Investigation of Flow Properties of Asphalt Binders Containing Polymer Modifiers

Yu-Hsun Nien,1 Pei-Hung Yeh,2 Wei-Chyum Chen,3 Wen-Tsung Liu,3 Jean-Hong Chen1

1Department of Polymer Materials, Kun Shan University, Yung-Kang City, Tainan Hsien 710, Taiwan
2Department of Applied Chemistry, Chaoyang University of Technology, Wufong Township, Taichung County 41349, Taiwan
3Department of Civil Engineering, Kao Yuan University, Lujhu Township, Kaohsiung County 82151, Taiwan

Modified asphalts using polymeric additives, including isotactic polypropylene (iPP) and different degrees of chlorinated polypropylene (CPP), have been prepared and examined using differential scanning calorimetry (DSC), scanning electron microscopy (SEM), rotational viscometer, and dynamic shear rheometer (DSR) in this study. The results of DSC studies show that different levels of the crystallization occur in asphalt/iPP blends because most of the crystallinity of iPP remain intact in the blend, while those asphalt samples having CPP tend to affect the thermal behavior in the DSC thermograms because of increasing the amorphous region of CPP. From the images of SEM, the distinctions of the phase distributions between the asphalt/iPP and asphalt/CPP blends illustrate that the chlorine content in CPP is a significant factor in controlling its compatibility with asphalt. The rheological studies show that the asphalt blends containing CPP (26 wt% of Cl) exhibit higher viscosity in terms of higher temperature to get Newtonian behavior and display superior rutting resistance to avoid permanent deformation at high temperature. POLYM. COMPOS., 29:518–524, 2008. © 2008 Society of Plastics Engineers

INTRODUCTION

Since the advent of asphalt blocks in paving industry, efforts to improve the properties of asphalt surfacing materials have centered on polymeric additives. Polymers typically employed in polymer modified asphalt (PMA) include plastics, elastomers, and fibers [1–5]. For instance, the addition of styrene-butadiene-styrene copolymers (SBS) block elastomer to asphalt increases the rutting resistance of PMA at high temperature and improves the ductility, elasticity, and cyclic loading properties of the mixture at low temperatures [6]. It is known that additions of polymers increase the cost of the corresponding PMA. Therefore, recyclable polymers have been chosen as the modifiers to provide the functions of improving the properties of asphalt surfacing materials within the past few years [7–10]. If price and availability of various polymers that have been proposed for asphalt modification are considered, it is obvious that polyolefins or vulcanized rubbers would be more economical than other polymeric candidates. Further, polyolefins, either from virgin material or from recycled waste, comprise approximately 60% of plastic solid wastes. Therefore, incorporation of these materials into PMA can be an effective method of polymer recycling and improve the properties of asphalt paving.

Thermoplastics, such as partially crystalline polyolefins, combine the advantages of rubber and fibers. The crystalline segments of polyolefins serve as high strength fillers in the asphalt/polymer blend and improve the blend properties and overall service conditions [11]. However, the polyolefins are only slightly compatible with asphalt; thus, the blends tend to separate at high temperature. On the contrary, amorphous segments of thermoplastics give asphalts improved flexibility to improve resistance to permanent deformation and increase elastic modulus [1, 12]. For instance, ethylene–vinyl acetate copolymers (EVA) are more compatible with asphalt, leading to the enhancement of the blends tested in the areas of fatigue resistance and field performance [13]. Unfortunately, the properties of EVA blends may vary substantially depending upon the asphalt used to prepare the PMA, and unsatisfactory mixes may be produced. For any specific asphalt cement (AC), the physical properties of the asphalt/polymer blend are affected by the amount of polymeric material added, its composition, its molecular weight, etc. However, the most important variable is the compatibility of the AC

Correspondence to: Pei-Hung Yeh; e-mail: phyeh@cyut.edu.tw
Contract grant sponsor: National Science Council, Taiwan.
DOI 10.1002/pc.20404
Published online in Wiley InterScience (www.interscience.wiley.com).
© 2008 Society of Plastics Engineers
with the admixed polymer. The ability to enhance the compatibility between asphalt and polymer is complicated further by source dependent variations in asphalt compositions within a given grade. Thus, the introduction of an incompatible polymer into such a system generally results in asphaltenene flocculation and oil bleeding, which is the main reason of a binder having no cohesion [14]. Even if the phase separation of the asphalt components is not apparent, extended mixing times will be required to achieve acceptable mixes [15].

One of the various approaches to improve the compatibility of asphalt/polymer blends is the utilization of chemical modification of polymer additive to enhance the interaction of the amorphous regions of the polymer with the asphalt phase [16–20]. In this article, isotactic polypropylene (iPP), with various degrees of chlorination, were designed to increase the interaction between polymer and polar components of asphalt. The asphalt blends were compared with the unmodified asphalt to assess the thermal transition properties of asphalt blends by differential scanning calorimetry (DSC), the phase structures of the asphalt blends by scanning electron microscopy (SEM), and the rheologic properties of asphalt blends by rotational viscometer and dynamic shear rheometer (DSR).

**EXPERIMENTAL METHODS**

**Materials**

Tank asphalt ACP-1 (tank asphalt) for the base material in this study was obtained from the Chinese Petroleum Corporation, Taiwan. The physical properties of this asphalt were as follows: penetration, 98 mm (25°C, ASTM standard D5); Flash Point, 219°C (ASTM standard D92); softening point, 48°C (ASTM standard D36); and solubility in trichloroethylene, 99.0% (ASTM standard D2042). Polymers investigated as asphalt additives were iPP (isotactic, spherical particles), and two types of chlorinated polypropylene (26 wt% of Cl and 32 wt% of Cl, noted as CPPA and CPPB, respectively), which were purchased from Aldrich Co.

**Preparation of PMA**

Unless specified otherwise, the modified asphalt was prepared by adding 3% or 5% by weight of polymer to the tank asphalt. The asphalt was preheated to 180°C in a small container, and then polymer was added into the tank asphalt while the blend was mixed with a high shear mixer. Then the polymer asphalt blend was stirred at 160°C for 40 min.

**Microscopic Behavior of Asphalt/Polymer Blends**

Images of asphalt samples were observed using a scanning electron microscope (JSM-5610, model E-3) to examine asphalt/polymer phase distribution. The structural formation of asphalt samples was recorded using a camera connected to the SEM. The SEM power was 0.5 keV and the magnification ranged up to 250.

**Measurement of DSC**

Thermal analyses of tank asphalt and asphalt blends were performed by DSC (Perkin Elmer Pyris Diamond) in a nitrogen atmosphere at a flow rate of 20 ml/min. The sample for DSC was sealed in aluminum sample pans using an empty aluminum sample with cap as a reference. The data were collected from −55°C to 200°C at a heating rate of 10°C/min. The glass transition temperatures ($T_g$), melting points ($T_m$), and heat of fusion ($H_f$) of the blend components were determined as previously described [21].

**Measurement of Rotational Viscometer**

Viscosity measured for each blend was based on ASTM D4402-02 and AASHTO TP48-97 by a rotational viscometer (Brookfield) at 60°C (spindle no. 29) and at 135°C (spindle no. 27), respectively.

**Measurement of DSR**

A dynamic shear rheometer, CSR-500 model (Carri-Med, Dorking, England), was used for dynamic mechanical analysis of asphalt binders with 1,000 Pa stress at a rate of 1.6 Hz to characterize the viscous and elastic behavior of asphalt binders at intermediate and high service temperatures. The samples were “sandwiched” between two parallel plates with a diameter of 25 mm and a gap of 1 mm and the temperature was raised from 30°C through one cycle before performing a temperature sweep to 80°C. The DSR measured the viscoelastic properties by measuring the strain response of the specimen to a fixed torque, measuring the $G^*$, $G''$, and $G'$ properties of asphalt binders as subjected to oscillator shear stress. Under these conditions, the asphalt samples were also run under a frequency sweep from 0.2 to 2.0 Hz at 30°C to comply with the asphalt binder specifications of Strategic Highway Research Program (SHRP).

**RESULTS AND DISCUSSION**

**Thermal Properties of Asphalt Binders**

The applications of DSC analysis of asphalt blend to evaluate the endothermic transitions from the melting/dissolution of crystalline and amorphous regions of asphalt blends have been reported [22]. A comparison of DSC thermograms of iPP, tank ACP-1, and ACP-1 blended with 3% of the iPP, and chlorinated iPP is shown in Fig. 1. In addition, the results of the effect of iPP, CPPA, and CPPB contents on the melting points ($T_m$), heat of
fusio... 

FIG. 1. DSC thermograms of ACP-1 (curve A), pure iPP (curve B), 3% iPP blend (curve C), 3% CPPA blend (curve D), and 3% CPPB blend (curve E).

The effect of the iPP and CPP additives with various concentrations on viscosity for each of the asphalt blends is listed in Table 2. The data illustrate that the viscosity of asphalt blends is higher than the asphalt matrix and enhancing the interaction between the polymer and the asphalt matrix.

Viscosity of Asphalt Blends

The morphology of asphalt blends was viewed with SEM to examine phase distribution and structure. Figure 2 shows the SEM images of tank asphalt ACP-1 without any modifier, 3%, and 5% of the asphalt/polymer blends. The SEM micrograph of ACP-1 blend with 3% iPP (Fig. 2B) or 5% iPP (Fig. 2C) shows that it obviously has two phases, the asphalt phase and polymer phase, either in 3% or 5% of the asphalt/iPP blends. It implies that phase separation severely occurs in the asphalt/iPP blend. However, Fig. 2D shows the SEM image of 3% asphalt/CPPA displays no clear-cut interface between CPPA and asphalt, while the ACP-1 blend with 3% CPPB shows a distinct boundary between asphalt and CPPB in Fig. 2F. This indicates that 3% of the asphalt/CPPA blend exhibits more compatibility with asphalt matrix than 3% of the asphalt/CPPB blend. In this study, the boundaries between asphalt and CPP additives in 5% of the asphalt/CPPA blend (Fig. 2E) as well as the asphalt/CPPB blend (Fig. 2F) are less observable than that in 3% of the asphalt/CPPA blend (Fig. 2D) and the asphalt/CPPB blend (Fig. 2F). A comparison of the SEM micrographs of asphalt/iPP blends and asphalt/CPP blends suggests that CPP additives in asphalt blends result in better dispersion of polymer into the asphalt phase than iPP additive and affect the phase distribution of asphalt blends. It had been reported that more highly dispersed polymer rich phases are expected to improve the toughness of brittle asphalt at low temperatures and reinforce asphalt at high temperatures [24]. Therefore, the distinctions of the phase distributions between the asphalt/iPP and asphalt/CPP blends illustrate that the chlorine content in CPP is a significant character in controlling its compatibility with asphalt.

The morphology of asphalt blends was viewed with SEM to examine phase distribution and structure. Figure 2 shows the SEM images of tank asphalt ACP-1 without any modifier, 3%, and 5% of the asphalt/polymer blends. The SEM micrograph of ACP-1 blend with 3% iPP (Fig. 2B) or 5% iPP (Fig. 2C) shows that it obviously has two phases, the asphalt phase and polymer phase, either in 3% or 5% of the asphalt/iPP blends. It implies that phase separation severely occurs in the asphalt/iPP blend. However, Fig. 2D shows the SEM image of 3% asphalt/CPPA displays no clear-cut interface between CPPA and asphalt, while the ACP-1 blend with 3% CPPB shows a distinct boundary between asphalt and CPPB in Fig. 2F. This indicates that 3% of the asphalt/CPPA blend exhibits more compatibility with asphalt matrix than 3% of the asphalt/CPPB blend. In this study, the boundaries between asphalt and CPP additives in 5% of the asphalt/CPPA blend (Fig. 2E) as well as the asphalt/CPPB blend (Fig. 2F) are less observable than that in 3% of the asphalt/CPPA blend (Fig. 2D) and the asphalt/CPPB blend (Fig. 2F). A comparison of the SEM micrographs of asphalt/iPP blends and asphalt/CPP blends suggests that CPP additives in asphalt blends result in better dispersion of polymer into the asphalt phase than iPP additive and affect the phase distribution of asphalt blends. It had been reported that more highly dispersed polymer rich phases are expected to improve the toughness of brittle asphalt at low temperatures and reinforce asphalt at high temperatures [24]. Therefore, the distinctions of the phase distributions between the asphalt/iPP and asphalt/CPP blends illustrate that the chlorine content in CPP is a significant character in controlling its compatibility with asphalt.

Viscosity of Asphalt Blends

The effect of the iPP and CPP additives with various concentrations on viscosity for each of the asphalt blends is listed in Table 2. The data illustrate that the viscosity of asphalt blends is higher than the asphalt matrix and enhancing the interaction between the polymer and the asphalt matrix.

Phase Distribution of PMA Blend

The morphology of asphalt blends was viewed with SEM to examine phase distribution and structure. Figure 2 shows the SEM images of tank asphalt ACP-1 without any modifier, 3%, and 5% of the asphalt/polymer blends. The SEM micrograph of ACP-1 blend with 3% iPP (Fig. 2B) or 5% iPP (Fig. 2C) shows that it obviously has two phases, the asphalt phase and polymer phase, either in 3% or 5% of the asphalt/iPP blends. It implies that phase separation severely occurs in the asphalt/iPP blend. However, Fig. 2D shows the SEM image of 3% asphalt/CPPA displays no clear-cut interface between CPPA and asphalt, while the ACP-1 blend with 3% CPPB shows a distinct boundary between asphalt and CPPB in Fig. 2F. This indicates that 3% of the asphalt/CPPA blend exhibits more compatibility with asphalt matrix than 3% of the asphalt/CPPB blend. In this study, the boundaries between asphalt and CPP additives in 5% of the asphalt/CPPA blend (Fig. 2E) as well as the asphalt/CPPB blend (Fig. 2F) are less observable than that in 3% of the asphalt/CPPA blend (Fig. 2D) and the asphalt/CPPB blend (Fig. 2F). A comparison of the SEM micrographs of asphalt/iPP blends and asphalt/CPP blends suggests that CPP additives in asphalt blends result in better dispersion of polymer into the asphalt phase than iPP additive and affect the phase distribution of asphalt blends. It had been reported that more highly dispersed polymer rich phases are expected to improve the toughness of brittle asphalt at low temperatures and reinforce asphalt at high temperatures [24]. Therefore, the distinctions of the phase distributions between the asphalt/iPP and asphalt/CPP blends illustrate that the chlorine content in CPP is a significant character in controlling its compatibility with asphalt.

Viscosity of Asphalt Blends

The effect of the iPP and CPP additives with various concentrations on viscosity for each of the asphalt blends is listed in Table 2. The data illustrate that the viscosity of asphalt blends is higher than the asphalt matrix and enhancing the interaction between the polymer and the asphalt matrix.

Viscosity of Asphalt Blends

The effect of the iPP and CPP additives with various concentrations on viscosity for each of the asphalt blends is listed in Table 2. The data illustrate that the viscosity of asphalt blends is higher than the asphalt matrix and enhancing the interaction between the polymer and the asphalt matrix.

The effect of the iPP and CPP additives with various concentrations on viscosity for each of the asphalt blends is listed in Table 2. The data illustrate that the viscosity of asphalt blends is higher than the asphalt matrix and enhancing the interaction between the polymer and the asphalt matrix.

Viscosity of Asphalt Blends

The effect of the iPP and CPP additives with various concentrations on viscosity for each of the asphalt blends is listed in Table 2. The data illustrate that the viscosity of asphalt blends is higher than the asphalt matrix and enhancing the interaction between the polymer and the asphalt matrix.

Viscosity of Asphalt Blends

The effect of the iPP and CPP additives with various concentrations on viscosity for each of the asphalt blends is listed in Table 2. The data illustrate that the viscosity of asphalt blends is higher than the asphalt matrix and enhancing the interaction between the polymer and the asphalt matrix.

Viscosity of Asphalt Blends

The effect of the iPP and CPP additives with various concentrations on viscosity for each of the asphalt blends is listed in Table 2. The data illustrate that the viscosity of asphalt blends is higher than the asphalt matrix and enhancing the interaction between the polymer and the asphalt matrix.

Viscosity of Asphalt Blends

The effect of the iPP and CPP additives with various concentrations on viscosity for each of the asphalt blends is listed in Table 2. The data illustrate that the viscosity of asphalt blends is higher than the asphalt matrix and enhancing the interaction between the polymer and the asphalt matrix.
increases with the varied modifiers and their concentrations at medium and high temperature ranges. Note that in no case does the viscosity of a blend exceed the SHRP criterion of 3 Pa s upper limit, in order to ensure that the binder can be adequately pumped and mixed with the aggregates at 135°C [25]. The asphalt with 5% of CPPA increases the viscosity significantly than those 3% and 5% of iPP as well as those CPPB at 60 and 135°C. Further, as confirmed by SEM observation, asphalt blends containing CPPA exhibit higher viscosity. However, the observation from Table 2 shows the presence of the higher chlorine content of CPPB in asphalt blends displays lower viscosity than the asphalt blends containing iPP and CPPA. Contrary to the expectation, the higher the chlorine content of modified iPP, the lower is the viscosity of the asphalt blend. Therefore, controlling the degree of chlorination in modification of iPP can be considered as an important factor to control the asphalt flow behavior.

### Rheological Properties of Asphalt Blends

The linear viscoelastic moduli of asphalt binder under oscillating conditions to yield dynamic mechanical properties can be measured by DSR to simulate traffic loading, because the data can be acquired within the linear range of the asphalt in a loading mode. In analyzing the rheologic properties of asphalt binder, $G^*$, the ratio of the peak stress to the peak strain, reflects the total stiffness. The in-phase component of $|G^*|$ is the shear storage

**TABLE 2. Influence of iPP, CPPA, and CPPB contents on the viscosity ($\eta$; Pa s at 60 and 135°C) of modified asphalts.**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Content (%)</th>
<th>60°C</th>
<th>135°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>ACP-1</td>
<td>0</td>
<td>116</td>
<td>0.31</td>
</tr>
<tr>
<td>ACP-1/iPP</td>
<td>3</td>
<td>274</td>
<td>0.74</td>
</tr>
<tr>
<td>ACP-1/iPP</td>
<td>5</td>
<td>407</td>
<td>1.13</td>
</tr>
<tr>
<td>ACP-1/CPPA</td>
<td>3</td>
<td>391</td>
<td>0.80</td>
</tr>
<tr>
<td>ACP-1/CPPA</td>
<td>5</td>
<td>921</td>
<td>1.58</td>
</tr>
<tr>
<td>ACP-1/CPPB</td>
<td>3</td>
<td>162</td>
<td>0.34</td>
</tr>
<tr>
<td>ACP-1/CPPB</td>
<td>5</td>
<td>230</td>
<td>0.51</td>
</tr>
</tbody>
</table>
modulus, $G'$ (the elastic portion), the out-phase component of $|G^*|$ is the loss modulus, $G''$ (the viscous portion), and tan is a mechanical damping or internal friction, i.e. $\tan \delta = G''/G'$, $\sin \delta = G'/G^*$, and $G^* = G' + iG''$. If the isochronal $\sin \delta$ value is 1, the system is considered as a Newtonian fluid, i.e. the storage modulus vanishes since $\sin \delta \cong 1$, $G^*/\sin \delta \cong G''$. The isochronal $\sin \delta$ curves are a function of temperature for asphalt blends, as illustrated in Fig. 3. It displays that $\sin \delta$ value of the tank ACP-1 approaches unity and the asphalt blends exhibit less than 1 at 52°C, while the other asphalt blends display the viscoelastic properties at 52°C. However, the isochronal $\sin \delta$ curves of the blends approach unity above 70°C as the viscoelastic asphalt binders begin to flow as Newtonian fluids except the asphalt/CPPA blends. In addition, as the polymer concentration increases, higher temperatures are required for all of asphalt blends to attain Newtonian behavior. As can be seen in Fig. 3, CPPA modified asphalt blends exhibit lower $\sin \delta$ than those of asphalt blends above 30°C. It illustrates that there is an increased viscoelastic component in the CPP modified asphalt blends over a wide temperature range, which could be responsible for the good interaction of chlorinated iPP with the polar fractions of asphalt matrix. Increasing viscoelastic properties in asphalt/CPPA blends at higher temperatures should improve their pavement performance at maximum in-service temperature.

The effect of frequency sweeps on the $\sin \delta$ values of asphalt binders at 30°C is illustrated in Fig. 4. The addition of polymeric additives leads to decrease in $\sin \delta$ values of asphalt blends than the tank asphalt at all frequency ranges. It suggests that all of the PMAs blends exhibit more elastic behavior than tank asphalt in frequency studies. In addition, it can be seen that the $\sin \delta$ values of asphalt binders decrease significantly with increasing both the polymer concentrations and frequencies. Note that there is no difference of $\sin \delta$ value between 3% and 5% of the asphalt/iPP blends at 0.2 Hz. However, the difference of $\sin \delta$ values between both 3% and 5% of asphalt/iPP blends changes gradually as the frequency increases. This phenomenon reveals that the flow property of the asphalt/iPP blend is not related to the polymer concentration at low frequency, but is associated with the volume of iPP at high frequency. Besides, the asphalt blend containing 5% CPPA has the lowest $\sin \delta$ value at all frequencies. It indicates that the blend has the best viscoelastic property among all of the asphalt binders in this study.

To evaluate the permanent deformation of asphalt binder, $G^*/\sin \delta$ is selected to express the contribution of asphalt binder to rutting properties. From the SHRP suggestions, the $G^*/\sin \delta$ value should be larger than 1 kPa at 10 rad/s for the original binder at a maximum pavement design temperature. Higher values of $G^*/\sin \delta$ are expected to result in high resistance to permanent deformation. Figure 5 shows the isochronal plot of $G^*/\sin \delta$ that reveals distinct differences because of the polymeric modifiers, which may interact with asphalt. The results of polymer additives leading to improvement on the performance grade of the PMAs are listed in Table 3. It can be observed that the in-service temperature of the asphalt blend is enhanced by the effects of polymeric modifier and concentration, with increasing the benefit of road performance. Note that all the modified asphalt binders are qualified according to SHRP suggestions of the stiffness parameter at 64°C, except the asphalt matrix. However, only the asphalt blends containing chlorinated iPP meet the SHRP suggestions at 70°C or higher service temperatures. As an in-service temperature of asphalt binder, when the temperature is below 70°C, the $G^*/\sin \delta$ value of the asphalt/CPPA binder to the qualification of the SHRP suggestions is up to 90°C, which is enough to avoid permanent deformation. These observations imply that introduction of chlorine atoms may enhance the
compatibility between the polymer additives and asphalt depending upon the degree of chlorination. Therefore, the rheological response of blends with CPPA may have important benefits as road paving binders on improving binder resistance to rutting.

Figure 6 displays the influence of frequency sweeps on the $G*/\sin \delta$ values of asphalt binders at 30°C. It exhibits that the $G*/\sin \delta$ values of all modified asphalt binders increase with frequency and concentration. An increase in frequency yields an increase in the $G*/\sin \delta$ value for all asphalt binders in this study. As may be seen in Fig. 6, the lowest $G*/\sin \delta$ value of the blend containing 3% CPPB in the whole frequency range, even when the CPPB concentration up to 5% exhibits the same performance of the asphalt matrix at high frequency range. Unlike the results of the rutting parameter under the temperature sweep at constant frequency (Fig. 5), the presence of CPPB modifier in tank asphalt does not expect to enhance the rutting resistance in the frequency studies.

CONCLUSIONS

Modified asphalts using polymeric additives, including iPP and different degrees of chlorinated polypropylene (CPP), have been prepared and reported in this study. Chlorination of polypropylene at different levels is an effective means to change the properties of the iPP as a modifier for improving the thermal and rheological properties of asphalt binder. The results of DSC studies show that different levels of the phase separations occur in asphalt/iPP blend, while those asphalt samples having CPPA or CPPB tend to disappear with the endothermic transition in the DSC thermograms because of the increase of the amorphous region of CPPA or CPPB. The images of the SEM, which show the phase distribution of asphalt blends, exhibit that the blends containing chlorinated iPP are more compatible than the corresponding blends with pure iPP. The viscosity of asphalt blend increases, resulting in improvement of its high temperature properties, in terms of higher temperature to attain Newtonian behavior. However, the asphalt blends containing CPPB (32 wt% of Cl) display lower viscosities and higher $\sin \delta$ values than the asphalt blends containing iPP and CPPA in this study. It is also found the enhancement
of the rutting resistance of the asphalt blend as reflected by $G*/\sin \delta$ parameter is correlated to the compatibility between the polymer modifier and the asphalt matrix at high temperatures. As results, the asphalt/CPPA blends exhibit superior rutting resistance in high temperature ranges to avoid permanent deformation, while the asphalt/iPP blends present better performance on the rutting resistance in the frequency studies.

REFERENCES