coating with a noticeable lower orange peel level (see Figure 2.16).

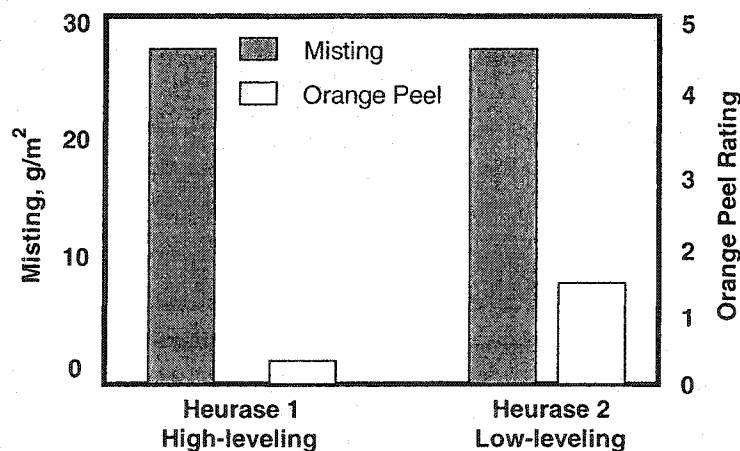


Figure 2.16 Effect of leveling (Roper et al, 1999).

This result suggests that a coating formulated with a thickener having a slower recovery after shearing gives better level and lower orange peel formation. As this figure shows, no effect is produced in misting amount.

Based on all of these results, they found that under certain conditions, there is a positive correlation between misting and orange peel formation, which means that factors that reduce misting at high speed also seem to reduce the degree of orange peel formation.

It is important to take into account the role that other formulation compounds play on the effects of the film splitting. The choice of a pigment depends on the required final properties of paper. However, it has been demonstrated that coating containing clay exhibit a lower degree of misting compared to those with calcium carbonate (Reimers et al, 1998, Drage and Hiorns, 1996 and Grön et al, 1996). Latex, for example, is the main binder that impacts the strength, optical and printability properties. It also improves the surface uniformity and misting resistance as shown in figure 2.17, where the two coatings without cobinders and high solids and latex content gave reduced misting. The rheological behavior of the coating is highly dependent on the choice of the cobinder.

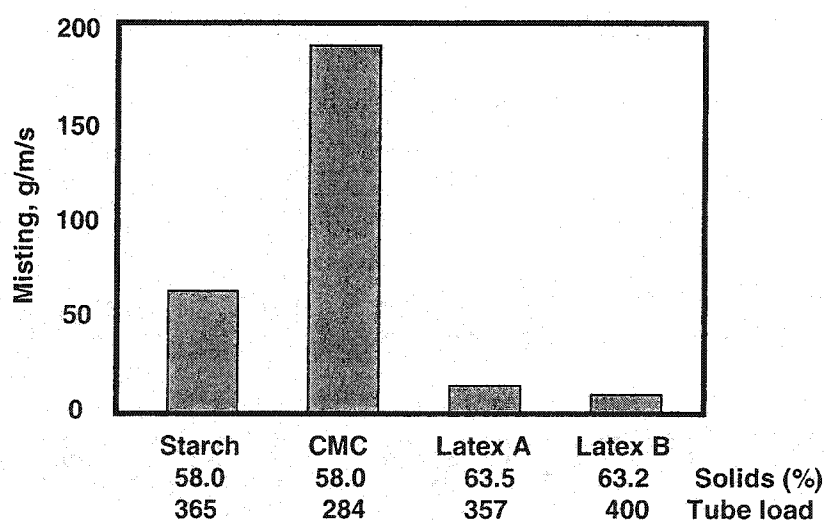


Figure 2.17. Effect of high solids, thickener and latex (Roper et al, 1998).

Latex is a good option specially when coating at high speed because it increases the rate of dewatering without significantly increasing the coating viscosity.

Another factor to consider in misting is the coat weight. It has found that when the coat weight increases a dramatic growth in the amount of coating color misting can be produced (Salminen et al, 1996). This effect can be observed in figure 2.18, where starch-containing coating color was used.

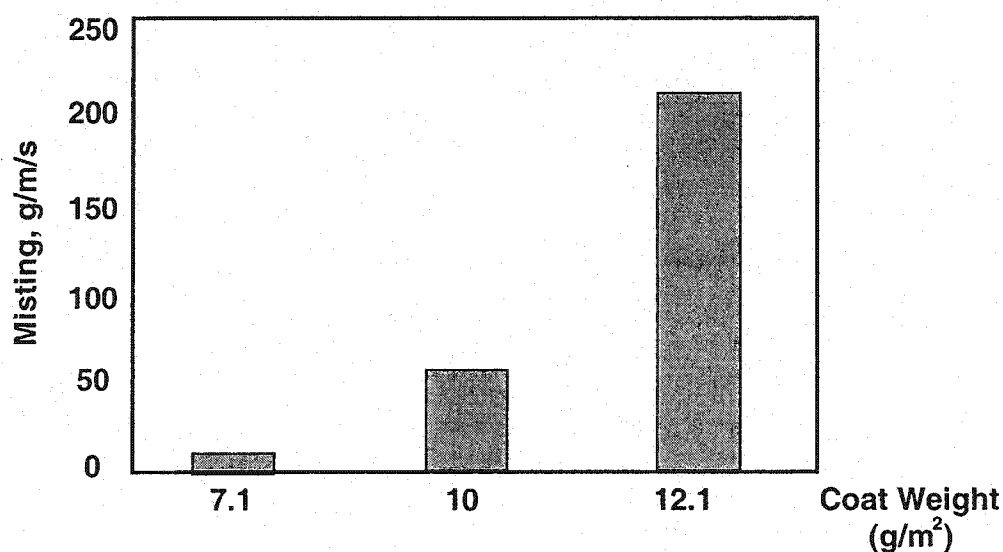


Figure 2.18 Effect of coat weight on misting (Salminen et al, 1996).

Coating formulation also plays an important role on web release in high speed in MFC coating. It has been demonstrated that the point of web release is dependent on the tension vectors created by the web tensions, penetration of coating into the web, and the internal strength of the coating color (Grön et al, 1998). Increasing the machine speed increases the distance between the nip outlet and the point of web release. When machine speed increases the nip dwell time is reduced which leads to a more "free" coating at the nip outlet and the amount of misting increases, and then paper adheres to the backing roll, as seen in figure 2.19.

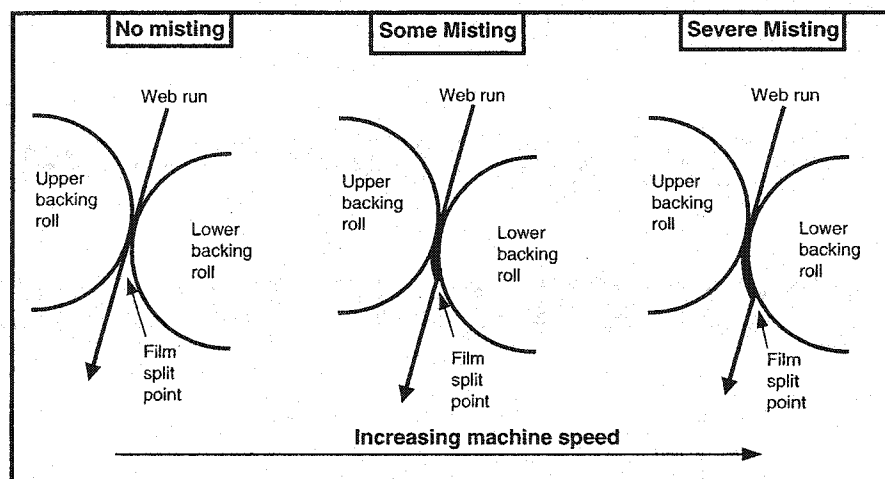


Figure 2.19. Coating adhesion dependence on misting and machine speed (Grön et al, 1998).

A crude way to determine the amount of misting is by mean of the transfer ratio, which is a value that can describe the coating color transfer to the paper (Grön et al, 1998). It is a relationship between the amount of premetered coating film to the amount of coating color transferred to the paper. The following equation defines the transfer ratio:

$$\text{Transfer ratio} = \frac{CW}{0.01Xm} \quad (2.13)$$

where

- CW = coat weight in g/m^2
- X = solids content in %
- m = premetered film amount in g/m^2

As shown in Figure 2.20, a transfer ratio between 0.3 to 0.4 will increase the misting value and result in severe misting. The cause of the tendency for misting during film split at the web release point may be an excessive amount of non-dewatered or penetrated bulk coating.

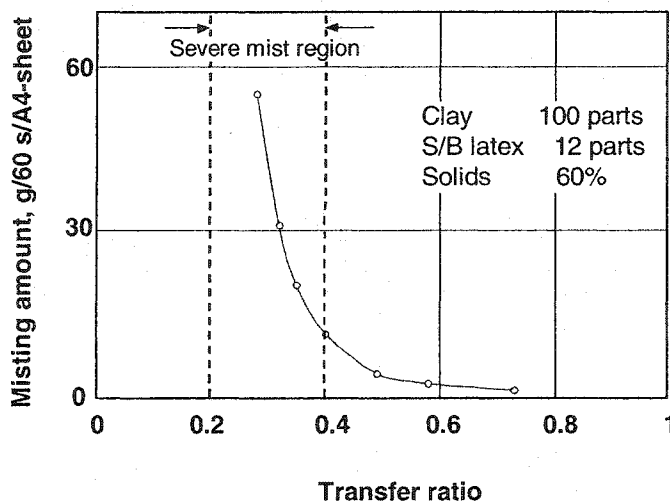


Figure 2.20 Misting amount as a function of transfer ratio at 1950 m/min (Grön et al, 1998).

Finally, the control of misting is not only important when coating paper. In order to avoid catastrophic effects of post-crash aircrafts fire and ground equipment, low concentrations of antimisting high molecular weight polymers have been used in Jet A and diesel fuels (Mannheimer, 1983). As these polymers have little or no effect on surface tension, the mechanism of antimisting has been related to rheological properties, in particular viscoelasticity (Mannheimer, 1983). At low viscosities, the ignition velocity is only slightly affected, but the dependence grows stronger as the viscosity increases. Matthys and Sabersky (1987) investigated antimisting kerosene in terms of its rheology, friction, and heat transfer properties. The additive used was designed to inhibit the formation of very small drops of kerosene when a mass of fuel is subjected to a rapid air stream, which is beneficial in the case of the rupture of a fuel tank during an airplane emergency landing for example. As a result, the flammability of the mist is dramatically reduced.

Almost all the machining operations require to cool or to lubricate the cutting tools in order to reduce the friction with the metal to be cut, so that a possible early wearing of cutting tool is avoided. One of the most common metalworking fluids is the mineral cutting oil. Mist and foul odors emanating from these fluids can be caused or

aggravated by both chemical and physical/mechanical factors (Lantz, 1997). Some substances such as polyisobutylene (PIB) have been used as antimisting additives in mineral oils, employing a coaxial air blast atomizer and an optical particle size to determine polymer effects on aerosol drop size distributions (Smolinski et al, 1996). In such cases, misting is mainly controlled by the fluid viscoelasticity. It has been demonstrated that these solutions increase significantly the average aerosol drop size (up to 200%), relative to pure mineral oil.

2.3 Rheology of suspensions and coating colors

Paper coating colors are concentrated suspensions made basically of water, pigments, thickener, binder and other additives. They are applied to the surface of the paper to improve its optical and printing properties by using several coating methods. Metered film coating is nowadays, one of the most used technologies for paper coating of low-weight coats or medium-weight coats at high speed. During the process, the coating colors are submitted to high shear rates in the metering nip and high extensional rates in the transfer nip (both higher than 10^6 s^{-1}). As stated before, the film splits at the exit of transfer nip and several runnability problems can occur and reduce the quality of coating.

For high-speed applications, Triantafillopoulos and Smith (1998) suggested to keep the elastic modulus (G') as low as possible and the viscous modulus (G'') as high as possible, which can be obtained by increasing the solid content and keeping low the level of binders. Although, this will increase the water retention rate, a special attention must be paid with the type of paper to be coated. A high-absorbency paper allows elongated filaments to be formed inside the immobilized layer, which leads to a permanent film pattern onto the paper and some misting droplets may be deposited on the coated surface. This can be alleviated by using a less absorbent paper and/or by increasing the water

retention with a higher level of water soluble binder. However, the coating elasticity must be minimized to avoid any permanent coating film patterns.

2.3.1 Shear rheology

The shear rheological properties of concentrated suspensions and coating colors have been widely studied by Carreau and Lavoie (1993), Roper and Attal (1993), Page (1999), Page et al (2002), Barbesta et al (2001), Yziquel et al (1999), Brian and Mackay (2002). A typical viscosity curve of concentrated suspensions exhibit a shear- thinning behavior following a power-law model for a range of shear rate relatively low. Then as the shear rate increases, suspensions reach a critical value and from this point a plateau viscosity known as high-shear rate viscosity (η_{∞}) is observed, which is true for the low-concentrated suspensions. The high-concentrated suspensions exhibit a different behavior. The particles tend to collide creating shear induced structures, which leads to shear thickening.

Yziquel et al (1999) proposed a power-law expression with a high-shear rate viscosity, η_{∞} , for describing the viscosity of coating colors:

$$\eta = m|\dot{\gamma}|^{n-1} + \eta_{\infty}$$

Roper and Attal (1993) measured the viscosity of paper coating colors at rates from 10^{-1} to 10^6 s^{-1} by using a combination of rheometers. They observed that some coating colors exhibit shear-thinning followed by dilatancy, which is more pronounced as the both the solid content and the particle size increase. For very high shear rates, a decrease of viscosity is observed, which could be due to viscous dissipation. The flow curve was fitted with the following five parameter model using an extension of the Cross model:

$$\mu = \frac{1000}{1 + k_1 \dot{\gamma}^n} + \mu_\infty + \frac{k_2 \dot{\gamma}}{(k_3 + \dot{\gamma})^2}$$

where k_1 and n are the power-law parameters in the shear-thinning region, μ_∞ is the high-shear rate viscosity, k_2 controls the magnitude of dilatancy and k_3 is the critical shear rate for the onset of dilatant behavior.

O'Brien and Mackay (2002) observed that a concentrated suspension first exhibit a shear-thinning behavior for relatively low shear rates. The suspension becomes dilatant as the shear rate increases and a plateau viscosity is reached for very high shear rates. It must be noted that this viscosity plateau obtained at about 10^5 s^{-1} is almost equal to that obtained at low shear rates (10^{-1} s^{-1}).

Carreau and Lavoie (1993) studied the structure build-up or breakup under constant applied shear rate. They tested coating color with a solid content ranging from 45 to 65 wt% with SBR latex as binder, low molecular weight Carboxymethyl cellulose (CMC) as thickener and a dispersing agent. They observed a very slow structure build-up at very low shear rates. When they suddenly stopped the test and restarted it within a few seconds at the same shear rate, a viscosity increase was observed due to the structure build-up; however an equilibrium value was obtained. A different trend was observed for a higher shear rate. The shear viscosity was much lower and slowly decreased with time. After stopping and restarting the test, a slowly decrease of viscosity with time was observed.

In regard to oscillatory shear tests, Yziquel et al (1999) observed a linear viscoelastic domain for small-amplitude oscillatory shear measurements, which can be related to a solid-like behavior. Yziquel et al (1999) found that the elastic modulus is constant up to critical strain and above this critical value, the elastic modulus decreases with strain and the viscoelastic behavior becomes non-linear. Carreau and Lavoie (1993) determined that

the linear viscoelastic properties are controlled by the strong particle interactions responsible for the solid-like behavior and the mechanism responsible for the flow, breakup of flocs, and eventually alignment of particles. Page et al (2002) performed oscillatory tests with industrial coating colors. They observed constant loss moduli for small oscillatory amplitudes. The loss modulus of the coating color containing carboxymethyl cellulose as thickener exhibited a higher value as deformation increases. This phenomenon, previously observed by Yziquel et al (1999), has been related to a flow induced microstructure change. If the strain is further increased the loss modulus tends to decrease.

2.3.2 Extensional rheology

The extensional viscosity is perhaps the most important material property that affects misting occurrence. However, most of the studies have been focused on shear viscosity or viscoelastic properties of suspensions and coating colors.

Della Valle et al (2000) determined the extensional properties of suspensions of delaminated kaolin clay with a solid contents ranging from 50-65 wt% by using an orifice flowmeter at rates from 10^3 - 10^4 s⁻¹. They found a Trouton ratio between 12-16 for suspensions made of kaolin dispersed in aqueous polyethylene glycol.

Using an opposed orifice rheometer, O'Brien and Mackay (2002) measured the extensional properties in a range from 10^1 to 10^4 s⁻¹ of suspensions made of very fine polydisperse paper coating grade kaolin. They found that the extensional thickening of concentrated suspensions having a solid content of 71 wt% became apparent at rate of 10^3 s⁻¹ while the shear thickening occurs at shear rates an order of magnitude larger. They also found a Trouton ratio of the order of 100, which can be due to the amount of dispersant dissolved in water.

Page (1999) studied the extensional properties of paper coating colors based on kaolin clay and three different thickeners: CMC and two associative polymers. They measured the extensional viscosity of the suspensions having a constant solid content of 65.5 wt% in a deformation rate range from 10^3 - 10^5 s⁻¹. Under these conditions they found a Trouton ratio up to 100.

2.4 Specific objectives

Practically all the information available in the literature about misting in MFC has been focused to propose strategies for reducing this runnability issue. The characteristics of the knowledge generated from these studies is mostly empirical and little scientific background has been developed. A good understanding of the mechanism of film splitting is the key for proposing appropriated strategies to improve the runnability of a MFC. Although many efforts have been made in studying flow behavior in the nip of two counter-rotating rolls with a scientific focus, the conditions used are very different from those ones in MFC. Capillary numbers up to 100, Reynolds numbers about 1 and flexible nips (two elastomer-covered rolls) are commonly used in MFC at high speed. However, no reports under these conditions were found in the literature.

We therefore propose to work on this issue and propose to tackle the following objectives:

Specific objective 1: to determine the main factors of the film splitting mechanism.

Specific objective 2: to understand the role of coating colors rheology on misting phenomenon.

This thesis is presented under the format of "thesis by papers", so that chapters 4, 5 and 6 correspond to three papers submitted to top journals in chemical engineering.

CHAPTER 3

EXPERIMENTAL SET-UP

3.1 Laboratory film coater

The film coater used for laboratory tests consists basically of three parts: the transfer the roll, the backing roll and the feeding system. Figure 3.1 shows a general view of the laboratory film coater. The transfer roll is a cylinder of 150 mm in diameter and 150 mm length with a rubber cover of 9.5 mm thickness and a hardness of 63 P&J (~60 shore A). This was the minimum thickness recommended by the supplier in order to avoid any interaction with the rigid core. The backing roll is a rigid cylinder of 150 mm diameter and 150 mm length with a smooth chrome plated surface having an average roughness of

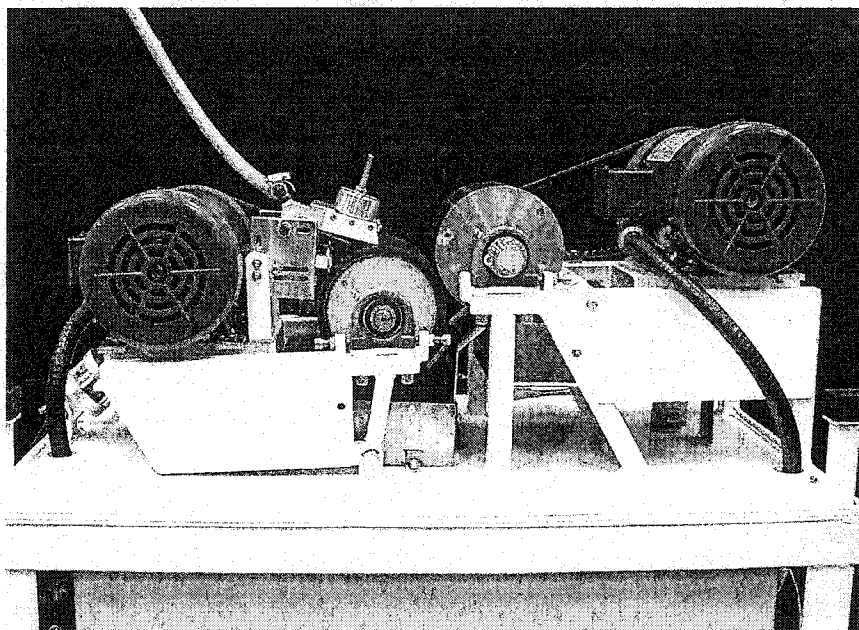


Figure 3.1 General view of the film coater

3 μm and a run out less than 2 μm . Both rolls are counter-rotating, supported by self-aligned journal bearings and driven by a pulley-belt array and an electrical motor each having a power of 746 W (1 hp), which speed is carefully controlled by an AC solid-state frequency changer receiving a feedback signal from a speed encoder. The speed control

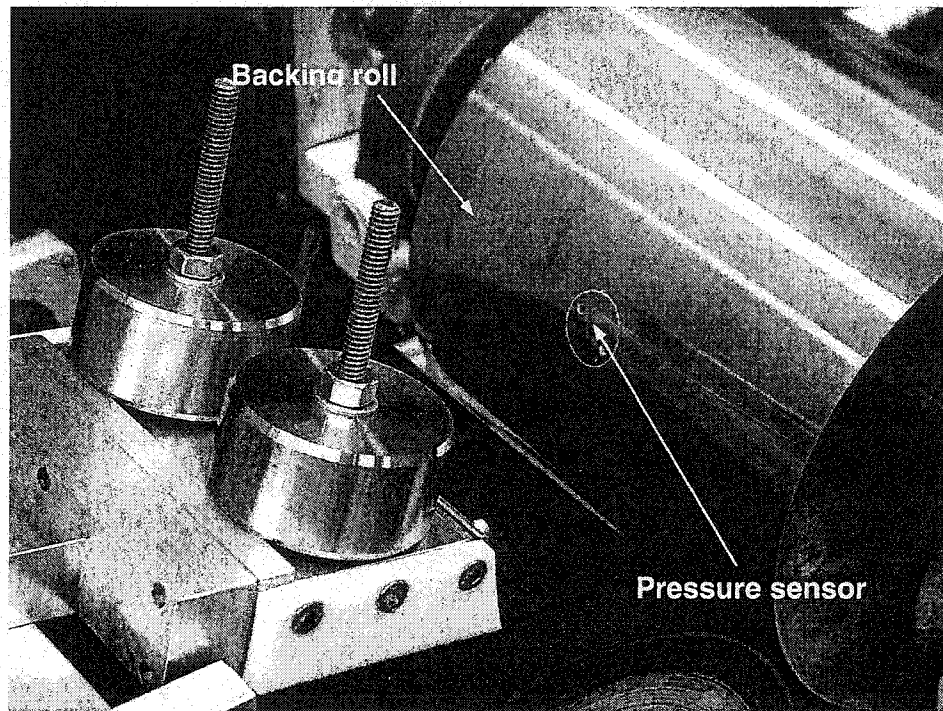
allows operating between 0-1600 m/min and also to vary the acceleration deceleration slopes in order to avoid a motor overload when they operate at high speed. Each roll was dynamically balanced in two planes at three different points up to 6250 rpm. A scrapping blade is used on each roll to remove the excess of fluid before entering the nip. The applied load between the roll can be varied from 2 to 4 kN/m by using a pneumatic cylinder attached to a rigid structure, in which the transfer roll is supported.

3.1.1 Pressure transducer

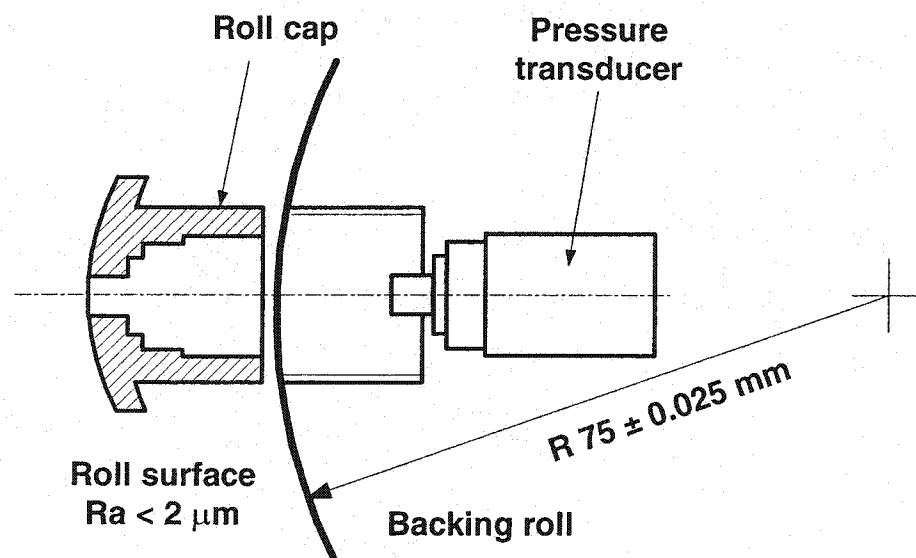
The pressure distribution of the fluid into the nip is measured by means of a subminiature dynamic pressure sensor of 2.5 mm in diameter mounted on the surface of the backing roll. Figure 3.2 shows a general view of the pressure sensor mounted on the backing roll and a diagram showing its mounting. The membrane of this sensor was carefully machined and polished to same backing roll radius in order to avoid intrusion effects. Different profiles are obtained at every backing roll revolution: one corresponding to the pressure into the nip and other for the pressure of the scrapping blade on the roll. The general specifications of the pressure transducer are presented in the following table.

Table 3.1 Performance specifications

Feature	Value
Measurement range (for ± 1 V output)	690 kPa
Sensitivity	1.45 mV/kPa
Maximum pressure	3500 kPa
Resolution	0.069 kPa
Frequency response	250 kHz
Rise time	≤ 2.0 μ s
Non-linearity	≤ 2.0 % FS



(a)



(b)

Figure 3.2 Pressure sensor: (a) General view; (b) Mounting.

3.1.2 Slip ring

The pressure distribution data are captured by a computer via a slip ring of four circuits connected to a data acquisition card. The slip ring brushes and ring are made of precious metals, which minimize noise and enable the assemblies to be used for low-level instrumentation signals. It was mounted to the end of the transfer roll shaft. The signal wires coming from the pressure transducer were routed through the center of the hollow shaft and an adapter was used for attaching the slip ring rotor to the roll shaft. The specifications of the slip ring are:

Circuits:	4
Current capacity per circuit:	500 mA
RPM rating:	12000
Maximum peak noise	0.1 Ω

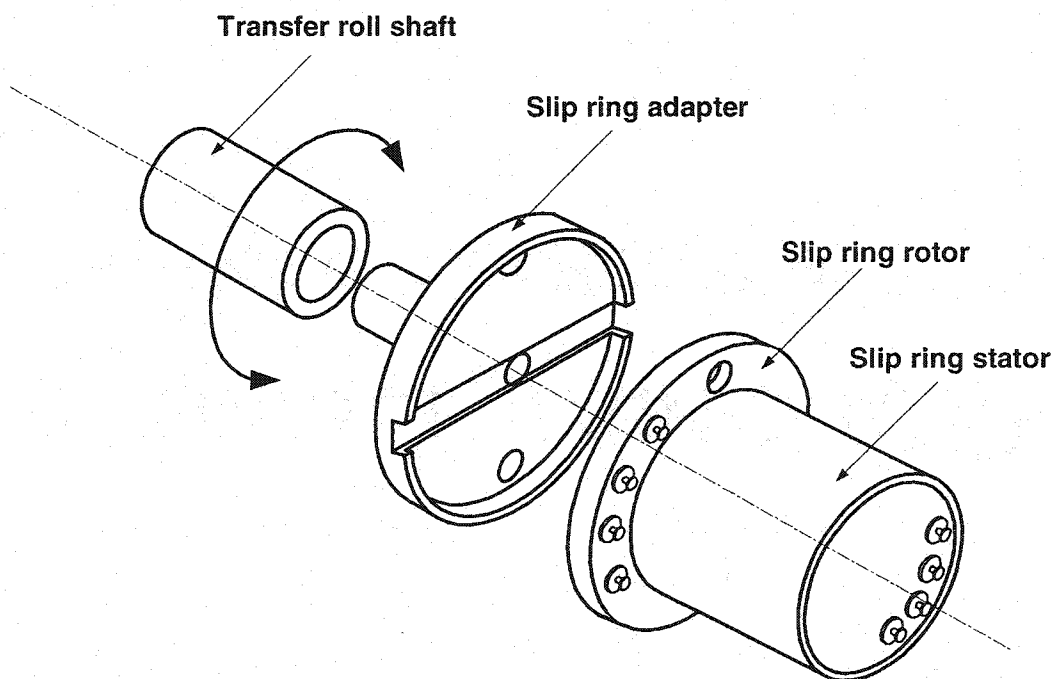


Figure 3.3 Slip ring mounting

3.1.3 Fluid delivery system

A 5 L reservoir containing the fluid to be delivered to the transfer roll is air pressurized, so that a constant flow rate is kept during the experiments. A metering ball valve allows setting carefully the flow rate required and a feeding chamber distributes uniformly the fluid along the coated strip. The thickness of the fluid on the transfer roll is controlled by means of a doctor blade placed at the chamber exit. As mentioned before, a scrapping blade was used for removing the excess of fluid before entering the feeding chamber and to avoid end effects during the experiments. (see Figure 3.4).

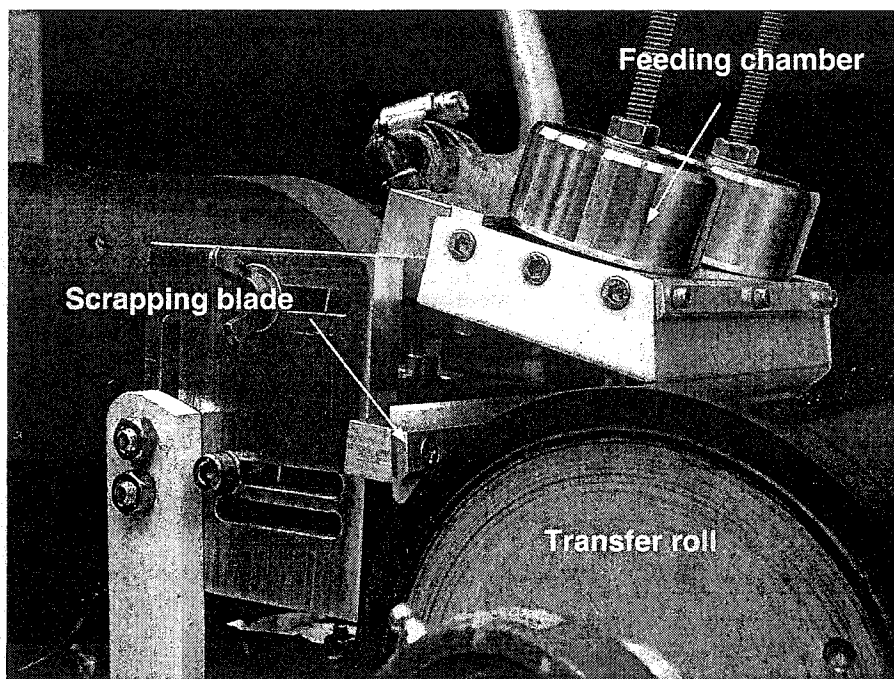


Figure 3.4 Feeding chamber

3.2 Orifice flowmeter

The extensional properties of suspensions and coating colors were determined by means of an orifice flowmeter based on the pressure entry technique, in which the extensional viscosity is a function of the flow rate and the pressure drop of a fluid passing through a small orifice. Although, several methods including the stagnation flows, filament stretching among others have been reported in the literature (Binding and Jones, 1989; Cohu and Magning, 1995; Titaatmadja and Sridhar, 1993), this technique was found to be the most suitable considering the difficulty of imposing a shear free flow at high strain rates ($\sim 10^4 \text{ s}^{-1}$).

The orifice flowmeter used for the present work consists of two identical reservoirs of 76.2 mm in diameter and 1.097 m length each and the pumping system. The fluid under test is pumped from the filled reservoir to the empty one by means of a hydraulic system, which consists of two different pumps drive by electrical motor. The speed of one of these pumps is carefully controlled by a AC solid-state frequency changer and the speed of the other one is fixed. A series of valves allow operating with the two pumps so that a broad range of flow rate is available. Each reservoir is connected to hydraulic piston large enough for testing under fully developed flow conditions. An orifice plate is mounted between the reservoirs and the pressure drop is measured by means of dynamic differential pressure transducer via small holes bored on each side of the orifice plate. The pressure drop can be measured in three different ranges: 0-70 kPa, 0-345 kPa and 0-1380 kPa and the corresponding resolution respect to the full scale are 1% for the first two ranges and 2% for the third one.

The speed of each piston is determined and by means of LVDT position transducers. All the instrumentation is connected to an analog-digital converter and to a data acquisition card so that the pressure drop and the flow rate are continuously recorded. An

acquisition program generates two files: one with the average values and the second one with the rough data.

CHAPTER 4

FORWARD DEFORMABLE ROLL COATING AT HIGH SPEED WITH NEWTONIAN FLUIDS

G. Ascanio^a, P.J. Carreau^a, E. Brito-De La Fuente^b and P.A. Tanguy^a

^aCRASP, Department of Chemical Engineering, Ecole Polytechnique
Montreal, Canada

^bFaculty of Chemistry, National University of Mexico
DF, Mexico

Keywords: high-speed forward roll coating, deformable gap, misting, nip pressure, hydrodynamics, Newtonian.

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4.1 Abstract

The flow of Newtonian fluids through the deformable nip of a high speed roll coater was experimentally studied. A digital video system was used for the observation of the formation, elongation and break up of fluid filaments at the exit of the nip and to investigate how misting droplets are ejected to the surroundings. The pressure profile in the nip was obtained by a miniaturized piezoelectric transducer mounted on one of the coater rolls. Experimental results show that both film splitting and air entrainment into the nip can be responsible for misting occurrence when a low load is applied between the rolls. When a high load is applied, misting can be eliminated and a periodic flow is generated in the nip gap.

4.2 Introduction

Forward roll coating is a process whereby liquid flows into a narrow nip between two counter rotating cylinders. The equipment used for that purpose performs the four basic functions of coating: to feed, to meter, to distribute and to apply a liquid film onto a continuous substrate.

In the paper industry, metered film coating is one of the most common technologies used for paper coating at high speed. A minimal rebuild, a relatively low investment, a good runnability (few process interruptions) and the possibility to have brand new product concepts such as coated newsprint are some reasons that justify the use of metered film coating (McAuley, 2000). Figure 4.1 shows the principle of a metered film coater, which consists basically of a metering rod, and a transfer roll and a backing roll driven in a counter-rotating mode. In low speed laboratory equipment, soft or rigid rolls can be used.

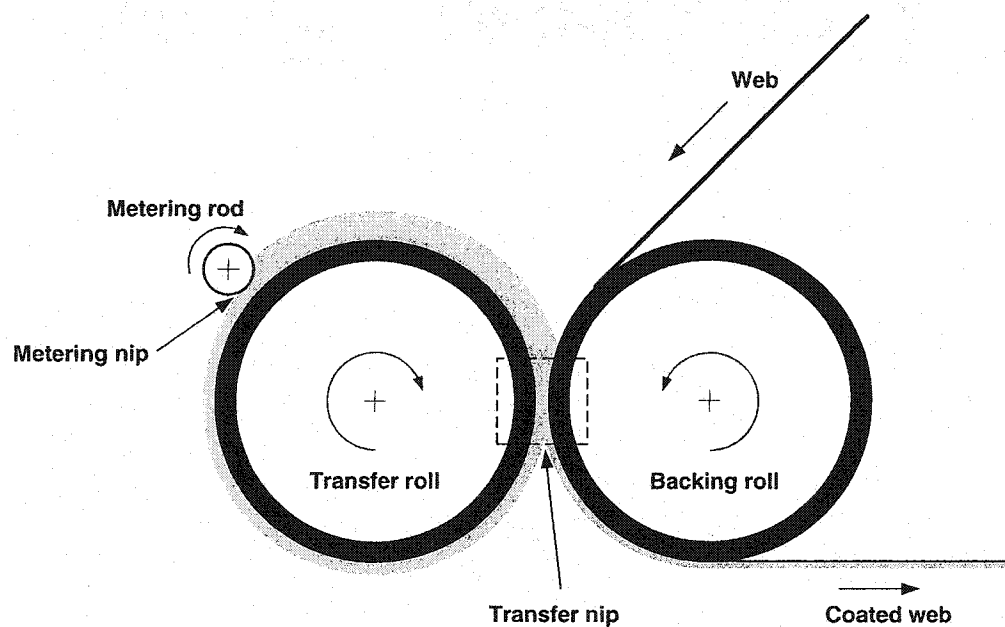


Figure 4.1 Metered film coater principle

However, in industrial equipment, a layer of deformable elastomer often covers one of the rolls. This allows pressing the rolls against each other to form thin films, and a negative gap is generated (Figure 4.2).

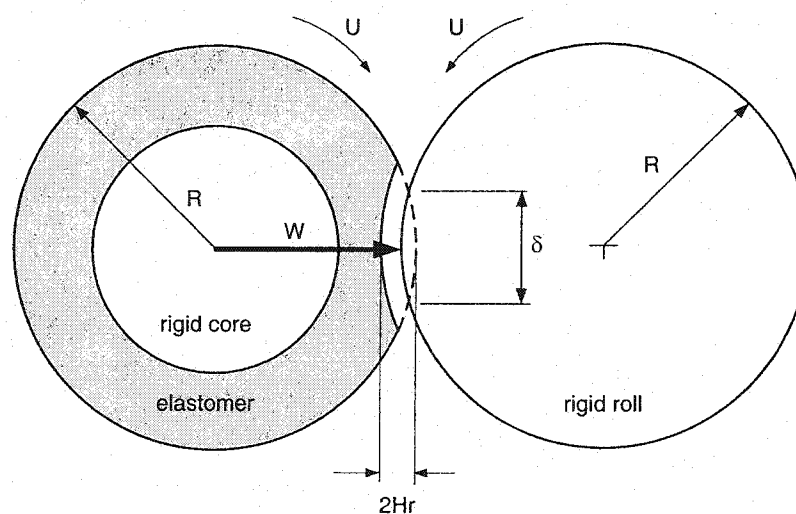


Figure 4.2 Forward deformable roll coating array with negative gap

The use of film coating technology has many advantages such as the process speed, the possibility of using high solid content fluids, etc. However, the process is prone to two major runnability issues related to the transfer nip: misting and orange peel (Roper *et al*, 1997). The first one is defined as the ejection of fluid droplet during the film splitting at the exit of the transfer nip. As stated by Roper *et al* (1997), misting occurrence depends on many factors, but it usually appears for speeds above 1000 m/min. Although, misting does not affect the quality of coating and reduces the coverage, it represents a very important source of mill air contamination and waste of material, and frequent machine washings are needed.

The first work on forward roll coating with a deformable gap was carried out by Coyle (1984, 1988). By means of dimensionless analysis, he studied the role of forces involved in the flow in the deformable nip, and he obtained a theoretical correlation for estimating the coating thickness. The validity of this correlation was experimentally confirmed by Cohu and Magnin (1997) in a study in which it was determined that the elastic modulus of rubber materials in cylinders may be time-dependent. Coyle (1984, 1988) proposed the following one-dimensional elastohydrodynamic model suitable for small deformations for predicting the pressure profile between forward rolls.

$$\frac{dp_e}{dx} = Ne \left(\frac{12}{h_n^2} - \frac{24q}{h_n^3} \right) \quad (4.1)$$

$$h_n = -2 + x^2 + p_e \quad (4.2)$$

where p_e is the pressure scaled to elastic forces, Ne is the modified elasticity number, q is the flow rate and h_n is the total roll gap. The dimensionless variables are defined by

$$p_e = \frac{PL}{H_r E} \quad (4.3)$$

$$q = \frac{\frac{1}{2} \int_{H_1}^{H_2} U dY}{UH_r} \quad (4.4)$$

$$x = \frac{X}{\sqrt{RH_r}} \text{ and } y = \frac{Y}{H_r} \quad (4.5)$$

where P is the pressure, L the thickness of deformable layer, E the Young modulus, U the roll speed, H_1 and H_2 are the roll surface profiles, X and Y the flow and cross-flow coordinates.

Carvalho and Scriven (1994, 1997) and Carvalho (1996) analyzed the effect of a deformable gap on roll coating. They confirmed the results obtained by Coyle (1984, 1988) with a one-dimensional model to describe the roll deformation. They also proposed a Hookean two-dimensional model for predicting the roll profile and the pressure distribution with both positive and negative gaps when the roll is highly deformed.

Because misting has its origin in the film splitting, a full understanding of this mechanism is the key for establishing strategies to improve runnability. MacPhee (1997) proposed a model for describing the film splitting process between two counter-rotating printing press rolls. He suggested that at low speed the splitting is smooth and free of spurious phenomena but as the roll speed increases the character of film splitting changes to ribbing, then to filamentation and cavitation, and finally to misting for the high speeds.

Not only does the film splitting depend on the roll speed but also on the fluid properties, the rigidity of surface rolls, etc., as pointed out by Cohu and Magnin (1997) and Carvalho (1996). Many efforts have been made for determining the factors governing the occurrence of misting (Roper et al, 1997; Grön, 1998). Roper et al (1997) used high-speed video techniques and image processing for assessing the severity of misting.

Results with coating colors revealed that as the solid content increased the amount of misting was drastically reduced. Owens et al (2002) observed the formation of curved thin sheets of liquid extending from the nip, known as septa, with both Newtonian fluids made of high molecular weight branched polymers and Boger fluids in a forward roll coater. At low roll speed, septa were found to be mainly responsible of misting generation because surface tension forces dominate over viscous ones. The role of pressure on nip flow stability has been analyzed in the literature (Carvalho and Scriven, 1994; Carvalho and Scriven, 1997; Carvalho, 1996; Young, 1997) but only at low coating speed. At the high coating speed used in the paper industry, little information is available and the relation between the nip pressure and misting, if any, is not documented. The purpose of the present work is to shed some light on this issue, by investigating the film splitting mechanism of a deformable nip with counter-rotating rolls at high speed.

4.3 Materials and Methods

4.3.1 Apparatus

The experimental set-up used for this work is shown in Figure 4.3, and the general specifications of the film coater are given in Table 4.1. The apparatus basically consists of three parts: a transfer roll, a backing roll and a feeding device. The transfer roll is a cylinder of 150 mm in diameter covered with a SBR layer (Variokote from Stowe Woodward) of 9.5 mm thickness. The backing roll is a rigid cylinder of 150 mm in diameter with a smooth chrome plated surface having a mean roughness less than 2 μm . Each roll is driven by a 746 W (1 hp) electrical motor, which speed is carefully controlled by a solid state AC drive in feedback mode receiving a feedback signal generated by a speed encoder. Both rolls have been dynamically balanced in two planes at three different points up to a speed of 6250 rpm. The fluid is fed to the transfer roll by means of a pressurized cylinder, a metering ball valve and a feeding chamber. A doctor blade is used

as metering device for controlling the amount of fluid to be transferred. A scraper is installed on both rolls for removing the excess of fluid before entering the nip. The applied load between both rolls (W) can be varied by a pneumatic cylinder and is measured by a pressure transducer.

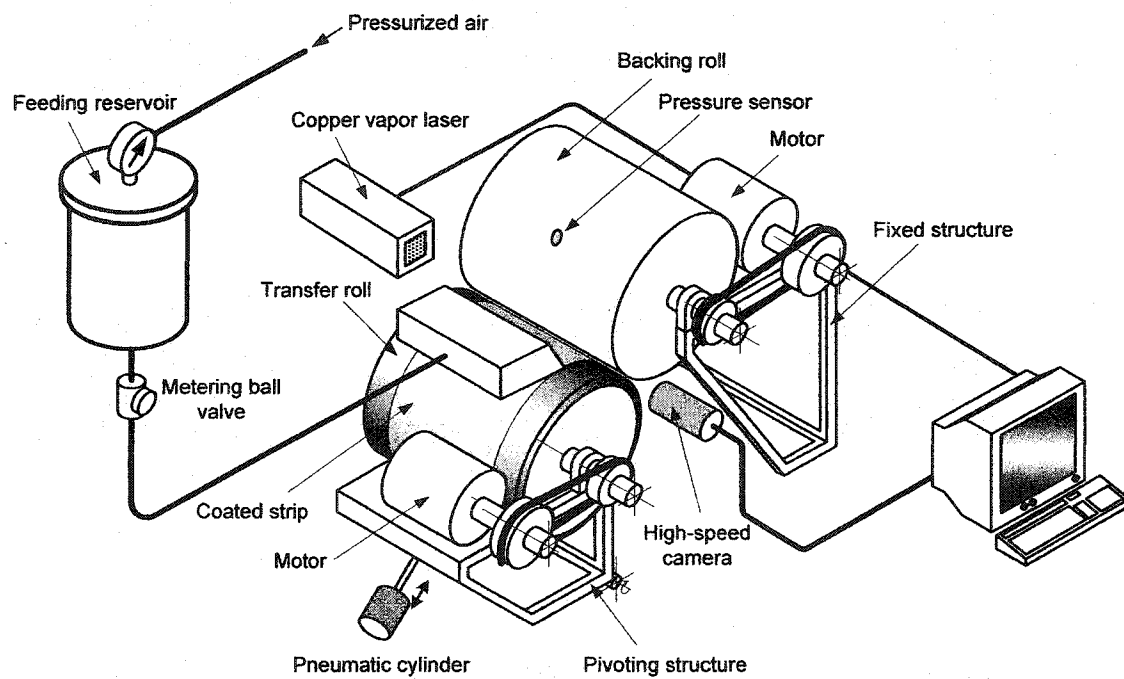


Figure 4.3 Experimental set-up

Table 4.1 Laboratory Coater Characteristics

Transfer roll	Diameter	150 mm
	Speed	0 to 1600 m/min
	Runout	Less than 5 μm
	Material	Steel with a SBR 63 P&J cover
	Applied load	0 to 4 kN/m
Backing roll	Diameter	150 mm
	Speed	0 to 1600 m/min
	Runout	Less than 2 μm
	Material	Steel with a chrome plated surface
Feeding system	Tank Capacity	5 L
	Max. flow rate	0.5 L/s @ 140 kPa

The pressure profile of the fluid passing through the transfer nip is measured with a high sensibility piezoelectric pressure sensor (diameter 2.5 mm) mounted on the surface of the backing roll. It should be noted that this technique has already been successfully used for obtaining the pressure profile in both forward and reverse modes (Young, 1997; Réglat and Tanguy, 1997). The measurement range is from 0 to 690 kPa, with a resolution of 0.01% f.s., a response time of 250 Hz and a non-linearity of 2.0% f.s.. The membrane of this sensor has been carefully machined and polished at the same radius as the backing roll in order to reduce intrusion effects. A single profile in the transfer nip region is acquired at every backing roll revolution (pressure along the nip length).

4.3.2 High-speed Imaging

A high-speed video system was installed for visualizing the fluid through the nip. A diode copper vapor laser was used as illumination system and delivered pulses of infrared laser light with variable pulse durations and frequencies. The laser was triggered by an external pulse from a camera strobe output. Imaging was performed with a high-speed video CMOS having a full resolution of 1024 x 1024 square pixels, which allows obtaining up to 462 frames/s at that resolution. This camera has an internal memory of 512 Mb so that 512 images can be stored at high speed and full resolution. A total of 150 frames were obtained for each experiment at a recording speed of 231 frames/s at full resolution. The shutter was set at 2.2 ms and a focal magnification of 2 was used for enlarging the view of the nip exit.

4.3.3 Test conditions

Newtonian aqueous solutions of polyethylene glycol ($M_w = 35000$ mol/g, from Clariant, Ltd.) at 20% and 30% (w/w) concentrations were used as inelastic model fluids. These formulations were chosen for their rheological stability. The fluids were allowed to settle during 24 hours before starting the experiments in order to eliminate air bubbles. The viscosity of the tested fluids was measured with a rheometer (model AR-2000, TA Instruments) using a Couette configuration.

The vapor pressure of the tested fluids was calculated by using the molar fraction at each concentration. The fluid measurements as well as the experiments were performed at room temperature (≈ 24 °C).

The flow rate of fluid fed to the transfer roll was kept constant at 0.2 L/s in all the experiments. The roll speed (U) range was from 500 m/min to 1100 m/min with an

applied load (W) from 2 kN/m to 4 kN/m. Under these conditions, the roll deflection (H_r) can be determined by⁵

$$H_r = R - \sqrt{R^2 - \left(\frac{\delta}{2}\right)^2} \quad (4.6)$$

where R is the roll radius and δ is the length of the contact surface between the two rolls, which was experimentally measured by comparing the images obtained statically at 0, 2 and 4 kN/m. The tests conditions used for this work are summarized in Table 4.2.

Table 4.2 Test conditions

Fluid viscosity	μ	0.13-0.63	Pa·s
Vapor pressure	P_v	2.68-2.86	kPa
Roll speed	U	500-1100	m/min
Applied load	W	2-4	kN/m
Roll deflection	$2H_r$	246-433	μm
Contact time	t_c	0.47-1.42	ms

4.4 Results

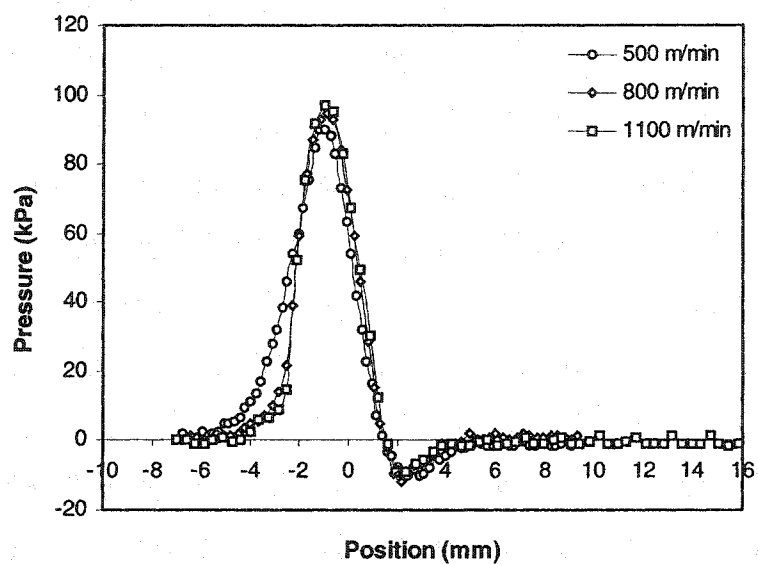
4.4.1 Pressure profiles

All the results reported in this work were obtained under a negative gap regime, in which the deformable roll was pressed against the rigid roll. The pressure profiles as a function of the roll speed, the fluid viscosity and the load between the rolls are plotted in Figures 4.4 and 4.5. As the fluid passes through the nip, it is sheared, elongated and submitted to a region of positive pressure and a region of negative pressure. At the nip exit, it splits to form filaments and under certain conditions these filaments can break up

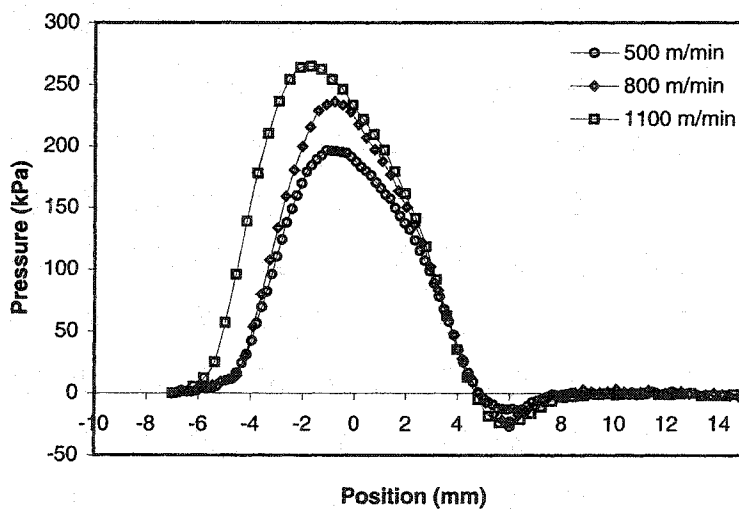
into two or more parts, and droplets can be ejected to the surroundings. The positive peak pressure (P_{max}) is a function of many factors such as the film thickness, the operating conditions, etc. The magnitude of the negative peak pressure (P_{min}) depends on both the rolls speed and the fluid properties (MacPhee, 1997). For interpreting the effect of U on the pressure profile, Young¹² proposed the integrated form of the Reynolds equation:

$$\frac{dP}{dX} = 12\mu U \left[\frac{H(X) - 2H^\infty}{H^3(X)} \right] \quad (4.7)$$

where $H(X)$ is the film thickness as a function of the flow coordinate and H^∞ is the eventual film thickness. Although, the Reynolds equation allows estimating the pressure profile in the positive region only, it was observed that as U increased, inertial effects became important and P_{max} increased. The same effect was observed for W . When the rolls were tightly brought together, $H(X)$ decreased and P_{max} increased also. The same trend would be expected for the negative region of the pressure profiles; in other words the higher U and W the lower P_{min} . However, as Figures 4.4a and 4.5a show, the trends in the negative region were quite different; U_{max} did not correspond to P_{min} at the exit of the nip.



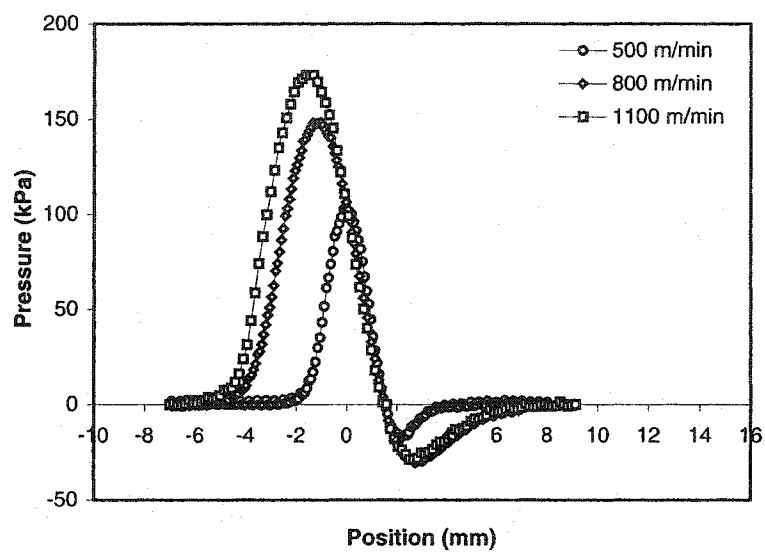
(a)



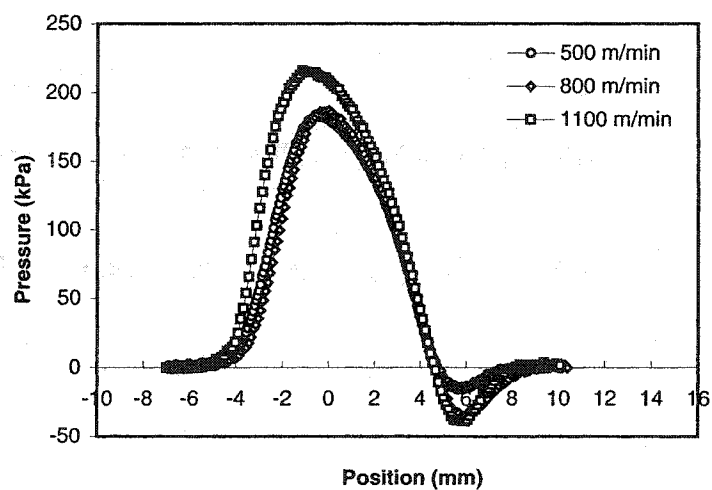
(b)

Figure 4.4 Pressure profiles for PEG20 ($\mu = 0.13$ Pa·s), $U = 500, 800$ and 1100 m/min:

(a) 2 kN/m; (b) 4 kN/m.



(a)



(b)

Figure 4.5 Pressure profiles for PEG30; $\mu = 0.63 \text{ Pa}\cdot\text{s}$, $U = 500, 800$ and 1100 m/min : (a) 2 kN/m ; (b) 4 kN/m .

It is important to point out that the deformable roll used for this work was covered with an elastomer having a hardness of 63 P&J, which is softer than the cover used in pilot coaters or industrial film coaters, in which the hardness can reach 45 P&J. In the present case, the applied load was not large enough to keep both rolls completely closed at U_{max} , and air was entrained into the nip; under these conditions the subatmospheric pressure increased.

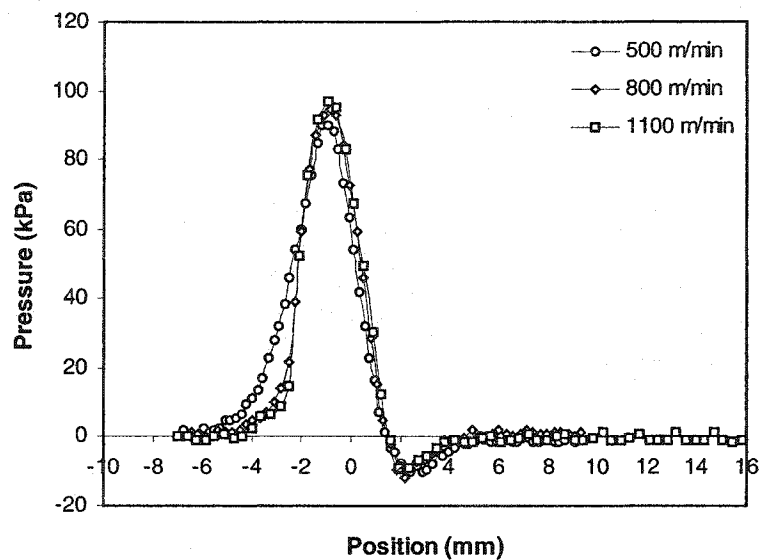
When the applied load was increased the trends in both positive and negatives regions were as expected (see Figure 4.4b and 4.5b). In other words the higher the roll speed the higher the maximum pressure into the nip and the lower the pressure at the exit. It must be noted that as the applied load increased the pressure at the exit of the nip decreased and if this pressure is low enough low some bubbles of air and water vapor can be formed into the liquid film. In the present case, the subambient pressure was not low enough to generate cavitation.

4.4.2 Imaging analysis

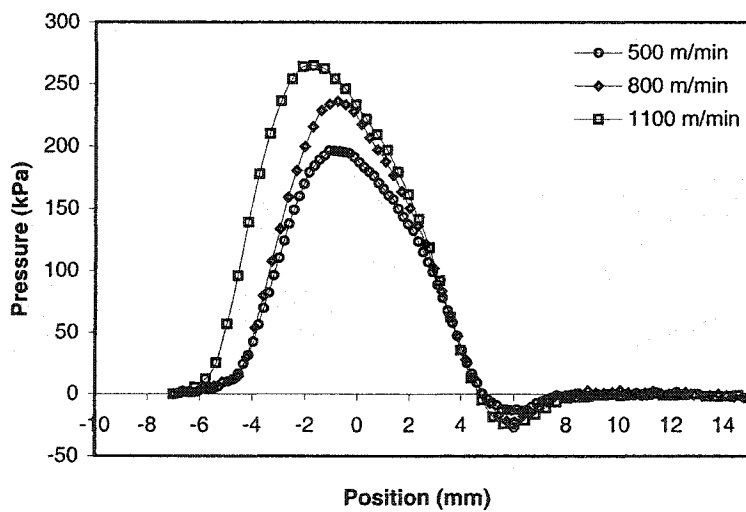
Figures 4.6 to 4.9 show the images obtained at the exit of the nip at the different test conditions, in which no septa were observed even for the lowest roll speed. For the low viscosity fluid (PEG 20, $\mu = 0.13 \text{ Pa}\cdot\text{s}$) and the lower level of applied load (2 kN/m) it is observed that filaments exhibit a long "c" shape when exiting the nip and only few droplets are ejected at low speed (500 m/min) (Fig. 4.6a). These filaments behave like a thread of a solid-elastic material attached at two parallel plates moving at high speed. Looking at Figure 4.6b, it can be seen that these filaments are less elongated and they break up closer to the nip and a higher amount of misting is generated as roll speed increases (800 m/min). Because no filaments are observed for the highest roll speed (1100 m/min) (Figure 4.6c), it is expected that they break up immediately at the exit of nip and only very small droplets are then ejected.

It has been stated that misting has its origin in the film splitting (MacPhee, 1997; Grön et al, 1998; Blayo et al, 1998). However, when a low applied load is used with a soft roll, air is entrained into the nip leading to a spray effect that can be partially responsible for the generation of misting. A similar effect is observed when a high viscosity fluid is used under the same conditions. The air entrainment into the nip can be also noted when looking at the negative pressure region of pressure profiles for both fluids at the lower level of the applied load. In both cases, the maximum negative peak pressure does not correspond to the highest roll speed. However, as Figure 4.7a evidences the filaments are wider and as they are elongated they become thinner and finally break up takes place far away from the nip. As expected, as the speed increases the filaments break up closer to nip and some droplet are ejected (Figure 4.7b). If the speed is further increased, once again both air entrainment and film splitting generate misting. As Fig. 4.7c shows, droplets of different sizes are generated. Under these conditions, small droplets are produced by film splitting, while the spray effect is mainly responsible for the large ones.

The spray effect can be avoided if the applied load is increased as Figure 4.8 shows. However, if the rolls are highly loaded the nip length extends and a smaller amount of fluid will pass through the nip, which can be observed at high speed (Figure 4.8c).



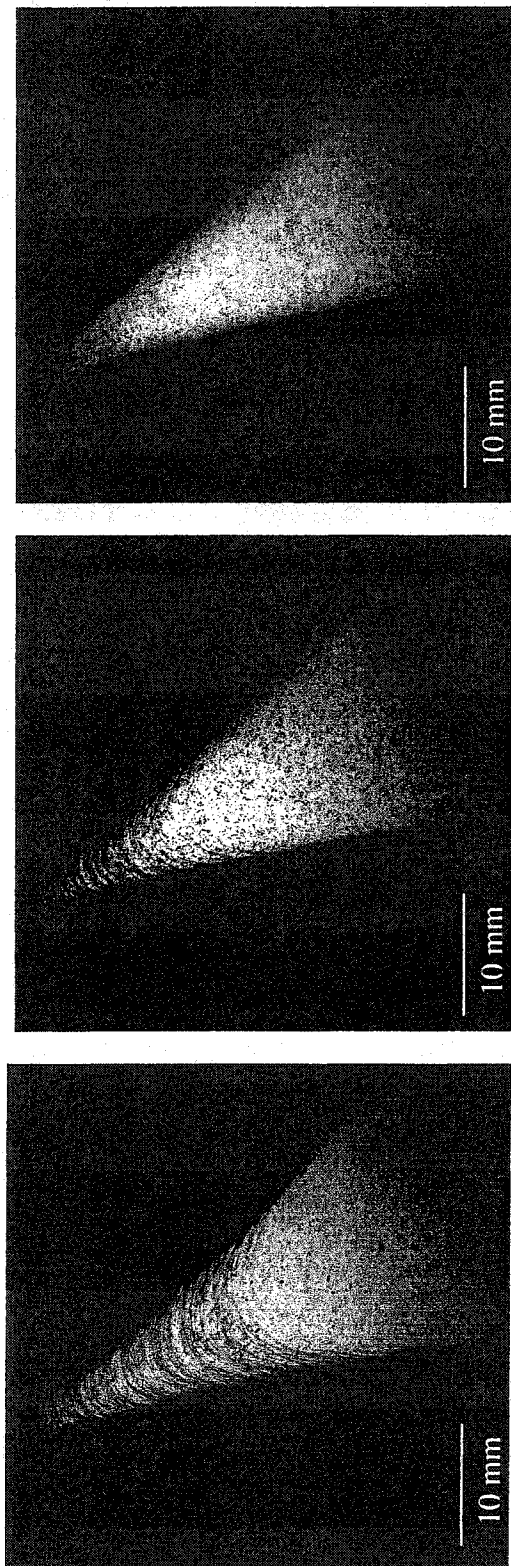
(a)



(b)

Figure 4.4 Pressure profiles for PEG20 ($\mu = 0.13 \text{ Pa}\cdot\text{s}$), $U = 500, 800$ and 1100 m/min :

(a) 2 kN/m; (b) 4 kN/m.

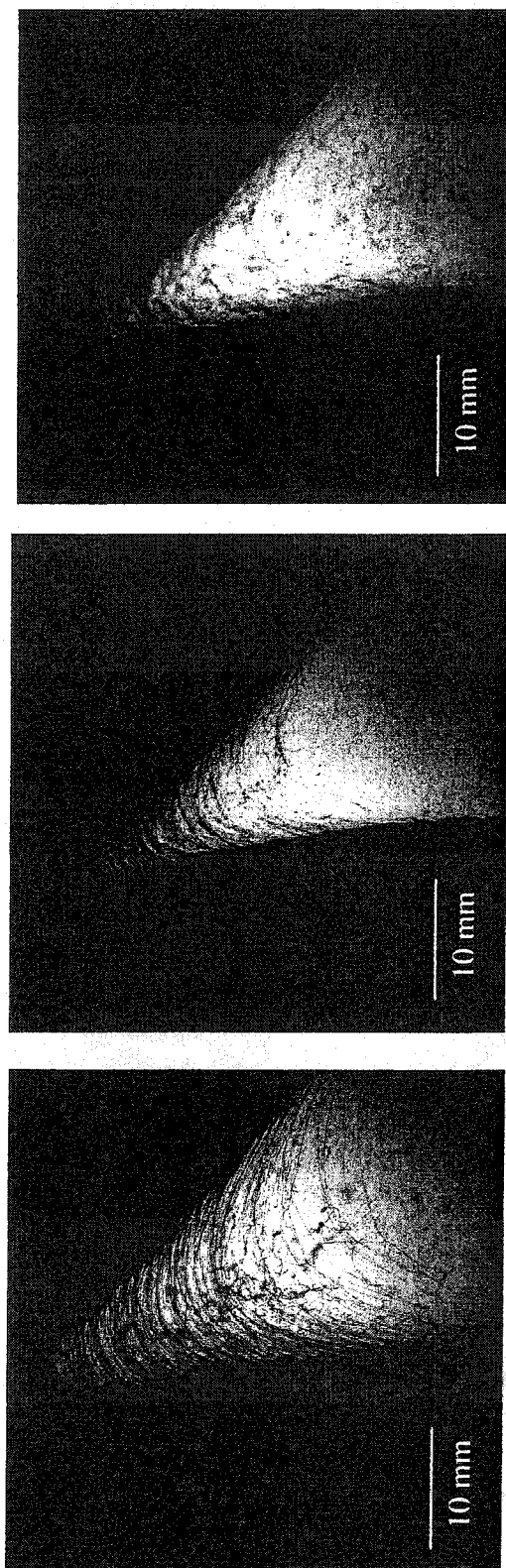


(a)

(b)

(c)

Figure 4.6 Images from the nip with PEG 20 ($\mu = 0.13 \text{ Pa}\cdot\text{s}$) at 2 kN/m : (a) 500 m/min ; (b) 800 m/min ; (c) 1100 m/min .

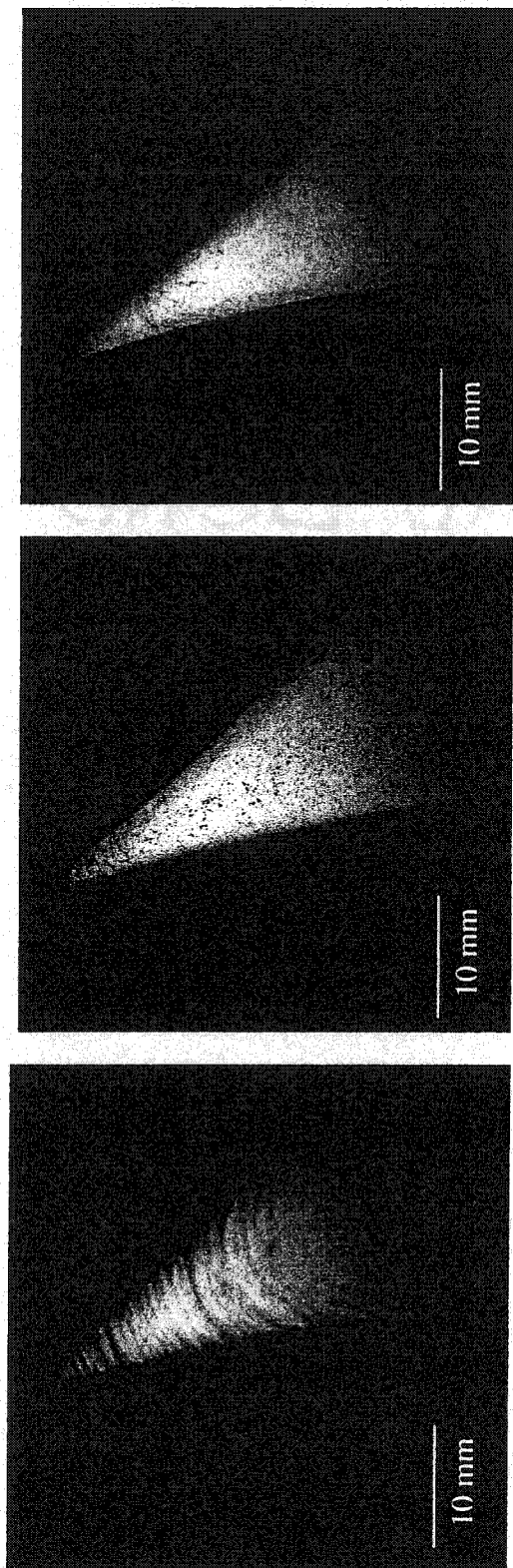


(a)

(b)

(c)

Figure 4.7 Images from the nip with PEG 30 ($\mu = 0.63$ Pa·s) at 2 kN/m: (a) 500 m/min; (b) 800 m/min; (c) 1100 m/min.

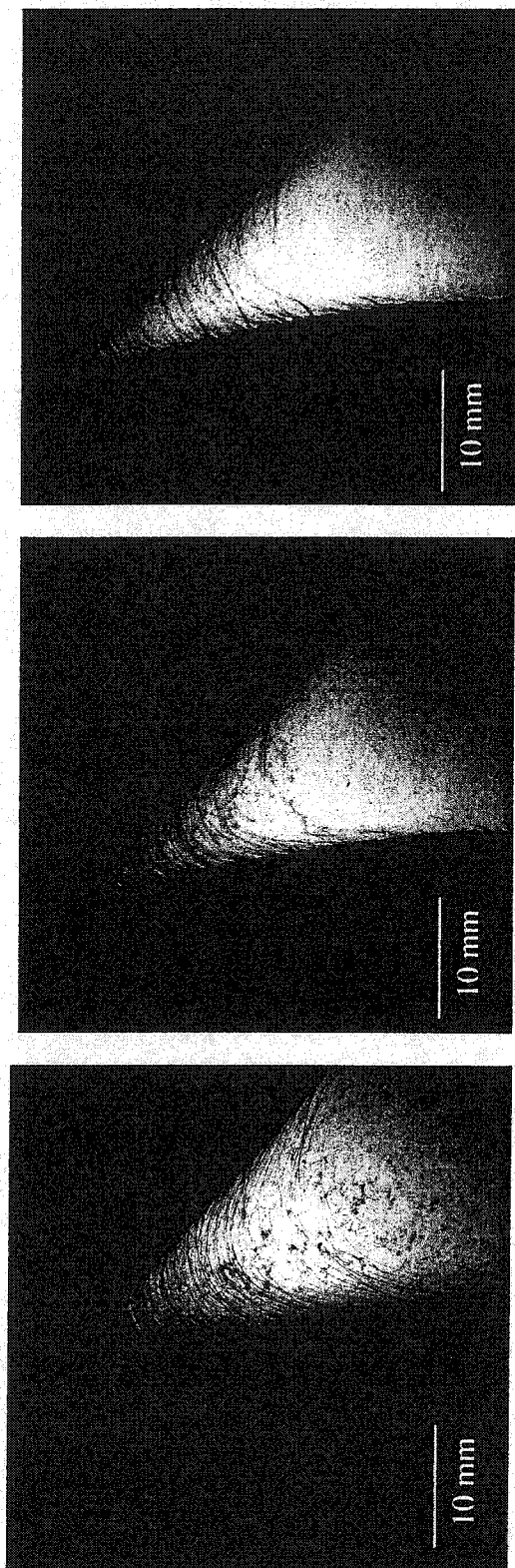


(a)

(b)

(c)

Figure 4.8 Images from the nip with PEG 20 ($\mu = 0.13 \text{ Pa}\cdot\text{s}$) at 4 kN/m : (a) 500 m/min ; (b) 800 m/min ; (c) 1100 m/min .



(a)

(b)

(c)

Figure 4.9 Images from the nip with PEG 30 ($\mu = 0.63 \text{ Pa}\cdot\text{s}$) at 4 kN/m : (a) 500 m/min ; (b) 800 m/min ; (c) 1100 m/min .

The trends observed for a high viscosity fluid at a high applied load are quite regular. As Figure 4.9 shows, the filaments are thinner than those obtained at a low applied load. They are also highly elongated and they break up at a long distance from the nip. It is important to point out that for the lowest roll speed either at low or high applied load, no misting is expected and the large droplets observed at the exit of the nip are broken filaments, which are smaller for a high applied load (see Figures 4.8a and 4.9a). Figures 4.8b and 4.9b show that as fluid viscosity increases misting tends to decrease even for high speeds confirming the results with coating colors, in which the higher the solid content of the coating color the lower the misting amount (Roper et al, 1997; Grön et al, 1998).

It has been demonstrated that under certain conditions (Banks and Mill, 1954; Pitts and Greiller, 1961) the character of the film splitting changes due to the onset of equally spaced ridges, parallel to the direction of travel, also known as ribbing, which is a typical hydrodynamic instability of roll coaters. It is important to point out that in our case, a ribbing phenomenon upstream the transfer nip was observed when a high applied load between the rolls was used. Figure 4.10 shows a sequence of six consecutive images with PEG 30 at 4 kN/m at 800 m/min. Even though the time between each image is very short (of the order of 4 ms), the flow appears periodic under these conditions. This can be also observed looking at Figure 11, in which the subambient region of the pressure profile for PEG30 at $W = 4$ kN/m was amplified. Because the amount of fluid passing through the nip changes with respect to the time, a similar behavior is observed with the pressure distribution. A jagged pressure profile is obtained for the subatmospheric region even though the signal has been electronically filtered. In such a case, both the fluid transferred to the web and the droplets ejection becomes time dependent. As a result of a periodic flow, some ridges in the cross direction of the web could appear leading to high orange peel level and as consequence a non-uniform coating would be obtained, which could be avoided if a harder roll covered is used.

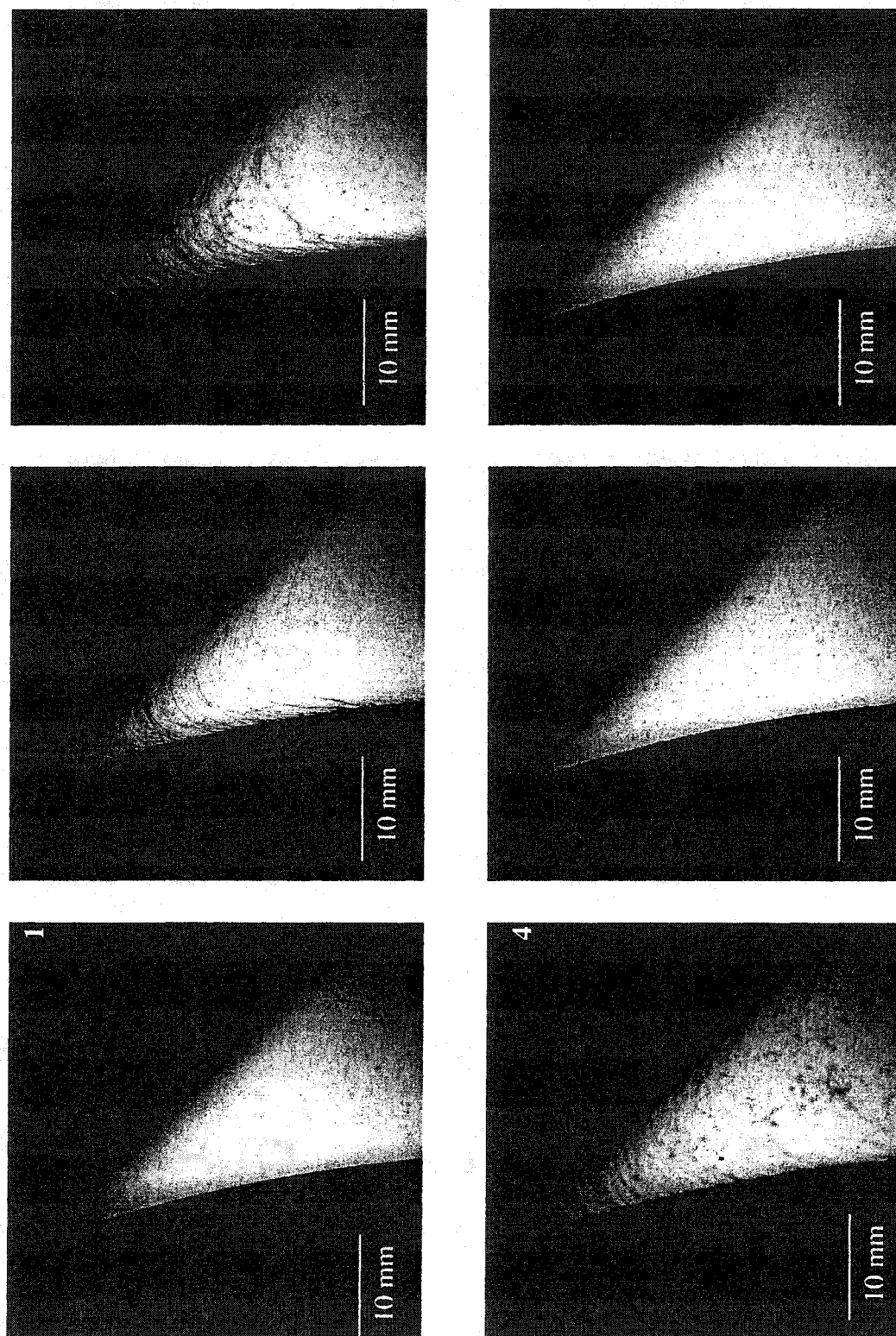


Figure 4.10 Sequence of six images downstream the nip with PEG 30 ($\mu = 0.63 \text{ Pa}\cdot\text{s}$) at 4 kN/m and 800 m/min

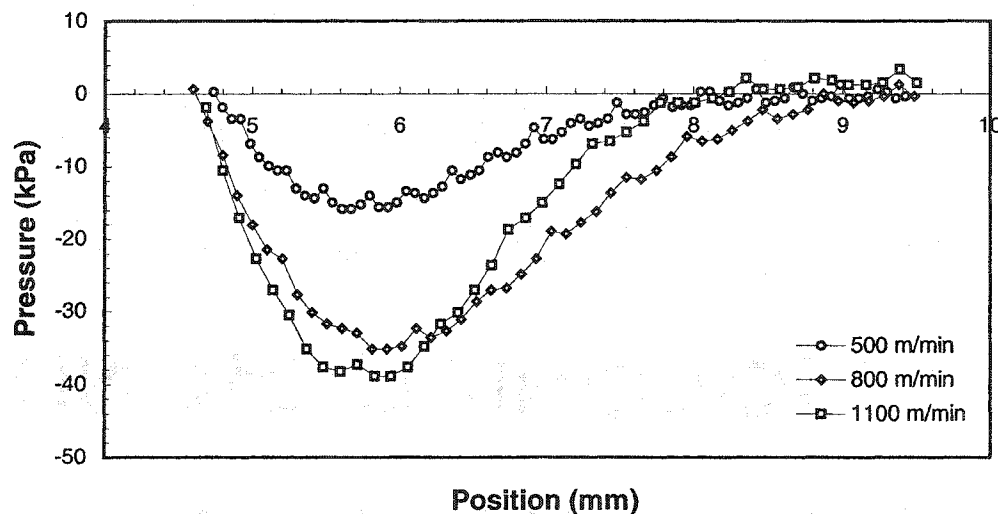


Figure 4.11 Negative region of pressure profile for PEG 30 at 4 kN/m.

4.5 Conclusions

A laboratory film coater was developed for studying the flow behavior of Newtonian fluids through a deformable nip. A high-speed video system allowed observing how the filaments are formed, stretched, broken up and how misting droplets are ejected to the surroundings. No septa were observed even for the lowest roll speed used in this work. It was observed that the negative peak pressure was not low enough compared with the vapor pressure of fluids, so that cavitation is not generated. When a low load applied between the rolls was used two different mechanisms were responsible for misting occurrence: film splitting and the spray effect due to air entrainment into the nip. At a higher load those phenomena could be avoided; however, the flow became periodic as the roll speed increases, which will lead to non-uniform coating. This can be avoided if harder elastomer covered rolls are combined with higher applied loads.

CHAPTER 5

NON-NEWTONIAN EFFECTS ON FORWARD DEFORMABLE ROLL COATING AT HIGH SPEED

G. Ascanio, P.J. Carreau and P.A. Tanguy*

CRASP, Department of Chemical Engineering, Ecole Polytechnique Montreal,
P.O. Box 6079, Stn. Centre-Ville, Montreal, QC H3C 3A7, Canada.

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5.1 Abstract

The flow of high solids content suspensions and coating colors through the deformable gap of counter-rotating rolls at high speed was investigated. Measurements of nip pressure profiles in a laboratory film coater were conducted and flow visualization experiments downstream the nip were carried out. A high-speed video system allowed showing the formation, elongation and break up of filaments that yield the misting droplets. The role of rheology on misting was also investigated using shear and extensional data. Results show that misting can be generated by film splitting and air entrainment into the nip when fluids exhibiting low viscosity at high shear rate flow through a highly deformed gap. From extensional rheological tests, it was also observed that the tendency to mist is reduced when the solids content and the extensional viscosity are both increased.

5.2 Introduction

Metered film coating is a versatile coating technology, which has generated considerable interest in the paper industry in recent years. It has been used for the production of many different coating grades such as precoated fine papers, low-weight coating for mechanical papers, etc. A metered film coater consists basically of a transfer roll and a backing roll, and a metering system. The two rolls are usually deformable and operated in a counter-rotating mode, while the metering system is composed of a delivery head and a co-rotating rigid bar. Metered film coating offers significant advantages over conventional blade coating as it allows the surface treatment of fragile webs. However, due to the film splitting phenomenon at the nip exit, there is a tendency toward the formation of "orange peel" especially at high solids typically above 62 wt% (Roper et al., 1998a). When the coating is applied at high speed, undesirable misting, i.e. the ejection of droplets from the application nip, also appears especially at high coat weights. These runnability issues are

very challenging to address and, as the coating equipment parameters and the web properties cannot be easily changed, the rheology of coating color formulations is the only practical factor that can be adjusted to control the runnability of film coaters.

Paper coating fluids also called colors are solids content aqueous suspensions composed mainly of mineral pigments, thickeners, latex and other binders, dispersing agents and water. The fluid properties have a strong influence on the performance of roll coaters. Liu et al. (1999) found from a experimental study that viscosity is the factor that determines the coated strip thickness when Newtonian and purely viscous non-Newtonian fluids flow through a rigid gap. In the case of deformable gaps, they observed that the flow could become unstable with viscoelastic fluids. In a film coater, coating colors are submitted to extreme shear rates and extensional rates ($\sim 10^6 \text{ s}^{-1}$) during a very short time in the metering nip and the transfer nip, respectively, which make the rheological effects very complex. Shear rheological properties of suspensions and coating colors have been studied in relation with coater runnability (Carreau and Lavoie, 1993; Yziquel et al., 1999; Kokko, 2001; O'Brien and Mackay, 2002, Page et al., 2002). As far as misting is concerned, the specific role played by rheology has been investigated by Triantafillopoulos and Smith (1998), Grön et al. (1997) and Roper et al. (1997, 1998b). It has been found that both the type and amount of thickener strongly influence the formulation viscosity. As the viscosity increases, it is believed that the rate of droplet ejection due to centrifugal forces decreases because of the viscous drag and the greater cohesive energy of the coating color (Triantafillopoulos and Smith, 1998; Grön et al., 1997). On the other hand, it has been found that latex has a minor influence on the viscoelastic properties (Fadat and Rigdhal, 1987). However, latex is a good option especially when coating at high speed because the rate of dewatering is enhanced without significantly increasing the coating viscosity (Roper et al., 1998b). The solids content also plays an important role on misting occurrence. It has been demonstrated that as the solids content increases misting decreases (Roper et al., 1998b).

Although extensional viscosity is likely to influence the generation of misting, attention in the literature has been rather focused on the shear rheological properties of coating colors and suspensions, and very little information on the extensional properties of suspensions and coating colors is available. From a theoretical point of view, at low strain rates the extensional viscosity of a fluid should be three times the shear viscosity (Trouton ratio). However, for rheologically complex fluids such as low solids content suspensions, the Trouton ratio can be of the order of 12 (Della Valle et al., 2000) and it has been reported to be as large as 30-60 for high solids content kaolin suspensions (O'Brien and Mackay, 2002).

The hydrodynamic conditions in film coaters also play a role on the generation of misting. While traveling through the nip, the fluid is subjected to a positive pressure in the center of the nip and a negative pressure at the exit. Although, the residence time in the nip is very short (of the order of 1 ms), it was found that if the negative pressure value (with respect to the atmospheric pressure) is sufficiently low, cavitation may appear in the fluid, which in turn tends to increase misting (MacPhee, 1997 a and b). CFD analysis of forward roll coating has also been carried out. Coyle (1984, 1988) and Coyle et al. (1987) performed a dimensionless analysis with Newtonian fluids, in which a one-dimensional elastohydrodynamic model suitable for small deformations was used for predicting the pressure profile between counter-rotating rolls. This model was further improved by Carvalho and Scriven (1994, 1997) and Carvalho (1996) who proposed a one-dimensional neo-Hookean model suitable for high roll surface deformations. They found also that the local displacement is a function of the whole pressure distribution, and a two-dimensional Hookean model was proposed to obtain a better prediction of the profile. Although the proposed models are very useful tools for predicting the nip pressure, the speed conditions used in these studies are significantly lower than those encountered in the paper industry.

The main objective of this work is to clarify the mechanisms governing misting occurrence in paper coating. This work will therefore describe the flow of rheologically complex fluids in a deformable gap of a forward roll coater at high speed. Flow visualization, the measurements of nip pressure distribution and shear and extensional rheology will be used to shed some new light on factors affecting misting occurrence.

5.3 Materials and Methods

5.3.1 Film coater

Figure 5.1 shows the high-speed laboratory film coater built to investigate misting. The set up consists of two counter-rotating rolls of 150 mm diameter and 150 mm long. The forward roll coating configuration used in this work is depicted in Figure 5.2. For illustration purposes the sketch is not to scale. The transfer roll is covered with an elastomer layer (Variokote, Stowe Woodward Inc.) of 9.5 ± 0.127 mm thickness and a

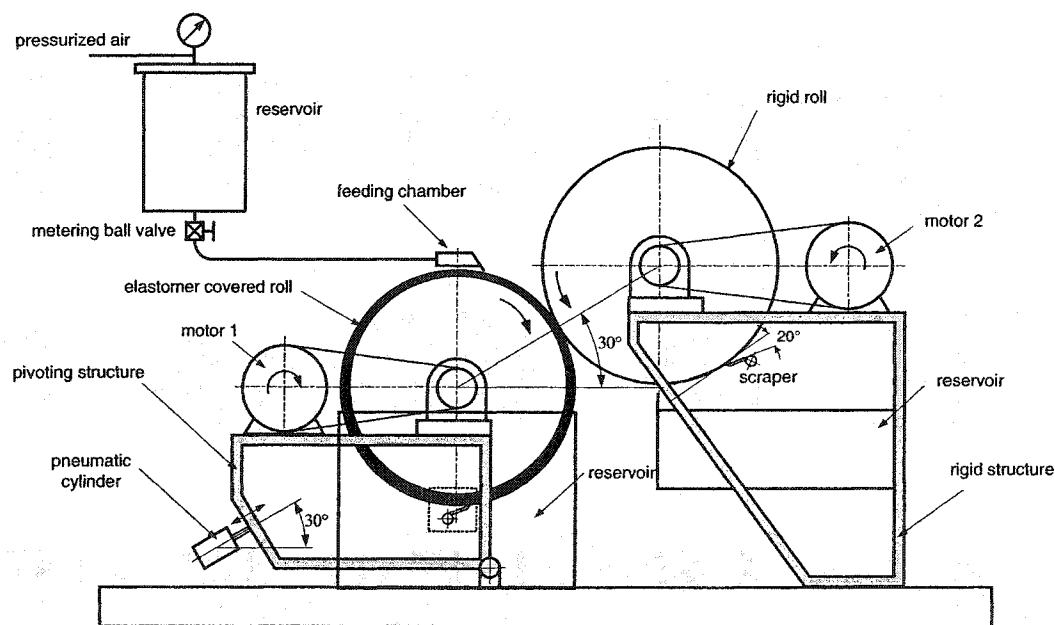


Figure 5.1 Laboratory film coater.

hardness of 63 P&J, allowing the transfer nip gap to be deformed. The backing roll is rigid with a smooth chrome plated surface to reduce wear when abrasive fluids such as coating colors are used. This roll has roughness of $2\text{ }\mu\text{m}$ and a run-out less than $3\text{ }\mu\text{m}$. Both rolls have been dynamically balanced in two planes at three points up to 6250 rpm. Each roll is driven by a 746 W electrical motor, which speed is carefully controlled by an AC solid-state variable frequency drive receiving a feedback signal from a speed encoder. The speed control allows operating in a range of 0 to 1600 m/min at constant torque. The applied load between the rolls can be varied from 2 to 4 kN/m by means of a pneumatic system. The fluid to be tested is contained in an air-pressurized reservoir, so that a constant flowrate can be delivered to the nip. A metering ball valve allows controlling the amount of fluid to be transferred by the feeding chamber. A doctor blade is used at the exit of this chamber for adjusting the fluid thickness and to avoid end effects. A scraping blade is mounted on each roll for removing the excess of fluid before re-entering the nip.

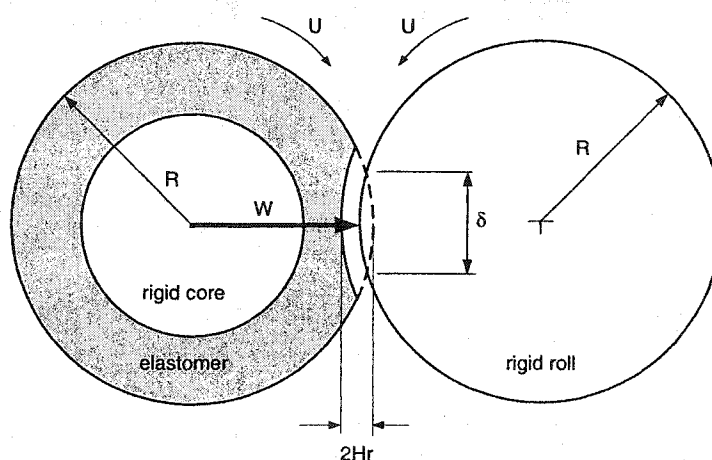


Figure 5.2 Forward deformable roll coating arrangement with negative gap.

The pressure profile in the transfer nip is measured with a high sensitivity piezoelectric pressure sensor (diameter 2.5 mm) mounted on the surface of the backing roll. It should be noted that this technique has already been successfully used for obtaining pressure profiles in both forward and reverse roll coating (Young, 1997; Réglat and Tanguy, 1997). The measurement range is from 0 to 690 kPa, with a resolution of

0.01% f.s., a response frequency of 250 kHz and a non-linearity of 2.0% f.s. The membrane of this sensor has been carefully machined and polished at the same radius as the backing roll in order to reduce intrusion effects. Several pressure profiles in the transfer nip region are acquired at every backing roll revolution (pressure along the nip length). The pressure transducer signal is transferred to the computer via a brushless slip ring. A PC analog-to-digital converter is used for recording the signals from the transducer for subsequent post-processing.

5.3.2 High-speed video

A high-speed video system was used for visualizing the fluid through the nip (Figure 5.3). The illumination system was based on a diode copper vapor laser providing pulses of infrared laser light with variable pulse durations and pulse frequencies. An external pulse from a camera strobe output triggered the laser. Imaging was performed with a high-speed video CMOS having a full resolution of 1024 x 1024 square pixels, which allows obtaining up to 462 frames/s at that resolution. The camera having an internal memory of 512 Mb, can store 512 images at high speed and full resolution. A total of 150 frames were obtained for each experiment at a recording speed of 231 frames/s at full resolution. The shutter was set at 2.2 ms and a focal magnification of 2 was used for enlarging the view of the exit of the nip region.

5.3.3 Fluids

Four suspensions were prepared with delaminated kaolin clay (Nuclay, Engelhard) and aqueous solutions of low solids content polyethylene glycol (PEG) having a molecular weight of 35,000 mol/g (Clariant). Carboxymethyl cellulose (CMC) Finnfix 10 and 30 (Noviant) having a molecular weight of 60,000 and 75,000 g/mol, respectively, were used as thickener in two suspensions. The pigments were fed into the tank at a rate of 9 kg/h and they were dispersed in water with a high-shear impeller (Cowles turbine)

with a tip speed of 7 m/s during 1 h. Then the tip speed was reduced to 4 m/s and the PEG was dissolved during 20 minutes. The final suspensions had a total solids content of 52 ± 0.2 wt% or 62 ± 0.2 wt% and the pH was about 7.5.

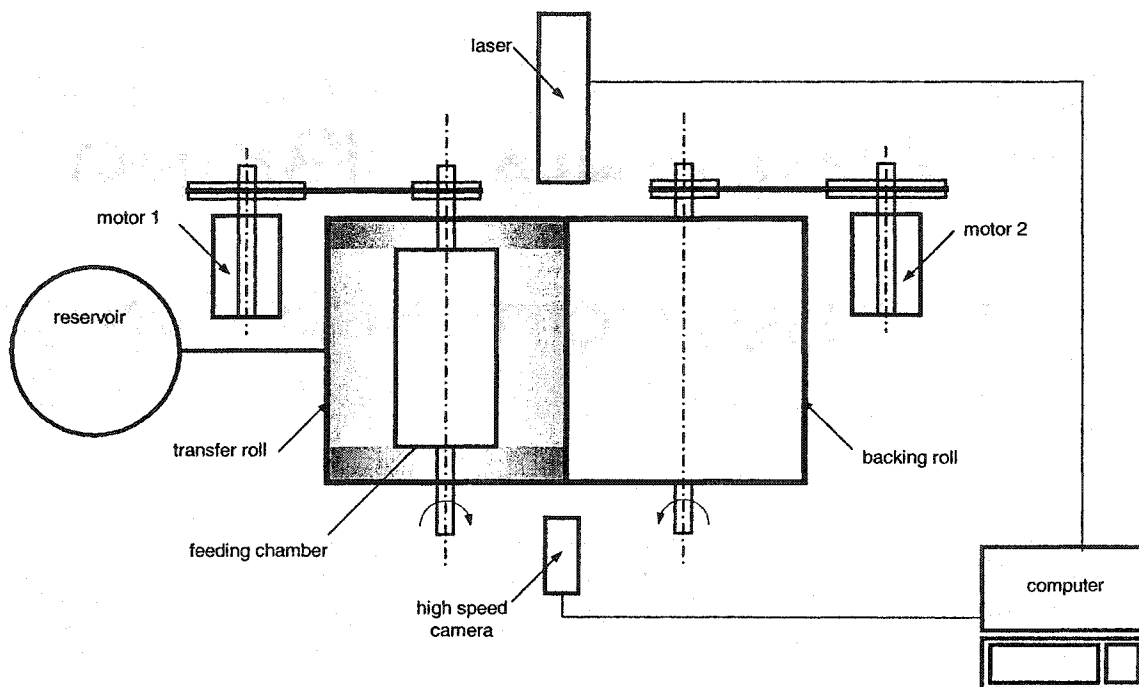


Figure 5.3 Experimental set-up.

Four coating colors were formulated with the same kaolin clay. The make-down process was as follows. The pigment slurries were first prepared under the same conditions as the suspensions. 0.15 parts per hundred (pph) of polyacrylate dispersant (Dispex N40V from Ciba) was then added to absorb the polyanions onto the surface of kaolin clay particles. This rendered the surface negatively charged creating repulsion between the particles and then a stable dispersion was formed. The next step consisted of stabilizing the viscosity by incorporating the thickener. Finnfix 10 and 30 was added to high and low solids content coating colors, respectively. Then a styrene-butadiene latex CP1030NA (Dow Chemical) was incorporated into the suspension. The pH was adjusted

between 8.0 ± 0.3 with sodium hydroxide (1 M) and the solids content of the final coating color was 52.0 ± 0.2 wt% and 62.0 ± 0.2 wt%.

The corresponding volume fraction ϕ was 30% and 35%, respectively for low and high solids content suspensions and coating colors. The particle size was measured with a particle size analyzer Mastersizer S (Malvern Instruments) and the surface tension was determined by the contact angle/surface tension analyzer FTÅ 200 (First Ten Ångstroms). Tables 1 and 2 show the recipes used for the suspensions and coating colors, respectively. The following nomenclature was used to distinguish the fluids. For suspensions the first two digits indicate the amount of PEG and the last two indicate the solids content followed by "CMC" for those containing thickener. For the coating colors, a letter "C" is followed by two digits indicating the solids content, a "L" and two digits for the amount of latex. All the amounts are expressed in wt%.

Both steady and oscillating shear rheological tests were performed with a controlled stress rheometer (Advanced Rheometer AR-2000, TA Instruments) in a Couette configuration (30 mm and 28 mm for the inside and outside cylinder diameter, respectively). The extensional viscosity data were obtained by using an orifice flowmeter based on the pressure entry technique (Della Valle, 2000; Ascanio et al., 2002a). All the fluids used for this work were allowed to settle at least 24 hours before performing the experiments in order to eliminate air bubbles. Shear and extensional viscosity measurements as well as all the experiments were carried out at room temperature (~ 24 °C).

Table 1. Composition and physical properties of suspensions.

Components	Suspensions			
	10PEG52	10PEG62	5PEG52-CMC	5PEG62-CMC
Delaminated clay (pph)*	100	100	100	100
PEG (pph)*	24	24	12	12
CMC Finifix 10 (pph)*	-	-	-	0.3
CMC Finifix 30 (pph)*	-	-	0.3	-
Physical properties				
Density (kg/m ³)	1355	1445	1380	1430
Solids content (wt%)	52±0.2	62±0.2	52±0.2	62±0.2
Mean particle size (µm)	3.44	3.36	3.06	3.38
Surface tension (mN/m)	22.5	16.5	22.0	17.2

*amounts based on 100 parts per hundred (pph) of kaolin clay

Table 2. Composition and physical properties of coating colors.

Components	Coating colors		
	C52-L8	C62-L8	C52-L12 C62-L12
Delaminated clay (pph)*	100	100	100
CMC Finifix 10 (pph)*	-	0.3	0.3
CMC Finifix 30 (pph)*	0.3	-	-
Latex FC 1030	8	8	12
NA(pph)*			
Physical properties			
Density (kg/m ³)	1350	1400	1330 1380
pH	8±0.3	8±0.3	8±0.3 8±0.3
Solids content (wt%)	52±0.2	62±0.2	52±0.2 62±0.2
Mean particle size (μm)	3.50	3.24	3.39 3.33
Surface tension (mN/m)	23.2	18.6	22.2 17.3

*amounts based on 100 parts per hundred (pph) of kaolin clay.

5.4 Rheology

5.4.1 Steady state shear

The steady state viscosity data of the suspensions and the coating colors are shown in Figure 5.4 for a shear rate ranging from 10^{-2} to $2.5 \times 10^3 \text{ s}^{-1}$. The viscosity curves are typical of high solids content suspensions: when increasing the shear rate, the viscosity first decreases according to a power-law model (shear-thinning) and then it tends to level off at high shear rates. No Newtonian plateau at low shear rates was observed in the shear rate range investigated. Figure 5.4a shows the effect of both the thickener and solids concentration on the viscosity data of the suspensions. The low solids content suspension with no thickener (10PEG52) reaches a long high-shear plateau for shear rates larger than 1 s^{-1} , which is not surprising considering that this suspension contains a large amount of polyethylene glycol (a Newtonian fluid). The 5PEG52-CMC suspension containing a smaller amount of PEG and CMC as thickener exhibits shear thinning up to 250 s^{-1} and from this point the viscosity remains constant. The trends are different for the high solids content suspensions. The suspension with no thickener (10PEG62) shows shear thinning up to 5 s^{-1} followed by a plateau and the viscosity slightly drops for high-shear rates, while the high solids content suspension containing CMC (5PEG62-CMC) follows the typical behavior of high solids content suspensions. The viscosity of these suspensions can be well described by a power-law expression with a high shear viscosity (η_{∞}), as proposed by Yziquel, et al. (1999):

$$\eta = m|\dot{\gamma}|^{n-1} + \eta_{\infty} \quad (5.1)$$

The parameters n , m and η_{∞} are reported in Table 3. Equation (5.1) is not suitable for fluids exhibiting a pronounced shear thickening, such as the low solids content coating colors (C52L8 and C62L12). As Figure 5.4b shows, all the coating colors exhibit shear thinning up to a shear rate of about 10^3 s^{-1} . The onset of shear thickening appears at a larger shear rate for the high solids content coating colors. It should be noted that a stronger effect of the latex type on the viscosity was observed for the low solids content

coating colors (C52L8 and C52L12) between 10^{-2} and 80 s^{-1} and from this point both fluids have a very similar behavior. The high solids content coating colors (C62L8 and C62L12) follow practically the same trend in the whole range of shear rate investigated. No significant effect of the latex type is observed for these suspensions.

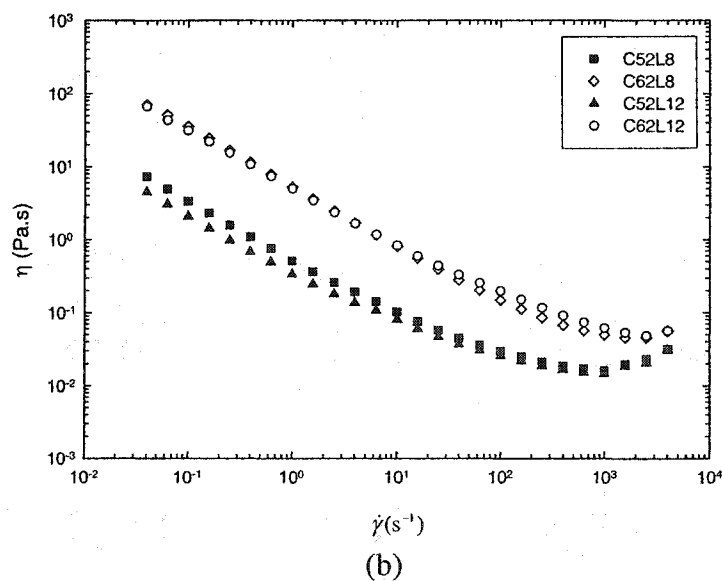
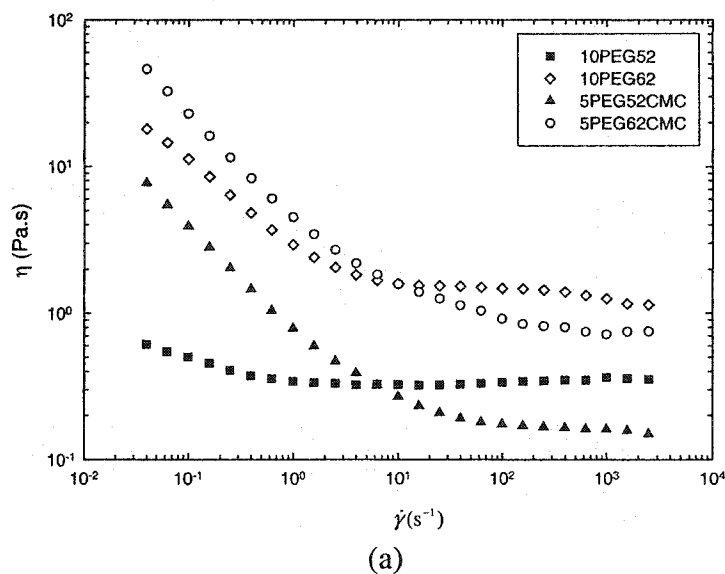


Figure 5.4 Steady shear viscosity as function of shear rate for (a) suspensions and (b) coating colors.

5.4.2 Oscillatory shear

Figure 5.5 reports the elastic modulus (G') and viscous modulus (G'') as functions of the strain amplitude at 1 Hz for the suspensions (Fig. 5.5a) and coating colors (Fig. 5.5b). All these suspensions exhibit a nonlinear viscoelastic behavior. For all fluids both moduli are constant at low strain amplitude up to a certain value known as the critical strain, and from this point the moduli decrease, then a sudden increase is observed at larger strain values (~ 2). This increase has been attributed to strain induced structure changes for strains of the order of the interparticle distances in the media (Carreau and Lavoie, 1993; Lavoie and Carreau, 1997). As Figure 5.5a shows, the low solids content suspensions present a larger critical strain than the high solids content ones. It is also observed that the viscous modulus is slightly smaller than the elastic modulus for the high solids content suspensions but larger for the low solids content ones. G'' is about 10 times larger than G' for suspension 10PEG52 and almost equal for suspension 5PEG52-CMC at a strain of 0.1. In the case of suspension 10PEG62 G'' is about 2 times larger than G' , while for 5PEG62-CMC G'' is smaller than G' for a strain up to 0.15 and above this point the tendency is inverted. A larger elastic contribution at low strain has been related to a solid-like structure (Lavoie et al., 1997). At large strains the ratio G'/G'' is much smaller than the unity, so that the elastic effects on the rheological behavior of the suspensions investigated can be neglected for coating processing. It is important to point out the effect of the solvent viscosity on the rheological properties of suspensions. The loss modulus G'' is about 2 times larger for the suspensions containing 10 wt% of PEG than those with 5 wt%. It is observed as well that G'' of suspensions 10PEG52, 10PEG62, 5PEG52-CMC and 5PEG62-CMC increases with deformation for values from 3 to 5, 2 to 3.5, 5 to 7 and, 2.5 to 4, respectively. This growth of G'' , previously observed by Yziquel et al. (1999) and by Page et al. (2002), has been related to flow induced microstructure changes. Beyond these values, G'' decreases with increasing deformation.

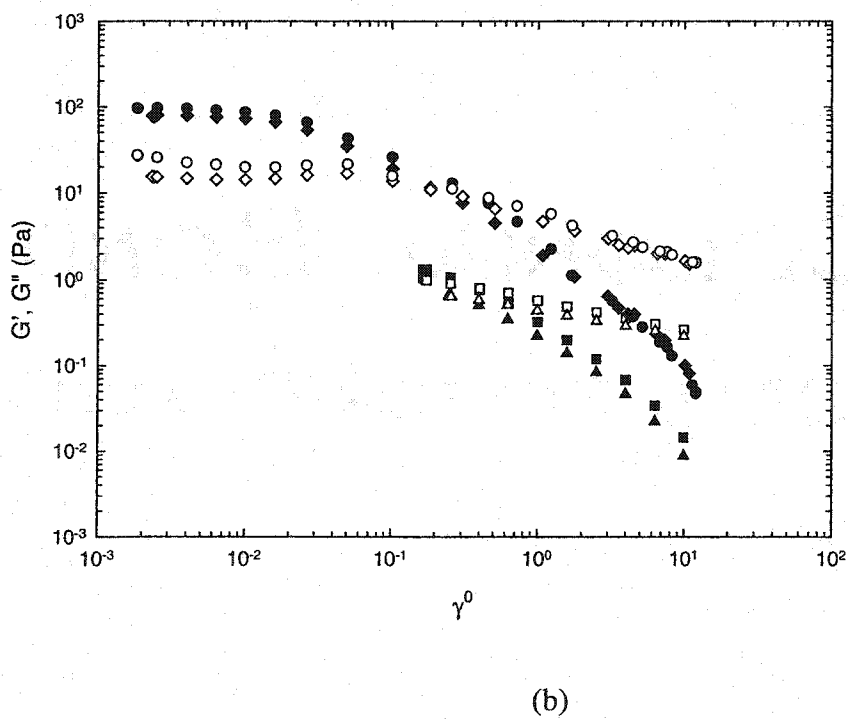
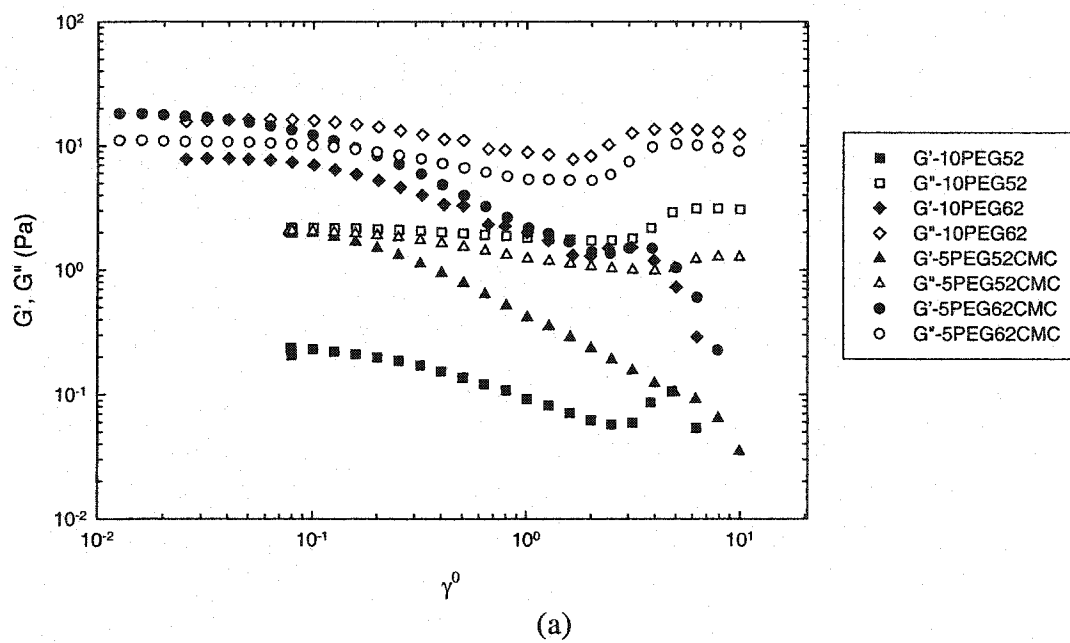


Figure 5.5 Dynamic moduli as functions of strain amplitude for (a) suspensions and (b) coating colors.

The trends for the coating colors investigated here are different. The critical strains for the low solids content colors are two or three orders of magnitude higher than that of the high solids content coating colors. It must be noted that the colors having a 62 wt% solids concentration exhibit a G' about 5 times larger than G'' but from a strain larger than 0.15 the tendency is inverted. For the low solids content coating colors, the level of elasticity is much lower compared to high solids content suspensions. The elastic modulus of suspensions with 52 wt% solids decreases above the critical strain, while a very slight increase of the loss modulus with deformation between 0.01 to 0.04 was observed for the high solids content suspensions only (C62L8 and C62L12) and then G'' decreases with increasing strain. The amount of latex does not appear to play a significant role on both G' and G'' in the viscoelastic nonlinear domain, confirming the observations of Fadat and Rigdahl (1987). However, we cannot be conclusive since the coating colors contain a different CMC, although of only slightly different molecular weight.

Table 3 also gives values of the storage and loss moduli in the linear regime at 1 Hz, a characteristic elastic time λ (Page et al., 2002). The characteristic elastic time is defined by

$$\lambda = \frac{G'}{G'' \omega} \quad (5.2)$$

where ω is the oscillating frequency, 6.28 rad/s. The critical strain was taken as the maximal strain for which G' was equal to 95% of the plateau elastic modulus.

In general terms, the characteristic elastic time (λ) is mainly a function of the volume fraction (ϕ). λ is 1.5 to 5 times longer for the high solids content suspensions, while λ is about 5 times longer for the coating colors with a high volume fraction. The thickener appears to have a stronger effect on λ for the suspensions. The characteristic elastic time of suspensions containing CMC is 7 times and 3.5 times longer for 52% wt solids and 62 %wt solids, respectively compared to the suspensions with no thickener. In the case of

the coating colors, it is observed that λ decreases when increasing the amount of latex. However, it is difficult to see the effect of CMC because of its different molecular weight.

5.4.3 Extensional rheology

An orifice flowmeter was used for measuring the extensional properties of the suspensions and the coating colors. The flowmeter was previously calibrated (pressure drop vs. flow rate) in terms of the Euler number versus Reynolds number with Newtonian fluids (Della Valle et al., 2000). This calibration curve was used to determine the extensional viscosity from flowrate (Q) and pressure drop (ΔP) data for fluids passing through a small orifice of diameter d_0 . The effective strain rate $\bar{\dot{\gamma}}$ is taken as half the square root of the second invariant of the rate-of-deformation tensor (Carreau et al., 1997), i.e.:

$$\bar{\dot{\gamma}} = \sqrt{3} \dot{\epsilon} \quad (5.3)$$

$\dot{\epsilon}$ is the extensional rate defined by

$$\dot{\epsilon} = k_s \left(\frac{32Q}{\pi d_0^3} \right) \quad (5.4)$$

where k_s is the classical Metzner-Otto proportionality constant (Metzner and Otto, 1957; Della Valle et al., 2000) adapted to calculate the extensional rate for non-Newtonian fluids. Della Valle et al. (2000) determined numerically that the extensional rate with purely shear-thinning fluids best fit was obtained with $k_s = 2.1$. The effective strain rate is then:

$$\bar{\dot{\gamma}} = 29.1 \left(\frac{v_0}{d_0} \right) \quad (5.5)$$

where v_0 is the velocity of the fluid in the orifice.

The apparent extensional viscosity is determined from the expression of calibration curve in the laminar regime ($Re < 20$)

$$Eu = \frac{38}{Re} \quad (5.6)$$

where the Euler number and Reynolds number are defined as

$$Eu = \frac{\Delta P}{\frac{1}{2} \rho v_0^2} \text{ and } Re = \frac{\rho v_0 d_0}{\eta} \quad (5.7)$$

From Eqs. (5.6) and (5.7), isolating η and using the theoretical Trouton ratio value of 3 ($\eta_E = 3\eta$) we get the following expression for the extensional viscosity:

$$\eta_E = \frac{\Delta P d_0}{19 v_0} \quad (5.8)$$

For $Re > 100$ the inertial forces are no longer negligible. Therefore we have corrected the pressure drop using:

$$\Delta P_{corr} = \Delta P - \frac{1}{2} \rho v_0^2 \quad (5.9)$$

As proposed by Ascanio et al. (2002a), a good approximation of the Trouton ratio can be obtained by:

$$Tr = 3 \left(\frac{Eu}{38} \right) Re \quad (5.10)$$

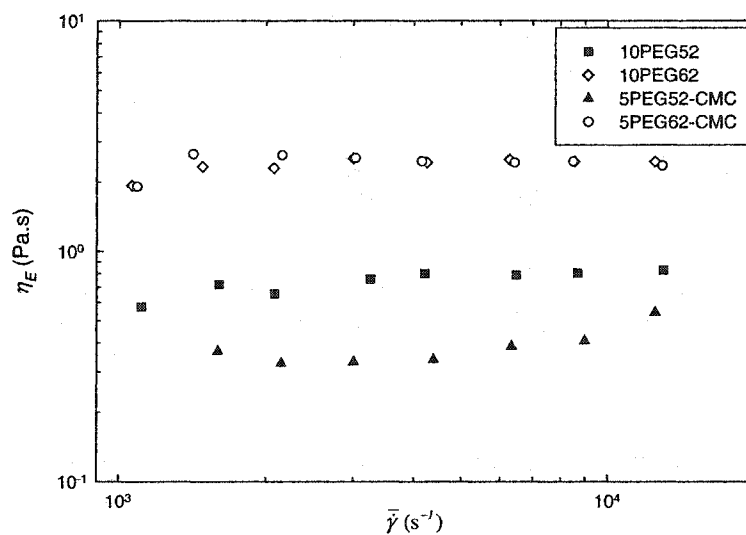
where Eu is the experimental Euler number and Re is the Reynolds number determined by:

$$Re = \frac{\rho v d_0}{\eta_\infty} \quad (5.11)$$

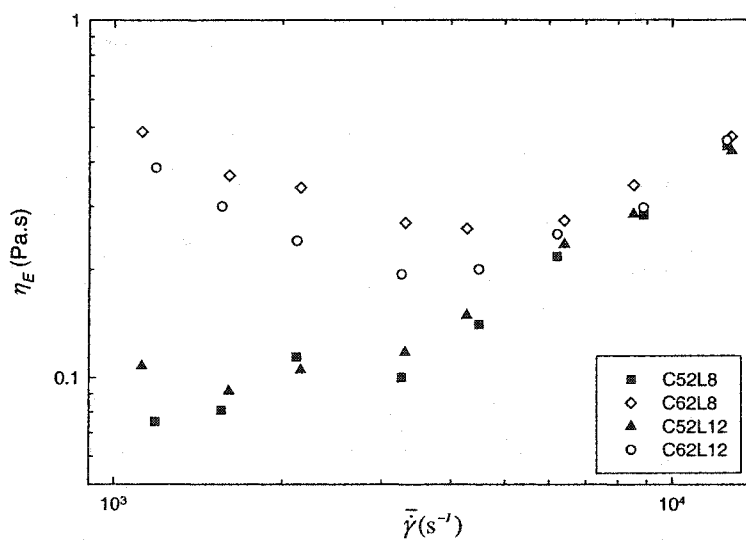
where η_∞ is the high-shear rate viscosity.

The apparent extensional viscosity of the suspensions and the coating colors obtained via Eq. (5.8) is plotted in Figure 5.6 as a function of the effective strain rate ranging from 10^3 to $12.5 \times 10^3 \text{ s}^{-1}$. Both high solids content suspensions (10PEG62 and 5PEG62-CMC)

have the same extensional viscosity and exhibit a slight strain hardening up to strain rates of the order of 10^3 s^{-1} and then a plateau viscosity of about $2.4 \text{ Pa}\cdot\text{s}$ is reached. The low



(a)



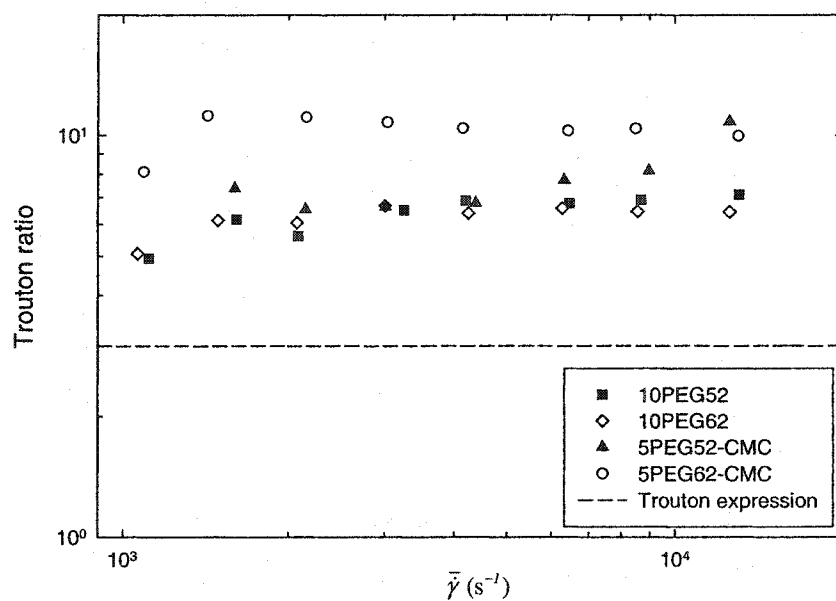
(b)

Figure 5.6 Extensional viscosity as function of strain rate for (a) suspensions and (b) coating colors.

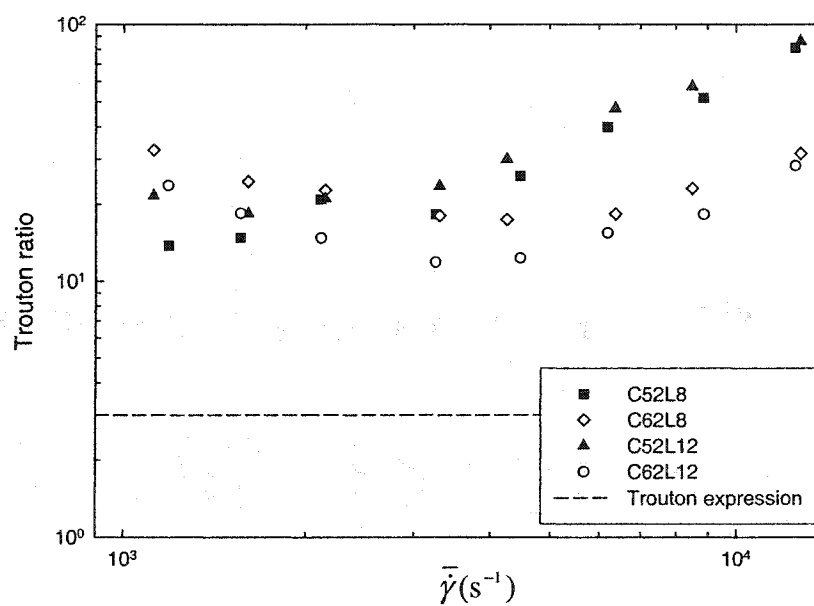
solids content suspension with no thickener (10PEG52) exhibits a similar behavior with a plateau viscosity. The extensional viscosity of the corresponding suspension containing CMC (5PEG52-CMC) is relatively constant at low strain rates, but of smaller value than that of 10PEG52. Above $2.5 \times 10^3 \text{ s}^{-1}$ this suspension exhibits a slight strain hardening.

The low solids content coating colors (C52L8 and C52L12) show a constant extensional viscosity up to $2 \times 10^3 \text{ s}^{-1}$ and above this point they exhibit strain hardening. The high solids content coating colors (C62L8 and C62L12) are strain thinning up to $4.5 \times 10^3 \text{ s}^{-1}$ and then strain hardening for larger extensional rates, confirming the results obtained by O'Brien and Mackay (2002) with high solids content suspensions. It is observed that the amount of latex combined with CMC has a direct effect on the extensional viscosity of coating colors. For the low solids content colors, the extensional viscosity is larger mainly in the plateau region when increasing the amount of latex. For the high solids content colors, a larger amount of latex tends to reduce the extensional viscosity mainly in the strain-thinning region. The latex used in this work was a styrene-butadiene specially formulated for reducing misting in roll coating. As observed by Kokko (2001) the use of certain latexes can largely affect the extensional viscosity. As far as the thickener is concerned, no important effect of the CMC type has been observed on the apparent extensional viscosity probably due to the similar molecular weight of both CMC.

Finally, it is interesting to compare the ratio of the extensional viscosity to the shear viscosity by plotting the Trouton ratio, Tr , defined by Eq. 5.11 as a function of the effective deformation rate, which is equal to the shear rate for simple shear and to $\sqrt{3} \dot{\epsilon}$ for extensional flows. Figure 5.7a shows that a fairly constant Trouton ratio is obtained for all the suspensions with values ranging from 5 to 12, which is in good agreement with the results obtained by Della Valle et al. (2000) for suspensions containing 50-65 wt% solids.



(a)



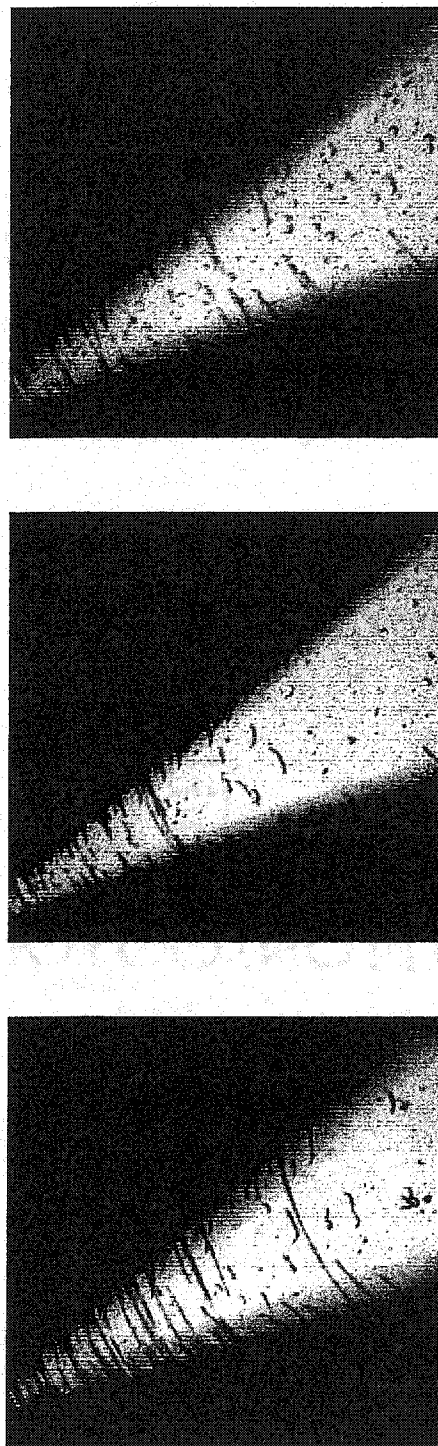
(b)

Figure 5.7 Trouton ratio for (a) suspensions and (b) coating colors.

As Figure 5.7b shows, a Trouton ratio ranging from 10 to 85 is obtained for the coating colors confirming the results of O'Brien and Mackay (2002) for larger solids content suspensions. The main reason for these quite different Tr values is likely due to the interaction of the components used in the make-down of the coating colors.

5.5 Imaging analysis

Figure 5.8 shows the images obtained at the exit of the nip with a low solids content suspension (10PEG52) at three different speeds. Under similar operating conditions with Newtonian fluids, Ascanio et al. (2002) observed that filaments with a "c" shape are formed at the exit of the nip. These filaments can be highly elongated depending on the fluid viscosity. In the literature it has been reported that viscous or viscoelastic fluids such as adhesives, may form septa, which are defined as a series of curved thin sheets of liquid extending from the nip (Owens et al., 2002; Vinjamur and Scriven, 2002). These septa find their origin in ribbing. Ribbing is extended from the film-split resulting in septa, which may be due, according to the authors, to the extensional thickening behavior. The same trends were observed by Vinjamur and Scriven (2002) with Boger fluids prepared with a high-molecular weight polymer (PEO, $M_w = 5 \times 10^6$ g/mol). The results in our case are different. Because the roll speed is considerably higher in the present work (1000 m/min versus 300 m/min), the capillary number ($Ca = \mu V / \sigma$) is significantly larger. However, it is well known that a lighter molecular weight polymer such as the PEG used for dispersing the kaolin clay in the suspensions, diffuses more rapidly to the interface causing compressive force to act on the surface, thereby reducing the surface tension. The magnification used for visualizing the flow downstream the nip does not allow to state if small septa were formed very close to the application nip. It is observed that the filaments of the suspension 10PEG52 become straight due to the low surface tension presumably as a result of the interaction of PEG and kaolin clay. These filaments could result from a hydrodynamically unstable process mainly due to ribbing.



(a)

(b)

(c)

Figure 5.8 Images downstream the nip with 10PEG52: (a) $Ca_E = 280$; (b) $Ca_E = 450$; (c) $Ca_E = 620$.

Because shear effects can be neglected when using rolls rotating at the same speed in forward mode, we propose to redefine the capillary number using the extensional viscosity (η_E) as follows:

$$Ca_E = \frac{\eta_E U}{\sigma} \quad (5.12)$$

where U is the roll speed, σ is the surface tension and η_E is extensional viscosity as a function of the extensional rate $\dot{\epsilon}$ defined by

$$\dot{\epsilon} = \frac{U}{H_R} \quad (5.13)$$

where H_R is the half nip gap.

Figure 5.8 shows how the filaments are stretched, break up and droplets are generated. Filaments can break up in more than two parts depending on the fluid rheological properties and the operating conditions. As evidenced in Figure 5.8a, a combination of large droplets and small pieces of filaments are both ejected at the lowest capillary number (Ca_E).

For the conditions used in this work, it is clear that the extensional forces are much larger than surface tension forces. The filaments break up closer to the nip and a larger amount of droplets is produced (Figure 5.8b) as Ca_E increases. For the largest Ca_E it is observed that only small droplets are ejected and some filaments recoil after breaking up, which can be associated to solid-like behavior (Figure 5.8c).

Figure 5.9 shows two images downstream the nip with the high solids content suspension (10PEG62) for two capillary numbers. It is observed that the filaments become thicker when high solids content suspensions are used. For a Ca_E of 120 (Fig. 5.9a) the filaments are highly elongated before breaking up in two or three parts. As the capillary number increases these filaments break closer to the nip and the first droplets are ejected. Under these conditions, no air entrainment is observed and misting is presumably generated by

film splitting. Because of technical difficulties involved in the visualization of coating colors flows, only images obtained with suspensions can be shown.

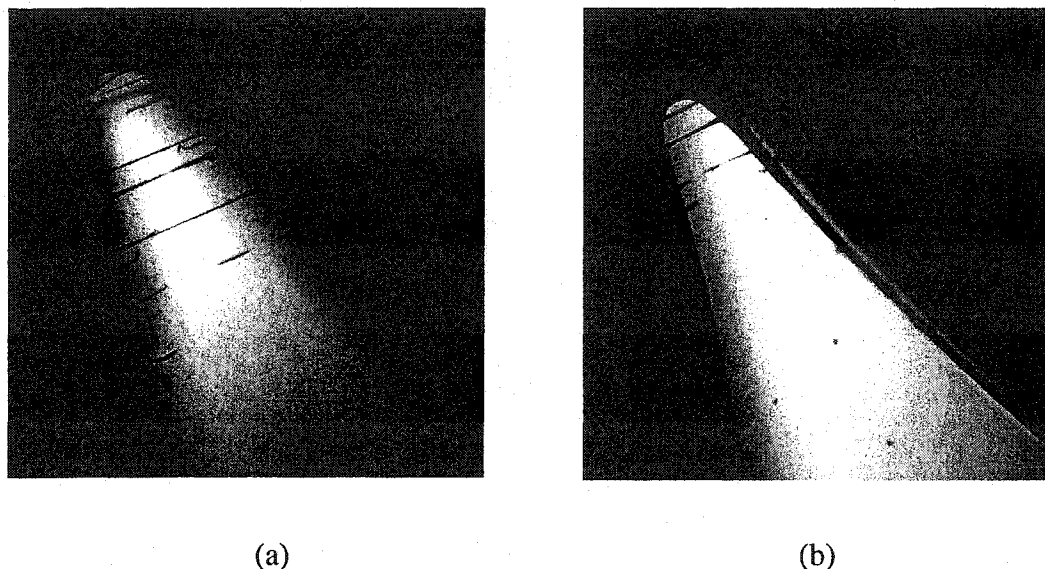


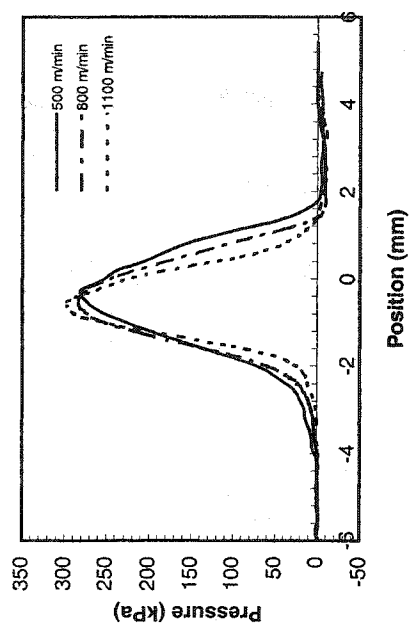
Figure 5.9 Images downstream the nip with 10PEG62: (a) $Ca_E = 120$; (b) $Ca_E = 240$.

5.6 Pressure profiles

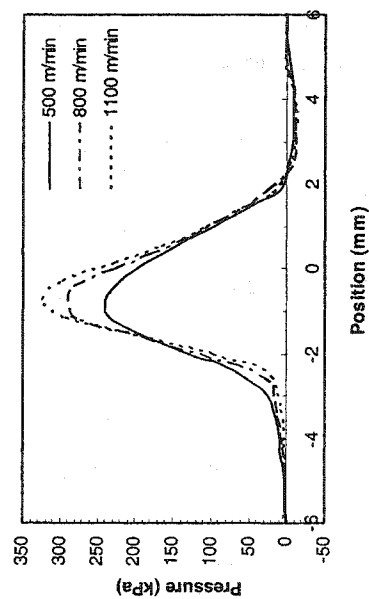
The pressure profiles reported here were obtained with negative gaps by pressing the deformable roll against the rigid roll, where the dynamic pressure sensor was installed. An applied load between the rolls of 3 kN/m set for all the experiments and the corresponding nip gap and length were 340 μm and 10.4 mm, respectively. The nip gap (H_r) was determined by (Cohu and Magnin, 1997)

$$H_r = R - \sqrt{R^2 - \left(\frac{\delta}{2}\right)^2} \quad (5.13)$$

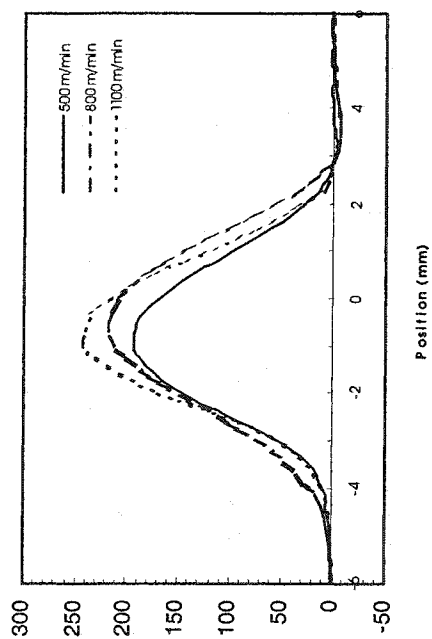
where R is the roll radius and δ is the nip length experimentally measured by image analysis. The roll speed was varied from 500 to 1100 m/min in the forward mode.



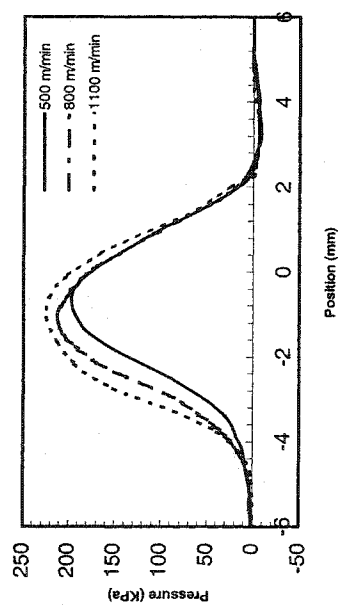
(b)



(d)



(a)



(c)

Figure 5.10 Pressure profiles for suspensions at $W = 3$ kN/m and $U = 500, 800$ and 1100 m/min: (a) 10PEG52; (b) 10 PEG62; (c) 5PEG52-CMC; (d) 5 PEG62-CMC.

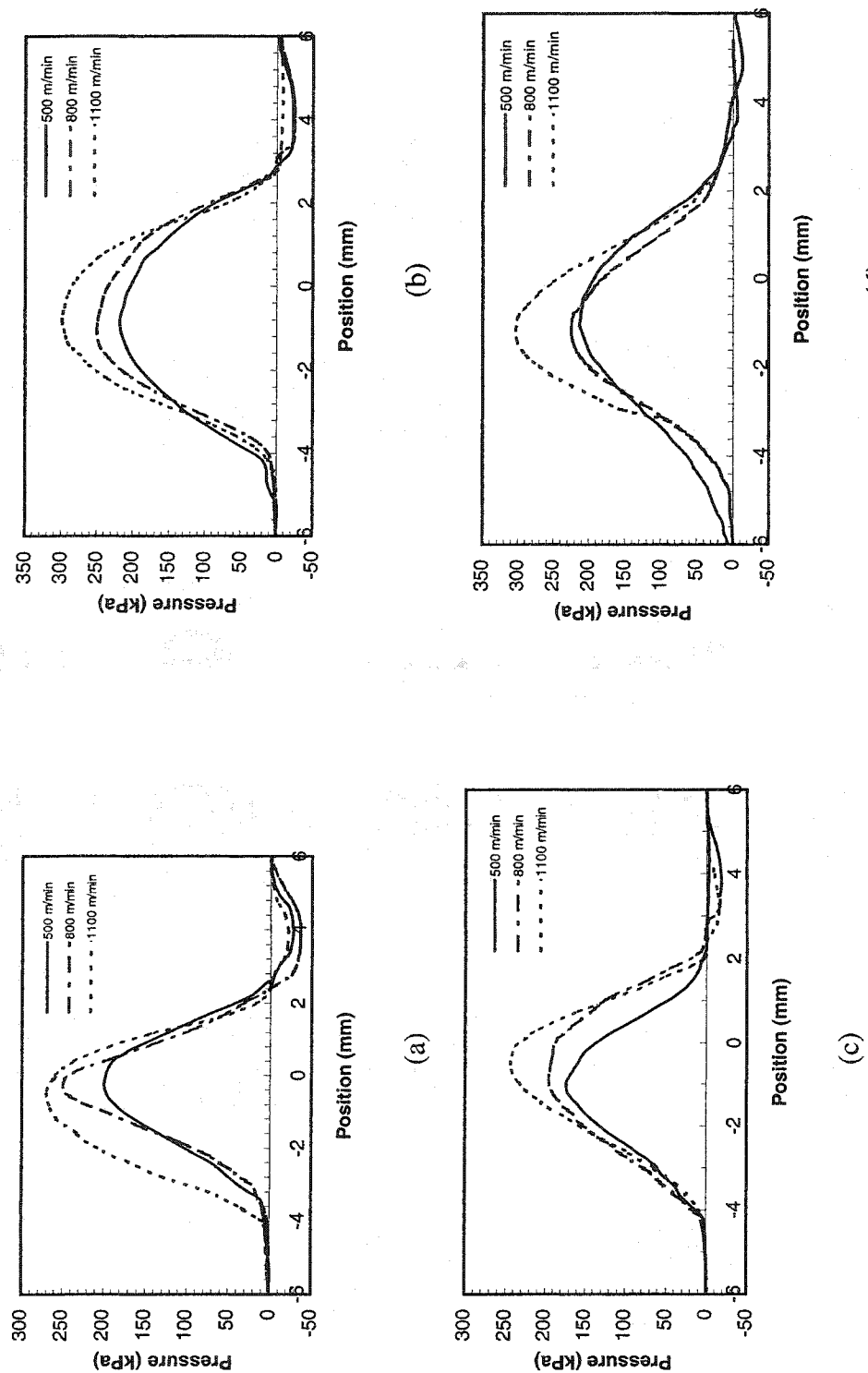


Figure 5.11 Pressure profiles for suspensions at $W = 3 \text{ kN/m}$ and $U = 500, 800$ and 1100 m/min : (a) C52L8; (b) C62L8; (c) C52L12; (d) C62L12.

Figures 5.10 and 5.11 show the pressure profiles obtained for the suspensions and the coating colors. As mentioned before, as the fluid passes through the nip, it experiences a region of positive pressure up to a maximum value (peak height) and, under certain conditions, it can be submitted to a sub atmospheric region (valley depth) at the exit of the nip.

As evidenced in Figure 5.10, both the peak height increases and the valley depth decreases with roll speed. However, the low solids content suspensions (10PEG52 and 5PEG52-CMC) exhibit a different behavior; the valley depth increases up to a certain roll speed and then it decreases when speed is further increased. We believe that this could be explained by air entrainment into the nip (Figures 5.10a and 5.10c). As the speed increases, some fluid can be ejected by the transfer roll due to centrifugal forces, so that a small amount of fluid will flow through the nip entraining air. This effect is more important when fluids with low infinite shear viscosity (η_{∞}) are used. Under these conditions, misting droplets can be generated by film splitting and spray effect due to the air entrainment. For suspensions with larger η_{∞} the results are as expected, or in other words the higher the roll speed the higher the peak height and the lower the valley depth at the exit of the nip (Figures 5.10b and 5.10d).

In the case of the coating colors, the peak height increases with the roll speed but the deepest valley does not correspond to the highest speed. This is also attributed to the entrainment of air into the nip. It is observed that coating colors yield lower pressure levels at the exit of the nip than suspensions. Although the pressure of the fluid exiting the nip can drop below the vapor pressure (MacPhee, 1997a, b), our results show that the valley depth is not large enough to believe that cavitation can induce misting. It must be noted that the amount of droplets ejected does not depend only on the level of the subambient pressure. However, as this pressure decreases the tendency to mist increases.

5.7 Analysis

It has been shown that both the suspensions and the coating colors exhibit a linear viscoelastic behavior at low strain amplitude and then the nonlinear behavior is observed at high strains. However, the storage modulus (G') decreases faster than the viscous modulus (G'') with the strain amplitude and the ratio G'/G'' is much smaller than one, which indicates that the elastic effects do not play any role under the conditions used in this work. From a rheological standpoint, the material property that mostly affects the flow through the application nip is the extensional viscosity. This property has a direct impact on the mechanism of filament break-up. It has been shown that viscous and viscoelastic fluids tend to form a thin sheet of fluid at the nip exit, known as septa (Owens et al., 2002; Vinjamur and Scriven, 2002). Because the flow through the nip is hydrodynamically unstable, the formation of septa is highly dependent on the capillary number. The nature of the filaments is different when suspensions are used. As Figure 5.8 shows, the filaments exhibit a little curved shape at low speed before they break up far away from the nip. As the roll speed increases they become straight and some misting droplets and small pieces of filaments are both ejected closer to the nip (Figure 5.8b). Although a low solids content suspension (10PEG52) exhibiting some extensional thickening was used for the visual assessment, it is observed that the filaments break as a rigid body and they recoil and smaller droplets are then generated.

On the other hand, it is important to observe in Figure 5.8c that some of the droplets are generated by air entrainment, which has been confirmed by the corresponding pressure profile (see Figure 5.10a). As mentioned before this is a result of a large deformation of the elastomer cover combined with a low solids content suspension. Figure 5.12 shows the predicted roll profile obtained with experimental data by using the one-dimensional elastohydrodynamic model developed by Coyle (1988) as a function of the elasticity number here defined as

$$Es_E = \frac{\eta_E U L}{E R^2} \quad (5.14)$$

where E is the Young modulus and L is the elastomer cover thickness.

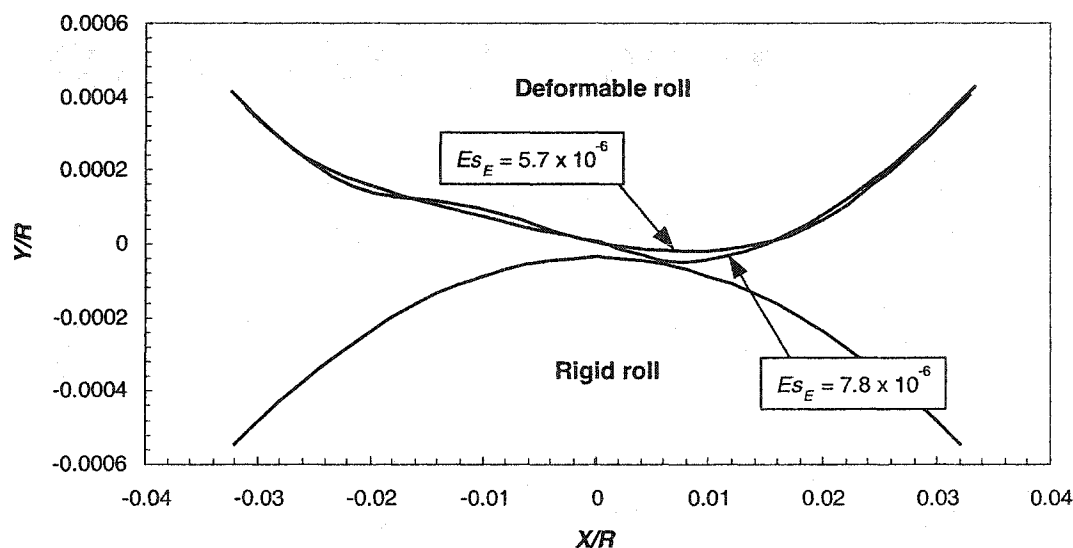


Figure 5.12 Roll profiles versus X/R as a function of the elasticity number (Es_E).

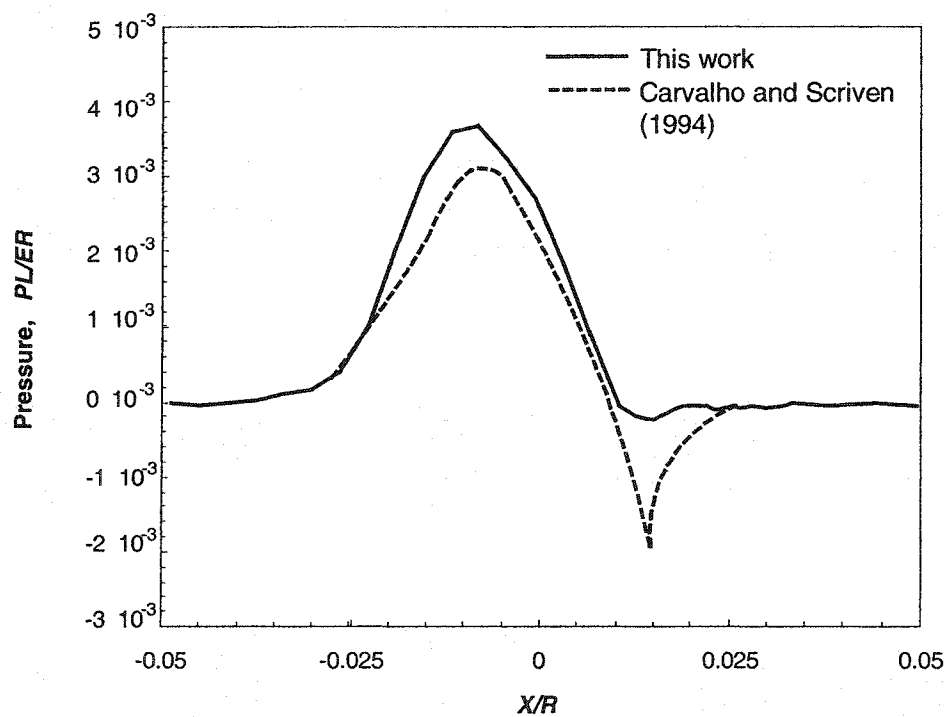
It must be noted that x and y coordinates have been scaled with the Young modulus and the roll radius, so that the roll profiles appear stretched in the x direction. At the highest speed (1100 m/min) it is observed that the flow generates a pressure high enough to deform largely the roll surface. In addition, if a low solids content suspension is used, some fluid will be lost before entering the nip due to the centrifugal action of transfer roll. Under these conditions, less fluid will flow through the nip allowing air bubbles to be entrained, which will be sprayed at the exit. The use of high solids content suspensions avoids air entrainment into the nip and the amount of misting is reduced. However, fluids exhibiting a pronounced strain hardening can lead to orange peel defects resulting in a low quality of the coated surface. Therefore, special attention must be paid to the extensional viscosity and particularly to strain hardening in order to reduce misting without increasing orange peel formation.

The role of the extensional rheology on the pressure profiles can be analyzed via Figures 5.6, 5.10 and 5.11. For a given roll speed, the maximal pressure (peak height) does not depend on the extent of strain hardening. In the case of the coating colors, although the low solids content colors exhibit a more pronounced strain hardening, the highest peak heights are obtained for the high solids content colors. In the case of the suspensions, the larger the solids concentration the larger the peak pressure. From the pressure profiles shown in Figures 5.10 and 5.11, it is difficult to clarify the influence of both peak height and valley depth on misting occurrence. However, a good idea of the tendency to mist can be obtained by analyzing the pressure ratio, here defined as the peak pressure divided by the valley pressure, both in absolute values. As Table 4 shows, the tendency of suspensions to mist decreases with increasing pressure ratio, which is in agreement with the fact that misting is reduced as the solids content increases. The trends for coating colors are different. The pressure ratio of high solids content colors with low latex content (C62L8) gives a slightly lower pressure ratio than the low solids content colors (C52L8). Coating colors having a larger latex content follow the same trends of the suspensions investigated, that is to say the tendency to misting decreases as the pressure ratio for a given roll speed increases.

Finally it is interesting to compare the pressure distribution experimentally obtained against the dimensional analysis of Carvalho and Scriven (1994). The two-dimensional Hookean model, in which the local displacement is a function of the whole pressure distribution, has been compared to the experimental profile of suspension 10PEG62 at 1100 m/min shown in Figure 5.10b. The dimensionless groups for a negative gap are PL/ER and X/R for the pressure and the x coordinate, respectively. An elasticity number (Es) of 7.8×10^{-6} was obtained by using $\eta_E = 2.5 \text{ Pa}\cdot\text{s}$, $E = 10 \text{ MPa}$ and $L = 9.52 \text{ mm}$. As Figure 5.13 shows the computed peak height is slightly underestimated with respect to that obtained from the experimental pressure distribution. However, in the negative region the model of Carvalho and Scriven considerably overestimates the subatmospheric pressure.

Table 5.4 Pressure ratio of suspensions and coating colors.

Fluid	Roll speed (m/min)		
	500	800	1100
10PEG52	2.98	3.34	3.59
10PEG62	4.01	4.17	4.41
5PEG52-CMC	3.15	3.39	3.49
5PEG62-CMC	3.67	4.44	4.61
C52L8	4.07	5.39	4.65
C62L8	4.05	4.57	4.31
C52L12	2.78	3.55	4.06
C62L12	3.31	3.66	4.55

Figure 5.13 Experimental and predicted pressure profiles for $E_{SE} = 7.8 \times 10^{-6}$.

This could be attributed to the use of a Newtonian fluid in the model. As pointed out before, under the conditions used in this work shear effects play a minimal role on the hydrodynamics and better results could be obtained if the extensional viscosity was introduced into the modified version of the elasticity number used by Carvalho and Scriven. The subatmospheric pressure would be reduced but the maximum pressure into the nip could increase considerably. This suggests that the lubrication theory cannot be applied here.

5.8 Concluding remarks

The flow of kaolin clay based suspensions and paper coating colors through a deformable gap of counter-rotating rolls at high speed was investigated. Visualization downstream the nip allowed to observe that the filaments could break up in two or more parts to form misting droplets. When the nip was highly deformed, these droplets could be generated by two different mechanisms: film splitting at the nip exit and spray effect due to the air entrainment into the nip. The roll profile was predicted by using an elastohydrodynamic model reported in the literature. The role that the rheology plays on misting occurrence was analyzed by shear and extensional tests. The suspensions and the coating colors investigated exhibited a typical viscosity behavior, or in other words the viscosity first decreased as the shear rate increased and they tended to level off at high shear rates. The tendency to mist was reduced with suspensions and coating colors exhibiting a large extensional viscosity. However, the extensional thickening must be kept as low as possible to avoid a high orange peel formation maintaining the amount misting to the lowest possible level.

CHAPTER 6

ORIFICE FLOWMETER FOR MEASURING EXTENSIONAL RHEOLOGICAL PROPERTIES

G. Ascanio¹, P.J. Carreau¹, E. Brito-De La Fuente² and P.A.Tanguy¹

¹CRASP, Department of Chemical Engineering, Ecole Polytechnique, P.O. Box 6079,
Stn. Centre-Ville, Montreal, QC, Canada, H3C 3A7

²Faculty of Chemistry, National University of Mexico, DF 04510, Mexico

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6.1 Abstract

Extensional rheological properties play an important role in processes in which the fluid is subjected to highly decelerated or accelerated flows. This paper describes an orifice flowmeter used to measure extensional properties of rheologically complex fluids at high strain rates. The operating principle of the flowmeter is based on the pressure drop due to the flow through a small size orifice. The flowmeter was first calibrated, by plotting the pressure drop-flow rate curve of the orifice, in terms of a dimensionless Euler number versus Reynolds number. Newtonian fluids consisting of aqueous solutions of corn syrup were used as calibration fluids. The calibration curve was then used to determine the apparent extensional viscosity of three different paper coating colors. The apparent extensional viscosity is compared to the shear viscosity in terms of the Trouton ratio. The Trouton ratio for one coating color is shown to exceed considerably the theoretical value of 3 expected for Newtonian fluids.

6.2 Introduction

Extensional or elongational deformations play a significant role in the product formulations encountered in several industrial processes such as blow molding, roll painting and paper coating, among others. Particularly, important extensional effects are found in flows through converging or diverging geometries. The importance of the extensional rheological properties is well recognized. However, nowadays little information is available on the extensional properties of complex fluids, because of the difficulty of imposing a purely extensional deformation on a fluid with no pre-shear or history of deformation (Everage and Ballman, 1978).

The first work reporting measurements of extensional viscosity was conducted by Trouton (1906) with very viscous materials (pitch, tar and shoemaker's wax) at low strain

rates. By comparing the extensional viscosity to the shear viscosity, Trouton determined the following relationship:

$$\eta_E = 3\mu \quad (6.1)$$

where η_E is the extensional viscosity and μ the shear viscosity. Relation (6.1) only holds for Newtonian fluids. For rheologically complex fluids such as suspensions and viscoelastic liquids, the Trouton ratio can be much larger than 3 (Della Valle et al., 2000; Macosko, 1994).

Several techniques have been proposed to generate steady extensional flows. Some of them have been adapted from methods used in the testing of solid samples and others have been developed specifically for low-viscosity liquids (Fuller et al., 1987). The most common methods for this type of liquids are stagnation flows (Binding and Jones, 1989; Winter et al, 1979; Cohu and Magnin, 1995; Ng et al, 1996; Cogswell, 1972) and entrance flows (Macosko, 1994; Binding, 1988 and 1991; Binding and Walters, 1988; Metzner and Metzner, 1970, Tremblay, 1989; Boger and Binnington, 1990). Fuller et al. (1987) proposed an opposed nozzle device, in which the liquid is either sucked or expulsed. The force required to keep the nozzles at a certain distance is measured and then the extensional viscosity can be determined as a function of the flow rate. Cohu and Magnin (1995) and by Ng et al (1996) used the same principle to investigate the extensional properties of paints and to compare the extensional flow resistance of several polymers, respectively. This technique allows operating with low viscosity fluids over a wide range of strain rates. However, wall effects and the instability of flow must be considered to interpret correctly the results. The filament stretching device developed by Tirtaatmadja and Sridhar (1993) is probably the most useful rheometer for measuring the extensional viscosity of viscoelastic fluids of low to moderate value viscosity. However, it is restricted to spinnable fluids under low extensional rates.

The simplest technique to determine extensional properties of low viscosity liquids and suspensions remains the entry flow method, in which a liquid flows from a large cross section into a small cross section. As the fluid experiences this cross-sectional area reduction, it dissipates a certain amount of energy related to the entrance pressure drop, ΔP_{en} (Fuller et al., 1987). The presence of an extensional flow is due to the convergence of the streamlines, but shear components must be also considered due to the presence of walls along the contraction (Binding, 1988; Binding and Walters, 1988).

Several approximate analyses can be found in the literature to determine the extensional viscosity from ΔP_{en} measurements. Cogswell (1972) analyzed converging flows in terms of their extensional and shear components to calculate the relationship between the volumetric flow rate and ΔP_{en} . He assumed that the power-law model could describe the shear viscosity over a limited range of shear stress. Knowing the shear viscosity, an apparent extensional viscosity can be calculated from ΔP_{en} and flow rate measurements. Padmanabhan and Macosko (1997) used a transient elongation technique with a commercial capillary rheometer for testing low-density polyethylene melts and they found good agreement with Cogswell's analysis. Binding (1988) presented an approximate analysis for the flow of fluids through planar and axisymmetric contractions, neglecting the radial or transverse component of the velocity. Binding and Walters (1988) estimated the extensional viscosity of polymer solutions using this analysis. In the axisymmetric case with a Boger fluid, vortices generated in the corners of the contraction increased in size and excess pressure losses were observed as the flow rate increased. Della Valle et al. (2000) also used the Binding analysis to measure the extensional properties of a Boger fluid. They found that both the apparent extensional and shear viscosities were approximately independent of the deformation rate, but the extensional viscosity was about 45 times the shear value. Although the complexities of contraction and converging flows are evident, entry flows are attractive given the experimental difficulties involved in measuring a true extensional viscosity. The objective of this work is to present the design and capabilities of an orifice flowmeter, built to investigate the

extensional properties of rheologically complex fluids such as paper coating colors at high strain rates ($>10^4 \text{ s}^{-1}$) in rheological conditions similar to those encountered in the paper coating industry.

6.3 Description of the Flowmeter

The orifice flowmeter consists of an orifice plate located between two identical cylindrical reservoirs of about 4 L each. The orifice plate is a disc of 127 mm diameter (D) and 12.7 mm thickness (e) with a small size hole (d) ranging from 0.6 mm to 4.79 mm with 45° converging and diverging sections (see Fig. 6.1). The test fluid can be

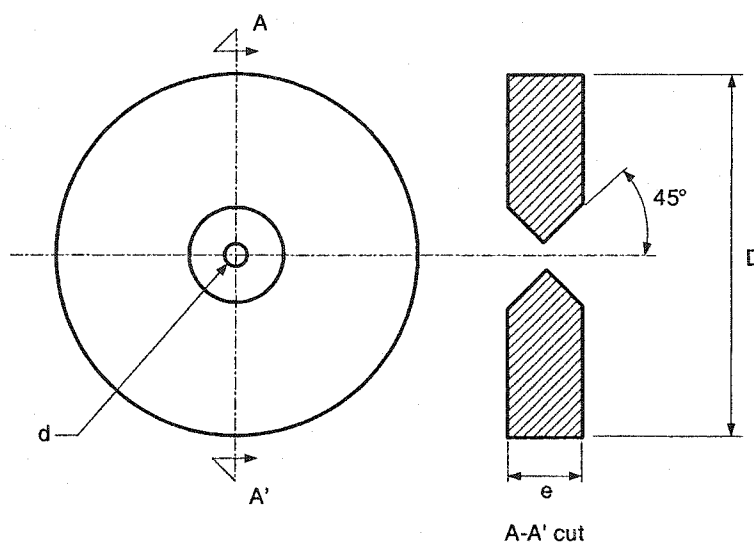
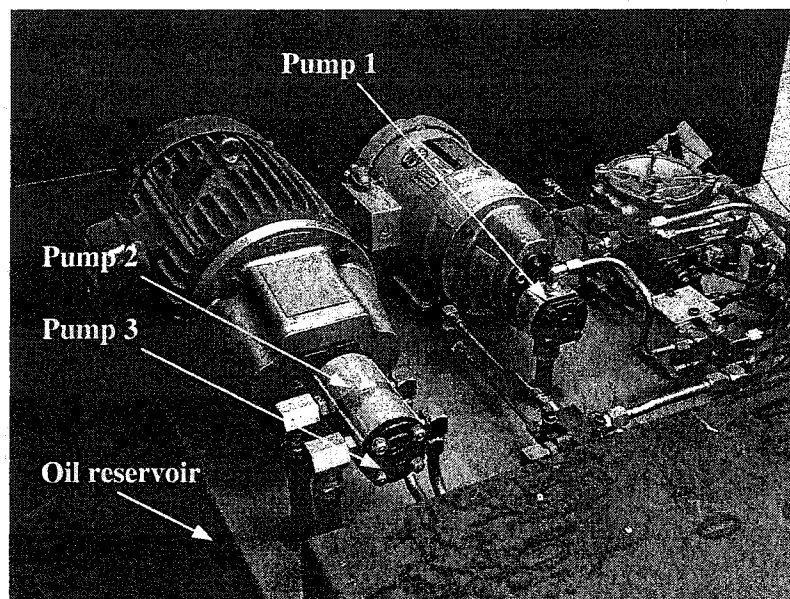


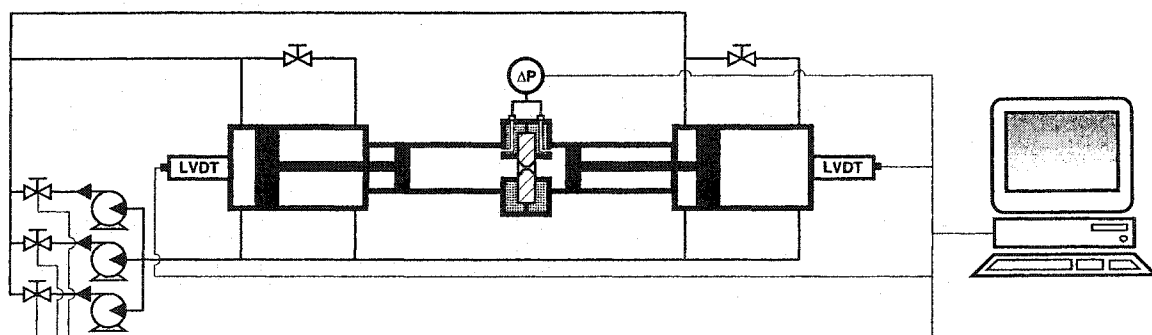
Figure 6.1 Orifice plate.

pumped from one filled reservoir to the empty one and then it can be pumped in the opposite direction. For that purpose, each reservoir is connected to a hydraulic piston large enough for testing under fully developed flow conditions and to investigate possible effects of entrance reservoir length. The hydraulic system was carefully designed to yield very stable flows. It basically consists of one piston for each reservoir with a stroke of

0.86 m, which speed is controlled by three different pumps (Fig. 6.2a). Pump # 1 is driven by an AC motor connected to a variable voltage/frequency drive so that a speed



(a)



(b)

Figure 6.2 Experimental set-up: a) Hydraulic system; b) General configuration of the orifice flowmeter.

between 0.5 mm/s and 3 mm/s can be obtained. On the other hand, pumps # 2 and 3 are both driven by a fixed speed motor, so that a piston speed of 5 mm/s and 10 mm/s can be obtained, respectively. A series of valves allows operating with one, two or the three pumps so that a broad range of flow is available. The speed of each piston is continuously recorded by means of a LVDT position transducer. The pressure drop across the orifice is measured by means of a differential pressure transducer via small holes bored on each side of the orifice plate. Three differential pressure transducers were available and chosen depending on the resolution and pressure range (1% @ 0-70 kPa, 1% @ 0-345 kPa and 2% @ 0-1380 kPa).

The configuration of this flowmeter has two interesting features: 1) As the motion is reversible, it is possible to use the same fluid sample for many runs; and 2) because the same fluid can be used indefinitely mechanical degradation under large deformation rate conditions can be investigated. The orifice flowmeter can be either manually or automatically operated from a computer and all the information about the pressure through the orifice can be obtained directly from the screen or as ASCII file. A general configuration of the orifice flowmeter is shown in Figure 6.2b.

6.4 Operating principle

Figure 6.3 shows a sketch of the conical die, which was considered for the analysis. The energy conservation of a fluid through this geometry (macroscopic energy balance) is the underlying principle for obtaining the two pressure drop components as proposed by Cogswell (1972):

$$E = (\Delta P_s + \Delta P_e) Q \quad (6.2)$$

where E is the dissipated energy, ΔP_S is the pressure drop due to shear flow, ΔP_E is the pressure drop due to extensional flow and Q is the volumetric flow rate. The following assumptions were made:

- 1) The shear viscosity obeys the power law model,

$$\eta = m\dot{\gamma}^{n-1} \text{ with } \dot{\gamma} = \left(\frac{3n+1}{4n} \right) \left(\frac{32Q}{\pi d^3} \right) \quad (6.3)$$

where d is the diameter at a given position in the convergent section.

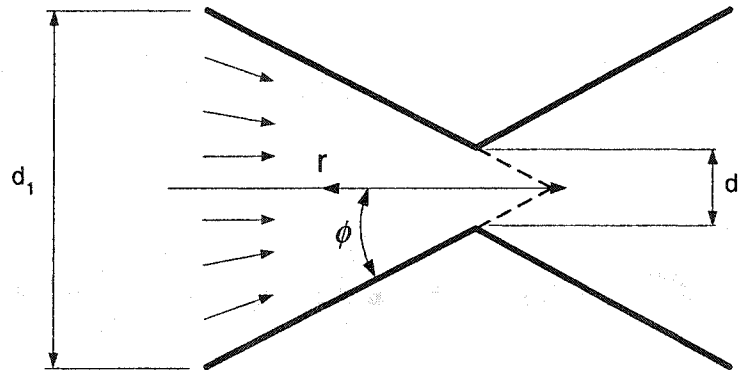


Figure 6.3 Flow through a conical die

- 2) The extensional viscosity is independent of the applied stress or extensional rate, i.e., the flow behavior index t is taken to be equal to 1 in the following power-law expression:

$$\eta_E = l\dot{\epsilon}^{t-1} \quad (6.4)$$

- 1) In the converging region, the flow is laminar, locally fully-developed under steady state and inertial effects are negligible.
- 2) The pressure drop in the divergent part of the orifice is negligible. Then, the pressure drop induced by the shear flow is

$$\Delta P_s = \frac{2m}{3n \tan \phi} \left(\frac{1+3n}{4n} \right)^n \left(\frac{32Q}{\pi d_0^3} \right)^n \left(1 - \left(\frac{d_0}{d_1} \right)^{3n} \right) \quad (6.5)$$

where d_0 and d_1 are the diameter at the orifice and at the pipe, respectively and ϕ is the half-angle of the convergent section (see Fig. 3). The pressure drop induced by the extensional flow is

$$\Delta P_E = \frac{l \tan \phi}{3} \left(\frac{32Q}{\pi d_0^3} \right) \left(1 - \left(\frac{d_0}{d_1} \right)^3 \right) \quad (6.6)$$

where l is equal to η_E for constant extensional viscosity. The average extensional rate and the maximum extensional rate are, respectively,

$$\dot{\epsilon}_{0ave} = \frac{l}{2} \tan \phi \left(\frac{32Q}{\pi d_0^3} \right) \quad (6.7)$$

and

$$\dot{\epsilon}_{0max} = \frac{3n+1}{n+1} \dot{\epsilon}_{0ave} \quad (6.8)$$

Binding (1988) extended the analysis of Cogswell (1972) to include strain-thinning effects for the extensional viscosity and inertial effects. The total pressure drop is then:

$$\Delta P = \Delta P_s + \Delta P_E + \Delta P_K \quad (6.9)$$

where ΔP_K is the pressure drop due to kinetic energy variation. ΔP_s is given by Equation (6.5) and ΔP_E is determined by

$$\Delta P_E = 2l \int_0^1 \frac{3n+1}{n+1} \left[\frac{3n+1}{(n+1)\pi} 8Q \tan \phi \right]^r \frac{\left(1 - \left(\frac{d_0}{d_1} \right)^{3r} \right)}{3td_0^{3r}} \quad (6.10)$$

with

$$Int = \int_0^1 \left[2 - \left(\frac{3n+1}{n} \right) \phi^{1+1/n} \right]^{r+1} \phi d\phi \quad (6.11)$$

The pressure drop due to the kinetic energy variation (ΔP_K) is given by

$$\Delta P_K = \frac{3}{2} \rho \frac{(3n+1)^2}{(2n+1)(5n+3)} \frac{16Q^2}{\pi^2 d_0^4} \left(1 - \left(\frac{d_0}{d_1} \right)^4 \right) \quad (6.12)$$

The maximum extensional rate at the orifice is given by the following relation

$$\dot{\epsilon}_{0max} = \frac{3n+1}{4n} \tan \phi \left(\frac{32Q}{\pi d_0^3} \right) \quad (6.13)$$

If this result is compared to Equation. (6.8), one obtains

$$\dot{\epsilon}_{0max \text{ Binding}} = \frac{n+1}{2n} \dot{\epsilon}_{0max \text{ Cogswell}} \quad (6.14)$$

For shear-thinning fluids ($n < 1$) $\dot{\epsilon}_{0max \text{ Binding}} > \dot{\epsilon}_{0max \text{ Cogswell}}$.

For the purpose of the present study, a good approximation can be obtained using the Binding analysis for a Newtonian fluid ($\eta_E = 3\mu$). This approach neglects the radial velocity component and assumes that the converging streamlines follow the actual conicity angle (ϕ), which is 45° for all the orifice plates, so that the pressure drop due to shear flow is

$$\Delta P_s = \frac{2}{3} \mu \left(\frac{32Q}{d_0^3} \right) \left[1 - \left(\frac{d_0}{d_1} \right)^3 \right] \quad (6.15)$$

For an axisymmetric contraction (Fig. 3) the velocity components are

$$v_r = \frac{Q}{A}; \quad v_\theta = v_\phi = 0 \quad (6.16)$$

The cross-sectional area, A , in the converging region is given by

$$A = 2\pi r^2 (1 - \cos \phi) \quad (6.17)$$

where ϕ is the half-angle of convergence. From Equations (16) and (17), the extensional rate is

$$\dot{\epsilon} = \frac{dv_r}{dr} = \frac{8Q \sin^3 \phi}{\pi d_o^3 (1 - \cos \phi)} \quad (6.18)$$

From Equations (6.1), (6.10) and (6.11) the pressure drop due to the extensional flow component is

$$\Delta P_E = \frac{16}{9} \mu \left(\frac{32Q}{\pi d_o^3} \right) \left[1 - \left(\frac{d_o}{d_i} \right)^3 \right] \quad (6.19)$$

Finally, the total pressure drop, neglecting the kinetic energy correction term, is

$$\Delta P = \frac{22}{9} \mu \left(\frac{32Q}{\pi d_o^3} \right) \left[1 - \left(\frac{d_o}{d_i} \right)^3 \right] \quad (6.20)$$

Equations (6.15), (6.19) and (6.20) indicate that the shear and the extensional contributions represent 27% and 73%, respectively, of the total pressure drop. To obtain this result we have assumed that the diverging flow downstream of the orifice does not contribute to the pressure drop. Also the analysis could be affected by the development of possible vortices in the convergent section of the orifice. Both the size and shape of vortices upstream of a sudden contraction have been previously studied for the flow of viscoelastic fluids (Binding, 1988; Binding and Walters, 1988; Tremblay, 1989; Boger and Binnington, 1990). Vortex enhancement and excess pressure loss have been both observed for large angles in the contraction region (i.e. 90°). The use of a small angle orifice (45° in our case) should reduce drastically the vortex size. However, we cannot rule out the possible influence of vortices on the pressure drop measurements even for inelastic fluids.

6.5 Materials and Methods

6.5.1 Calibration

The pressure transducer was first calibrated in terms of the output voltage by means of a transducer calibration machine. With this system, one end of the pressure transducer is kept at atmospheric pressure and the other one is connected to a hydraulic piston, which is subjected to known pressures by adding different balance weights. Both the slope and the offset from the calibration curve must be entered in the control/data acquisition program to obtain the pressure drop actual value. The next step consists of calibrating the orifice flowmeter by plotting the variation of the pressure drop with the flow rate through the orifice, in terms of an Euler number versus Reynolds number master curve. The Reynolds number is defined as

$$Re = \frac{d_o v_o \rho}{\mu} \quad (6.21)$$

and the Euler number as

$$Eu = \frac{\Delta P}{\frac{1}{2} \rho v_o^2} \quad (6.22)$$

where v_o is the average velocity of the fluid at the orifice.

By combining Equations (6.20), (6.21) and (6.22) and considering that $d_1 \gg d_o$ the orifice curve can be described by the following relation:

$$Eu = \frac{39.11}{Re} \quad (6.23)$$

This relation represents the theoretical expression of the Euler number as a function of the Reynolds number for the laminar regime. Relation (6.23) will be assessed using an orifice of 2.314 mm diameter, a flow rate ranging from 2.5 to 28 mL/s and a fluid viscosity from 0.01 to 1 Pa.s.

6.5.2 Fluids

Newtonian aqueous solutions of corn syrup (65 wt% and 90.5 wt%) were used as calibration fluids. The behavior of three different coating colors based on 100 pph (parts per hundred) of pigment was investigated. Their formulations are shown in Table 1. Two polymers were chosen as thickeners for the make down of the paper coating colors: carboxymethylcellulose (CMC) and a rheology modifier gum (Alco L-283). CMC allows the control of the viscosity without gelling. Alco L-283 gum is an associative alkali soluble viscosifier suitable for using in film and metering size units. It is important to point out that the pH has a stronger influence on the coating color viscosity prepared with the Alco gum. The shear viscosity was measured for all the fluids with an Advanced Rheometer model AR-2000 (TA Instruments) in a Couette configuration of 30 mm and 28 mm for the inside and outside diameter, respectively. Both shear and extensional viscosity measurements were carried out at room temperature ($\approx 24^\circ\text{C}$).

Table 1. Coating color composition.

Compound	Coating colors		
	CC-1	CC-2	CC-3
Nuclay (pph)	100	100	100
CMC Finnfix 5 (pph)		0.6	
CMC Finnfix 30 (pph)	0.3		
Alco L283 (pph)			0.1
Latex FC 620 NA (pph)	12		
Dispex N40V (pph)	0.15	0.15	0.15
Solids content (wt%)	60*	64.7	64.7
pH	8.24	8.21	8.11
Density (kg/m^3)	1370	1617	1617

* (Nuclay + latex)

6.6 Results and discussion

6.6.1 Calibration

Figure 6.4 reports the pressure drop-flow rate results (Euler number versus Reynolds number curve) for the two Newtonian fluids. It can be seen that at low Reynolds numbers ($Re < 20$) a laminar region exists (slope of -1) where viscous forces dominate the flow. The calibration results in the laminar region can be correlated by

$$Eu = \frac{37.99}{Re} \quad (6.24)$$

This is not significantly different from the theoretical expression obtained using the Binding analysis (Eq. 6.23) and quite close to the expression obtained by a finite-element

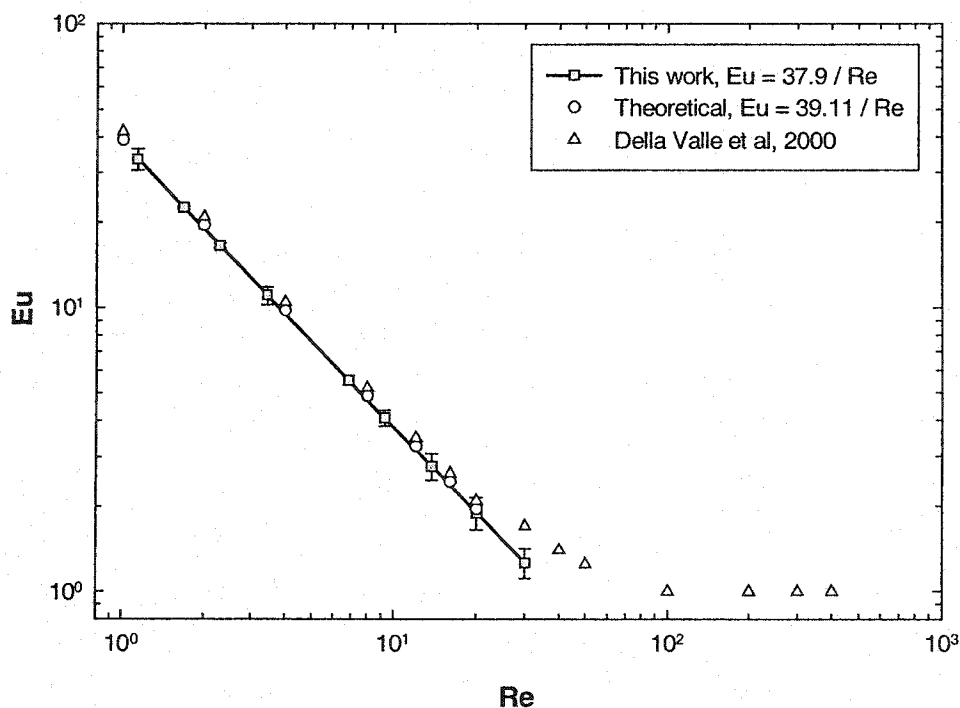


Figure 6.4 Calibration curve for Newtonian fluids.

method (Della Valle et al., 2000). Hence, we believe that the assumptions on which the analysis is based are valid. At high Reynolds numbers, inertial effects become important and the Euler number tends to reach a plateau (the pressure drop becomes independent of the viscosity). For a Reynolds number of about 20, an inflexion point is observed corresponding to the transition regime.

6.6.2 Coating Color Characterization

The shear viscosity data in a stress range from 0.015 Pa to 2000 Pa for the coating colors used in this work are plotted in Figure 6.5. The shear viscosity essentially obeys a power-law expression for which the parameters are presented in Table 2. The studied coating colors did not exhibit low-shear viscosity plateau, suggesting yield stress and solid-like behavior under small deformation (see Yziquel et al., 1999; Page et al., 2002).

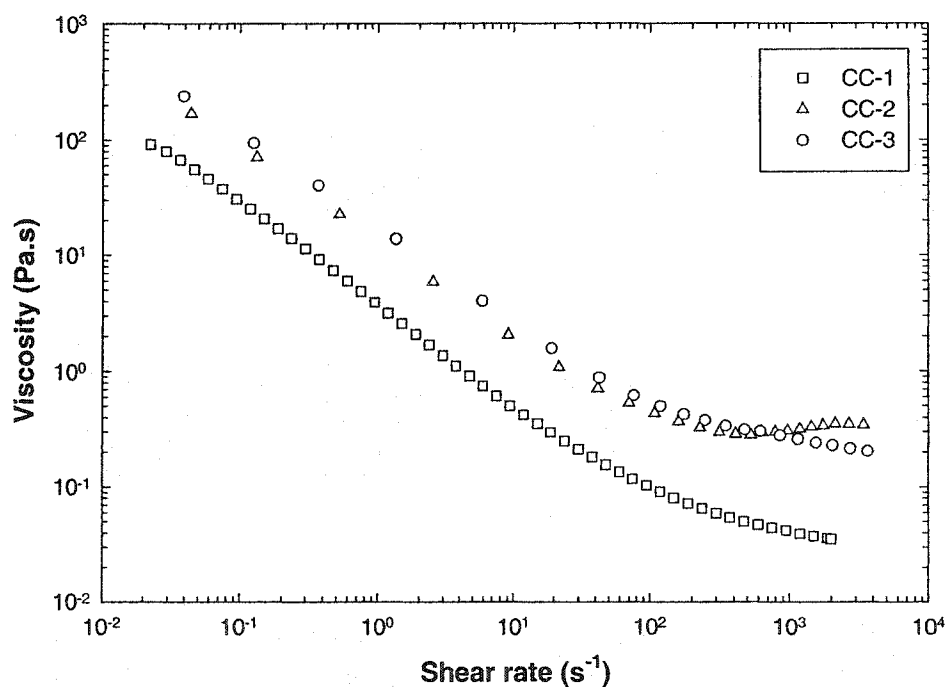


Figure 6.5 Shear viscosity data for the coating colors.

The behavior is slightly different for coating color CC-1 having a solid content of 60% compared to 64.7% for the two other colors. As Table 2 shows, the smaller values for the power-law parameters (m and n) correspond to color CC-1, while the largest ones to color CC-3. The tested suspensions are highly shear-thinning at shear rate ranging from 0.001 to 100 s⁻¹ and become slightly shear-thinning at high shear rates (> 100 s⁻¹), except coating color CC-2, which exhibits some shear thickening at a shear rate exceeding 500 s⁻¹ and reaches a high-shear plateau ($\eta_{\infty} \approx 0.35$ Pa.s) at 1500 s⁻¹. From Figure 5 one can expect that the high-shear plateau viscosity for the other two colors, CC-1 and CC-3 would be reached at a shear rate somewhat larger than 10⁴ s⁻¹, unfortunately outside the range of the rheometer. However, high-shear plateau viscosities of 0.035 Pa.s and 0.20 Pa.s were considered for CC-1 and CC-3, respectively, for other calculations.

Table 2. Power-law parameters and infinite shear viscosity for the coating colors.

Coating color	Consistency index (m)	Flow behavior	η_{∞}
	(Pa s ^{n})	index (n)	(Pa.s)
CC-1	7.566	0.1196	0.035
CC-2	13.007	0.1657	0.35
CC-3	17.531	0.1848	0.20

The apparent extensional viscosity data are plotted in Figure 6.6. The apparent extensional viscosity was calculated via the Reynolds number in Equation (25) using the pressure drop/flow rate measurements assuming that the extensional viscosity is equal to three times the shear viscosity (Trouton relation for Newtonian fluids). The effective strain rate $\bar{\dot{\gamma}}$ is taken as half the square root of the second invariant of the rate-of-deformation tensor, i.e.:

$$\bar{\dot{\gamma}} = \sqrt{3} \dot{\epsilon} \quad (6.25)$$

In simple shear flow the effective strain rate is equal to the shear rate. Using the Metzner-Otto (1957) concept the extensional rate can be expressed as

$$\dot{\epsilon} = k_s \left(\frac{32Q}{\pi d_o^3} \right) \quad (6.26)$$

where k_s is proportionality constant. Della Valle et al. (2000) determined by numerical simulation that purely shear-thinning fluids could be described with $k_s = 2.1$. Then, combining Equations (25) and (26) yields:

$$\bar{\gamma} = 29.10 \left(\frac{v_o}{d_o} \right) \quad (6.27)$$

It must be noted here that as the strain rate increases inertial effects of a fluid passing through the orifice become more important and the data need to be corrected. As shown Della Valle et al. (2000) with Newtonian fluids the pressure forces are equal to inertial forces for Reynolds numbers larger than 100, so that the Euler number becomes the unity. Hence, the Euler number used to calculate the apparent extensional viscosity was corrected as follows:

$$Eu_{corr} = Eu - 1 \quad (6.28)$$

All data obtained for $Re > 20$ have been corrected using Equation (6.28). The behavior of two coating colors, CC-2 and CC-3, is quite similar showing initially a rapid increase of the apparent extensional viscosity with increasing effective strain rate and then leveling off at large strain rates. The behavior of CC-1 is completely different: its extensional viscosity decreases first and then reaches a plateau as the strain rate increases. This suspension is hence strain-thinning up to 6500 s^{-1} .

It is important to point out that color CC-1 was prepared with a CMC having a larger molecular weight than that used for CC-2 and a modified styrene-butadiene latex FC 620 NA from Dow Chemical was added to that color as a binder. The presence of the

latex particles could also be responsible for the different extensional rheology. A more in-depth investigation of the rheology of coating colors containing different binders must be carried out to elucidate the phenomenon.

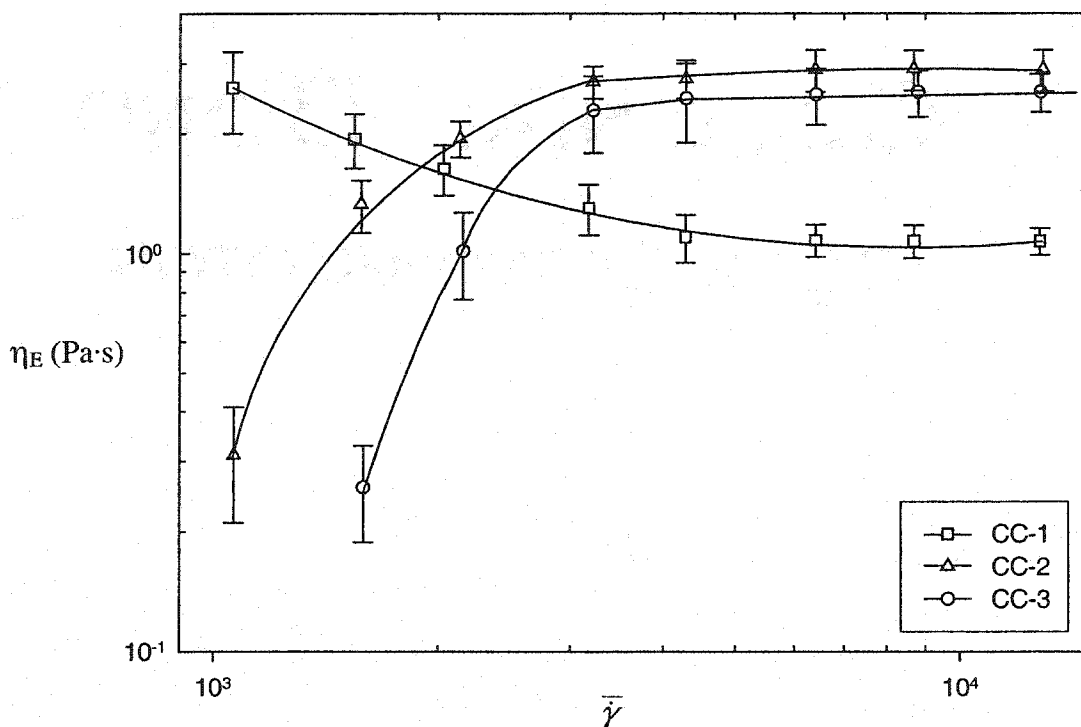


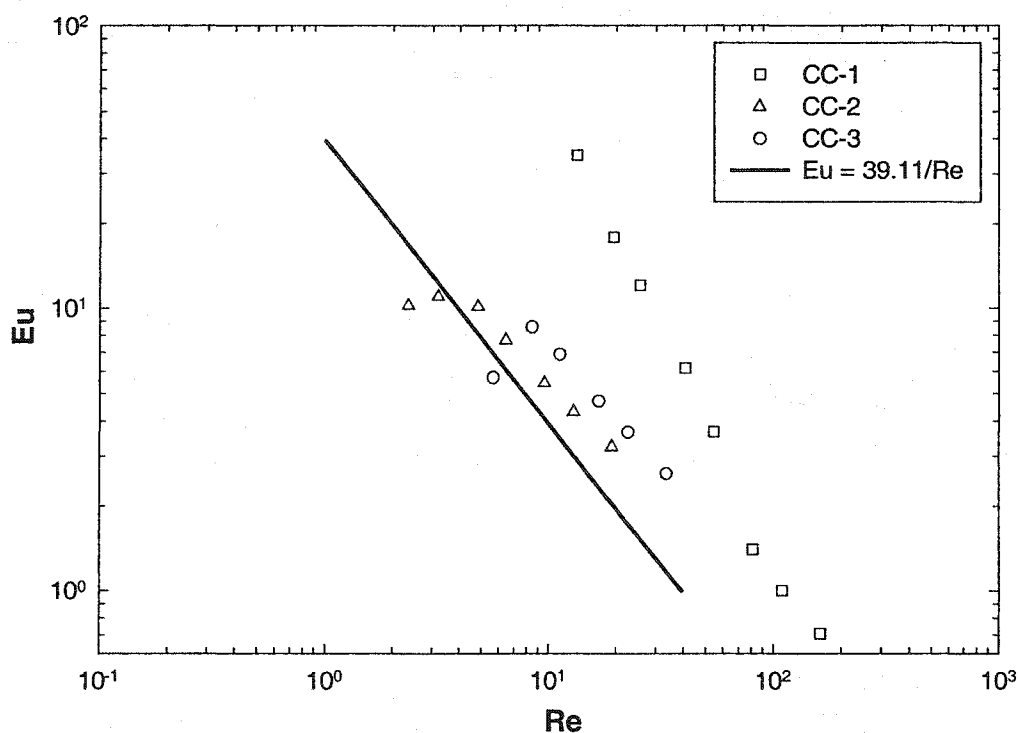
Figure 6 Extensional viscosity data for the coating colors.

The orifice flow data for the coating colors used in this work are shown and compared to the theoretical or calibration curve in Figure 7. The Reynolds number was calculated using the high-shear plateau viscosity from the shear viscosity data presented in Table 2. Coating color CC-1 is far above the theoretical curve. CC-2 and CC-3 curves are both close to the Newtonian one. The Trouton ratio as a function of effective strain is plotted in Figure 8. Two different approaches were used for estimating the Trouton ratio. The first one consisted simply of dividing the extensional viscosity data plotted in Figure 6 by the shear viscosity of Figure 5 at the same value of the effective strain rate (open

symbols in the figure). In the second approach, the Trouton ratio was calculated by the following expression:

$$Tr = 3 \frac{Eu}{38} Re \quad (6.29)$$

where Eu is the experimental Euler number plotted in Figure 6.7. That is the Trouton is taken as three times the vertical shift shown in Figure 6.7 with respect to the theoretical



reached a plateau at large shear rates. Coating colors CC-2 and CC-3 show a similar tendency. However, it is observed that CC-3 reaches slightly larger Trouton ratios than CC-2, which could lead to think that L-283 gum has a stronger influence than CMC. It is also observed that using the first approach the Trouton ratio tends to a constant value of 9 for CC-2 and 13 for CC-3 as the strain rate increases. This could be very useful information for the development of constitutive equations of coating colors. The Trouton ratio for coating color CC-1 is considerably larger than 3 especially at small strain rates, showing, as mentioned before, strain-thinning followed by a plateau at about 19 at large strain rates using the first approach. The value of the Trouton ratio for CC-1 is comparable to that obtained by Della Valle et al. (2000) for suspensions of clay in a Newtonian fluid without thickener nor binder. However, for those suspensions, the Trouton ratio was found to be constant.

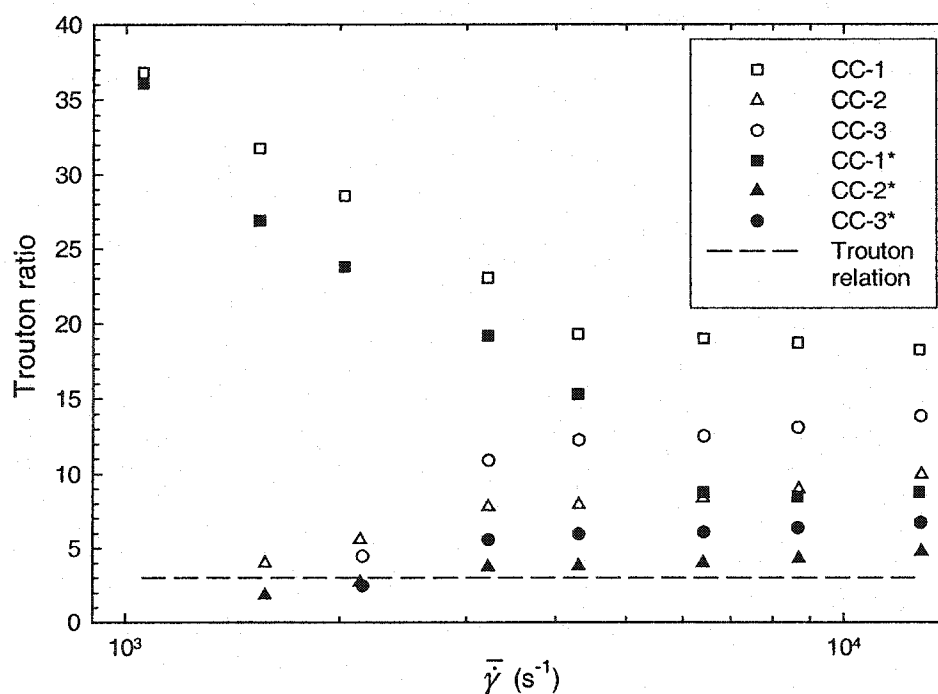


Figure 6.8 Extensional viscosity/shear viscosity ratio as a function of effective strain rate.

6.7 Conclusions

An orifice flowmeter was developed to determine the extensional properties of rheologically complex fluids at high strain rates. The apparent extensional viscosity of fluids and suspensions was determined from the pressure drop due to the flow through a small size orifice. For that purpose a master curve in terms of a dimensionless Euler number versus Reynolds number was used as a calibration curve. Two aqueous solutions of corn syrup were used as Newtonian calibration fluids and the experimental results were found to be in good agreement with the theoretical result based on the Binding (1988) analysis. The extensional properties of three different coating colors have been determined. The behavior of the coating color containing a higher molecular weight thickener and a binder was found to be quite different from the other two, with a much larger extensional viscosity and strain-thinning behavior at large strain rates.

Although it is practically impossible to obtain a shear-free extensional flow at high strain rates, the pressure entry concept used in the orifice flowmeter is considered as the simplest extensional method. From pressure drop/flow rate measurements, extensional data over a wide range of strain and viscosity can be obtained. However, it is important to consider the complexities of the entrance flow, especially for concentrated suspensions and viscoelastic fluids. For that reason, more work is needed to assess the hypotheses on which the analysis for the flow through a small orifice is based and to prove the interest of this simple method.

CHAPTER 7

CONCLUSIONS AND RECOMMENDATIONS

The objective proposed for this research was to develop a state of knowledge on the film splitting mechanism in the transfer nip of a metered film coater for a better understanding of misting occurrence. A laboratory film coater was developed specially for this research. It was used for studying the flow behavior of both Newtonian and non-Newtonian fluids through a deformable nip of counter-rotating rolls. A miniaturized pressure transducer was installed on one roll for determining the pressure distribution of the fluid through the nip. A high-speed video system was used for observing the flow downstream the nip focusing on the stretching and breaking up of filaments and how misting droplets are generated. The role that the rheology plays on misting occurrence was investigated by steady and oscillatory shear and extensional test. An orifice rheometer was used for determining the extensional properties of rheology complex fluids such as suspensions and paper coating colors.

In regard to the main factors governing misting occurrence in a metered film coater, we found that:

- Septa were observed for both Newtonian and viscoelastic fluids at low capillary numbers (Owens et al, 2002; Vinjaumur and Scriven, 2002). However, the filaments observed here with Newtonian fluids exhibit a long “C” shape when exiting the transfer nip. They behave like a thread of a solid-elastic material attached to two parallel plates moving at high speed.
- The filaments become straight with suspensions and coating colors.
- Because the flow through the nip is hydrodynamically unstable, the formation of possible septa immediately at the nip exit is highly dependent of the capillary number.

- No septa were observed, at least with the magnification used with the video system, for the roll speed range investigated. No evidence of septa at high speed has been reported in the literature (Owens et al , 2002).
- As the roll speed increases the elastomer cover is highly deformed when a low load between the rolls is used allowing the air to be entrained into the nip, so that two different mechanisms are responsible for misting occurrence: film splitting and the spray effect due to air entrainment into the nip. This was confirmed by the corresponding pressure distribution.
- Air entrainment can be avoided when a high load between the rolls is applied; however, the flow becomes time dependent as the roll speed increases.
- Although it is believed that cavitation can induce misting (MacPhee, 1997), the pressure at the exit of the nip (valley depth) was not low enough compared to the vapor pressure of the fluids investigated, so that cavitation does not account for in the film splitting mechanism.
- The higher the roll speed and the solid concentration the higher the peak pressure and the deeper the valley at the exit of the nip.
- Although it is difficult to predict misting from the pressure profiles, the pressure ratio gives a good idea of misting occurrence. As this ratio increases misting tends to decrease.
- The lubrication theory cannot be applied for the conditions used for this research, which was confirmed by comparing the experimental pressure distribution with the predicted profiles reported in the literature.

Concerning to the role that the rheology plays on misting, we could mention the following findings:

- The storage modulus (G') decreases faster than the elastic modulus (G'') and the ratio G'/G'' is much smaller than one, which suggests that elastic effects do not account for the rheological behavior of suspensions and coating colors, at least for the roll speed range investigated.

- Although coating colors having a low solid content exhibit a more pronounced extensional thickening, a high solid content color is preferred for improving the process runnability.
- The extensional thickening must be reduced to avoid a high orange peel formation maintaining the amount of misting at the lowest possible level.
- Because the extensional viscosity is the material property that most affect misting, a modified version of the capillary number was proposed.
- The Trouton ratio for both suspensions and coating colors exceed considerably the value expected for Newtonian fluids.

The results obtained from this thesis give new perspectives of research. A very interesting point to deal could be the study of the hydrodynamics in the transfer nip and the role that viscoelasticity plays on roll coating. In this case a larger magnification could be used with the visualization system looking for observing how the filaments are formed immediately at the exit of the nip. Another interesting research subject could be the numerical study of roll coating at high speed. This could be used to associate both the pressure distribution and the velocity profiles to misting occurrence. However, it must be noted the boundary conditions to be imposed on two free surfaces is a real challenge.

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